



ARBEITSBERICHT NAB 23-26

Diffusion Measurements of HTO, $^{36}\text{Cl}^-$ and $^{22}\text{Na}^+$ on Rock Samples of Opalinus Clay and Confining Geological Units from Deep Boreholes of Northern Switzerland:
Jura Ost, Nördlich Lägern and
Zürich Nordost

July 2024

**Nagra | National Cooperative for the
Disposal of Radioactive Waste**

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Diffusion, Opalinus Clay, confining geological units, deep geological repository, radionuclides, EDL model

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Nagra Arbeitsberichte ("Working Reports") present the results of work in progress that have not necessarily been subject to a comprehensive review. They are intended to provide rapid dissemination of current information.

The report titled "Diffusion Measurements of HTO, $^{36}\text{Cl}^-$ and $^{22}\text{Na}^+$ on Rock Samples of Opalinus Clay and Confining Geological Units from Deep Boreholes of Northern Switzerland: Jura Ost, Nördlich Lägern and Zürich Nordost" presents the fundamental laboratory data derived from Nagra's extensive deep drilling campaign, and represents the basis for the model approaches chosen in the compilation of the diffusion database.

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Abstract

This summary report focuses on through-diffusion experiments of rock samples with different mineralogical compositions taken from several new deep boreholes in the Mesozoic rock sequence of Northern Switzerland. The borehole campaign was organised and conducted by Nagra (National Cooperative for the Disposal of Radioactive Waste). The Opalinus Clay, a Jurassic-age argillaceous shale with > 100 m thickness, is the primary target rock for hosting a planned deep geological repository for radioactive waste in Switzerland. Argillaceous rocks are currently being examined worldwide as potential host rocks for repositories for radioactive waste and spent fuel due to their favourable characteristics, such as the self-sealing capacity and limited diffusive transport of fluids, solutes or gases, for safe, long-term disposal.

This study summarises the results of the diffusion measurements, during which diffusion parameters of tritiated water (HTO), $^{36}\text{Cl}^-$ and $^{22}\text{Na}^+$ tracers perpendicular to the bedding plane of the rock samples were determined and both effective diffusion coefficients and rock capacity factors were calculated from the data obtained. Accessible porosities were derived from the rock capacity factors in the case of HTO and $^{36}\text{Cl}^-$, and sorption distribution factors in the case of $^{22}\text{Na}^+$. Rock samples from seven different boreholes distributed across the three siting regions defined by Nagra were used, involving four different types of synthetic porewaters. Their composition was considered to be representative with respect to in-situ measurements on similar rock samples. For the Opalinus Clay, the effective diffusion coefficients (D_e) and accessible porosity values (ε) of the neutral HTO within and across all three siting regions are very consistent ($D_e = 8.8 \pm 1.9 \times 10^{-12} \text{ m}^2 \text{ s}^{-1}$; $\varepsilon = 0.12 \pm 0.02$), indicating that the material can be considered homogeneous in terms of diffusion properties. The rocks above and below the Opalinus Clay, up to the next aquifer (also termed confining geological units), consist of tight sedimentary units (carbonates, siliciclastics including argillaceous rocks) with a wider range of effective diffusion coefficients, indicating a higher level of heterogeneity. A direct comparison of the effective diffusion coefficients of the Opalinus Clay with the different heterogeneous rock units across the three siting regions is based on HTO data for which the diffusion behaviour is unaffected by the chemical composition of the porewater.

The effective diffusion coefficients of anions such as $^{36}\text{Cl}^-$ tend to be smaller than those of HTO due to anion exclusion effects, while those of cations such as $^{22}\text{Na}^+$ tend to be larger due to surface-enhanced diffusion. The extent of these effects is largely determined by the composition of the porewater and, in the case of cations, by the extent of sorption which depends also on the sorption capacity – indicated by the cation exchange capacity – of the solid. As a result, variations in the effective diffusion coefficient of $^{36}\text{Cl}^-$ and $^{22}\text{Na}^+$ are observed between different rock types (i.e. composition, fabric) as well as between the different siting regions where differences in the chemical composition of the porewaters in contact with the solid phase are present.

It is widely recognised that diffusive transport of charged species in charged clay media must be consistently described for ions with different charge signs and numbers, as well as with pore diffusion schemes for inert solutes that do not undergo interactions with the clay surface. A modelling approach that meets these requirements for the diffusion of HTO, $^{22}\text{Na}^+$ and $^{36}\text{Cl}^-$ tracers in heterogeneous Mesozoic sediment sequences of multiple deep boreholes in Northern Switzerland is also presented here. The model uses an electrical double layer (EDL) approach and a discrete Donnan potential description of cation enrichment and concomitant anion depletion in the Donnan layer close to the basal (planar) clay surfaces to predict effective diffusion coefficients and rock capacity factors of the charged tracers. It incorporates the diffusion data of HTO to determine the parameters related to the functional relationships between the accessible porosity and geometry factors with the total clay content as the main input variable. It also calculates concentration enrichment or depletion factors for mobile cationic and anionic species using

relationships with the total clay content, such as the average density of surface sites or the thickness of the Donnan layer and the related volume fraction of free porewater. These factors are subsequently used to derive effective diffusion coefficients that include surface diffusion of cations and anion exclusion effects and the related rock capacity factors. Despite the different lithologies of the rock samples, where significant variability between carbonates and siliciclastic contents are observed, the model shows good agreement with experimental data. Discrepancies between experimental and modelled data are mainly due to inadequacies in the geometric description of diffusion pathways, rather than by potential bias in the calculation of concentration enhancement or depletion factors. However, the model approach assumes that diffusion in clay rock occurs via the pore space of clay minerals and treats all related parameters as simple average values, neglecting heterogeneity of mineral constituents and the unknown statistical distribution of the parameter values.

The analysis of 130 samples shows that the effective diffusion coefficients of the Mesozoic units in Northern Switzerland exhibit a significant correlation with the total clay content of the rocks. These findings resulted in a novel empirical relationship that paved the way for the establishment of a diffusion database (NTB 23-08) (Glaus et al. 2024a) for the relevant elements under consideration in the Swiss radioactive waste disposal programme and the safety case.

The model has identified the chemical composition of the equilibrium porewater as a significant influencing variable for predicting concentration enhancement or depletion factors and related effective diffusion coefficients. Experimental data obtained for different synthetic porewater compositions support this finding. The robustness of the model predictions across various types of lithologies and formations, all of which exhibit diffusion as the dominant transport mode, is important for the generic application of the EDL diffusion model to predict diffusion parameters for elements with no or limited experimental data.

This report is a compilation of two peer-reviewed publications (Van Loon et al. 2023, Glaus et al. 2024b), extended with appendices containing detailed additional information that has not been published so far.

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List of Acronyms

BET	Brunauer–Emmett–Teller (theory to determine surface area)
Bq	Becquerel
BOZ	Bözberg
BUL	Bülach
CEC	Cation exchange capacity
CT	Computed tomography
DAO	Dogger above Opalinus Clay
DDB	Diffusion database
DL	Diffuse layer (DL may also denote the Donnan layer in cases where a discrete potential distribution is assumed, cf. section 4.2.2)
EDL	Electrical double layer
HPAEC	High-performance anion exchange chromatography
HPCEC	High-performance cation exchange chromatography
HTO	Tritiated water
JO	Jura Ost
LSC	Liquid scintillation counting
MAR	Marthalen
MT	Mont Terri
NAB	Nagra Arbeitsbericht (Nagra Work Report)
NAGRA	National Cooperative for the Disposal of Radioactive Waste
NL	Nördlich Lägern
NTB	Nagra Technical Report
OPA	Opalinus Clay
pH	$-\log_{10}(a_{\text{H}})$, negative logarithm of proton activity
PSI	Paul Scherrer Institute
RWI	Rock-water interaction
SCK-CEN	Belgian Nuclear Research Centre
SGT	Sectoral Plan for Deep Geological Repositories
SPW	Synthetic porewater
STA	Stadel

TBO	Deep borehole (campaign)
TRU	Trüllikon
URL	Underground Rock Laboratory
ZNO	Zürich Nordost

List of Symbols

A_p	pore surface area [m^2]
A_{sp}	specific surface area [$\text{m}^2 \text{kg}^{-1}$]
CEC	cation exchange capacity [mol kg^{-1}]
CEC_{100}	cation exchange capacity extrapolated to 100% clay [mol kg^{-1}]
CEC_{vol}	cation exchange capacity on a volumetric basis [mol m^{-3}]
D_a	apparent diffusion coefficient [$\text{m}^2 \text{s}^{-1}$]
D_e	effective diffusion coefficient [$\text{m}^2 \text{s}^{-1}$]
$D_{e,par}$	effective diffusion coefficient parallel to bedding plane [$\text{m}^2 \text{s}^{-1}$]
$D_{e,per}$	effective diffusion coefficient perpendicular to bedding plane [$\text{m}^2 \text{s}^{-1}$]
D_w	diffusion coefficient in free water [$\text{m}^2 \text{s}^{-1}$]
F	Faraday constant [C mol^{-1}]
F_{an}	anisotropy factor [-]
G	geometrical factor [-]
I	ionic strength [M]
J	diffusive flux [$\text{mol m}^{-2} \text{s}^{-1}$]
L_s	clay sample length [m]
R	molar gas constant [$\text{J mol}^{-1} \text{K}^{-1}$]
R_d	(sorption) distribution factor [$\text{m}^3 \text{kg}^{-1}$]
T	absolute temperature [K]
V_c	volume of the clay phase in rock [m^3]
V_p	pore volume of the total rock [m^3]
$V_{p,c}$	pore volume of the clay phase in rock [m^3]
V_T	volume of the total rock [m^3]
W_c	total clay content in rock [wt.%]
W_r	weight loss of the resin [kg]
W_T	total weight loss [kg]
$c, c_{solution}$	tracer concentration in the free porosity [mol m^{-3}]
c_{sorbed}	concentration of sorbed species on the solid phase [mol kg^{-3}]
c_{DL}	concentration in the Donnan layer [mol m^{-3}]
d_{DL}	Donnan thickness [m]
d_p	pore width [m]
f_{an}	anion porosity fraction [-]
f_{clay}	weight fraction of the clay phase [-]
f_{DL}	pore volume fraction of Donnan layer [-]

f_{free}	pore volume fraction of free porosity [m]
m_c	mass of the clay phase [kg]
m_r	mass of the total rock [kg]
n_{DL}	ratio between Donnan thickness and Debye length [-]
q_η	ratio of viscosity of free and Donnan layer water [-]
r_{SL}	solid:liquid ratio [kg m ⁻³]
t	time [s]
x	distance [m]
z_i	charge number of species i [-]
α	rock capacity factor [-]
δ	constrictivity [-]
Δ_r	residual [-]
ε	(diffusion) accessible porosity, total porosity [-]
ε_{an}	anion accessible porosity [-]
ε_c	porosity of the clay phase in rock [-]
ε_{max}	maximum total porosity [-]
ε_{tot}	total porosity [-]
ε_{wl}	weight loss porosity [-]
κ	reciprocal value of Debye length [m ⁻¹]
ρ_{bc}	bulk dry density of the clay phase in rock [kg m ⁻³]
ρ_{br}	bulk dry density of the total rock [kg m ⁻³]
ρ_{gc}	grain density of the clay phase [kg m ⁻³]
ρ_{gr}	grain density of the total rock [kg m ⁻³]
ρ_w	water density [kg m ⁻³]
σ	standard deviation
τ	tortuosity [-]
$\varnothing_s$	clay sample diameter [m]
ψ	electric potential [volt]
ψ_D	Donnan potential [volt]

1 Introduction

1.1 Background

For Stage 3 of the Sectoral Plan for Deep Geological Repositories (SGT Stage 3), Nagra drilled several (9) new deep boreholes (2018 – 2022) during its extensive geoscientific investigation programme (i.e. Nagra «TBO» campaign) (Nagra 2021) to meticulously characterise the Mesozoic sedimentary sequence of Northern Switzerland (Fig. 1-1). The aim of the investigations was to expand the knowledge base and scientific understanding of both the Opalinus Clay and the confining (over- und underlying) rock units. Core recovery of the new boreholes was significant (~ 60%) and the cores provided the material for fully characterise important geological and hydrogeological properties of the geological units (Mazurek et al. 2023).

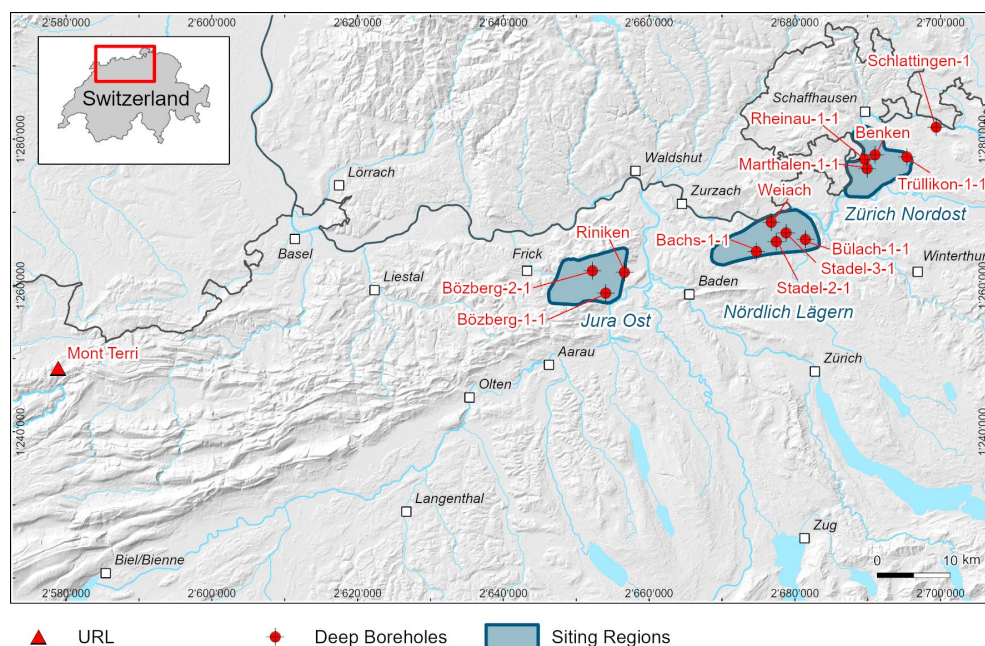


Fig. 1-1: Cartographic representation illustrating the geographical region of Northern Switzerland, including the three siting regions Jura Ost, Nördlich Lägern and Zürich Nordost

The map features the deep borehole (TBO) sites referred to in this study, as well as the location of the Mont Terri Underground Rock Laboratory (MT-URL), which is under the guidance of Swisstopo. MT-URL (since 1995), Benken (1999), and Schlattingen (2012) provided Opalinus Clay samples for previous diffusion experiments.

The geological sequence from the Mesozoic era within the vicinity of all three siting regions (Jura Ost, Nördlich Lägern, Zürich Nordost) (Fig. 1-1) can be divided into a simple scheme of three units, with the Opalinus Clay forming the core unit and the primary target for hosting the potential waste repository. The Jurassic-age Opalinus Clay (OPA) is a clay-rich rock unit that is over 100 m thick. The second sedimentary package consists of Dogger and Malm, which are tight clay-, carbonate-, and silt-/sandstone rocks that extend up to the next aquifer in the Malm and overlay the Opalinus Clay. The third sequence is located below the Opalinus Clay and is composed of Lias- and Keuper-/Triassic aged sediments of mostly tight clay-, carbonate-, anhydrite, and silt-/sandstone rocks up to the next aquifer (either in the Keuper or the Triassic Muschelkalk, see Mazurek et al. 2023). Both over- and underlying units have a high proportion of clay-rich rocks,

and the subunits with variable rock types also vary in thickness (Fig. 1-2). Further information about thickness and composition variability can be found in Mazurek et al. (2023).

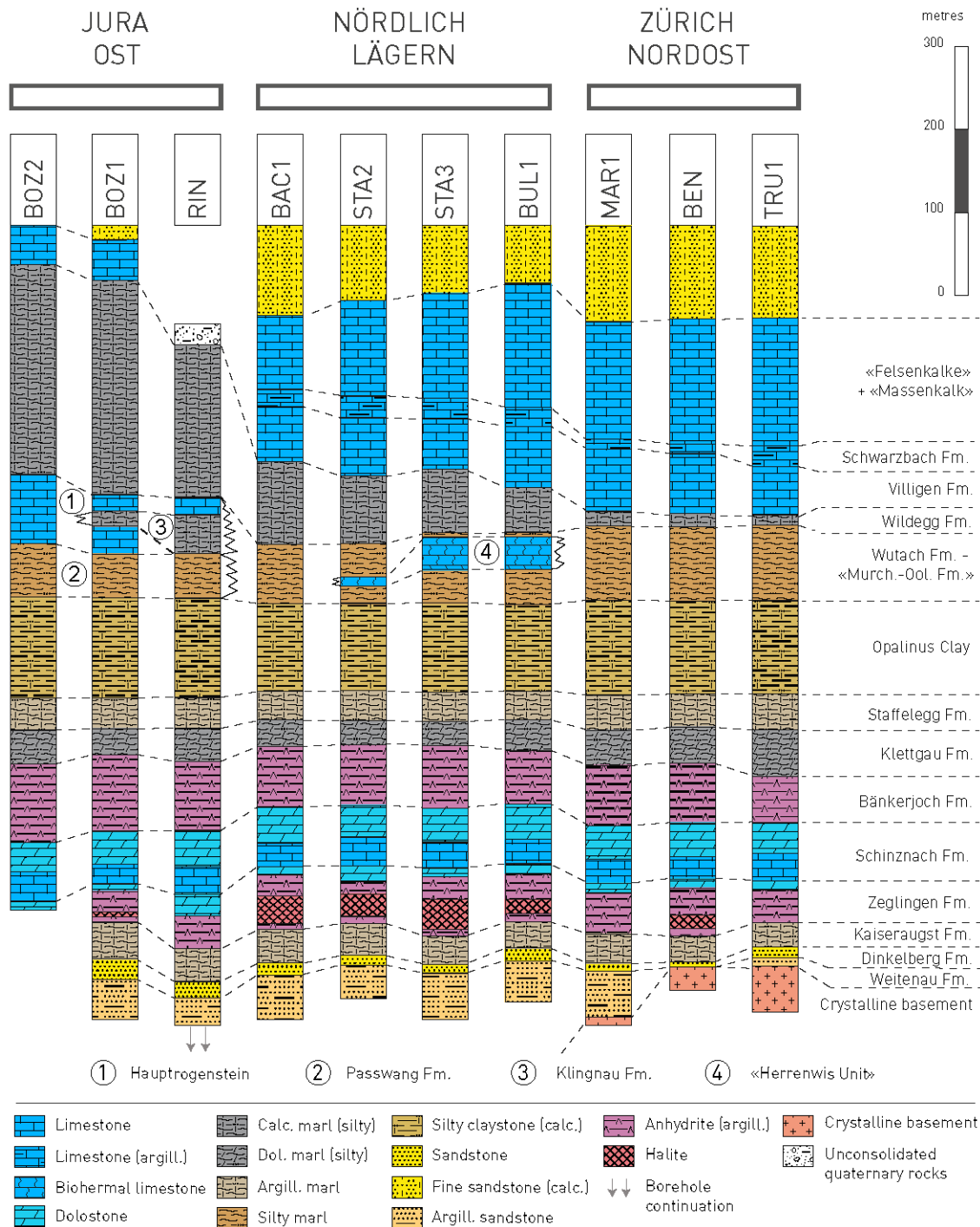


Fig. 1-2: Lithological overview correlation of Mesozoic sediments in the boreholes of the three siting regions

Simplified lithological legend according to Naef et al. (2019). Lithostratigraphy on the right side according to Jordan & Deplazes (2019).

Claystones have been considered as a potential host rock for the Swiss radioactive waste disposal programme since the late 1980s (Nagra 2002), with a focus on the Jurassic-age Opalinus Clay. The Paul Scherrer Institute (PSI) has played a leading role in investigating diffusion in the Opalinus Clay, with studies conducted on samples from the Mont Terri Rock Laboratory, located in the northwestern part of Switzerland (Bossart & Thury 2007) and the deep boreholes of Benken in the Zürich Nordost siting region (Van Loon & Soler 2003) and Schlattingen (Wersin et al. 2013) (Fig. 1-1). Despite the limited number of sample points, the studies obtained reliable results of the effective diffusion coefficients (D_e) and accessible porosities (ε) for neutral species (HTO) that were confirmed by other studies on MT-URL Opalinus Clay samples (Wu et al. 2009, Joseph et al. 2013, Kaplan 2013, Lübke 2015). In addition, the laboratory diffusion studies clearly demonstrated the existence of anion exclusion in Opalinus Clay (Van Loon & Soler 2003). This phenomenon, caused by anions being repelled from the negatively charged surface of clay minerals, was first described by Dutt & Low (1962) in clay-rich soils and was subsequently applied in various other studies (Bazer-Bachi et al. 2006, Aertsens et al. 2008, Descostes et al. 2008, Cavé et al. 2009). A comprehensive discussion and newly developed approaches based on the introduction of interlayer equivalent pores, a Donnan pore space and uncharged pores were later presented by Wigger & Van Loon (2017). Further progress and details of the anion exclusion effect and new evidence for differences in the exclusion of anions in argillaceous rocks from the new borehole campaign is now also documented by Zwahlen et al. (2024).

During the early studies of diffusion properties of the Opalinus Clay, measurements using $^{22}\text{Na}^+$, $^{85}\text{Sr}^{2+}$ and $^{134}\text{Cs}^+$ provided evidence of surface-enhanced diffusion (Appelo et al. 2010). This effect could be successfully modelled using a multispecies geochemical transport model implemented in PHREEQC (<https://www.usgs.gov/software/phreeqc-version-3>) that also incorporated the Donnan model to account for anion exclusion and surface diffusion (Appelo & Wersin 2007). The model was subsequently expanded to include surface diffusion of transition metals (Glaus et al. 2015a) and lanthanides (Glaus et al. 2020), as well as diffusion competition (Glaus et al. 2021). Another significant milestone was achieved during the early phase of working with Opalinus Clay when it was demonstrated that sorption in compacted systems (Opalinus Clay) is comparable to sorption on loose (crushed) material (Van Loon et al. 2009).

The early work also investigated temperature and anisotropy effects on the diffusion behaviour. Temperature-dependent diffusion tests with HTO, $^{36}\text{Cl}^-$ and $^{22}\text{Na}^+$ revealed that the temperature effect on diffusion can be appropriately described by the Arrhenius equation within a temperature range between 5 – 70 °C (Van Loon et al. 2005) using an activation energy of $20 \pm 2 \text{ kJ mol}^{-1}$. Savoye et al. (2011) came to a similar conclusions for diffusion of HTO, $^{36}\text{Cl}^-$ and $^{22}\text{Na}^+$ in Callovo-Oxfordian argillite, but not for diffusion of $^{137}\text{Cs}^+$, for which the activation energy was only 9 kJ mol^{-1} . Since shales and clay-rich rocks are characterised by clay platelets with a preferred horizontal orientation, anisotropic behaviour of different properties is expected (Winhausen et al. 2023). Van Loon et al. (2004) conducted diffusion measurements in both parallel (par) and perpendicular (per) orientation to the bedding plane of the Opalinus Clay. With the results, the diffusion anisotropy factors, $F_{\text{an}} = D_{e,\text{par}} / D_{e,\text{per}}$, for Opalinus Clay ranged from 4 (Mont Terri) to 6 (Benken borehole). A follow-up field experiment conducted in Mont Terri found that the in-situ anisotropy ratios for diffusion were larger for HTO and $^{22}\text{Na}^+$ (~ 5) than for anions ($\sim 3 - 4$ for Br and I) (Gimmi et al. 2014). These results provide evidence of the presence of diffusion anisotropy, which is expected given the layered structure of these clay-rich rocks of the Opalinus Clay (Wenk et al. 2008). Similar conclusions were made by Xiang et al. (2013) for Ordovician sedimentary rocks from the Michigan Basin in southwest Ontario (Canada). It could also be demonstrated that confining pressure had a small effect on the effective diffusion coefficient, but no measurable effect on the accessible porosity (Van Loon et al. 2003a, Xiang et al. 2016). In the remaining part of this report, all values of diffusion coefficients mentioned are perpendicular to the bedding plane.

For Stage 2 of the SGT (Van Loon 2014), only a few Opalinus Clay samples from the Schlattingen borehole (close to the Zürich Nordost siting region) (Fig. 1-1) were evaluated for effective diffusion coefficients of HTO, $^{36}\text{Cl}^-$ and $^{22}\text{Na}^+$, and their values were in good agreement with those samples from the Opalinus Clay of the Mont Terri URL and the Benken borehole (Van Loon 2014) (see Fig. 1-1 for locations). In addition to laboratory diffusion work on cm-scale samples, several field diffusion studies in the Opalinus Clay were performed in the Mont Terri URL on a large scale of several decimetres (Leupin et al. 2017 and references therein). The results of these field studies demonstrate excellent agreement with laboratory measurements. Such field- to laboratory-scale comparisons increase confidence in the reliability of the early laboratory work. Also, extensive studies on natural tracer profiles showed that diffusion is the dominant transport process over several million years (Mazurek et al. 2011).

The main objective of previous diffusion studies was to obtain initial insights and understanding of diffusion processes in the Opalinus Clay, and to use this knowledge to develop a diffusion database (DDB) that can be applied in long-term safety assessment studies. The first DDB (Van Loon 2014) was created using Archie's relationship, which links effective diffusion coefficients to accessible porosity. Van Loon & Mibus (2015) developed an extended version of Archie's law (e-Archie) primarily based on literature data from different sedimentary rocks. However, only a limited amount of data covered the Opalinus Clay and even less data were derived from the adjacent over- or underlying units for the development of the e-Archie approach.

More recent studies have shown that molecular diffusion is also the main transport mechanism for radionuclides in clay-rich layers adjacent to the Opalinus Clay (Van Loon 2014, Gimmi & Waber 2004, Wersin et al. 2018). In 2018, Nagra started a new geoscientific investigation programme aimed at acquiring an extensive and comprehensive dataset on diffusion, encompassing the Opalinus Clay as well as the over- and underlying sedimentary packages, and expanding the dataset to the subsequent aquifers in all three siting regions (Fig. 1-1 and Mazurek et al. 2023). Previous studies have suggested that the Opalinus Clay may have a uniform diffusion behaviour, while the overlying layers, such as the Dogger and Malm sequences, and the underlying layers, such as the Lias, Keuper/Triassic sequences, show considerable heterogeneity (see Mazurek et al. 2023).

The main objective of this study is to measure the diffusion properties of HTO, $^{36}\text{Cl}^-$ and $^{22}\text{Na}^+$ in samples from all the different rock types across the lithostratigraphic profile of interest (Opalinus Clay and sedimentary units that occur between the overlying Malm and underlying Keuper or Triassic aquifers). This approach assesses the spatial distribution of diffusion properties across different geological units and helps to determine the degree of heterogeneity of diffusion properties. Additionally, another aim of the study is to identify any correlations between diffusion parameters and material properties, such as mineral composition or microfabric. These correlations can provide valuable information for understanding transport of dissolved species through heterogeneous sedimentary sequences (Bollermann et al. 2022).

The diffusion properties are basically expressed as the effective diffusion coefficients and rock capacity factors. The rock capacity factors are subsequently interpreted in terms of accessible porosity in the case of HTO and $^{36}\text{Cl}^-$ and in terms of accessible porosity and sorption distribution ratios in the case of $^{22}\text{Na}^+$.

1.2 Theoretical background of one-dimensional (planar) through-diffusion

For a one-dimensional diffusion process through a plane, the flux J (mol m⁻² s⁻¹) is given by Fick's first law (Van Loon et al. 2003a, Van Loon & Soler 2003):

$$J = -D_e \cdot \frac{\partial c}{\partial x} \quad (1-1)$$

Where D_e represents the effective diffusion coefficient (m² s⁻¹), c is the concentration (mol m⁻³) of the diffusing species in free solution and x distance (m). Free solution is the solution that has a sufficient distance from the pore walls.

If the concentration within the system changes, Fick's second law applies:

$$\frac{\partial c}{\partial t} = D_a \cdot \frac{\partial^2 c}{\partial x^2} \quad (1-2)$$

where t represents time (s) and D_a the apparent diffusion coefficient (m² s⁻¹) and can be written as:

$$D_a = \frac{D_e}{\alpha} \quad (1-3)$$

with α representing the rock capacity factor defined as:

$$\alpha = \varepsilon + R_d \cdot \rho_{br} \quad (1-4)$$

ε is the (diffusion) accessible porosity, ρ_{br} the bulk-dry density (kg m⁻³) and R_d the (sorption) distribution factor defined as:

$$R_d = \frac{c_{sorbed}}{c_{solution}} \quad (1-5)$$

where c_{sorbed} represents the number of species sorbed on the solid phase (mol kg⁻¹) and $c_{solution}$ is the species concentration in solution (mol m⁻³) in equilibrium with the sorbed amount ($c_{solution} = c$).

D_e is related to the diffusion coefficient in free water, D_w (m² s⁻¹), by:

$$D_e = \frac{\varepsilon \cdot \delta}{\tau^2} \cdot D_w \quad (1-6)$$

where δ (-) is the constrictivity and accounts for the fact that the pore diameter varies along the diffusion pathway, and τ (-) is the tortuosity of the sample and takes account of path lengthening (van Brakel & Heertjes 1974). Because constrictivity and tortuosity cannot be determined independently, they are combined in the so-called geometrical factor G (-):

$$G = \frac{\tau^2}{\delta} \quad (1-7)$$

Eq. (1-6) can be written as:

$$D_e = \frac{\varepsilon}{G} \cdot D_w \quad (1-8)$$

2 Materials and methods

2.1 Rock samples

Sample material was collected from seven deep boreholes drilled in the three siting regions, Jura Ost (JO), Nördlich Lägern (NL), and Zürich Nordost (ZNO) in Northern Switzerland (see Fig. 1-1 and also Mazurek et al. 2023). Upon core retrieval, the material was cleaned, photographed, and immediately sealed in vacuum bags (plastic- and aluminium-lined) and then transferred to a core storage facility. The samples for this study were only further treated upon selection for diffusion analysis. In total, over 130 samples were investigated. An overview of selected sample analyses and number of samples analysed (geological formation) is given in Fig. 2-1.

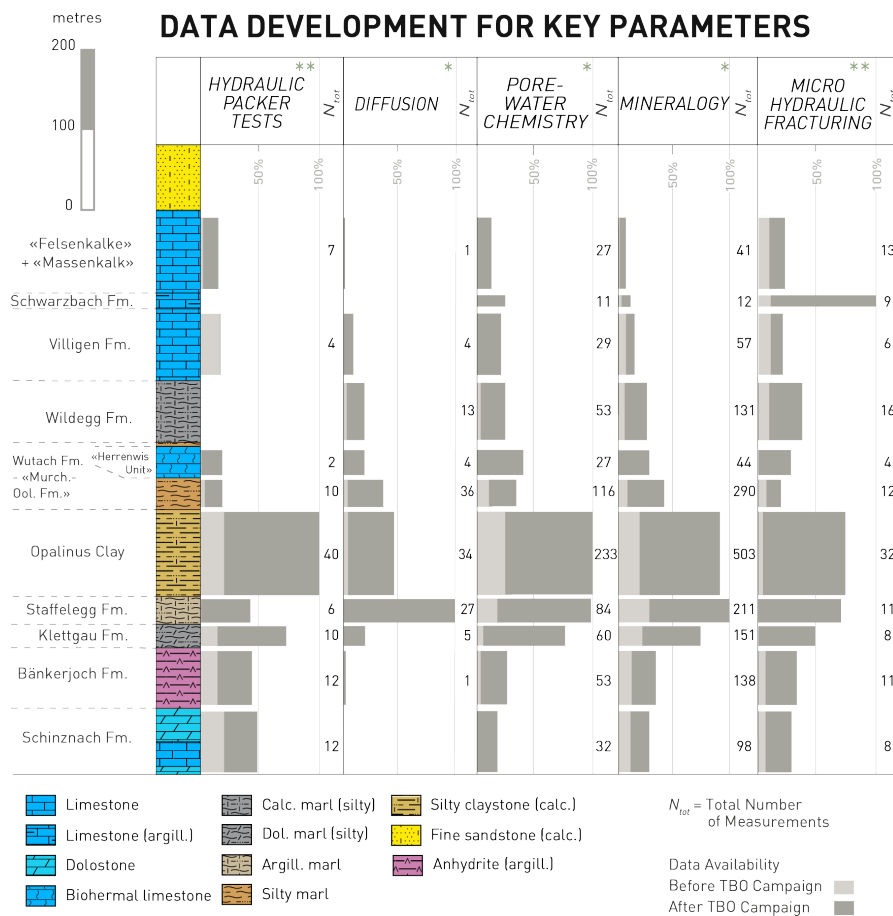


Fig. 2-1: Abstract profile (Stadel-3-1 borehole) providing an overview of data analysis for key parameters including diffusion measurements in connection with the deep borehole campaign during Stage 3 of the Sectoral Plan (2018 – 2022) (from Nagra (2024))

This figure shows data obtained from the 9 boreholes drilled within the framework of the recent deep borehole campaign as well as from the older boreholes Benken, Riniken and Weiach-1. Data are projected to the Stadel-3-1 borehole, which is shown as a reference on the left. Data are normalised to the average formation thickness. Percentage readings (horizontal bars) refer to the formation with the highest normalised values (sample numbers normalised to thickness in m, usually Opalinus Clay). Parameters with one asterisk indicate laboratory experiments, while parameters with two asterisks represent in-situ measurements. The figure reflects the focus of the investigations on the host rock and the surrounding confining units.

The core material was derived from the three siting regions as follows:

- Bözberg-1-1 (BOZ1-1) (Wersin et al. 2022) and Bözberg-2-1 (BOZ2-1) (Gimmi et al. 2022) located in JO
- Bülach-1-1 (BUL1-1) (Mazurek et al. 2021), Stadel-2-1 (STA2-1) (Zwahlen et al. 2022), and Stadel-3-1 (STA3-1) (Aschwanden et al. 2022) located in NL
- Trüllikon-1-1 (TRU1-1) (Aschwanden et al. 2021) and Marthalen-1-1 (MAR1-1) (Mäder et al. 2021) located in ZNO

Details of the locations are provided elsewhere (Mazurek et al. 2023). In the case of BOZ1-1, BUL1-1 and TRU1-1, samples of the Opalinus Clay (OPA), and samples from the overlying Dogger and Malm and the underlying Lias, Keuper and Triassic sediments were studied. In the case of BOZ2-1, STA2-1, STA3-1 and MAR1-1, only samples of the overlying and underlying sedimentary sequences (*i.e.*, confining units) were used. A summary of the samples is given in Tab. A-1, Tab. A-2 and Tab. A-3 in App. A.

Furthermore, this investigation builds upon previous studies and findings from isotopic diffusion research conducted as part of earlier exploration initiatives. In 1995, the search for a host rock in which to construct a deep geological repository for radioactive waste in Switzerland transitioned towards the Opalinus Clay. This led to the planning and drilling of an exploratory borehole in Benken (1998 – 1999), situated close to Schaffhausen (Fig. 1-1), that penetrated the subsurface to a depth of 1,007 m (Nagra 2001). In 2012, an additional borehole, Schlattingen-1, classified as another exploratory borehole (with the purpose of extracting geothermal energy), also provided sample material for diffusion tests of the Opalinus Clay (Wersin et al. 2013, Van Loon 2014).

2.2 Sample preparation

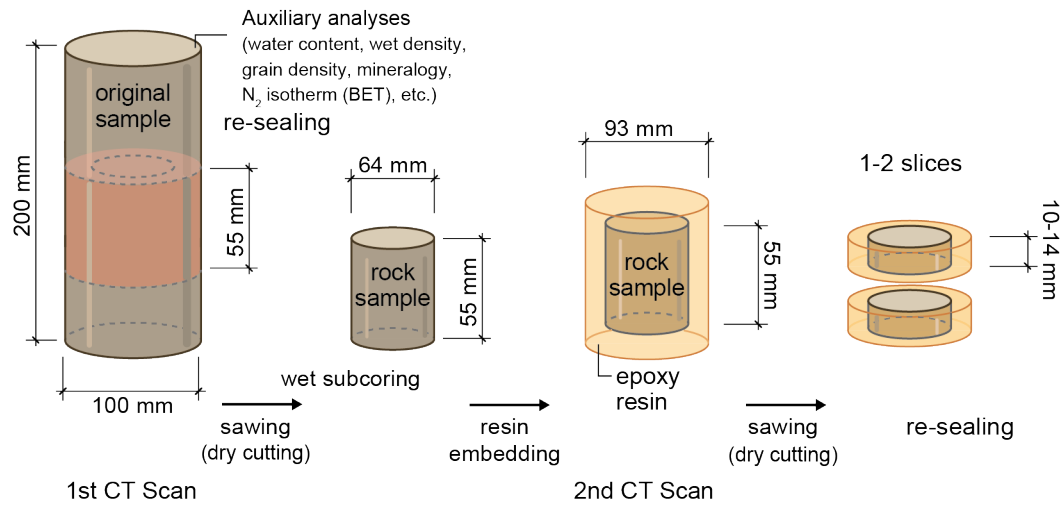
The samples used for the diffusion experiments in this study were selected and prepared at the Institute of Geological Sciences, University of Bern. From each sealed 200-mm-long core section, a 55-mm-long cylinder with a diameter of 64 mm was subcored after initial CT scan analysis. This helped to identify homogeneous rock portions (Fig. 2-2). Details of these image analyses are provided in App. G.

The remainder of the material was used for auxiliary investigations, including the quantification of porosity, mineralogy and microfabric, and the sample material was sealed immediately after the preparatory steps.

For the diffusion samples, the cylinders were drilled perpendicular to the bedding plane by wet (tap water) drilling using a Hilti® drill. After drying, the subcores were embedded in epoxy resin (Epofix, Struers, Germany). After hardening at 40 °C overnight, the samples were cut dry into one or two slices with a thickness of 10 – 14 mm (Fig. 2-2). Exposure of the samples to atmospheric (oxic) conditions was limited during sample preparation, and any unused material was resealed immediately. Material requiring preparatory steps was resealed immediately. The samples were packed in aluminium-lined vacuum bags and shipped to PSI for the diffusion analysis.

At PSI, the samples were unpacked, photographed (Fig. 2-3) and visually inspected for mechanical damages (see also App. G). After polishing the samples using a P420 abrasive paper, the exact dimensions (thickness and diameter) were determined by means of a digital calliper. Because the thickness was not consistently equal across the sample disks, it was measured at four positions as indicated by the yellow dots in Fig. 2-3. The average value was used in later data processing.

A: Sample preparation



B: Diffusion experiments

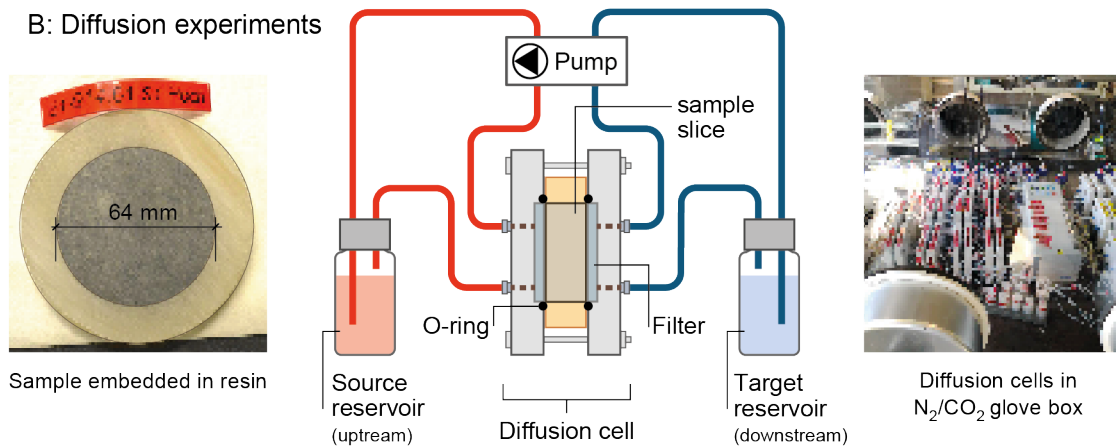


Fig. 2-2: A: Schematic representation of the sample preparation procedure employed by the University of Bern. B: The diffusion setup scheme is depicted in the middle, with accompanying images of an embedded sample (left) and diffusion cells in the glove box (right)

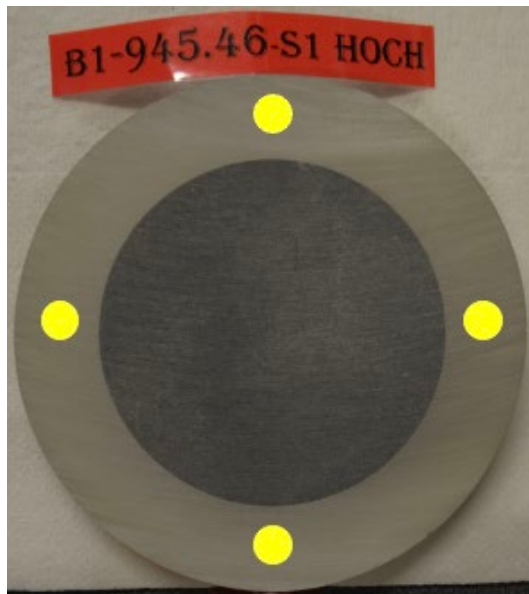


Fig. 2-3: Picture of a sample slice embedded in epoxy resin

The yellow dots indicate the positions where the thickness of the samples was measured, and the average value was used for the thickness calculations. It was assumed that the thickness of the resin equals the thickness of the clay.

2.3 Porewaters

The experimental porewater compositions used in this study were derived from preliminary data obtained through advective displacement laboratory analyses of samples sourced from the corresponding boreholes situated within the siting regions (BUL1-1: Mazurek et al. 2021; TRU1-1: Aschwanden et al. (2021); BOZ1-1: Wersin et al. 2022). The concentrations of initial aliquots were modified via geochemical equilibrium modelling to relevant mineral equilibria and an atmospheric $p\text{CO}_2$, using chemical equilibration calculations in PHREEQC based on the PSI-Nagra Thermodynamic Database (Hummel & Thoenen 2023). A distinct composition was established for each siting region to account for regional variability, as presented in Tab. 2-1. While this approach does not account for vertical variability across the sedimentary units within a borehole, it is suitable because the in-situ porewaters were exchanged almost completely with the synthetic porewaters during the saturation phase of the samples.

MAR1-1, TRU1-1 and BUL1-1 were the first boreholes with an identified porewater composition, of which TRU1-1 and MAR1-1 had identical porewaters (Tab. 2-1), while BUL1-1 had a markedly elevated salt concentration ($\sim 13,000 \text{ mg dm}^{-3} \text{ Cl}$). Although BUL1-1 lies within the NL siting region (Fig. 1-1), the porewaters of both STA2-1 and STA3-1 were more closely aligned with TRU1-1 and MAR1-1, having $8,000 \text{ mg dm}^{-3} \text{ Cl}$ and slightly lower Ca and SO_4 concentrations. The BOZ1-1 and BOZ2-1 boreholes in the western Jura Ost siting region contained the lowest salinity porewater composition ($\sim 2,000 \text{ mg dm}^{-3} \text{ Cl}$).

The synthetic porewaters (SPW) (Tab. 2-1) were prepared by dissolving Ca-, Mg-, Na- and K-salts in ultrapure water. The water was degassed by boiling and purging nitrogen (N_2) through it during two hours prior to usage. Subsequently, the heated water was then transferred to a glovebox (MBraun, Germany) that had an O_2 content $< 0.1 \text{ ppm}$ and a CO_2 content of $400 \pm 10 \text{ ppm}$, where it was allowed to cool and equilibrate with the glovebox atmosphere for two days. After this period, specific quantities of salt were dissolved in the predetermined volume of water (slightly

less than the final volume). After one day, the pH of the solution was adjusted to the target value by adding small amounts of a 1 M NaOH solution, and water was added to achieve the desired volume.

Tab. 2-1: Composition of the porewaters used in the diffusion measurements

The concentrations of initial aliquots determined during advective displacement methods of Opalinus Clay core material were modified via geochemical equilibrium modelling to relevant mineral equilibria and an atmospheric $p\text{CO}_2$, using chemical equilibration calculations in PHREEQC based on the PSI-Nagra Thermodynamic Database (Hummel & Thoenen 2023).

Element	Concentration (M)			
	¹ SPW-BOZ (BOZ1-1/BOZ2-1)	¹ SPW-BUL (BUL1-1)	¹ SPW-STA (STA2-1/STA3-1)	¹ SPW-TRU (TRU1-1/MAR1-1)
Na	1.40×10^{-1}	3.43×10^{-1}	1.56×10^{-1}	2.44×10^{-1}
K	7.76×10^{-4}	1.91×10^{-3}	1.91×10^{-3}	1.64×10^{-3}
Ca	9.24×10^{-3}	3.83×10^{-2}	3.50×10^{-2}	2.22×10^{-2}
Mg	3.94×10^{-3}	1.76×10^{-2}	1.76×10^{-2}	1.56×10^{-2}
Sr	1.15×10^{-4}	3.33×10^{-4}	3.33×10^{-4}	2.89×10^{-4}
Cl	8.28×10^{-2}	4.02×10^{-1}	2.19×10^{-1}	2.72×10^{-1}
S (as SO_4)	4.21×10^{-2}	2.75×10^{-2}	2.22×10^{-2}	2.43×10^{-2}
C (as HCO_3)	7.05×10^{-4}	4.13×10^{-4}	4.13×10^{-4}	4.87×10^{-4}
pH	7.90	7.56	7.56	7.68
² I	0.190	0.505	0.306	0.354

¹ Four types of SPW were used, abbreviated as SPW-BOZ, SPW-BUL, SPW-STA and SPW-TRU.

² Ionic strength (I), as calculated from speciation calculations with PHREEQC

2.4 Diffusion

A detailed description of the diffusion setup is provided in Van Loon (2014), with a scheme presented in Fig. 2-2. The prepared diffusion samples were positioned between two porous stainless-steel filters (thickness 1.2 mm; diameter 65 mm; porosity 0.4, pore sizes approx. 10 μm) and fixed within the diffusion cells, with the bedding plane perpendicular to the direction of diffusion. The use of end plates and porous filters provides a rigid confinement for the clay sample, thus limiting the possibility of swelling and potential disintegration of the samples (Fig. 2-4). The use of the porous filters provides a homogeneous contact for the fluid and the clay surface, thus ensuring a homogeneous concentration boundary condition for diffusion. The end plates were secured by a set of four bolts, without the control of confining pressure. Previous studies revealed that confining pressure has a minimal, insignificant effect on diffusion (Van Loon et al. 2003a, Xiang et al. 2016). The cells were connected to two reservoirs, namely a 100 ml glass flask and a 20 ml polyethylene β -vial, both containing 20 ml of synthetic porewater that was circulated using a peristaltic pump (IPC, Ismatec, IDEX corporation, USA).

In a first stage, the samples were saturated with the borehole-specific synthetic porewaters (Tab. 2-1) for a period of ca. 4 – 5 weeks. The solutions were exchanged once per week and analysed for main anions and cations by ion chromatography (see Section 2.6.2). The saturation process continued until the composition of the porewater in both source (upstream) and target (downstream) reservoirs no longer changed significantly. More information on the evolution of the porewater composition during the saturation phase can be found in App. C.

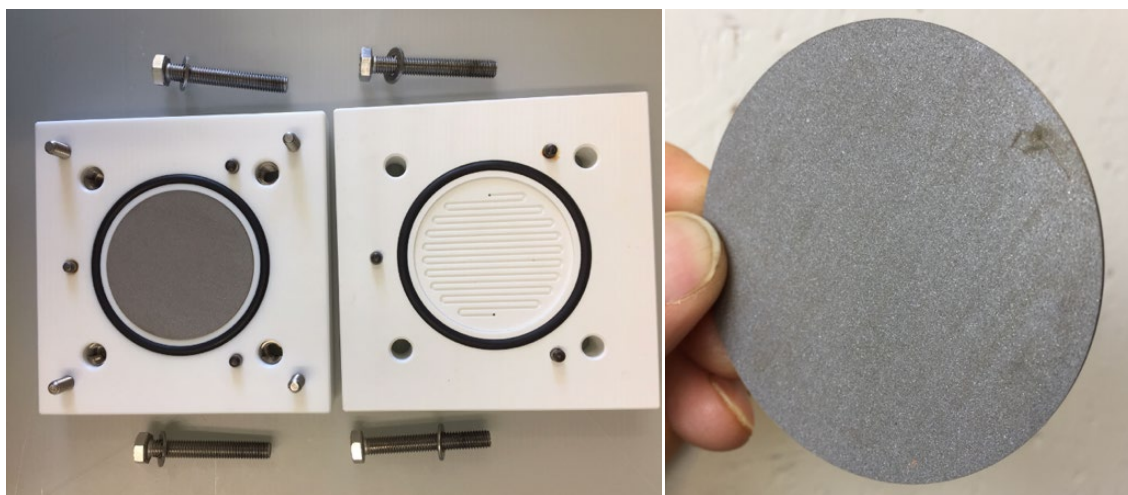


Fig. 2-4: Picture of the diffusion cells (left) and stainless-steel filter (right) used

In a second stage, the diffusion experiments started when the source reservoir (100 ml glass flask) was filled with 100 ml SPW spiked with HTO and $^{36}\text{Cl}^-$. The activity in this reservoir was approximately 500 Bq cm^{-3} for ^3H (ca. $4.5 \times 10^{-10} \text{ mol dm}^{-3}$) and 500 Bq cm^{-3} for $^{36}\text{Cl}^-$ (ca. $1 \times 10^{-5} \text{ mol dm}^{-3}$). The target reservoir (20 ml polyethylene β -vial) was filled with 10 ml SPW. The solution at the low concentration side (target reservoir) was regularly replaced after a time interval, Δt , of typically 2 – 3 days to keep the concentration of the tracer in this compartment as low as possible. The radiotracers in the exchanged solution of the target reservoir were analysed. The solution of the source reservoir was sampled once a week to analyse the radiotracers. The sampled volume (100 μl) was not replaced because the volume decrease due to sampling was taken into account in the modelling of the data. HTO and $^{36}\text{Cl}^-$ were measured with β/β discrimination by liquid scintillation counting (see Section 2.6.1). All diffusion measurements were performed in an inert gas glove box (Fig. 2-5) under anoxic conditions ($\text{O}_2 < 0.1 \text{ ppm}$; $\text{CO}_2 = 400 \pm 10 \text{ ppm}$). The temperature in the glove box was $26.7 \pm 0.8 \text{ }^\circ\text{C}$.

In a third stage, the experiments with ^{22}Na were started as soon as the diffusion of $^{36}\text{Cl}^-$ and HTO had reached the quasi-steady state phase indicated by an approximately constant flux of the tracers over time. A given amount of $^{22}\text{Na}^+$ tracer, resulting in an activity of about 500 Bq cm^{-3} (ca. $9.5 \times 10^{-11} \text{ mol dm}^{-3}$), was added to the same source reservoir. The target side was connected to a 20 ml polyethylene β -vial (target reservoir) containing 10 ml SPW. The experimental protocol was identical to the one described for HTO/ $^{36}\text{Cl}^-$. The $^{22}\text{Na}^+$ activity in the sampled solutions and source reservoir was measured by γ -counting (see Section 2.6.1).

Duplicate measurements on a few twin samples of the Trüllikon borehole were performed by SCK-CEN in Belgium. The main results of these comparison measurements can be found in App. G and in Van Laer et al. (2024).



Fig. 2-5: MBraun glovebox (N₂/CO₂) with diffusion cells

2.5 Water content of the samples after diffusion measurements

After finishing the diffusion experiments, the diffusion cells were dismantled and the water content (kg) of the samples with known thickness L_s (m) and diameter $\varnothing_s$ (m) was measured. To this end, the samples were dried in an oven at 105 °C until constant weight was reached. The total weight loss, W_T (kg), was corrected for the weight loss, W_r (kg), caused by heating of the resin. The weight loss by the resin was measured by heating a specimen of resin at 105 °C until constant weight was reached. Because the amount of resin was identical for all samples, the weight loss of the resin could be normalised to the sample thickness (0.02 g/mm). The water loss porosity (ε_{wl}) was calculated as:

$$\varepsilon_{wl} = \frac{4 \cdot (W_T - W_r)}{\rho_w \cdot \pi \cdot L_s \cdot (\varnothing_s)^2} \quad (2-1)$$

with the density of water $\rho_w = 998 \text{ kg m}^{-3}$ at 20 °C.

2.6 Analytical methods

2.6.1 Radioactivity measurements

The $^{22}\text{Na}^+$ activity in the sampled solutions was measured by γ -counting. 7 ml of solution was placed in a counting vial and the activity measured in a γ -counter (Minaxi- γ , Autogamma 5000 series, Canberra-Packard). HTO and $^{36}\text{Cl}^-$ were measured with β/β discrimination by liquid scintillation counting (LSC). A detailed description of the β/β discrimination is given in Glaus et al. (2017). To this end, 10 ml of solution was mixed with 10 ml of Ultimagold AB in a 20 ml polyethylene β -vial and measured by LSC (TRICARB 2250 CA, Canberra-Packard).

2.6.2 Main cations and anions in aqueous solutions

The main cations and anions in solution were monitored by ion chromatography (high-performance anion exchange chromatography: HPAEC; high-performance cation exchange chromatography: HPCEC). HPCEC measurements were performed using an ICS5000+ system (Thermo Fisher Scientific, Switzerland) equipped with a 2 mm × 50 mm Ionpac CG12A precolumn and a 2 mm × 250 mm Ionpac CS12A analytical column. A 18 mM methanesulphonic acid eluent was used which was produced electrochemically using an eluent generator. The flow rate was 0.25 cm³ min⁻¹. The injection volume was 10 µl, injected from a 2-position valve with 6 ports. Analytes were quantified by conductivity detection, after electrochemical suppression of the eluent ions in auto-suppression mode. Calibration was performed using commercial calibration standards diluted to three different concentrations matching the matrix of the diluted samples.

HPAEC measurements were also performed using an ICS5000+ system equipped with a 2 mm × 50 mm Ionpac AG16 precolumn and a 2 mm × 250 mm Ionpac AS16 analytical column. The 18 mM KOH eluent, which was used with a flow rate of 0.25 cm³ min⁻¹, was produced electrochemically using an eluent generator. The injection volume was 10 µl and was also injected from a 2-position valve with 6 ports. Analytes were quantified by conductivity detection, after electrochemical suppression of the eluent ions in auto-suppression mode. Calibration was performed using commercial calibration standards diluted to three different concentrations matching the matrix of the diluted samples.

2.6.3 pH

The pH of the solutions was measured in the glovebox using a pH meter (pH197i, WTWTM) equipped with a combined glass electrode (Semi-Micro, Mettler Toledo). The electrode was calibrated each time using standard buffer solutions of pH = 4 and pH = 10 (VWRTM).

2.7 Uncertainty estimations

Uncertainty estimations on the values of the flux and total diffused mass are based on the uncertainty of the parameters involved in calculating these values. The procedure to estimate uncertainties is described in detail in Van Loon & Soler (2003).

2.8 Parameter estimation using COMSOL Multiphysics

Modelling of the diffusion data is performed with Comsol Multiphysics® using the “Transport of Diluted Species in Porous Media” interface involving a 1D-linear geometry for representing the filter and clay domains (Glaus et al. 2017). The concentration change in the source solution reservoir is reflected in variable boundary conditions at the interfaces between filters and solution, while a zero-concentration boundary condition was applied for the target side. The calculation of the variable source boundary concentration involves the difference between the total mass present at time zero and the mass diffused from the source domain as a function of time. Best-fit parameter values and the respective parameter uncertainties (95% confidence level) are obtained from a parameter optimisation routine (using the lsqnonlin function in Matlab®), in which the Comsol Multiphysics® model is used as a Matlab® script. Glaus et al. (2017) provided a comprehensive description of the methodology, with additional information below.

To fit the parameters, the cumulated diffused mass at the target side and the source concentration of the diffusing species as a function of time are used as the source data. Using average target flux data instead of accumulated mass yields similar results with discrepancies that typically fall within the fitting uncertainties. Both the cumulative mass and the average target flux are derived from the measured activity which accumulated in the target reservoir solution between sample

exchanges. In that sense, those quantities contain the same basic information, and only negligible numerical differences between the evaluation methods related to these quantities would be expected. In reality, the average target flux data inherently comprise systematic errors owing to inadequacies related to averaging sampling times. The comparison between the two evaluation methods (cumulative mass versus average target flux) demonstrates that the sampling regime applied is adequate for keeping the introduced errors at an acceptable level.

The adjustable parameters for all three nuclides include the effective diffusion coefficient in the clay rock (D_e) and the initial concentration at time zero. The rock capacity factor (α) was used as a third adjustable parameter.

HTO and $^{36}\text{Cl}^-$ are treated as non-sorbing species ($R_d = 0$). Thereby, Equation (1-4) reduces to $\alpha = \varepsilon$, where ε is assumed to represent the total porosity (ε_{tot}) in the case of HTO and the anion-accessible porosity (ε_{an}) in the case of $^{36}\text{Cl}^-$. For technical reasons, ρ_{br} is treated as the adjustable parameter in the case of HTO instead of α . ρ_{br} was related via a generic (formal) material (or grain) density (ρ_{gr}) of $2,800 \text{ kg m}^{-3}$ to the total porosity (ε_{tot}) according to the relationship:

$$\varepsilon_{\text{tot}} = 1 - \frac{\rho_{\text{br}}}{\rho_{\text{gr}}} \quad (2-2)$$

For this reason, the best-fit parameter value of ρ_{br} depends on the choice of ρ_{gr} and cannot be directly compared with the sample specific data presented in Mazurek et al. (2023). However, the difference between the measured ρ_{br} using pycnometry and the optimised value in this work was at most 12%.

In the case of the $^{36}\text{Cl}^-$ data, the anion porosity fraction (f_{an}):

$$f_{\text{an}} = \frac{\varepsilon_{\text{an}}}{\varepsilon_{\text{tot}}} \quad (2-3)$$

is used as the third adjustable parameter in combination with the given total porosity for HTO from the parent experiment. Similarly, R_d is used as the third adjustable parameter in the case of the $^{22}\text{Na}^+$ data via the definition of a 1-site Langmuir isotherm in which the affinity constant is chosen in a way to keep the experimental data in a “linear sorption range” (Glaus et al. 2020). The diffusion-accessible porosity of $^{22}\text{Na}^+$ is assumed to equal the diffusion-accessible porosity of HTO. For this reason, the total porosity value is selected based on the best-fit parameter value obtained from the HTO data.

While the initial concentration is an experimentally determined value, it is treated as an adjustable parameter (within a rather narrow range of possible values) if there is a discrepancy between the measured value at time zero and the extrapolation from the subsequent measurements at the following points in time.

To estimate the uncertainties of the diffusion properties of the confining filters on the best-fit parameter values of the adjustable parameters, a sensitivity analysis was conducted, because there were no pertinent values determined for the diffusivity in the filters. As the effect of the confining filters on the evaluation of the diffusion properties of the clay rock samples were rather small in these experiments, values from other filters were chosen, based on experience (Glaus et al. 2008, Aldaba et al. 2014) were chosen. Comparatively large uncertainty ranges for the values were selected to cover the potential variability among different production batches (Glaus et al. 2008).

For HTO, the uncertainty bounds were $5 \times 10^{-11} \text{ m}^2 \text{ s}^{-1} < D_{e \text{ fil}} < 1.5 \times 10^{-10} \text{ m}^2 \text{ s}^{-1}$ with a preferred value of $1.0 \times 10^{-10} \text{ m}^2 \text{ s}^{-1}$. These upper and lower limits were used for two “bounding” parameter optimisation scenarios (in Matlab), which then generated upper and lower bounds for the adjustable parameter values (with regard to filter uncertainties).

The best-fit parameter values for the preferred filter value were then calculated by averaging each of the two bounding “filter scenarios”. The maximum and minimum uncertainty values obtained in the two scenarios were used as the upper and lower bounds of the best-fit parameter values. This approach is a pragmatic means of accounting for the effect of filter diffusion uncertainties on the clay diffusion parameters. For $^{22}\text{Na}^+$, the preferred effective diffusion coefficient for the filters is $6 \times 10^{-11} \text{ m}^2 \text{ s}^{-1}$, with $3 \times 10^{-11} \text{ m}^2 \text{ s}^{-1}$ and $9 \times 10^{-11} \text{ m}^2 \text{ s}^{-1}$ as the lower and upper bounding values, respectively. In the case of $^{36}\text{Cl}^-$, the chosen effective diffusion coefficient for the filters is $1 \times 10^{-10} \text{ m}^2 \text{ s}^{-1}$. As a result of the very low effective diffusivities of this tracer in the clay, no sensitivity analysis was carried out for the filter diffusivities for this tracer. For the porosity of the filter, a value of 0.4 was taken for all three tracers.

It should be noted that the diameter of the confining filters in the experiments is slightly larger ($65 \pm 0.5 \text{ mm}$) than the diameter of the samples ($63.6 \pm 0.1 \text{ mm}$). However, a 2D simulation (see App. D) was conducted which demonstrated that the overlapping domains of the filters did not have a significant influence on the best-fit parameter values and their uncertainties.

Diffusion tests with HTO in small Opalinus Clay (OPA) samples embedded in resin clearly show the signature of the resin being available as a porous medium for diffusion. Scoping calculations (App. E) showed that this effect was of minor importance and could be neglected in the modelling.

3 Results

3.1 Effective diffusion coefficients and diffusion-accessible porosities

The diffusion data of the Mesozoic sedimentary material of Northern Switzerland present a unique and significant database of HTO, $^{36}\text{Cl}^-$ and $^{22}\text{Na}^+$ in ~ 130 rock samples. All samples have auxiliary geological data, including sediment composition. The aim is to identify relationships between geological and diffusion parameters (see also Mazurek et al. 2023). The findings indicate that the diffusion parameters are significantly impacted by the mineralogical composition and rock parameters.

The investigations of the effective diffusion coefficients (D_e) for HTO, $^{36}\text{Cl}^-$ and $^{22}\text{Na}^+$, the corresponding accessible porosities (ε , ε_{an}) and, in the case of $^{22}\text{Na}^+$, the distribution coefficients (R_d) were the focus of this study. The complete set of results for all tracers, D_e values and quantities related to the rock capacity factors (accessible porosities, distribution factors) are available in App. A (data) and App. F (graphical illustration of data per species and borehole). A graphical illustration of the distribution of the D_e values of HTO, $^{36}\text{Cl}^-$ and $^{22}\text{Na}^+$ for the siting regions is also compiled in Fig. 3-1. There, data from the boreholes investigated for each individual siting region (Fig. 1-1) were projected onto each siting region's reference borehole, i.e. BOZ1-1 for Jura Ost, STA2-1 for Nördlich Lägern and TRU1-1 for Zürich Nordost (Fig. 3-1). This approach was adopted because in some boreholes, priorities for diffusion analysis were given to Opalinus Clay samples, while in neighbouring boreholes in the same siting region, samples were selected from the over- and underlying rock sequences. As the variation in diffusion parameters is partly linked to rock composition, the mineralogical profiles (derived from a multi-mineralogical, i.e. MultiMin, dataset, e.g. Marnat & Becker 2021) representing the downhole rock variability (see Mazurek et al. 2023) are also presented. Since the uncharged HTO has no electrostatic interactions with solid surfaces (Van Loon et al. 2003a, Van Loon et al. 2003b, Van Loon & Soler 2003), its diffusion behaviour is considered as the reference case, primarily influenced by the mineral composition and rock porosity structure. In the three siting regions (Fig. 1-1), the effective diffusion coefficients of HTO are lowest in the Opalinus Clay and in a few other samples in both the overlying and underlying units that are rich in clay or carbonate (Fig. 3-1).

Importantly, the diffusion behaviour of charged species is significantly influenced by both the pore structure and physico-chemical factors. For example, anions such as $^{36}\text{Cl}^-$ are repelled by negatively charged clay surfaces, a phenomenon known as anion exclusion (Dutt & Low 1962). Consequently, the effective diffusion coefficient of charged species is lower than that of neutral species (as shown in Fig. 3-1), which is similar to the characteristically lower accessible porosity observed in previous studies (Appelo et al. 2010, Wigger & Van Loon 2017) and discussed further here and in a recent publication (Zwahlen et al. 2024). The composition of the porewater has a considerable effect on anion exclusion. For example, in a comparison of the results of OPA/BOZ1-1 (porewater with an ionic strength of 0.19 M) with those of OPA/TRU1-1 (porewater with an ionic strength of 0.35 M) and OPA/BUL1-1 (porewater with an ionic strength of 0.505 M), the D_e values of $^{36}\text{Cl}^-$ are on average 9.1 ± 1.4 times lower than those for HTO in the case of BOZ1-1, 5.6 ± 1.4 times lower in the case of TRU1-1 and only 4.6 ± 0.5 times lower in the case of BUL1-1.

Conversely, cations such as $^{22}\text{Na}^+$ preferentially reside near negatively charged clay surfaces, resulting in their enrichment in the solid/liquid boundary zone. As indicated in Fig. 3-1, the effective diffusion coefficient of $^{22}\text{Na}^+$ is, when normalised with the respective diffusion coefficients in free water, larger than that of HTO (and $^{36}\text{Cl}^-$) due to the contribution of adsorbed cations to the overall diffusive flux. This process is known as surface diffusion or surface-

enhanced diffusion and has been extensively investigated in the past (Glaus et al. 2007, Appelo et al. 2010). Further details are found in Section 4.2 and in Gimmi & Kosakowski (2011) and Krejci et al. (2021).

Unlike for $^{36}\text{Cl}^-$, the composition of the porewater has an opposite effect on the diffusion of $^{22}\text{Na}^+$. In the case of OPA/BOZ1-1, the D_e of $^{22}\text{Na}^+$ are – after normalisation for the diffusion coefficients in free water – on average 2.0 ± 0.8 times larger than those of HTO, whereas in the case of OPA/TRU1-1 and OPA/BUL1-1, the D_e are only 1.7 ± 0.4 and 1.9 ± 0.1 times larger, respectively.

The diffusion behaviour of $^{36}\text{Cl}^-$ seems to be more sensitive to ionic strength than that of $^{22}\text{Na}^+$. A more detailed discussion on effects of porewater composition on anion exclusion and surface diffusion is given in Section 4.2 of this report.

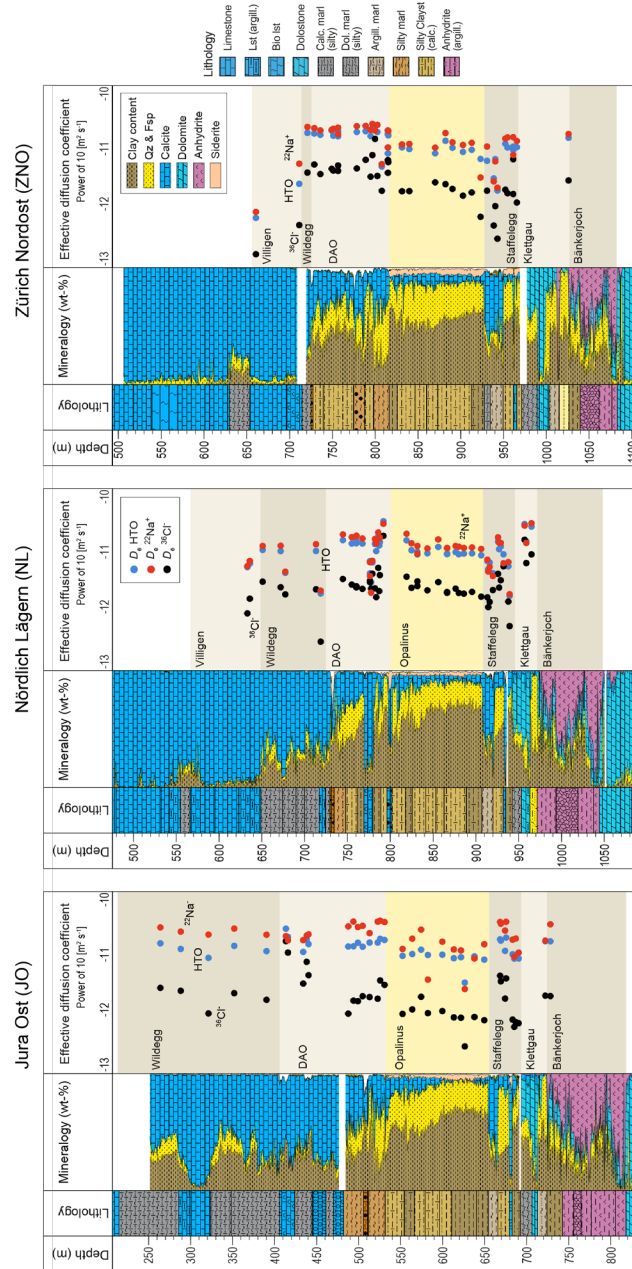


Fig. 3-1: Comparison of the effective diffusion coefficients of HTO, $^{36}\text{Cl}^-$ and $^{22}\text{Na}^+$ for Opalinus Clay (OPA), the Dogger above the Opalinus Clay (DAO) and Malm, as well as the underlying Lias, Keuper and Triassic sediments for each siting region (JO, NL, ZNO)

Lithological units (see Mazurek et al. 2023 for detailed lithological data) and mineralogical compositions (dominant mineral phases only) based on a multi-mineralogical log approach (calibrated using core data) are also provided to illustrate the main compositional changes as a function of depth. For each siting region, the reference borehole was used, and the data of the other boreholes were projected into the reference borehole. It is important to note that such a projection does not account for the porewater differences observed in some areas (important for $^{22}\text{Na}^+$ and $^{36}\text{Cl}^-$). Individual downhole plots with specific lithological and multi-mineralogical data are provided in App. F.

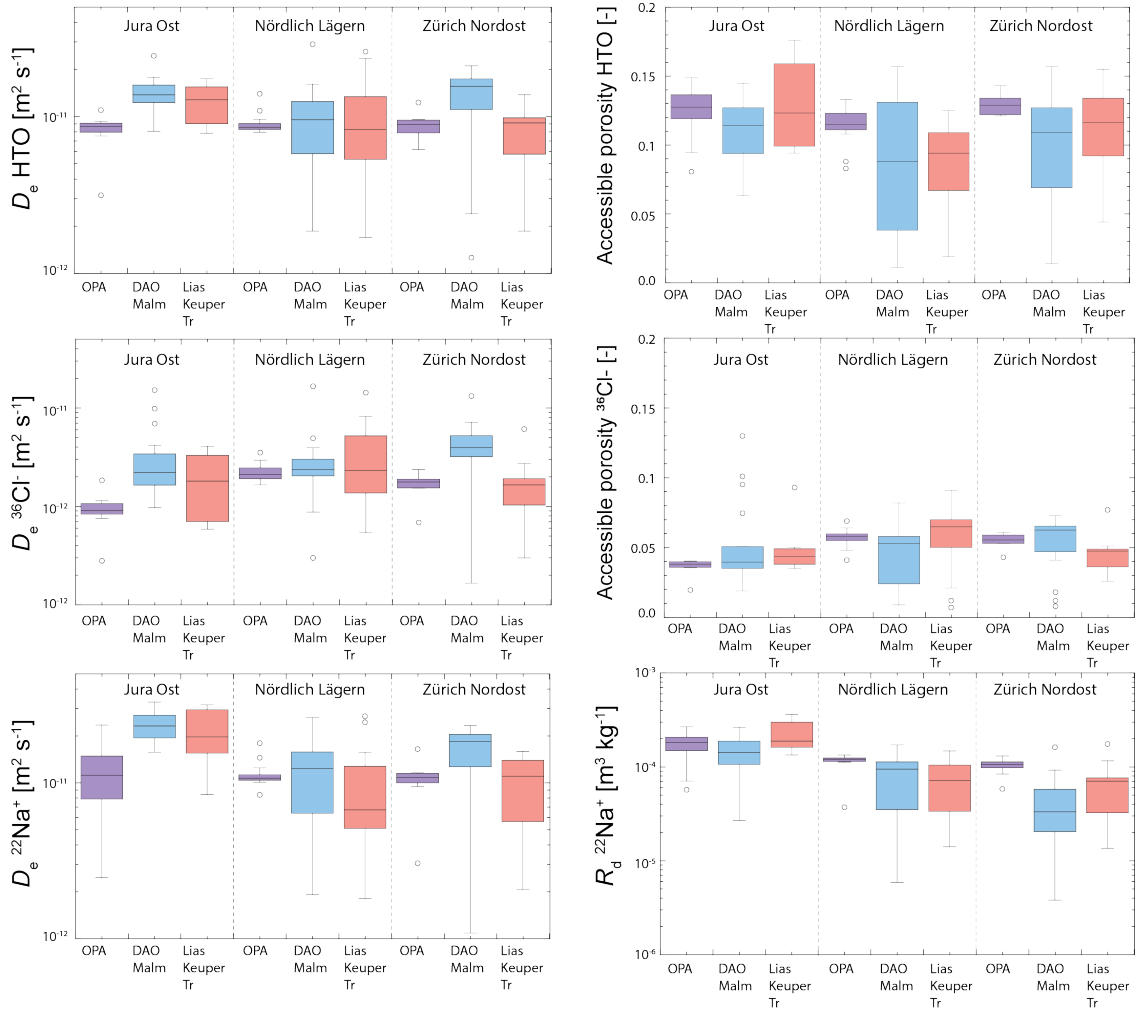


Fig. 3-2: Comparison of effective diffusion coefficients of HTO, $^{36}\text{Cl}^-$ and $^{22}\text{Na}^+$, of accessible porosities of HTO and $^{36}\text{Cl}^-$, and of R_d values for $^{22}\text{Na}^+$ for the Opalinus Clay (OPA) and for the samples from the overlying Dogger above the Opalinus Clay (DAO) and Malm, as well as for the underlying Lias, Keuper and Triassic sediments for each siting region (JO, NL, ZNO)

The boxes represent the median (line inside the box) and those data within the first and third quartiles (upper and lower ends of the box). The whiskers indicate the variability outside the upper and lower quartiles (i.e. maximum and minimum values).

Fig. 3-2 presents box plots that summarise the measured effective diffusion coefficients and accessible porosity values (or R_d in the case of $^{22}\text{Na}^+$) for the siting regions. The results shown in Fig. 3-2 reveal that the effective diffusion coefficients for HTO and $^{22}\text{Na}^+$ in the Opalinus Clay are similar across all three siting regions, i.e. approximately $1 \times 10^{-11} \text{ m}^2 \text{ s}^{-1}$, as is the effective diffusion coefficient of $^{36}\text{Cl}^-$ (approximately $1 \times 10^{-12} \text{ m}^2 \text{ s}^{-1}$). Furthermore, the distribution of the D_e values for the three tracers in the Opalinus Clay is relatively narrow, indicating that this layer has highly homogeneous diffusion properties. The accessible porosity values for HTO in the Opalinus Clay have a value of approximately 0.12 and those for $^{36}\text{Cl}^-$ approximately 0.06.

In contrast, the units above (Dogger, Malm) and below (Lias, Keuper, Trias) the Opalinus Clay exhibit greater variability in diffusion properties, indicating a more heterogeneous composition (Fig. 3-2). This heterogeneity can be attributed to diverse sedimentary facies, varying deposition rates, and different diagenetic overprints. Consequently, these units have a more heterogeneous mineralogical composition, porosity and pore-space architecture. Nevertheless, the diffusion parameters of these rock units are still within the same order of magnitude as those observed in the Opalinus Clay.

A comparison was also made between previous and current diffusion data of the Opalinus Clay, with a specific focus on the effective diffusion coefficients of HTO. This analysis involved examining data from multiple previously and recently drilled boreholes and the Mont Terri Underground Rock Laboratory (<https://www.mont-terri.ch/en/home.html>). The results show a significantly narrow range of values (Fig. 3-3), i.e. from $5 \times 10^{-12} \text{ m}^2 \text{ s}^{-1}$ to $1.5 \times 10^{-11} \text{ m}^2 \text{ s}^{-1}$. The samples from the MT-URL (Fig. 1-1) have the highest values (Fig. 3-3), which may be due to the lower maximum burial depth that the rocks experienced in the past compared to those in the three siting regions (Mazurek et al. 2006). Furthermore, the data show that the samples from the NL (BUL1-1), JO (BOZ1-1) and ZNO (Benken, TRU1-1) siting regions and from the Schlattingen-1 borehole (Fig. 1-1) yield similar values, within a factor of two. There is also no discernible impact of the current sample depth on the values of the effective diffusion coefficients.

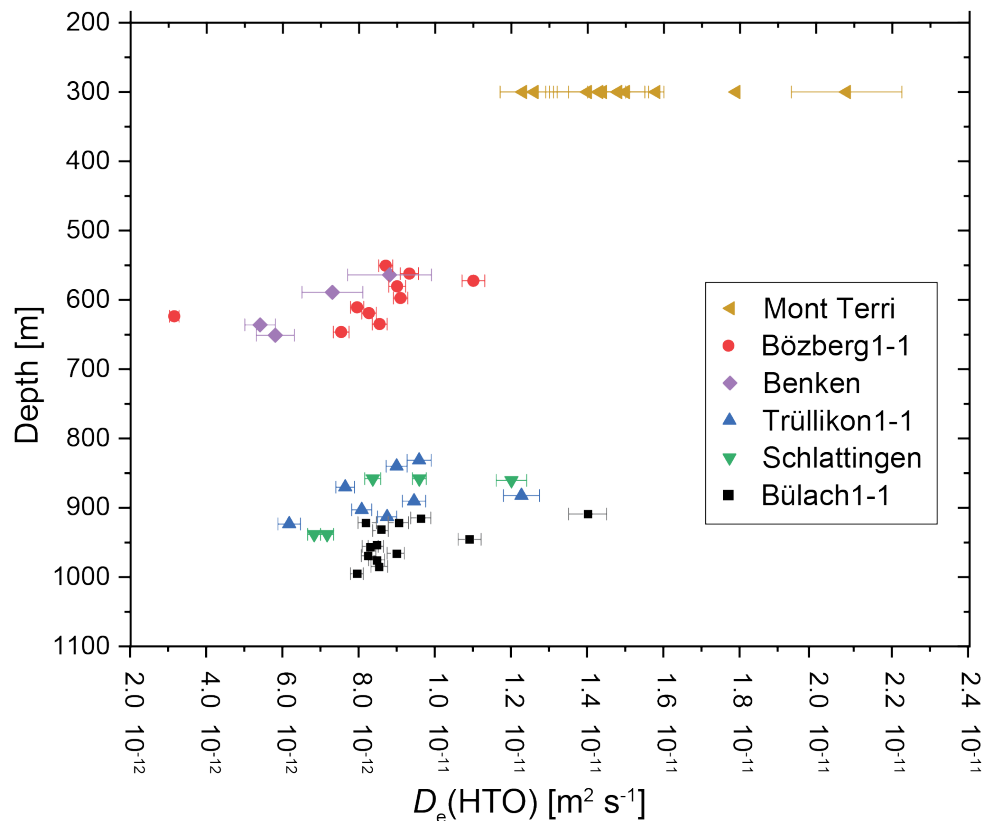


Fig. 3-3: Overview of the effective diffusion coefficients (perpendicular to the bedding plane) of HTO in OPA as a function of depth for samples from different boreholes and siting regions

Data for OPA/Mont Terri are taken from Van Loon et al. (2003a), Van Loon et al. (2005), Joseph et al. (2013) and Wu et al. (2009). Data for Benken (Fig. 1-1) are taken from Van Loon et al. (2018) and those of Schlattingen-1 from Jacops et al. (2017) and Van Loon (2014).

It must also be mentioned that diffusion measurements of clay-rich samples performed by Van Laer et al. (2024) at SCK-CEN in Belgium on five selected twin samples from the TRU1-1 borehole are in good agreement with the data presented in this study (see also App. G). Both the experimental techniques as well as the modelling approach used by the two institutes, SCK-CEN and PSI, resulted in comparable values of the effective diffusion coefficients and diffusion-accessible porosities. The difference between the effective diffusion coefficient, D_e , and the rock capacity factor, α , for each of the three tracers was – except for one outlier – consistently below a factor of 2. This level of difference is comparable to the intrinsic variability observed in the new TBO diffusion measurements between diffusion values obtained from through-diffusion experiments performed on twin samples.

3.2 Porosities from water-loss measurements

After completing the diffusion measurements, the dry weight of the samples was measured, and total porosity was calculated based on this. An overview of this water-loss porosity measurement is given in Tab. A-4, Tab. A-5 and Tab. A-6 in App. A, together with the water-loss porosity measured by the University of Bern on pristine samples, and the diffusion-accessible porosity for HTO as derived from through-diffusion measurements. A graphical overview of the data is given in Fig. 3-4, Fig. 3-5, Fig. 3-6 and Fig. 3-7.

In general, the water-loss porosity measured by the University of Bern (u^b) is somewhat lower than the diffusion-accessible porosity for HTO and the water-loss porosity measured on samples used for diffusion experiments. There could be several reasons for this, one of which is probably the fact that the samples swell (in particular the clay material) and this swelling cannot be prevented by the relatively soft diffusion cells. The swelling results in the uptake of additional water that leads to a slightly higher water-loss porosity determination.

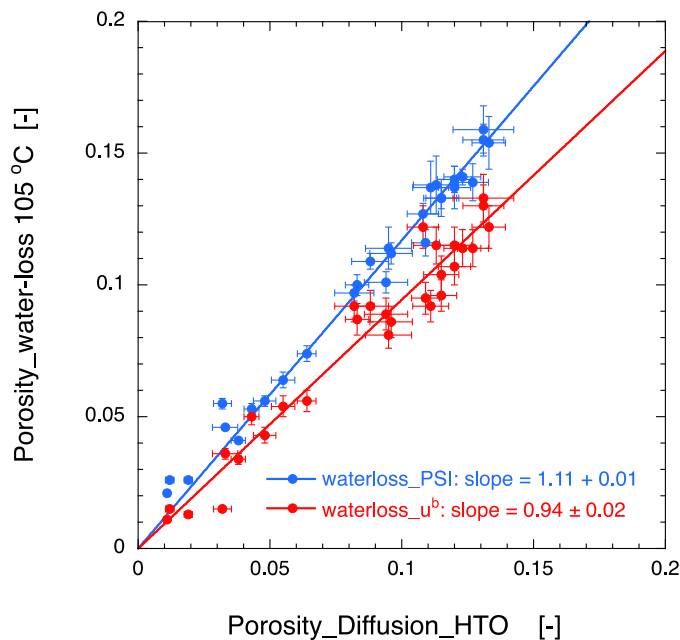


Fig. 3-4: Water-loss porosity vs. diffusion-accessible porosity for samples from BUL1-1
Water-loss porosity was measured by PSI and by the University of Bern (u^b).

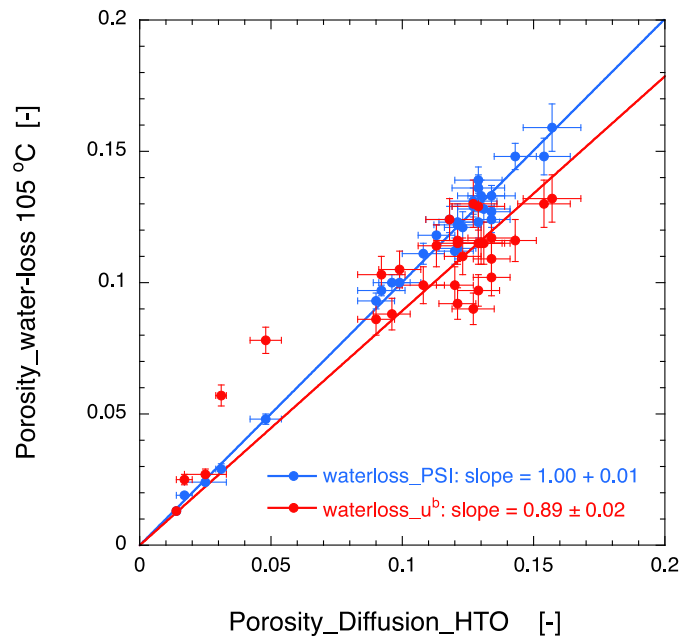


Fig. 3-5: Water-loss porosities vs. diffusion-accessible porosity for HTO for samples from TRU1-1

Water-loss porosity was measured by the PSI and by the University of Bern (u^b).

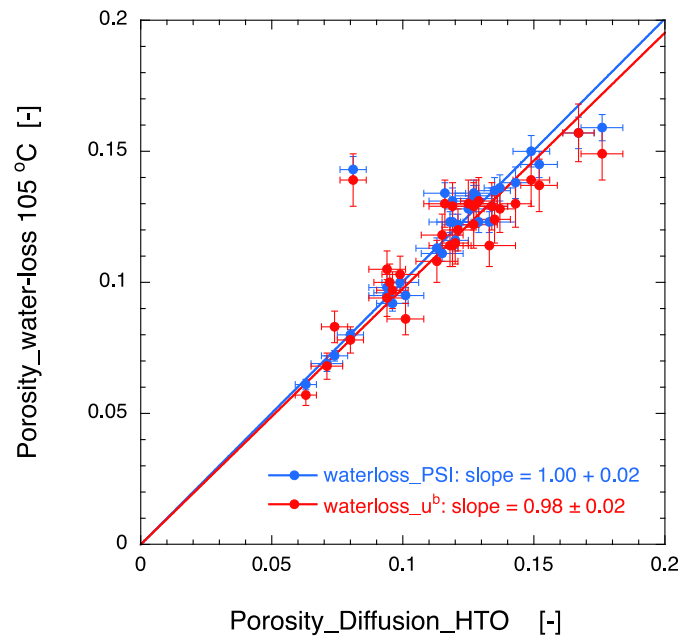


Fig. 3-6: Water-loss porosities vs. diffusion-accessible porosity for HTO for samples from BOZ1-1

Water-loss porosity was measured by the PSI and by the University of Bern (u^b).

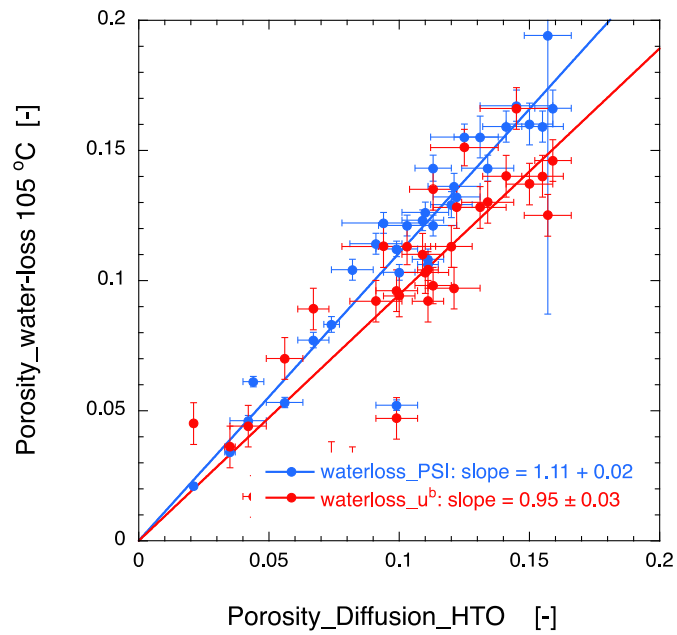


Fig. 3-7: Water-loss porosities vs. diffusion-accessible porosity for HTO for samples from the BOZ2-1, MAR1-1, STA2-1 and STA3-1 boreholes

Water-loss porosity was measured by the PSI and by the University of Bern (u^b).

4 Discussion

4.1 Diffusion of HTO

4.1.1 Relationship between porosity, geometrical factor and clay content

Diffusion experiments from the Opalinus Clay and other rock formations (e.g. Van Loon 2014) often refer to the close relationship between total porosity and the effective diffusion coefficient (analogous to Archie's law) (see also Boving & Grathwohl 2001). Consequently, the tortuosity factor is often expressed as a function of total porosity. Hence, Archie's relationship and extensions of it were often used in previous studies (Bestel et al. 2018, Glaus et al. 2010, Van Loon & Mibus 2015) for a simplistic empirical description of the effective diffusivity as a function of the total porosity of the porous medium. This relationship could also be explored by means of the additional 130 new diffusion data of the highly heterogenous Mesozoic rock sequence in Northern Switzerland and is correspondingly plotted in Fig. 4-1.

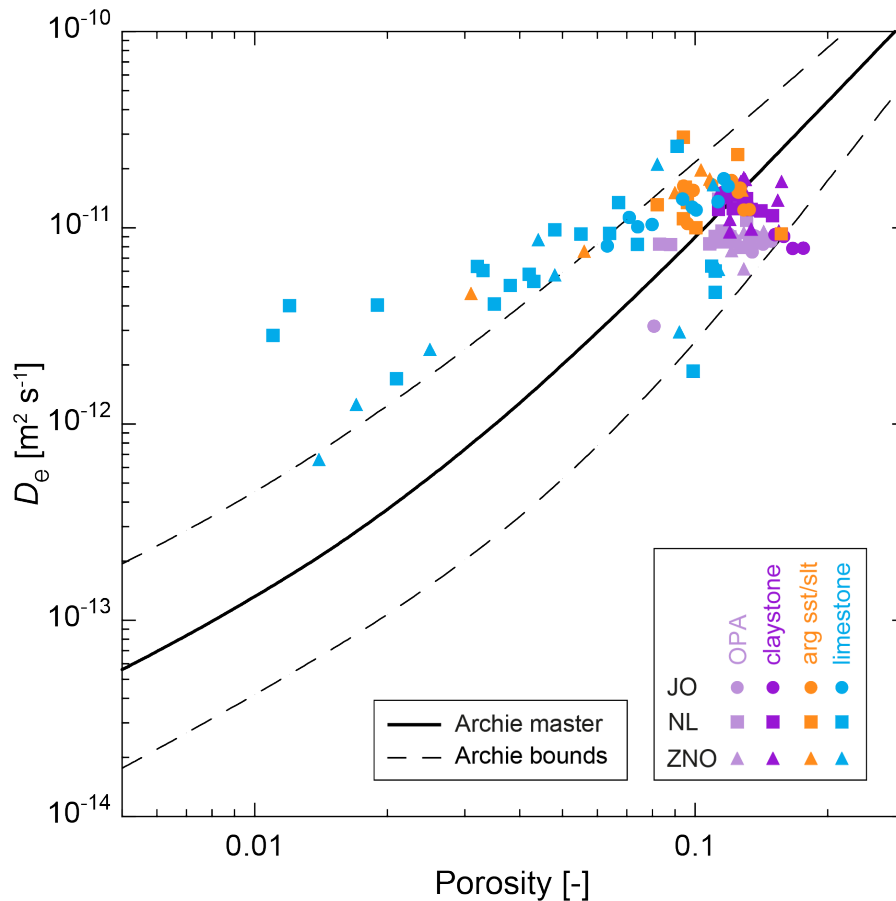


Fig. 4-1 Relationship between accessible porosity and the effective diffusion coefficient of HTO (Archie's plot) showing that data of samples with low clay content deviate markedly from the ideal Archie model

The lines were calculated using the e-Archie equation and parameters given in Van Loon & Mibus (2015).

From the data obtained (Fig. 4-1), it is rather obvious that this approach is not sufficiently accurate given that the majority of the porosity values of the rock samples cluster around a value of ~ 0.1 with a rather large variability of the D_e values for HTO. Furthermore, the best-fit parameter values for the extended Archie's relationship provided in Van Loon & Mibus (2015) did not produce a valuable consistency with the results obtained for the additional, newly acquired data of carbonate-rich rock samples (Fig. 4-1). For this reason, alternative empirical model descriptions are proposed here.

As the presence of clay minerals determines several rock properties important for radionuclides migration processes (e.g., specific surface area, pore-size distribution, cation-exchange capacity), the correlation between diffusion parameters and total clay content is explored using the diffusion parameters of HTO as it is a neutral species that is not significantly affected by the chemical composition of the porewater. Equation (1-8) is used as the basis to test the parameter dependence on the total clay content. Fig. 4-2 displays the correlation between clay content and HTO porosity (derived from diffusion of HTO), indicating that porosity is mainly associated with the clay phase of the rock, consistent with the findings of Mazurek et al. (2023).

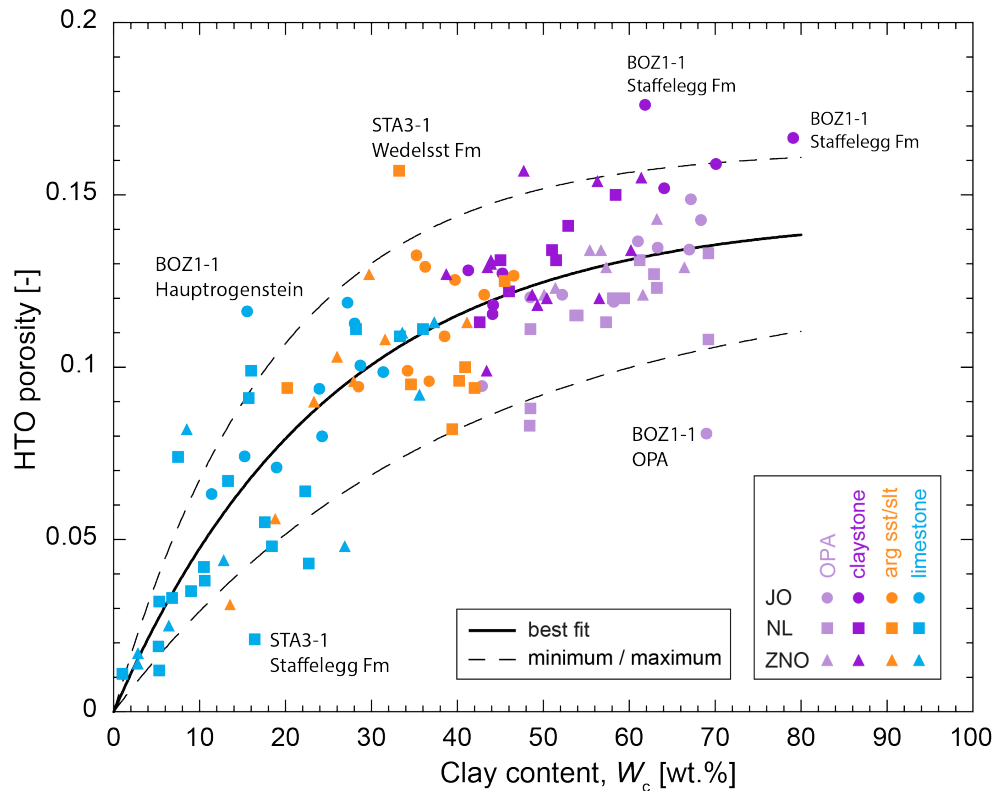


Fig. 4-2: Correlation between the HTO porosity derived from diffusion of HTO and the clay content (W_c)

The fit curves were obtained with Eq. (4-2) using the parameter values and uncertainties given in Tab. 4-1.

$$G = \frac{\varepsilon}{D_e} \cdot D_w \quad (4-1)$$

In agreement with the box plots shown in Fig. 3-2, there is no discernible trend with respect to the siting regions in the data shown in Fig. 4-2 and Fig. 4-3.

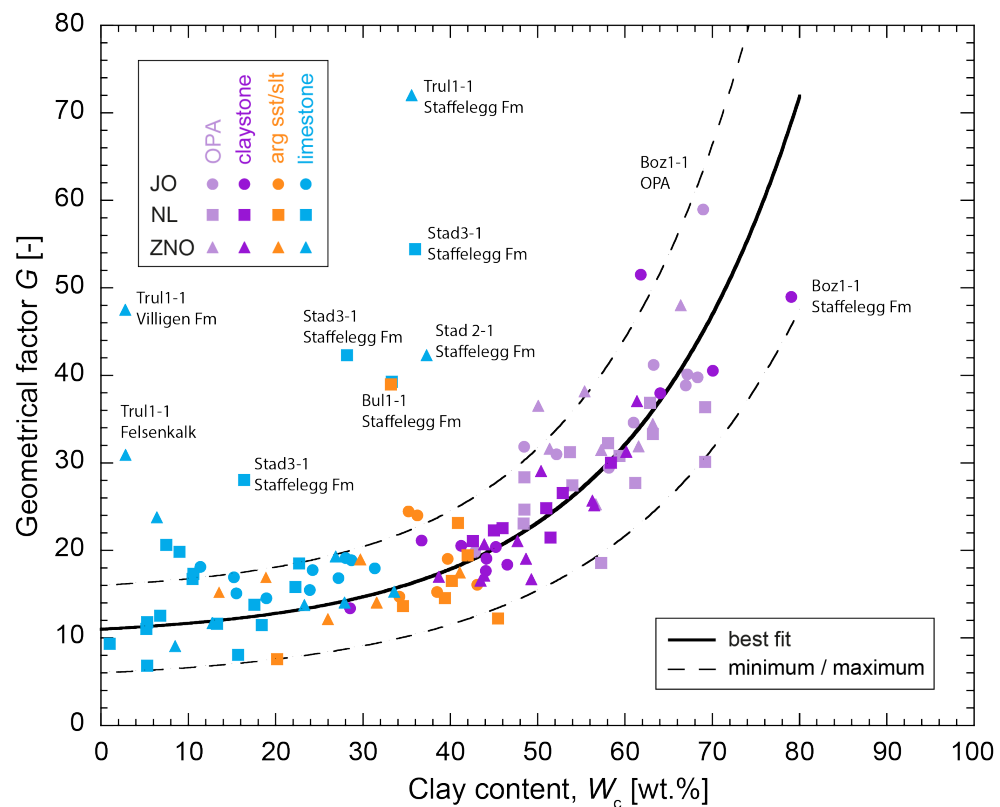


Fig. 4-3: Correlation between the geometrical factor of HTO and the clay content (W_c)

The fit curves were obtained by Eq. (4-3) using the parameter values and uncertainties given in Tab. 4-2.

The correlation between clay content and accessible porosity can be modelled by an empirical exponential function:

$$\varepsilon = m_1 \cdot (1 - e^{-m_2 \cdot W_c}) \quad (4-2)$$

Where m_1 and m_2 are fitting constants and W_c represents the total clay content in weight percent (wt.%). The parameter m_1 represents a maximum total porosity value ($\varepsilon_{\max}$) in the clay phase. A maximum total porosity of ca. 0.14 results from high clay content. The solid black line represents the best fit, whereas the upper and lower curves represent upper and lower bounds at 3σ . The values of the parameters to calculate these curves are given in Tab. 4-1.

Tab. 4-1: Values and uncertainties (1σ and 3σ confidence level) of the parameters m_1 and m_2 used in Eq. (4-2) to calculate the curves given in Fig. 4-2

Parameter	Best value	1σ	3σ
m_1	0.1444	0.0063	0.0188
m_2	0.0398	0.0045	0.0134

Mazurek et al. (2023) observed a similar dependency between the total clay content and the water-content porosity and observed a quasi-linear increase of the porosity at low clay content and a flattening of the curve for higher clay contents. They used a power-law function to fit the experimental data. The reason for the flattening of the curve will be explained later.

The geometrical factor G significantly increases with increasing clay content beyond a clay content larger than approximately 30 wt.%. Below this threshold, G remains relatively constant at a value of approximately 11 ± 4 . According to the conceptual shale and microstructure model of Bourg (2015) shown in Fig. 4-4, the threshold value should be at $W_c = 34 \pm 10\%$, which supports the selected threshold of 30 wt.% in this report¹. The outliers observed for limestones could be correlated with a large textural heterogeneity observed for these rock types and bi- or multi-modal pore-size distributions (Mazurek et al. 2023). Empirical analysis of the data suggests a correlation between the geometrical factor and the total clay content that can be described by an empirical exponential function:

$$G = a_1 + e^{a_2 \cdot W_c} \quad (4-3)$$

where a_2 is a fitting parameter and W_c represents the total clay content in weight percent (wt.%). The value of a_1 was arbitrarily fixed at 10. The solid black line represents the best fit, whereas the upper and lower curves represent upper and lower bounds at 3σ . The values of the parameters to calculate these curves are given in Tab. 4-2.

¹ Note that the critical clay content for the transition should be regarded as a smooth range rather than a discrete value. For this reason, values that deviate slightly from this threshold are also taken into account below

Tab. 4-2: Values and uncertainties (1σ and 3σ confidence level) of the parameters a_1 and a_2 used in Eq. (4-3) to calculate the curves given in Fig. 4-3

Parameter	Best value	1σ	3σ
a_1	10.0	2.0	6.0
a_2	0.0515	0.0016	0.0047

4.1.2 Relationship between the rock microfabric and clay content

Argillaceous rocks are composed of a range of mineral grains of varying sizes (see also Mazurek et al. 2023, with clay minerals mainly falling in the range of 10 nm to 20 μm or larger (illite, kaolinite, chlorite, mica), and non-clay minerals such as quartz, carbonates and feldspars commonly in the order of 5 μm to larger than several millimetres. The grain size, rock fabric and texture (including diagenetic cements or recrystallised grains), and thus pore size and space depend on depositional environment, source material, burial depth and diagenetic overprint, which can significantly alter the fabric and composition of the rocks (Wüst et al. 2018). The diffusive behaviour of solutes in sedimentary rocks often shows a strong dependency on the rock composition, with the clay phases playing a significant role due to their large surface area and surface charge. Hence, clay and non-clay mineral aggregates, sizes, grain arrangement, inter-granular pore space or pore connectivity are parameters determined by the rock type and diagenetic overprint (Mazurek et al. 2023). Thereby, in simple terms, the load-bearing phase in clay rocks depends on the composition, with either the clay phase or the non-clay phase forming the load-bearing phase. This has significant implications for the rock's properties, including permeability, diffusivity (Dabat et al. 2019, Dabat et al. 2020, Ferrage et al. 2023), mechanical strength (Bourg 2015), and the anisotropy of these properties.

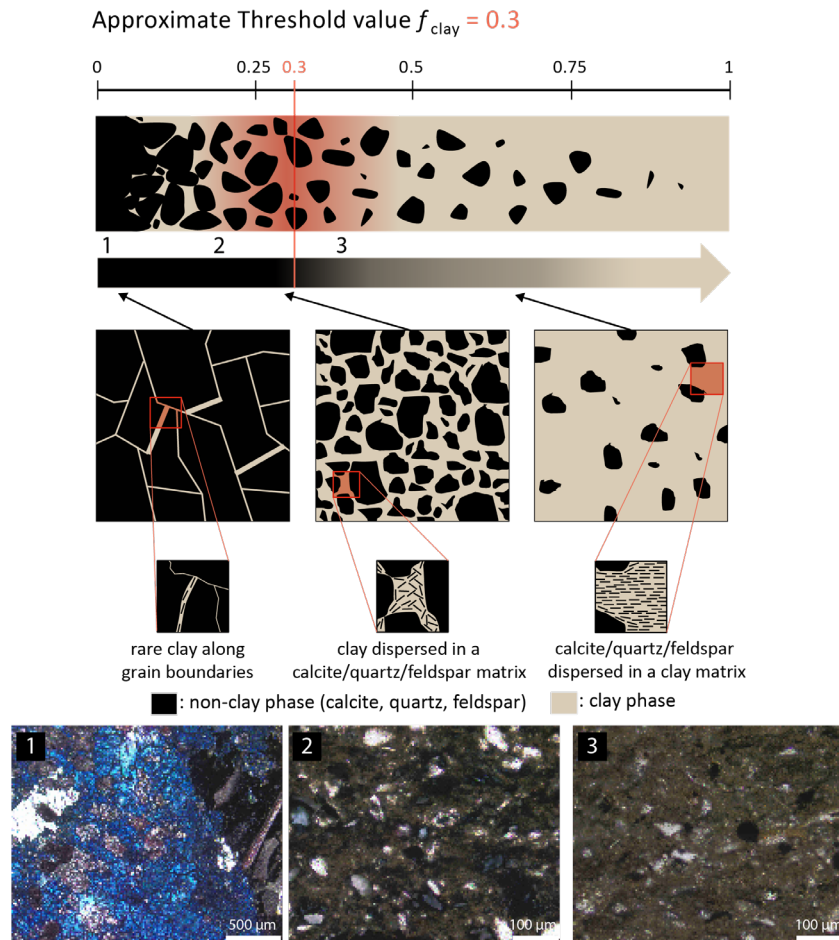


Fig. 4-4: Conceptual model of clay rocks with different amounts of fine-grained clay and coarse-grained calcite, quartz and feldspar

Figure (top) modified from Crawford et al. (2008), Houben et al. (2014), and Bourg (2015). Thin-section images (bottom from left to right) display recrystallised carbonates, and silt- and claystones (all DAO).

In clay-rich rocks, the clay minerals are preferentially oriented perpendicular to the direction of sedimentation or rock compaction, whereas for clay-poor rocks, the orientation is more random and depends on mineral sizes and aggregate fabric. According to Bourg (2015) and Bourg & Ajo-Franklin (2017), the critical clay content where the change of preferential clay and clay-mineral orientation occurs is approximately 33 wt.% (weight fraction of clay $f_{\text{clay}} \approx 0.33$). In view of diffusion properties perpendicular to the bedding plane, it can be expected that the geometrical factor is larger in the case of clay-rich rocks (clay content > 30 wt.%) because of the larger tortuous diffusion pathways (τ is larger in Eq. (1-7)). In the case of clay-poor rocks (clay content ≤ 30 wt.%), the geometrical factor is lower because of the more random orientation of the clay platelets between the grains (shelter effect of the larger grains during compaction of the sediments; see also Mazurek et al. 2023) resulting in a smaller tortuosity. Additionally, the G value is comparatively constant for clay poor rocks because the non-clay minerals form the load-bearing phase (Fig. 4-4), which limits the effect of compaction on the orientation of clay platelets.

4.1.3 Relationship between partial clay density and clay content

For clay contents ≥ 30 wt.% ($f_{\text{clay}} \geq 0.3$), the clay minerals form the load-bearing phase (Fig. 4-4), thus, the larger the clay content, the more pronounced the preferred orientation of the clay platelets and the larger the G value (Fig. 4-3). Additional evidence for such a behaviour is given in a previous study by Lash & Blood (2004). The authors investigated the orientation of clay platelets in black shale of the Upper Devonian (Frasnian) Rhinestreet shale of western New York State with ellipsoidal carbonate concretions. These concretions changed the elastic stress field in such a way that the sediments in the immediate near-field at the lateral edges of the concretions were shielded from the overburden pressure by the creation of a so-called pressure shadow. This resulted in a random distribution of the clay platelets, in a so-called ‘house-of-cards’ fabric. At several tens-of-cm distance from the pressure shadows, the sediment was fully affected by the overburden stress, leading to a preferential orientation of the clay or a ‘strongly oriented microfabric’ (Lash & Blood 2004).

It is important to point out that not only does the orientation of the clay platelets differ with different clay contents, but porosity and bulk dry density of the clay phase also change. Such changes are also associated with the progressive burial of the sediments during which clay-mineral crystallinity can increase. If we assume that porosity within these diffusion-driven tight rocks of different origin within the Mesozoic sequence is associated with the clay phase, the porosity can be estimated from the weight fraction of clay minerals. The pore volume of the total rock (V_p) equals the pore volume of the clay phase ($V_{p,c}$):

$$V_p = V_{p,c} \quad (4-4)$$

The pore volume can be calculated from the porosity and the volume of the rock and/or minerals:

$$\varepsilon \cdot V_T = \varepsilon_c \cdot V_c \quad (4-5)$$

where ε is the total porosity of the rock, ε_c is the porosity of the clay phase, V_T and V_c are the volumes of the total rock and the clay phase, respectively.

Eq. (4-5) can be written as:

$$\varepsilon \cdot \frac{m_r}{\rho_{br}} = \varepsilon_c \cdot \frac{m_c}{\rho_{bc}} \quad (4-6)$$

where m_r and m_c are the masses of the rock and the clay phase, respectively, and ρ_{br} and ρ_{bc} represent the bulk dry densities of the rock and the clay phase. Rearranging Eq. (4-6) results in:

$$\varepsilon = \varepsilon_c \cdot \frac{m_c}{m_r} \cdot \frac{\rho_{br}}{\rho_{bc}} \quad (4-7)$$

with $f_{clay} = m_c/m_r$ (weight fraction of the clay phase). Note that $f_{clay} = W_c / 100\%$

$$\varepsilon = \varepsilon_c \cdot f_{clay} \cdot \frac{\rho_{br}}{\rho_{bc}} \quad (4-8)$$

The porosity of the clay phase can be written as:

$$\varepsilon_c = 1 - \frac{\rho_{bc}}{\rho_{gc}} \quad (4-9)$$

where ρ_{gc} is the grain density of the clay phase. Replacing ε_c in Eq. (4-8) with Eq. (4-9) gives:

$$\varepsilon = \left(1 - \frac{\rho_{bc}}{\rho_{gc}}\right) \cdot f_{clay} \cdot \frac{\rho_{br}}{\rho_{bc}} \quad (4-10)$$

Or, after rearranging:

$$\varepsilon = f_{clay} \cdot \rho_{br} \cdot \left(\frac{1}{\rho_{bc}} - \frac{1}{\rho_{gc}}\right) \quad (4-11)$$

Thus, the porosity of the rock can be calculated from the weight fraction of the clay, the total bulk dry density of the rock, and the bulk dry density and the grain density of the clay phase. The weight fraction of the clay is a variable and was varied from 0.05 to 0.8 in the model. An average value of the bulk dry density of the rock was taken to be $2,600 \text{ kg m}^{-3}$. This is an approximation because the bulk dry density of the sedimentary rocks also varies with the clay content. At low clay contents, rock densities are about $\rho_{br} \approx 2,700 \text{ kg m}^{-3}$ whereas at high clay contents they are about $\rho_{br} \approx 2,400 \text{ kg m}^{-3}$ (Fig. B-5 in App. B). For the grain density of the clay phase, a value of $2,750 \text{ kg m}^{-3}$ was selected (Fig. B-5 in App. B).

Two scenarios were considered for the bulk dry density of the clay phase. In the first scenario (Fig. 4-5), a constant value of $\rho_{bc} = 1,900 \text{ kg m}^{-3}$ was assumed regardless of the clay content. This scenario was expected to result in a linear increase of the calculated porosity as a function of the clay content (shown by the blue curve in Fig. 4-5), which disagrees with the data shown in Fig. 4-2. However, the results in Fig. 4-5 show that for clay contents up to approximately 20 – 25 wt.%, the calculated and measured porosity values are in good agreement when a constant ρ_{bc} is used. The choice of $\rho_{bc} = 1,900 \text{ kg m}^{-3}$ is based on a trial-and-error approach giving the best results, at least for the lower clay content. For clay contents > 25 wt.%, the porosity is clearly overestimated when using $\rho_{bc} = 1,900 \text{ kg m}^{-3}$, suggesting that the assumed value is too low. In a second scenario (Fig. 4-5), the bulk dry density of the clay phase was kept constant up to a clay content of approximately 25 wt.% and was continuously increased (linearly) for clay contents > 25 wt.% to values between $\rho_{bc} = 2,000 \text{ kg m}^{-3}$ and $\rho_{bc} = 2,340 \text{ kg m}^{-3}$. The use of variable bulk dry density resulted in a good agreement between the calculated porosity of the rocks and the data in Fig. 4-5. This indicates that the porosity of clay rocks is strongly influenced by the clay content (i.e. clay-content dependent) and the load-bearing nature of the clay phase.

As there is no experimental way to determine ρ_{bc} in a rock sample, it is difficult to judge whether there is a continuous change of the dry density with clay content ($\rho_{bc} = f(W_c)$) or whether ρ_{bc} is constant up to a given threshold value of clay content, e.g. 30 wt.%, and changes for clay contents larger than this threshold value. However, according to the observed porosity values, it seems that ρ_{bc} at low and high total clay content must be different, i.e. the value at low clay content is lower than at high clay content, though the exact dependency on the clay content remains unknown. The clay bulk dry density might also affect the pore-size distribution. It is expected that at lower bulk dry densities, the pore sizes are larger than in the case of higher bulk dry densities. In that sense, it can be expected that the pore size in low clay rocks is larger than in the case of clay-rich rocks, because – as discussed earlier – the clay bulk dry density is lower. This agrees with the pore-size distribution measurements given in Mazurek et al. (2023). Beside tortuosity, the pore size could also affect the geometrical factor. A larger pore size results in a larger pore constrictivity, δ , in Eq. (1-7) and vice versa.

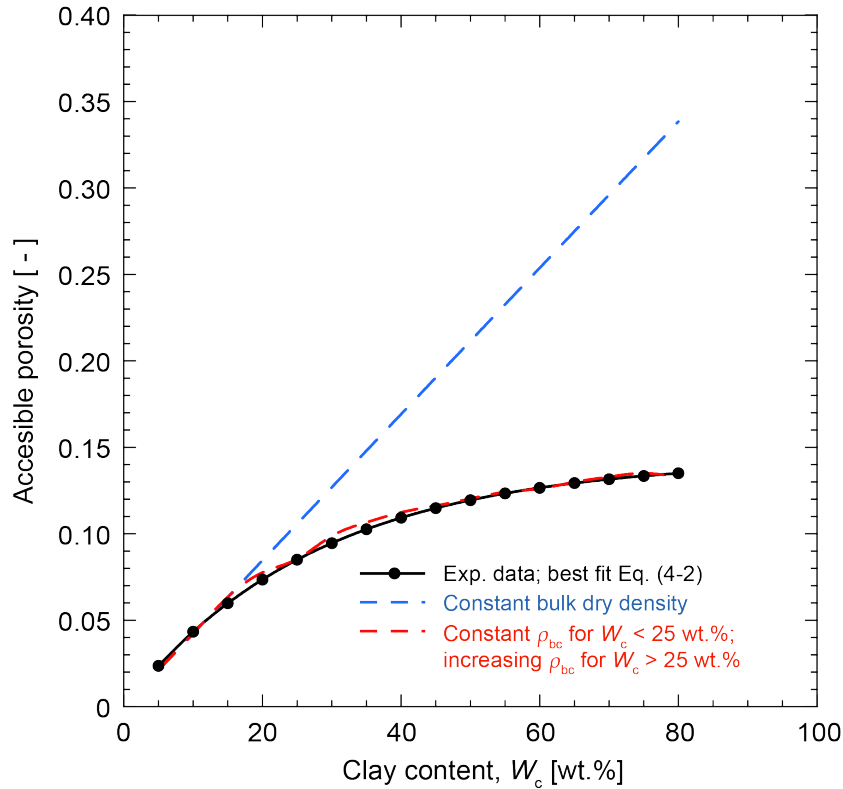


Fig. 4-5: Calculated and observed porosity values for samples with different mineral compositions

The blue line shows porosities calculated with Eq. (4-11) using a clay bulk dry density of $1,900 \text{ kg m}^{-3}$, irrespective of the clay content of the samples. The red curve shows porosities calculated with Eq. (4-11) using $\rho_{bc} = 1,900 \text{ kg m}^{-3}$ for a clay content $< 25 \text{ wt. %}$, and an increasing ρ_{bc} for clay contents $> 25 \text{ wt. %}$.

4.1.4 Relationship between effective diffusion coefficients and clay content

Combining Equations (1-8), (4-2) and (4-3) results in an empirical equation that relates the effective diffusion coefficient to the total clay content:

$$D_e = \frac{m_1 \cdot (1 - e^{-m_2 \cdot W_c})}{a_1 + e^{a_2 \cdot W_c}} \cdot D_w \quad (4-12)$$

Fig. 4-6 visualises the correlation between the effective diffusion coefficient of HTO and the clay content of a sample. Between a clay content of approximately 20 and 50 wt.%, the dependency of the effective diffusion coefficient on the clay content is comparatively weak. Below a clay content of 20 wt.% and above a clay content of 50 wt.%, the effective diffusion coefficient decreases with decreasing or increasing clay content.

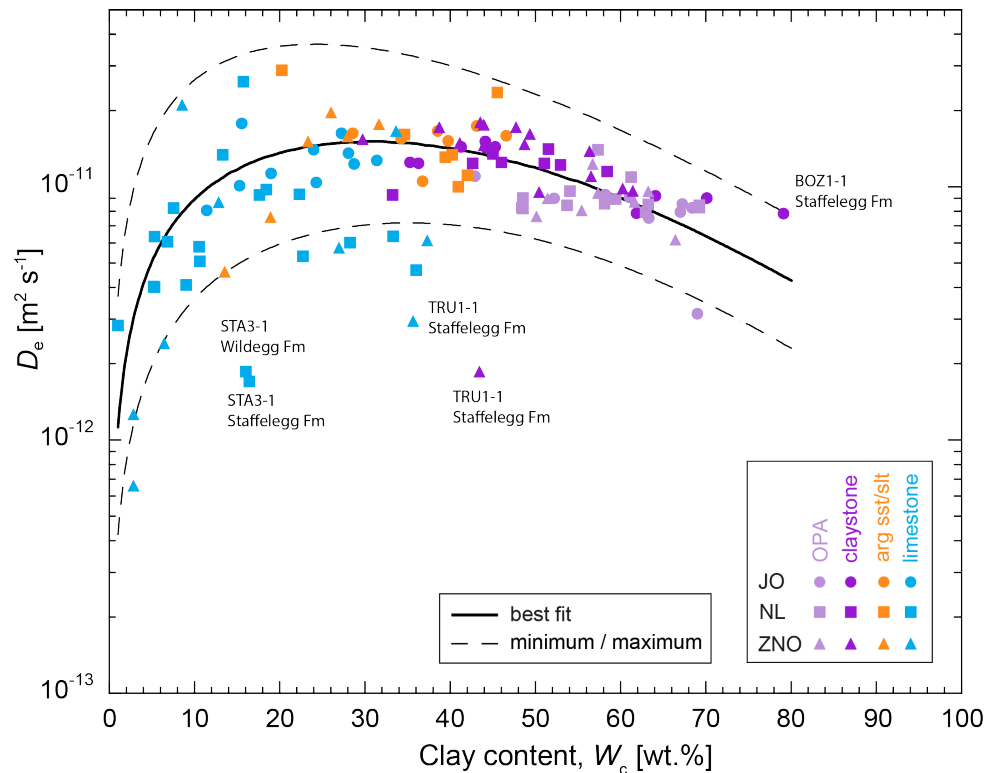


Fig. 4-6: Correlation between the effective diffusion coefficient of HTO and the total clay content (W_c) for all samples from the three siting regions

The solid line represents the best fit using Eq. (4-12) and the parameters summarised in Tab. 4-1 and Tab. 4-2. The upper and lower curves represent the upper and lower bounds at a 3σ confidence level. Some of the outliers are labelled and are clay-rich samples with high amounts of organic carbon (4.7 and 7.8 wt.%) from the Staffelegg Formation of the TRU1-1 borehole (ZNO). Two other outliers are clay-poor samples with high calcite (79 and 74 wt.%) but low organic carbon contents (0.36 and 0.28 wt.%) and are from the Staffelegg and Wildegg Formation of the STA3-1 borehole (NL).

4.1.5 Effect of the mineralogy of the rock on diffusion parameters

A model that includes more detailed information regarding mineralogical and textural properties beyond what is presented in Eq. (4-12) would necessitate additional fundamental data. This is exemplified in Fig. 4-7, which shows a Füchtbauer diagram (Füchtbauer 1988). This is a graphical representation of the sedimentary rocks in terms of their clay minerals, carbonates, and quartz-feldspar assemblages. Within the ternary diagram, sample distribution is displayed together with diffusion parameter values, such as G , porosity and D_e for HTO. The diagram reveals several distinct trends regarding the correlations of mineralogy and diffusion parameters.

The Füchtbauer diagram (Fig. 4-7) shows that an increase in clay minerals leads to a rise in both the geometrical factor and porosity (as well as external surface area), regardless of the presence of calcite and quartz.

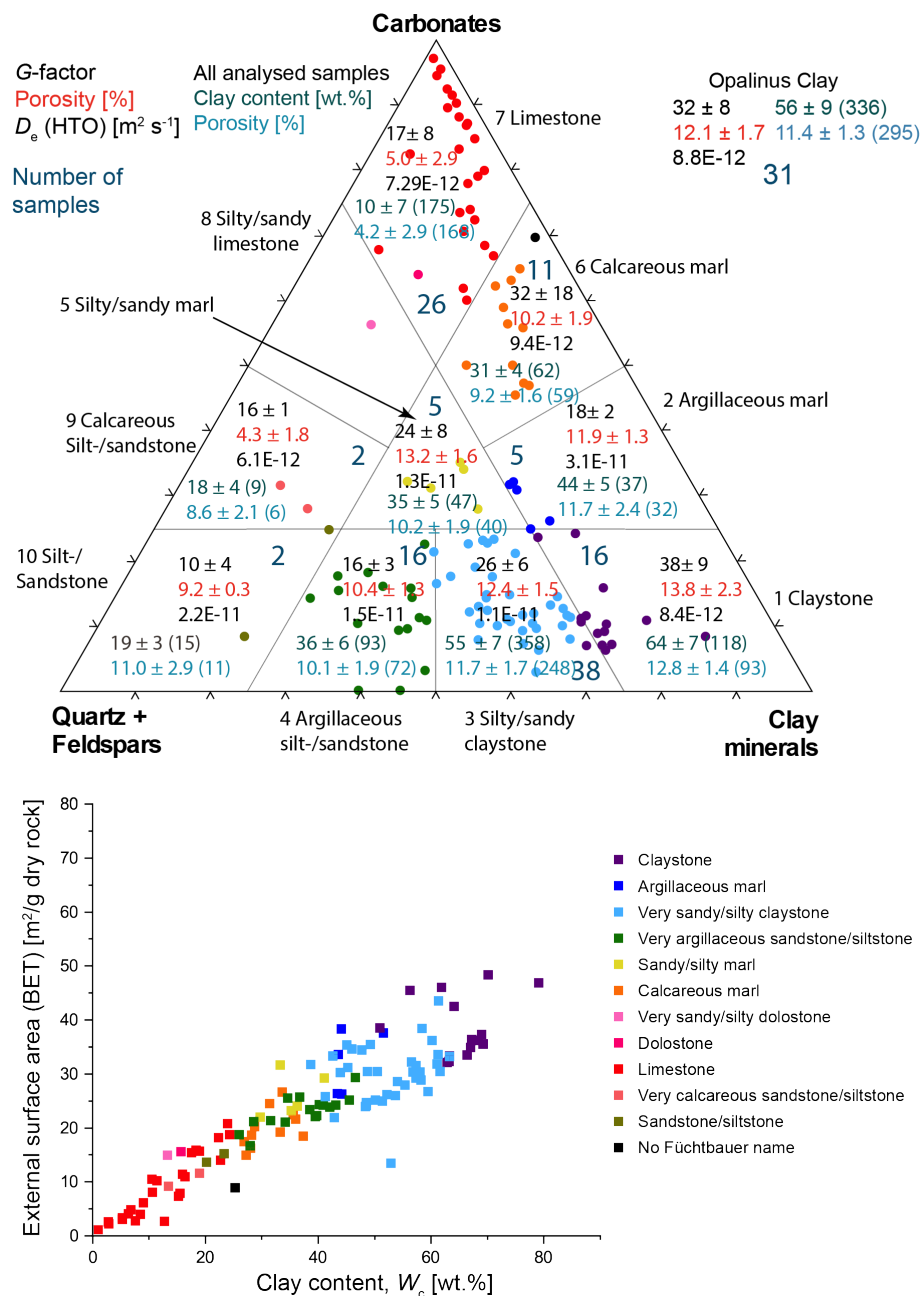


Fig. 4-7: Füchtbauer diagram (top) illustrating diffusion parameters within the ternary diagram of carbonates (calcite, dolomite, ankerite), quartz and feldspars (K-feldspar, plagioclase), and clay minerals (illite, smectite, kaolinite, chlorite-end-members) as end-members

These parameters include the geometrical factor, accessible porosity, and effective diffusion coefficient of HTO. The diagram demonstrates that the G -factor and porosity generally increase from both quartz and feldspars and carbonate end-members towards the clay mineral end-member. Conversely, D_e , the effective diffusion coefficient of HTO, is lowest in limestones. The relationship between sample surface area (BET) and total clay content (bottom) illustrates that high carbonate content within these Mesozoic sequences characterise low BET and thus apparent and effective diffusion coefficients.

The geometrical factor increases from approximately 10 in the siltstones/sandstones to 38 in the claystones, while the carbonate line shows a value of 17 for limestones, 32 for calcareous marls, and 18 for argillaceous marls. These findings align with those presented in Fig. 4-3. The impact of quartz and calcite on pore microstructure differs depending on the clay content, with an increase in quartz content resulting in a decrease in the geometrical factor but an increase in porosity when the clay content is below 30 – 40 wt.% (Fig. 4-2). The concurrent trends of porosity and geometry factors from carbonates and quartz/feldspars, respectively, to clay minerals partially offset each other in terms of the structure of Eq. (1-8) and the individual parameter dependencies. As a result, porosity and the geometrical factor are more informative and useful for a detailed and far-reaching description of the data in general.

These observed trends (Fig. 4-7) suggest that, in addition to mineralogy, particle size and diagenesis may have a significant influence on pore microstructure (see also Mazurek et al. 2023). Quartz and carbonate minerals differ primarily in terms of their grain-size distribution, with carbonates having a broader range including recrystallised microfossils as well as having a greater likelihood of pressure solution or matrix cementation (i.e. micrite, sparite) (see also Wüst et al. 2018). However, without relevant data and supporting arguments, such hypotheses remain speculative.

To investigate the impact of mineral composition, diagenesis, and pore structure on diffusion in sedimentary rocks, further petrographic or microscopic (including scanning electron microscopy) studies are necessary in the future. These studies should focus on rock samples with higher quartz content, such as silt/sandstone and calcareous silt/silty sandstone samples and provide additional textural and structural information.

The effective diffusion coefficient (D_e) of HTO exhibits minimum variation along the line of the clay-carbonate end-members but increases from clay towards the quartz end-member. However, this trend is based on varying and, occasionally, few sample numbers within the different categories and therefore only has limited statistical significance.

4.2 Diffusion of anions and cations

4.2.1 Electrical double layer

The permanent negative charge on the clay surface of mainly 2:1 clay minerals (illite, montmorillonite) causes a repulsion of negatively charged species and an attraction of positively charged species (Tournassat et al. 2015). This results in an area close to the surface, which is partially depleted in anions and enriched in cations. This area is also called the diffuse layer (DL). The area closest to the surface consisting only of cations that are strongly bound to the surface is called the Stern layer. The Stern layer together with the diffuse layer forms the electric double layer (EDL) (Fig. 4-8). In the EDL, the cation (or anion) concentration continuously decreases (or increases) from the clay surface to its value in the free solution (for sufficiently wide pores). In the Donnan approximation of the EDL, which is taken for the following purposes, two homogeneous domains, each with a constant concentration, are assumed: a central “free” domain, where the cation/anion concentrations equal the concentrations in free water, and a Donnan layer near the pore boundaries. The concentrations of anionic and cationic species in the Donnan layer ($c_{DL,i}$) are related to the respective concentrations in the free water (c_i) by Boltzmann’s law:

$$c_{DL,i} = c_i \cdot e^{-\frac{z_i F \psi_D}{RT}} \quad (4-13)$$

where z_i is the charge of species i , F is the Faraday constant (C mol^{-1}), T the absolute temperature (K), R the molar gas constant ($\text{J mol}^{-1} \text{K}^{-1}$) and ψ_D is the so-called Donnan potential (Volt), which is assumed to take a homogeneous average value that is representative of the potential distribution in the Donnan layer. The thickness d_{DL} (m) of the Donnan layer is assumed to be a multiple of the Debye length (see later).

In the case of anions, the concentration in the DL is lower than that in the free water (anion depletion). In the case of cations, the concentration in the DL is higher (enrichment of cations). For neutral species, the concentration in both the DL and free water is identical.

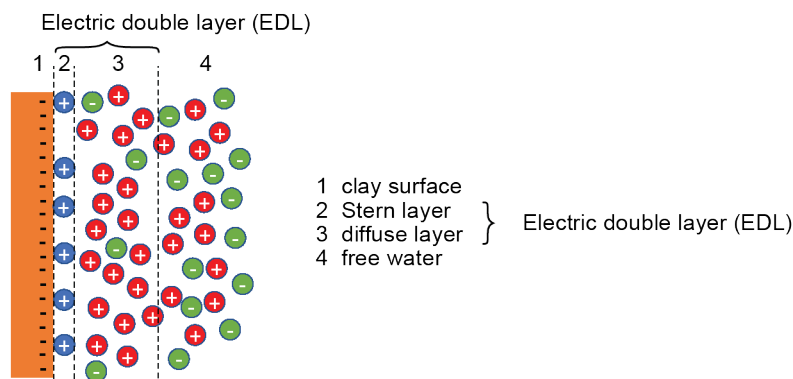


Fig. 4-8: Schematic view of the electric double layer (EDL)

The diffuse layer represents a charged aqueous phase where the number of anions is smaller than the number of cations. The net positive charge of the EDL neutralises the net negative charge on the surface. The free water represents an electro-neutral aqueous phase where the number of anions equals the number of cations.

4.2.2 Diffusion in the EDL

The aim of the following section is to evaluate the applicability of current models in describing the interaction of cationic and anionic species at planar clay surfaces using the above data obtained from diffusion measurements. Previous studies, including van Schaik et al. (1966), Dufey & Laudelout (1975), Jensen & Radke (1988), and Molera & Eriksen (2002), have established that concentration gradients of mobile surface-associated cationic species in negatively charged clays also contribute to overall observed fluxes. These gradients can explain the “unexpectedly large” effective diffusion coefficients of such elements because the definition of the effective diffusion coefficient relates the overall observed flux to the concentration gradients of the aqueous phase species only (see Eq. (1-1)).

In the case of compacted smectites, the concentration gradients of mobile surface-associated cationic species are even the dominant driving forces for diffusion (Glaus et al. 2007), which has been confirmed in diffusion experiments carried out under gradient conditions of the main electrolyte, in which gradients of the electro-potential act as an additional driving force (Glaus et al. 2013). Under such situations, the concentration gradients of mobile surface-associated cationic species were identified as the dominating driving force for diffusion. Similar findings were also made in the case of diffusion of transition metal cations in compacted illite (Glaus et al. 2015a). This latter study used the Donnan approach involving immobile Stern layer cations and mobile diffuse-layer cations for a quantitative description of the effective diffusion coefficients as a function of the properties of the clay surface and the solution composition. The present study

focuses on the same conceptual model. Fig. 4-9 shows a similar graphical representation as Fig. 4-8, but adds the different qualities of the cations with regards to their mobility. Further, the Stern and the Donnan layers are represented for simplicity as a single domain. The cations in free water (blue charge signs) and the cation swarm in the Donnan layer (green charge signs) are assumed to be mobile, while cationic species in the Stern layer (red charge signs) are immobile surface complexes. The blue domain represents free water with bulk properties where charge neutralisation is fulfilled by solution anions and cations. The green domain represents the Donnan layer (including the Stern layer), a charged solution phase exhibiting the opposite net charge of the clay surface. The local concentration gradients parallel to the clay surface in the different domains result in individual flux contributions (J_i) if there is a concentration difference of a tracer cation species.

This model has been applied successfully to describe diffusion and retardation properties of a wide range of chemically different elements in compacted illite (Glaus et al. 2015a, 2021) and Opalinus Clay (Appelo et al. 2010). The concept of the “Donnan layer” is considered as a simplifying model for the more general “diffuse layer” approach, in which the potential curve is a function of space coordinates (mostly assumed as exponential according to the Poisson-Boltzmann equation (cf. the black dotted line in Fig. 4-9). In the Donnan approach, the potential curve is defined as a homogeneous discrete step function (cf. the red line in Fig. 4-9). The concentration ratios between the species in the diffuse layer and free porewater are determined, among other factors, by the (mean) Donnan potential resulting from the excess free negative surface charges. The overall charge neutralisation is achieved by the balance between the sum of charges of all cations and anions present in the Stern and in the Donnan layer and the sum of permanent negative lattice charges.

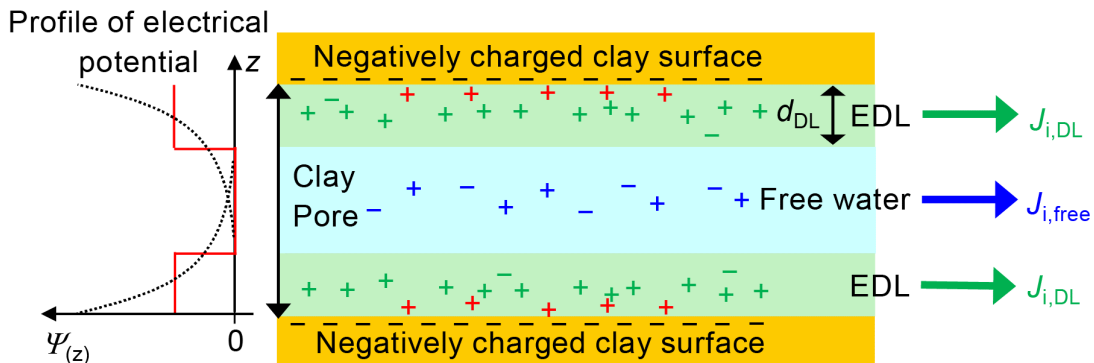


Fig. 4-9 Schematic representation of the Donnan approximation for an electrical double layer (EDL) for a planar pore geometry

Details on the symbols and colours are given in the main text (sketch adapted from Maes et al. 2021).

The main scientific question addressed in this research is whether the EDL model for illite, which was shown to be transferable to the Opalinus Clay in experiments with $^{57}\text{Co}^{2+}$ tracer (Glaus et al. 2021), can generally be applied to sedimentary rocks with variable clay mineralogical compositions, carbonates and other silicates. The EDL model comprises parameters that cannot be directly measured or which may differ in a specific manner for the rock samples from different lithologies, such as the average pore diameter or the thickness of the Donnan (or EDL) layer (d_{DL}) at the surfaces of a clay pore. Another objective of this study is to assess whether generic average parameter values can adequately describe the experimental results. The predictions of the model

primarily involve the effective diffusion coefficient (D_e , [$\text{m}^2 \text{s}^{-1}$]) and the rock capacity factor (α , [–]), which are related to the apparent diffusion coefficient (D_a) by Eq. (1-3), or, after rearranging:

$$D_e = D_a \alpha \quad (4-14)$$

The rock capacity factor, α , can be further expanded according to Eq. (1-4), which allows for secondary information, related to the diffusion behaviour and to the distribution of a diffusing species between the aqueous and the clay phases, to be derived from α . In the case of uncharged tracers such as HTO, it is normally assumed that α equals the total porosity, ε , ($R_d = 0$ in Eq. (1-4)). In the case of anionic species, α may be represented by the “anion-accessible” porosity (ε_{an}) for the assumption of exclusion of anions from parts of the total porosity. “Anion exclusion” is different from the situation shown in Fig. 4-9, where anions are present in both the diffuse layer and the free porosity. A more detailed discussion of anion exclusion effects and their formal treatment in an EDL model can be found in Section 4.2.4 of this document and in Zwahlen et al. (2024).

4.2.3 Model definitions and parametrisation

The EDL model (cf. Fig. 4-9) is an extension of the classic cation-exchange model for the uptake of cations at the planar clay surfaces. There, the negative charges near the planar clay surfaces are completely neutralised by charge-compensating cations (Baeyens & Bradbury 2004, Stumm & Morgan 1996). The EDL extension assumes that only a fraction of the planar clay charges is directly compensated by cations at the surface (Stern layer), leaving several free charges that give rise to electrostatic effects (Appelo et al. 2010). These free charges are counterbalanced by loosely bound cations which are assumed to be mobile, while those in the Stern layer are treated as immobile (Glaus et al. 2020). The Stern layer species are thermodynamically treated as surface complexes, the stability of which is given by a surface-complexation constant and an additional surface-potential term (Appelo & Postma 2005).

The EDL model using a constant Donnan potential in a given volume near the surface (the Donnan layer) has been successfully applied to describe the effects of enhanced diffusion rates of cationic species in charged clays (Glaus et al. 2015a, 2020). D_e is calculated from the superposition of parallel diffusive fluxes in the free porewater and in the Donnan layer as

$$D_e = \frac{\varepsilon}{G} D_w \left(f_{\text{free}} + f_{\text{DL}} \frac{c_{\text{DL}}}{c} q_\eta \right) \quad (4-15)$$

The c_{DL}/c ratio represents the ratio of total concentrations of charged species between the Donnan layer and the free aqueous phase. ε is the total porosity, G [–] the geometrical factor, f_{free} [–] the volume fraction of free porewater, f_{DL} [–] the volume fraction of the Donnan layer, D_w [$\text{m}^2 \text{s}^{-1}$] the diffusion coefficient in bulk (free) water and q_η [–] the ratio of viscosity of free and EDL water. Note that the application of Eq. (4-15) is restricted to a series of prerequisites, such as a fast thermodynamic equilibrium between the Donnan layer and free water species compared to the dynamics of diffusion, and the condition of linear sorption. For the application of Eq. (4-15) to the diffusion data of the charged radioisotopes, it is assumed for the sake of simplicity that G has the same value as measured for HTO, both in the free aqueous pore phase and in the Donnan layer, and that q_η equals 1. For uncharged species, such as HTO, the concentration ratio c_{DL}/c equals 1 and Eq. (4-15) is reduced to Eq. (1-8) because $f_{\text{free}} + f_{\text{DL}} = 1$. Thus, the water-accessible porosity (ε) is equal to the total porosity (ε_{tot}), justifying the use of ε instead of ε_{tot} from now on. For anions, the notation ε_{an} will be employed.

With fixed assumptions for G and q_η as described previously, D_e values depend on all parameters that could affect f_{free} , f_{DL} and the concentration ratio c_{DL}/c . The latter expression is calculated for all charged species from an electrical potential term assuming equal activity coefficients for species in the free aqueous phase and in the Donnan layer according to Eq. (4-13). Ψ_D is obtained by iteratively solving the defining equations comprising (i) the overall charge neutrality for solution and solid and (ii) Eq. (4-13) for all charged species present in the system (Appelo et al. 2010). According to the conventions regarding the signs of charge numbers and the involved potential, cationic species are enriched in the Donnan layer compared to their concentrations in the bulk aqueous phase ($c_{\text{DL}}/c > 1$) while anionic species are depleted ($c_{\text{DL}}/c < 1$). For the present application of the EDL diffusion model, the parameters affecting f_{free} , f_{DL} and the concentration ratio c_{DL}/c can be grouped into different categories. This compilation also provides the derivation of the parameters for the purpose of application to clay rock and the values chosen as default settings.

4.2.3.1 Geometric properties of the pore space

This section is based on the assumption of infinitely long planar pores, restricted to clay domains only. The pore width (d_p [m]) for a planar pore with much longer lateral dimensions compared to thickness is approximately proportional to the ratio of the pore surface area (A_p [m²]) and pore volume (V_p [m³]) and thus to the specific surface area (A_{sp} [m² kg⁻¹]) and the solid:liquid ratio (r_{SL} [kg m⁻³]), according to:

$$d_p = \frac{2V_p}{A_p} = \frac{2}{A_{\text{sp}} \cdot r_{\text{SL}} \cdot f_{\text{clay}}} \quad (4-16)$$

Note that r_{SL} is related to the mass of clay rock per pore volume. The restriction that porosity is present only in clay minerals is realised in Eq. (4-16) by the introduction of the weight fraction of the clay phase (f_{clay}). The Donnan thickness (d_{DL} [m]) is assumed to be a multiple (n_{DL}) of the Debye length ($1/\kappa$), which is, among other factors, a primary function of the ionic strength, cf. Appelo & Postma (2005). Thus:

$$d_{\text{DL}} = \frac{n_{\text{DL}}}{\kappa} \quad (4-17)$$

A commonly used default value for n_{DL} is 2, as established in previous research (Glaus et al. 2015a), but other values can be taken as well (e.g. $n_{\text{DL}} = 4$ in Soler et al. (2019)). Based on the simplified illustration in Fig. 4-9, it is assumed that there are no domains where complete anion-exclusion exists, even though such areas have been identified in dense clay media (Chagneau et al. 2015, Wigger et al. 2018). Therefore, the volume fraction of free porewater f_{free} can be calculated as follows (note that there are two Donnan layers on each side of a planar pore):

$$f_{\text{DL}} = \frac{2d_{\text{DL}}}{d_p} \text{ and } f_{\text{free}} = 1 - f_{\text{DL}} \quad (4-18)$$

For the implementation of the model in PHREEQC (Appelo & Wersin 2007), it is important to prevent situations where $2d_{\text{DL}} > d_p$, as this can result in negative values of f_{free} . To address this issue for such conditions, f_{free} is limited arbitrarily to 0.01 and d_{DL} to $0.99 d_p/2$.

The geometric properties of the rock samples are derived from the same empirical relationships of ε and G on W_c as described in Section 4.1.1 (Eq. (4-2) and Eq. (4-3), Tab. 4-1 and Tab. 4-2).

The bulk-dry density (ρ_{br} [kg m^{-3}]) and r_{SL} were calculated, assuming an average grain (or material) density (ρ_{gr} [kg m^{-3}]) of the solid rock of 2700 [kg m^{-3}], which was chosen as an approximate average from all rock samples obtained from the deep borehole campaign (Fig. B-2 of App. B). Both quantities thus depend on W_c according to the dependence of ε on W_c (cf. Eq. (4-2)):

$$\rho_{br} = \rho_{gr}(1 - \varepsilon) \text{ and } r_{SL} = \frac{\rho_{br}}{\varepsilon} \quad (4-19)$$

A_{sp} is set to 10^5 [$\text{m}^2 \text{kg}^{-1}$ of clay], a typical value for illite, which is, for the sake of simplicity, assumed to be the representative clay mineral in the rock samples. Alternatively, it would also be possible to use a representative value ($\sim 6 \times 10^4$ [$\text{m}^2 \text{kg}^{-1}$ of clay]) of all samples of the deep borehole campaign used for the purpose of diffusion measurements (Fig. B-2 of App. B). This choice is somehow irrelevant because it cannot be resolved based on the choice of the assumptions regarding n_{DL} .

4.2.3.2 Mineralogical properties of the solid

The mean value of cation-exchange capacity (CEC [mol kg^{-1}]) measurements, obtained using both the Ni-ethylenediamine and the Cs isotope dilution methods on the rock samples collected during the deep borehole campaign, has been extrapolated to 100% clay (CEC_{100}). The estimated value of CEC_{100} is approximately 0.21 mol kg^{-1} (as shown in Fig. B-3 of App. B). To facilitate the practical implementation in PHREEQC, the CEC is converted to a volumetric basis (CEC_{vol}) using Eq. (4-20), which expresses CEC as moles of cations per volume of porewater:

$$\text{CEC}_{vol} = \text{CEC}_{100} \cdot f_{clay} \cdot r_{SL} \quad (4-20)$$

4.2.3.3 Chemical properties of the solution and the clay surfaces

Four different compositions of synthetic porewater (SPW) were employed to conduct the diffusion measurements. Their compositions (Tab. 2-1) were derived from the results of advective displacement studies of selected laboratory-scale samples to provide a maximum of representativity for the adjacent layers. The abbreviations in their acronyms correspond to the names of the locations of the drill sites.

The primary aim of using SPWs was to prevent electrolyte concentration gradients between the pore solutions of the rock samples and the free solutions during the phase of tracer through-diffusion measurements. To attain such conditions, the rock samples were pre-equilibrated with the SPWs for sufficient periods before adding the tracer.

Tab. 4-3, as reported by Glaus et al. (2021), provides the surface complexation constants for the Stern layer species. These constants serve as the primary variables that determine the negative excess surface charge and, consequently, the species distribution between the free aqueous phase and the Donnan layer.

Tab. 4-3: Surface complexation constants for the formation of dominant Stern layer species

The constant for H^+ has been omitted because such complexes are hardly formed at the pH of the experiments. The constant for Sr^{2+} has been adjusted to that of Ca^{2+}

Site types	$\log^{Su}K$
Na^+	-0.7 ^a
K^+	-1.0 ^b
Ca^{2+}	0.104 ^b
Mg^{2+}	0.107 ^b

^a Taken from Appelo & Wersin (2007) and references cited therein

^b Adapted values that produce a similar cation loading as a pure cation exchange model (Glaus et al. 2021)

4.2.3.4 Calculation of diffusion and sorption/depletion parameters using the EDL model

For the simulations of D_e values (cf. Eq. (4-15)), the species-specific D_w values ($D_{w,HTO} = 2.2 \times 10^{-9} \text{ m}^2 \text{ s}^{-1}$, $D_{w,Cl} = 2.0 \times 10^{-9} \text{ m}^2 \text{ s}^{-1}$, $D_{w,Na} = 1.3 \times 10^{-9} \text{ m}^2 \text{ s}^{-1}$) are taken from the literature (Woolf 1975, Li & Gregory 1974), while the total porosity (ε) and geometrical factor (G) are derived, as discussed above, from empirical relationships with W_c (cf. Eq. (4-2) and Eq. (4-3)) and the parameter values specified in Tab. 4-1 and Tab. 4-2. The base thermodynamic equilibrium data were obtained from the PSI/Nagra TDB 2020 (Hummel & Thoenen 2023). A summary of all parameters, constants and variables used for the EDL simulations is provided in Tab. B-1 of App. B.

The parameter predictions of the EDL model are not only restricted to D_e values which are derived from the quasi-steady-state flux behaviour in a through-diffusion experiment (Glaus et al. 2015b). The retardation behaviour is characterised by the rock capacity factor α which is related to the R_d value according to Eq. (1-4). The simulation of R_d values for cationic species using the EDL model is based on the definition of R_d as the ratio of the total number of species bound to the clay divided by the total number of species present in solution, normalised for the mass of the clay and volume of the solution. The bracket expression in Eq. (4-15) cannot be directly applied for the calculation of R_d , because the c_{DL}/c ratio only represents the equilibrium conditions of the Donnan layer species. To accurately calculate R_d , the contribution of the Stern layer species must also be considered and included in the calculation. All these characteristics can be obtained directly from the output data of PHREEQC. The calculation of R_d values according to Eq. (1-4) for cationic tracers such as $^{22}Na^+$ further involves the assumption that the accessible porosity of these species equals the accessible porosity of HTO.

Eq. (1-4) is also formally applied within the scope of an anion-exclusion model, in which a fraction of the pore volume is assumed to be completely devoid of anions. The anion exclusion volume is not identical to the Donnan volume because anions distribute both to free porewater and Donnan volume in the EDL model (cf. Fig. 4-9). Nevertheless, the results obtained from the EDL model can be readily converted to an anion exclusion model. With regards to Eq. (4-15) ($q_\eta = 1$), the conversion is achieved by:

$$\alpha = \varepsilon_{an} = f_{free} + f_{DL} \frac{c_{DL}}{c} \quad (4-21)$$

This conversion allows for comparison of the present model to different conceptual approaches to predict Cl-accessible porosity fractions assuming full anion exclusion (Zwahlen et al. 2024). Using the accessible porosity data of the $^{36}\text{Cl}^-$ tracer derived from the EDL model, the respective anion-accessible porosity fractions (f_{an}) were calculated according to Eq. (4-22), where ε_{HTO} denotes the accessible porosity of HTO.

$$f_{\text{an}} = \frac{\varepsilon_{\text{an}}}{\varepsilon_{\text{HTO}}} \quad (4-22)$$

4.2.4 Modelling results and discussion

As shown in Section 3.1, the composition of the porewater has a significant impact on the effective diffusion parameters of $^{36}\text{Cl}^-$ and $^{22}\text{Na}^+$ tracers. For any given type of lithology, the porewater composition has a greater influence on these parameters than the exact location from where the samples were obtained within the siting regions. Consequently, it is considered a practical approach to model the entire set of diffusion data using generic parameters for pore geometry and clay properties, while accounting for the specific composition of the porewater. Therefore, the main focus of the investigations is on the impact of the porewater on the properties of the Donnan layer in terms of Donnan potential and geometric expansions for different clay contents.

As shown in Section 4.2.3.1, the dependence of the pore geometry on the clay content has a pivotal impact on the equilibrium distribution of charged species between the free aqueous phase and the Donnan layer. Fig. 4-10 shows the spatial extension of the Donnan thickness in comparison to the pore width.

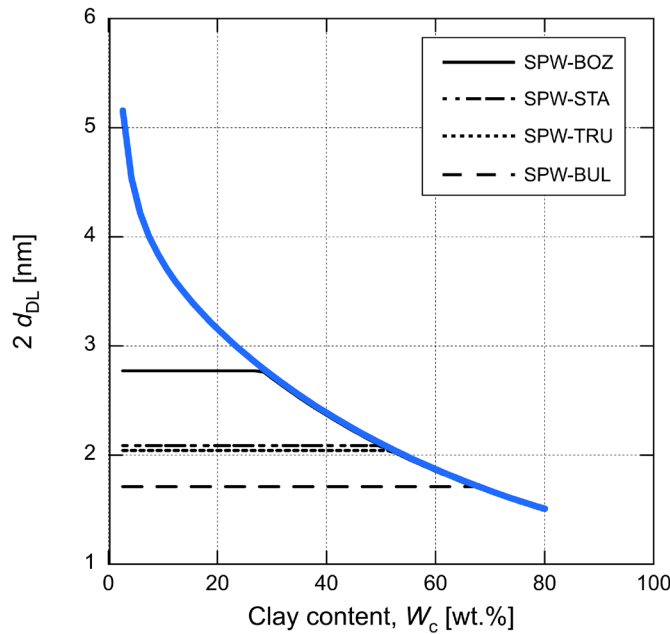


Fig. 4-10: Twice the Donnan thickness ($2d_{\text{DL}}$) as a function of clay content calculated for the various SPWs using the default model settings (black lines)

The blue line represents the pore width (d_p). The points at which the black lines reach the blue line indicate that d_{DL} has been truncated to $0.99 d_p$ according to the condition $2d_{\text{DL}} > d_p$.

The dependence of these two quantities on the total clay content is governed by the assumption that the porosity of the clay rock is limited to the domains containing the clay minerals (cf. Eq. (4-16)). This is clearly an oversimplification of reality, which is made here for the purpose of reducing the model description to a minimum of adjustable parameters. According to Eq. (4-2), the limiting total porosity would tend towards zero for a zero total clay content. This is certainly outside the scope of realistic conditions and for this reason, the model cannot be indiscriminately adapted to such cases. Accordingly, the model curves in Fig. 4-10 and the subsequent diagrams are interrupted arbitrarily at very low total clay contents. As can be seen from Fig. 4-10, the pore diameter decreases with increasing clay content. This trend agrees with the hypothesis in Mazurek et al. (2023) and this work (Section 4.1.1) stating that the clay compaction (and clay platelet orientation) increases with increasing clay content. The Donnan thickness is further governed by the porewater properties, with regard to the ionic strength of the porewater. In order to avoid situations of overlapping double layers, the limitation of $2 d_{DL} > d_p$ has been introduced (cf. Section 4.2.3). At the transition point, at which this condition is reached, the Donnan thickness is truncated to $0.99 d_p/2$. These situations are represented in Fig. 4-10, where the black curves reach the blue line.

Fig. 4-11, Fig. 4-12, and Fig. 4-13 provide a visual comparison between the model predictions (lines) for $^{36}\text{Cl}^-$ and $^{22}\text{Na}^+$ (including those of HTO for completeness) and the experimental data grouped into selected different lithologies. The plots are arranged based on the different porewater compositions used in the experiments, starting with the lowest ionic strength (JO, Fig. 4-11) to the highest ionic strength (NL in Fig. 4-13). The model curves are calculated according to the information given in Section 4.2.3.

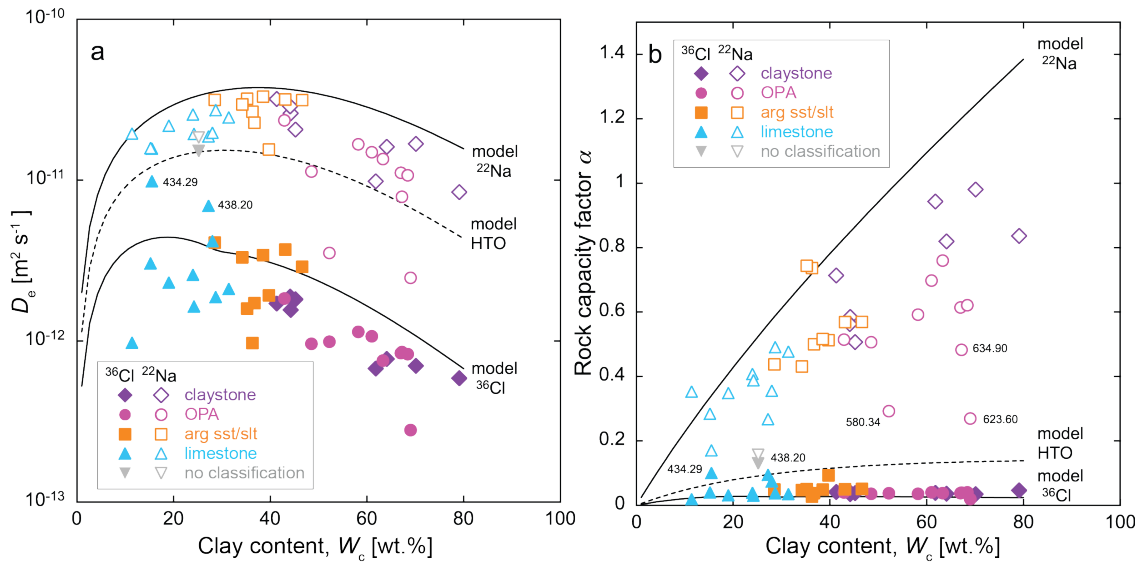


Fig. 4-11: Comparison between experimental data and the EDL simulations involving samples from the JO siting region (Bözberg-1/2) using SPW-BOZ

Note that α equals the anion-accessible porosity for the $^{36}\text{Cl}^-$ tracer (Eq. (4-21); R_d values can be derived from α in the case of the $^{22}\text{Na}^+$ tracer according to Eq. (1-4).

The data from Marthalen-1 and Trüllikon-1 (ZNO) and Stadel-2/3 (NL), which used similar porewater compositions, are shown together in a single plot (Fig. 4-12). Lithologies are categorised as clay-, quartz/feldspar- and carbonate-rich rock types. The clay-rich Opalinus Clay is represented by a lighter variant of violet. No experimental uncertainties are shown in these plots

for clarity reasons. The uncertainties on the best-fit parameter values were evaluated in the course of the Matlab parameter optimisation routine on a 95% confidence level (Section 2.8). For most tracers, the uncertainties on the values of the effective diffusion coefficients, D_e , are below 10%. The calculated relative uncertainties on the α values (accessible porosities) of HTO are in the order of less than 10%, while these are significantly larger in the case of $^{36}\text{Cl}^-$ (average of $\sim 40\%$, single cases up to almost 100%). The calculated uncertainties on the R_d values of $^{22}\text{Na}^+$ are in the order of 10 – 15%. Any measurement data that deviate significantly from the model are marked for the discussion by their sample identity (drilling depth). Note that the α values for HTO represent the diffusion-accessible porosity which is commonly assumed to be identical to the total porosity.

In terms of providing an overall description of the average system behaviour, the model predictions demonstrate a satisfactory agreement with the experimental data, although certain biases are observed in various aspects. This reaffirms the suitability of the fundamental data for illite to be applied to the Opalinus Clay, as previously established in earlier research (Glaus et al. 2021). The choice of the model parameters does not have to be adjusted specifically for the rock samples investigated. The model can also be applied to a broad range of other formations and lithologies within the Mesozoic sediments of Northern Switzerland where diffusive transport dominates. There, it is even applicable at low clay contents. This is unexpected as the model only considers clay-mineral properties and does not take into account other mineral constituents like carbonates or other silicates. However, as demonstrated earlier, the diffusion data (D_e values) of the clay- and quartz-rich rock samples exhibit smaller variability than those of the carbonate-rich samples. This is also evident in Fig. 4-12 that shows the diffusion data from the Marthalen and Stadel boreholes, where the focus of the sample analysis was on the over- and underlying geological units rather than on the Opalinus Clay formation.

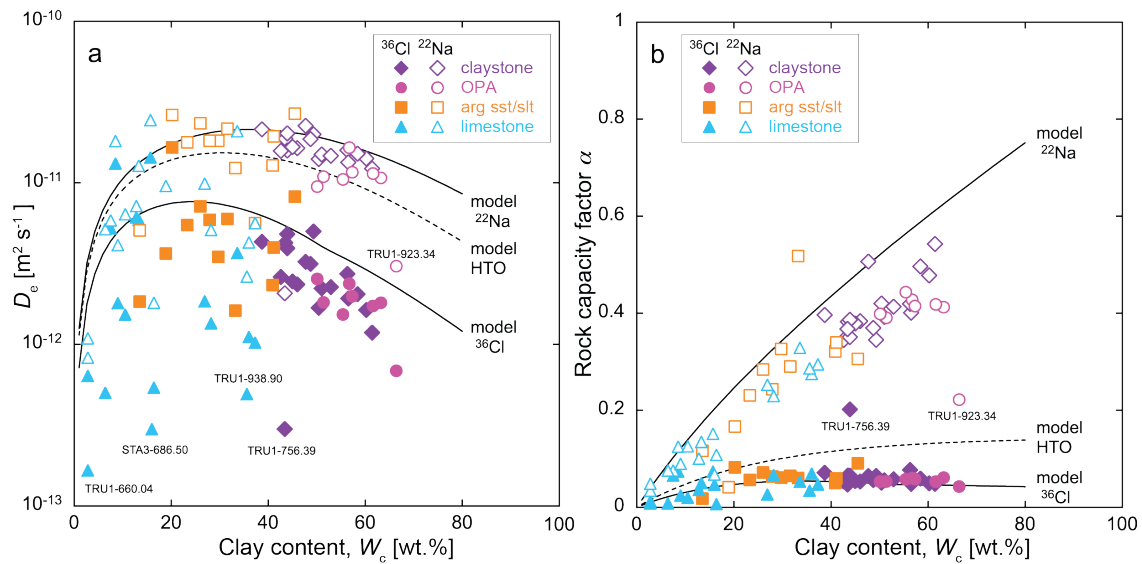


Fig. 4-12: Comparison between experimental data and the EDL simulations involving samples from NL (Stadel-2/3) and ZNO (Trüllikon-1, Marthalen-1) using SPW-STA and SPW-TRU, respectively

No systematic differences could be found for the diffusion behaviour of samples of the different siting regions. Note that α equals the anion-accessible porosity for the $^{36}\text{Cl}^-$ tracer (Eq. (4-21)); R_d values can be derived from α in the case of the $^{22}\text{Na}^+$ tracer according to Eq. (1-4).

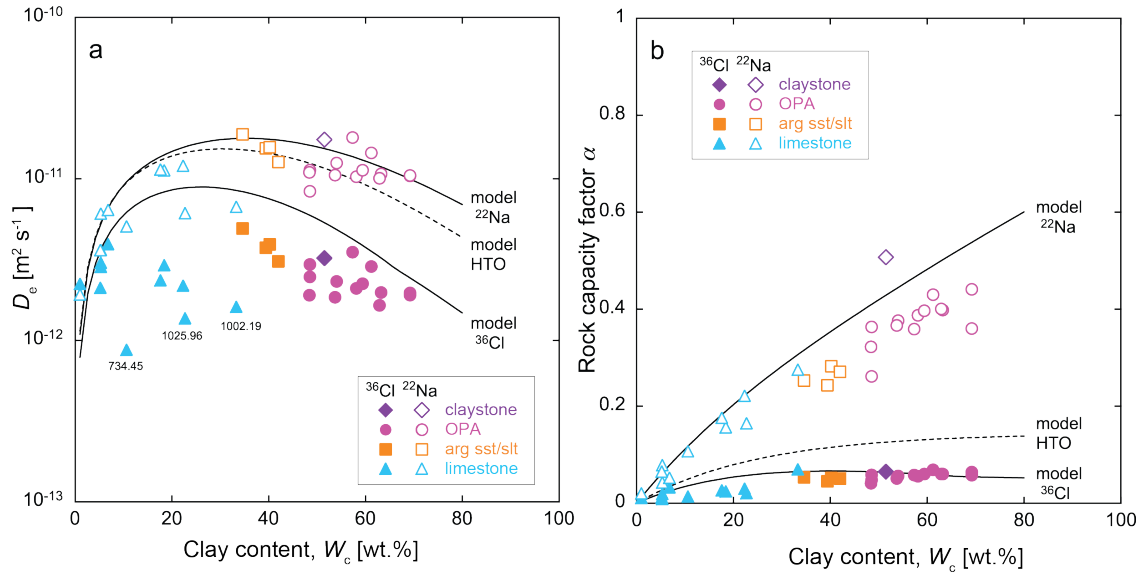


Fig. 4-13: Comparison between experimental data and the EDL simulations involving samples from NL (only Bülach-1) using SPW-BUL

Note that α equals the anion-accessible porosity for the ³⁶Cl⁻ tracer (Eq. 4-21); R_d values can be derived from α in the case of the ²²Na⁺ tracer according to Eq. 1-4.

Fig. 4-11, Fig. 4-12, and Fig. 4-13 demonstrate the impact of surface diffusion, which is active in the case of ²²Na⁺. This can be seen by comparing the model curves for ²²Na⁺ and HTO. The effect decreases as ionic strength increases. It should be noted that the diffusion coefficient of HTO in free water is larger (2.2×10^{-9} m² s⁻¹) than that of ²²Na⁺ (1.3×10^{-9} m² s⁻¹). To show an unbiased effect of surface diffusion, the model curves should be normalised by the ratio of these free water-diffusion coefficients. The reason for increased model discrepancies in some experimental data is intrinsic inconsistencies of the underlying diffusion measurements. This is the case in the marked ²²Na⁺ cases shown in Fig. 4-11, Fig. 4-12 and Fig. 4-13 by specifying the drilling depth. In some cases, a significant mass discrepancy between the tracer depletion in the source reservoir and the mass transferred to the target reservoir and clay was noted. The reason for this issue could not be resolved.

As expected, the diffusion data of ³⁶Cl⁻ exhibit the opposite trend, showing an increasing alignment to the D_e values of HTO with increasing ionic strength. The ³⁶Cl⁻ results marked by depth in Fig. 4-11 are not due to experimental inconsistencies, but rather to samples with a high content of goethite. This raises a broader discussion of the α values for anions, which are commonly assumed to be non-sorbing species, as to whether the observed α values for anions are representative of the anion-accessible porosity. However, it has been established that anions may sorb significantly on clay minerals, in particular on kaolinite and, to a lesser extent, on illite (Chen et al. 2018). Although the underlying mechanism is not fully understood, anions can sorb via electrostatic forces on protonated aluminol sites, which are abundant on the oxidic surfaces of kaolinite. Sorption of anions increases as the pH decreases. As a result, the α values will be higher than the anion-accessible porosity in such cases. A high content of goethite (or other iron oxides) in clay rock samples could particularly enhance anion sorption.

The elevated discrepancies between experimental data and model prediction for other cases of ³⁶Cl⁻ results as shown in Fig. 4-12 and Fig. 4-13 are not due to obvious experimental inconsistencies or distinct differences in rock properties. The geometrical factors are notably high in some

carbonate-rich rock samples and in some shales with high organic carbon content. However, no generic correlation can be found between the organic carbon content and the geometrical factors for HTO (Fig. B-1 of App. B).

In some cases (Fig. 4-11a, Fig. 4-12a, and Fig. 4-13a), the experimental effective diffusion coefficients of $^{36}\text{Cl}^-$ exhibit more significant outliers than those of $^{22}\text{Na}^+$. The significance of this difference can be evaluated using statistical methods, such as the chi-square analysis. However, a quantitative statistical analysis of the data deviations from the model curve may not be informative given the nature of the model curves representing near-blind predictions instead of error-minimised best fits. Additionally, the logarithmic scale of the representation of the D_e values in Fig. 4-11a, Fig. 4-12a, and Fig. 4-13a can be misleading as negative errors can appear larger than positive ones. To avoid bias, Fig. 4-14a shows a representation of the relative residuals (Eq. (4-23)) on absolute, rather than logarithmic scale.

$$\Delta_r y = \frac{y_{exp} - y_{mod}}{y_{mod}} \quad (4-23)$$

In the present case, $\Delta_r y$ represents the relative residuals of D_e values with subscript *exp* denoting the experimental values and *mod* the model values. A $\Delta_r y$ of 1 thus denotes a deviation of 100%. Fig. 4-14a indicates that the deviations from the model are approximately similar for all tracers, with a tendency towards lower deviations for clay contents exceeding $\sim 50\%$. Fig. 4-14b shows the relationship between the relative residuals of D_e values of the charged tracers as a function of the relative residuals of HTO, revealing correlated datasets with similar slopes. This finding suggests that bracket expression inadequacies in Eq. (4-15) are not the primary reason for the model discrepancies. The assumption of an average pore width, which may be biased, and the generalisation that porosity is only linked to clay minerals can contribute to these inadequacies. Zwahlen et al. (2024) provide a more detailed discussion of this topic.

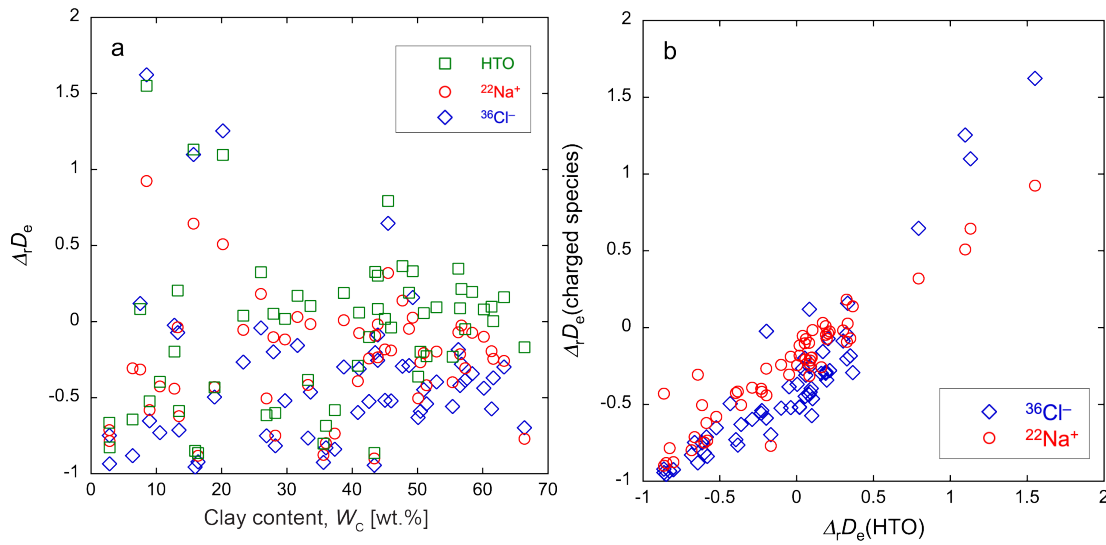


Fig. 4-14: Representation of relative residuals (Eq. (4-23)) of effective diffusion D_e values shown in Fig. 4-12a (left-hand plot) and correlation between the relative residuals of charged species as a function of the relative residuals for HTO (right-hand plot)

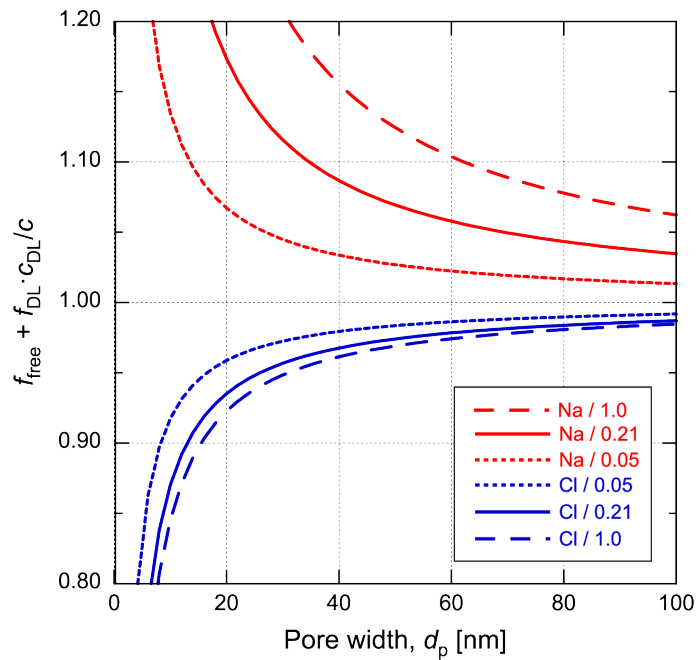


Fig. 4-15: Enhancement/depletion factors (bracket expression in Eq. (4-15)) for Na^+ and Cl^- as a function of pore width

The values in the figure legend denote the number of sites (mol kg⁻¹). In agreement with the simulations shown in Fig. 4-11, Fig. 4-12, and Fig. 4-13, q_η is assumed to take a value of 1.

Fig. 4-15 shows the dependence of the bracket expression of Eq. (4-15), the “concentration enhancement/depletion factor”, as a function of pore width, which is calculated using simple average values based on Eq. (4-16). If non-clay minerals constitute a significant portion of the porosity, this equation may not be suitable to adequately evaluate concentration enhancement/depletion effects. Despite the opposite behaviour of the model curves of $^{36}\text{Cl}^-$ and the $^{22}\text{Na}^+$ tracer, Fig. 4-14b shows an almost identical correlation. Consequently, for the majority of the data, it can be concluded that the geometric effects rather than the “chemical contributions” are the main reasons for the deviations between experimental data and model curves. Also note that the behaviour of positively and negatively charged species in Fig. 4-14b is not precisely symmetrical. Nevertheless, these nuances are lost in the context of the typical data uncertainties and the minor role of the electrostatic factors to the overall uncertainties.

The sole systematic deviation in the model is an overestimation of the rock capacity factors for the $^{22}\text{Na}^+$ data. This can be attributed to an overestimation of the occupancy of the planar clay surfaces with Na^+ by the thermodynamic data shown in Tab. 4-3, which was derived from illite data. The formal cation selectivities, obtained by summing up the amounts of cationic species present in the Stern layer species and those in the Donnan layer, are not in full agreement with the data shown in Tab. 4-3. However, generic values are still used for the modelling and in view of the tolerable model discrepancies, no further adjustment of those thermodynamic base data is made.

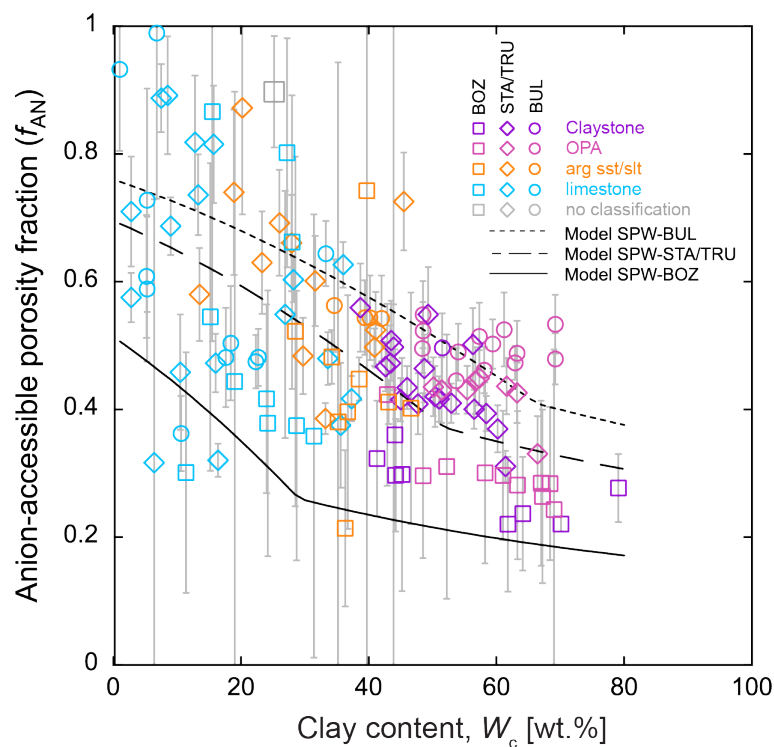


Fig. 4-16: Anion-accessible porosity fraction for $^{36}\text{Cl}^-$ versus clay content showing the data of all siting regions and the modelled output due to the different SPW compositions

The data related to SPW-BUL (highest ionic strengths) are shown as circles, those related to SPW-STA and SPW-TRU as diamonds, and those to SPW-BOZ (lowest ionic strength) as squares.

An important parameter in evaluating palaeogeological stable-isotope concentration profiles (Aschwanden et al. 2024, Wersin et al. 2023) is the porosity fraction accessible for different anions, which is shown for $^{36}\text{Cl}^-$ in Fig. 4-16 along with experimental uncertainties for all siting regions. The data are calculated according to Eq. (4-21) and Eq. (4-22). The propagated experimental uncertainties are quite large in many cases due to the high experimental uncertainties related to the anion-accessible porosity for $^{36}\text{Cl}^-$. The effect of porewater composition on f_{an} is clearly visible in the clay-rich rock samples, which have less experimental uncertainty and are very well represented by the model curves. The distinction becomes less noticeable in samples with low clay contents, however, most of the data agree with the model predictions despite the large uncertainties generally involved. Fig. 4-16 highlights that the model assumptions considered in the simulations of palaeogeological isotope profiles of charged species must take into account the composition of the in-situ porewaters such as the ionic strength.

Zwahlen et al. (2024) present an empirical model that relates the Cl-accessible porosity fraction, derived from various approaches, to the clay fraction. At clay contents $> 30\%$, the modelled accessibilities are close to constant at approximately 0.32 for samples from the JO siting region (BOZ) and at 0.47 for the ZNO and NL siting regions. This difference is attributed to the significantly lower ionic strength of the in-situ porewater of JO compared to that of the two other siting regions.

5 Conclusions

This study investigated the diffusion properties of HTO, $^{36}\text{Cl}^-$ and $^{22}\text{Na}^+$ in 130 samples with varying mineral compositions collected from Nagra's recent TBO campaign characterising the Mesozoic sediment sequence of Northern Switzerland. Through-diffusion experiments were conducted to calculate effective diffusion coefficients and diffusion-accessible porosities. The results showed that all D_e HTO data of the Opalinus Clay are within a factor of two (average $D_e = 8.8 \pm 1.9 \times 10^{-12} \text{ m}^2 \text{ s}^{-1}$) and thus have a very narrow range. The Opalinus Clay appears homogenous in terms of effective diffusion coefficients and accessible porosities ($\varepsilon = 0.12 \pm 0.02$). In contrast, the diffusion characteristics of the over- and underlying rock units, between the Malm and Keuper/Triassic aquifers, exhibit a wider range (considering the effective diffusion coefficient (D_e) and the porosities available for HTO, which are independent of the chemical composition of the porewater). The variance reflects the more diverse nature of sediment origin, depositional processes, and diagenetic alterations, which collectively contribute to the heterogeneity in these rock formations.

Anion-exclusion effects led to generally smaller effective diffusion coefficients and accessible porosities of anions like $^{36}\text{Cl}^-$. Cations such as $^{22}\text{Na}^+$ showed larger effective diffusion coefficients due to surface-enhanced diffusion. The extent of anion-exclusion and surface-diffusion effects depends on the composition of the porewater and, in the case of cations, the extent of sorption, which depends also on the sorption capacity of the solid. As a result, differences in the effective diffusion coefficient of $^{36}\text{Cl}^-$ and $^{22}\text{Na}^+$ between different areas can be observed due to the different chemical compositions of the porewaters in contact with the solid phase.

Comparison of the results for the Opalinus Clay from BOZ1-1 (where the porewater has the lowest ionic strength), TRU1-1 (where the porewater has an intermediate ionic strength) and BUL1-1 (where the porewater has the highest ionic strength) reveals that the average effective diffusion coefficient of $^{36}\text{Cl}^-$ is lower than the one of HTO by a factor of 9.1 ± 1.4 in the case of BOZ1-1, 5.6 ± 1.4 in the case of TRU1-1, and 4.1 ± 0.5 in the case of BUL1-1. The average effective diffusion coefficient of $^{22}\text{Na}^+$ is, after normalisation for the respective diffusion coefficients in bulk water, a factor of 2.0 ± 0.8 larger than the one of HTO in BOZ1-1 and 1.9 ± 0.1 and 1.7 ± 0.4 larger in BUL1-1 and TRU1-1, respectively. Anion diffusion is more sensitive to changes in water composition than cation diffusion.

A novel type of correlation was established between the effective diffusion coefficients and the total clay content of the samples. The results suggest the possibility of developing procedures for calculating effective diffusion coefficients for units with varying clay content. However, in samples with low clay content, the model and derivation of its dependency will require further investigations. This approach differs from the diffusion correlations (to porosity) in Stage 1 and Stage 2 of the Sectoral Plan (Van Loon 2014). However, despite the different approach of calculating the effective diffusion coefficients, the overall differences between the values of the diffusion parameters are relatively small (see Glaus et al. 2024a).

An appropriate prediction of the effective diffusion coefficients and rock capacity factors for the $^{22}\text{Na}^+$ and $^{36}\text{Cl}^-$ tracers was achieved using an electrostatic double layer (EDL) model in which the assumption is made that the cationic species in the double layer near the planar clay surfaces are mobile and also contribute to overall diffusive fluxes. The EDL model's prediction for these tracers shows remarkable consistency with the experimental diffusion data. The settings for the model were previously used for illite systems and found to be transferable to the Opalinus Clay (i.e. using similar values for the parameter settings in terms of CEC and specific surface area) (Glaus et al. 2021). The settings are applicable also for other types of argillaceous rocks in which illite is not exclusively the dominating clay mineral. The robustness of the model predictions for these different types of lithologies and formations, all of which show diffusion as the dominant

transport mode, is very important for a generic application of the EDL diffusion model to predict diffusion parameters for elements for which no experimental data exist. Despite a slight bias in the Na^+ sorption data, which is mainly due to an overestimation of the population of Na^+ in the Stern layer sites, the model's performance is not significantly impaired. A bias in the population of the Stern layer species is not critical for the prediction of the sorption properties of strongly sorbing radioelements, such as transition metals, lanthanides or actinides. Surface complexes at the clay-edge surfaces are the dominant sorbing species for these elements at trace conditions and overrule the contribution from Stern layer species. These surface complexes are not relevant with respect to surface diffusion because they are treated as immobile. Previous work (Glaus et al. 2015a, 2020) has demonstrated the model's applicability to a broad variety of cations with different chemical behaviours in clay minerals, and this work further supports the EDL model's capability to predict the diffusion behaviour of charged species in different sedimentary rocks.

The diffusion-analysis comparison of the three different siting regions demonstrates that accurately describing the diffusion and retardation properties of charged species in sedimentary rocks relies heavily on the chemical composition of the porewater. The clay-rock samples used in the experiment were pre-equilibrated with synthetic porewaters that represent an average in-situ porewater of the Opalinus Clay (as determined by porewater extraction of the Opalinus Clay samples). However, it is important to note that this porewater composition may not be identical to the in-situ conditions and that the ionic strength of the porewater can impact the effective diffusion coefficients (i.e. ratio of mobile surface species versus mobile aqueous species). The significance of ionic strength is evident in the spread between the model curves for $^{36}\text{Cl}^-$ and $^{22}\text{Na}^+$, which decreases with increasing ionic strength (e.g., Jura Ost versus Nördlich Lägern). Interestingly, the porewater composition of SPW-STA and SPW-TRU differ significantly in terms of the content of bivalent alkaline earth cations and sulphate and the monovalent Na^+ and Cl^- . Nonetheless, the calculated Donnan potential for these two porewaters is very similar, indicating that all types of charged solutes are important for the resulting enrichment or depletion factors between the Donnan layer and the bulk aqueous phase. Therefore, the ionic strength appears to be an ideal indicator for the description of the global effect of all cations and anions.

6 References

- Aertsens, M., Van Gompel, M., De Cannière, P., Maes, N. & Dierckx, A. (2008): Vertical distribution of $\text{H}^{14}\text{CO}_3^-$ transport parameters in Boom Clay in the Mol-1 borehole (Mol, Belgium). *Physics and Chemistry of the Earth, Parts A/B/C* 33, S61-S66. DOI: 10.1016/j.pce.2008.10.060.
- Aldaba, D., Glaus, M., Leupin, O.X., Van Loon, L.R., Vidal, M. & Rigol, A. (2014): Suitability of various materials for porous filters in diffusion experiments. *Radiochimica Acta* 102, 723-730, available online at <https://api.semanticscholar.org/CorpusID:101745859>.
- Appelo, C.A.J. & Postma, D. (2005): *Geochemistry, groundwater and pollution*. 2nd edition. CRC Press, London.
- Appelo, C.A.J., Van Loon, L.R. & Wersin, P. (2010): Multicomponent diffusion of a suite of tracers (HTO, Cl, Br, I, Na, Sr, Cs) in a single sample of Opalinus Clay. *Geochimica et Cosmochimica Acta* 74/4, 1201-1219. DOI: 10.1016/j.gca.2009.11.013.
- Appelo, C.A.J. & Wersin, P. (2007): Multicomponent diffusion modeling in clay systems with application to the diffusion of tritium, iodide, and sodium in Opalinus Clay. *Environmental Science & Technology* 41/14, 5002-5007.
- Aschwanden, L., Camesi, L., Gaucher, E., Gimmi, T., Jenni, A., Kiczka, M., Mäder, U., Mazurek, M., Rufer, D., Waber, H.N., Wersin, P., Zwahlen, C. & Traber, D. (2022): TBO Stadel-3-1: Data report Dossier VIII Rock properties, porewater characterisation and natural tracer profiles. Nagra Arbeitsbericht NAB 22-01 VIII.
- Aschwanden, L., Camesi, L., Gimmi, T., Jenni, A., Kiczka, M., Mäder, U., Mazurek, M., Rufer, D., Waber, H.N., Wersin, P., Zwahlen, C. & Traber, D. (2021): TBO Trüllikon-1-1: Data report dossier VIII Rock properties, porewater characterisation and natural tracer profiles. Nagra Arbeitsbericht NAB 20-09 VIII.
- Aschwanden, L., Waber, H.N., Eichinger, F. & Gimmi, T. (2024): Isotope diffusive exchange experiments for deriving porewater isotope composition in low-permeability rocks – Improvements in experimental procedure and data processing. *Applied Geochemistry* 160, 105844. DOI: 10.1016/j.apgeochem.2023.105844.
- Baeyens, B. & Bradbury, M.H. (2004): Cation exchange capacity measurements on illite using the sodium and cesium isotope dilution technique: effects of the index cation, electrolyte concentration and competition: modeling. *Clays and Clay Minerals* 52/4, 421-431.
- Bazer-Bachi F., Tevissen E., Descostes M., Grenut B., Meier P., Simonnot M.-O. & Sardin, M. (2006): Characterization of iodide retention on Callovo-Oxfordian argillites and its influence on iodide migration. Geckeis H.; Savenije H.H.G.: Migration '05: Chemistry and migration behaviour of actinides and fission products in the geosphere: 10th international conference, Avignon, France, September 18-23, 2005.- *Physics and Chemistry of the Earth* 31/41913, 517-522.
- Bestel, M., Glaus, M.A., Frick, S., Gimmi, T., Juranyi, F., Van Loon, L.R. & Diamond, L.W. (2018): Combined tracer through-diffusion of HTO and ^{22}Na through Na-montmorillonite with different bulk dry densities. *Applied Geochemistry* 93, 158-166. DOI: 10.1016/j.apgeochem.2018.04.008.

- Bollermann, T., Yuan, T., Kulenkampff, J., Stumpf, T. & Fischer, C. (2022): Pore network and solute flux pattern analysis towards improved predictability of diffusive transport in argillaceous host rocks. *Chemical Geology* 606, 120997. DOI: 10.1016/j.chemgeo.2022.120997.
- Bossart, P. & Thury, M. (2007): Research in the Mont Terri Rock Laboratory: Quo vadis? Physics and Chemistry of the Earth: 2nd international meeting on clays in natural and engineered barriers for radioactive waste confinement, Tours (France), 14-18th March 2005. Part 1, 32, 19-31.
- Bourg, I.C. (2015): Sealing shales versus brittle shales: A sharp threshold in the material properties and energy technology uses of fine-grained sedimentary rocks. *Environmental Science & Technology Letters* 2/10, 255-259. DOI: 10.1021/acs.estlett.5b00233.
- Bourg, I.C. & Ajo-Franklin, J.B. (2017): Clay, water, and salt: Controls on the permeability of fine-grained sedimentary rocks. *Accounts of Chemical Research* 50/9, 2067-2074. DOI: 10.1021/acs.accounts.7b00261.
- Boving, T.B. & Grathwohl, P. (2001): Tracer diffusion coefficients in sedimentary rocks: correlation to porosity and hydraulic conductivity. *Journal of Contaminant Hydrology* 53/1-2, 85-100.
- Cavé, L., Al, T., Xiang, Y. & Vilks, P. (2009): A technique for estimating one-dimensional diffusion coefficients in low-permeability sedimentary rock using X-ray radiography: Comparison with through-diffusion measurements. *Journal of Contaminant Hydrology* 103/1, 1-12. DOI: 10.1016/j.jconhyd.2008.08.001.
- Chagneau, A., Tournassat, C., Steefel, C.I., Bourg, I.C., Gaboreau, S., Esteve, I., Kupcik, T., Claret, F. & Schäfer, T. (2015): Complete restriction of $^{36}\text{Cl}^-$ diffusion by celestite precipitation in densely compacted illite. *Environmental Science and Technology Letters* 2/5, 139-143.
- Chen, Y., Glaus, M.A., Van Loon, L.R. & Mäder, U. (2018): Transport of low molecular weight organic compounds in compacted illite and kaolinite. *Chemosphere* 198, 226-237. DOI: 10.1016/j.chemosphere.2018.01.137.
- Crawford, B.R., Faulkner, D.R. & Rutter, E.H. (2008): Strength, porosity, and permeability development during hydrostatic and shear loading of synthetic quartz-clay fault gouge. *Journal of Geophysical Research* 113, B03207, doi:10.1029/2006JB004634.
- Dabat, T., Hubert, F., Paineau, E., Launois, P., Laforest, C., Grégoire, B., Dazas, B., Tertre, E., Delville, A. & Ferrage, E. (2019): A general orientation distribution function for clay-rich media. *Nature Communications* 10/1, 5456. DOI: 10.1038/s41467-019-13401-0.
- Dabat, T., Porion, P., Hubert, F., Paineau, E., Dazas, B., Grégoire, B., Tertre, E., Delville, A. & Ferrage, E. (2020): Influence of preferred orientation of clay particles on the diffusion of water in kaolinite porous media at constant porosity. *Applied Clay Science* 184, 105354. DOI: 10.1016/j.clay.2019.105354.
- Descostes, M., Blin, V., Bazer-Bachi, F., Meier, P., Grenut, B., Radwan, J., Schlegel, M.L., Buschaert, S., Coelho, D. & Tevissen, E. (2008): Diffusion of anionic species in Callovo-Oxfordian argillites and Oxfordian limestones (Meuse/Haute-Marne, France). *Applied Geochemistry* 23/4, 655-677.

- Dufey, J.E. & Laudelout, H.G. (1975): Self-diffusion of sodium on clay surfaces as influenced by two other alkali cations. *Journal of Colloid and Interface Science* 52/2, 340-344. DOI: 10.1016/0021-9797(75)90208-8.
- Dutt, G.R. & Low, P.F. (1962): Diffusion of alkali chlorides in clay-water systems. *Soil Science* 93, 233-240, available online at <https://api.semanticscholar.org/CorpusID:92949623>.
- Ferrage, E., Hubert, F., Dabat, T., Asaad, A., Dazas, B., Grégoire, B., Savoye, S. & Tertre, E. (2023): Anisotropy in particle orientation controls water diffusion in clay materials. *Applied Clay Science* 244, 107117. DOI: 10.1016/j.clay.2023.107117.
- Füchtbauer H. (1988): *Sedimente und Sedimentgesteine*. 4. Aufl. Sediment-Petrologie, Teil II. Schweizerbart, Stuttgart.
- Gimmi, T., Aschwanden, L., Camesi, L., Gaucher, E., Jenni, A., Kiczka, M., Mäder, U., Mazurek, M., Rufer, D., Waber, H.N., Wersin, P., Zwahlen, C. & Traber, D. (2022): TBO Bözberg-2-1: Data Report Dossier VIII Rock Properties, Porewater Characterisation and Natural Tracer Profiles. Nagra Arbeitsbericht NAB 21-22 VIII.
- Gimmi, T. & Kosakowski, G. (2011): How mobile are sorbed cations in clays and clay rocks? *Environmental Science & Technology* 45/4, 1443-1449. DOI: 10.1021/es1027794.
- Gimmi, T., Leupin, O.X., Eikenberg, J., Glaus, M.A., Van Loon, L.R., Waber, H.N., Wersin, P., Wang, H., Grolimund, D., Borca, C.N., Dewonck, S. & Wittebroodt, C. (2014): Anisotropic diffusion at the field scale in a 4-year multi-tracer diffusion and retention experiment – I: Insights from the experimental data. *Geochimica et Cosmochimica Acta* 125, 373-393.
- Gimmi, T. & Waber, H.N. (2004): Modelling of tracer profiles in pore water of argillaceous rocks in the Benken borehole: Stable water isotopes, chloride, and chlorine isotopes. Nagra Technical Report NTB 04-05.
- Glaus, M.A., Aertsens, M., Appelo, C.A.J., Kupcik, T., Maes, N., Van Laer, L. & Van Loon, L.R. (2015a): Cation diffusion in the electrical double layer enhances the mass transfer rates for Sr^{2+} , Co^{2+} and Zn^{2+} in compacted illite. *Geochimica et Cosmochimica Acta* 165, 376-388.
- Glaus, M.A., Aertsens, M., Maes, N., Van Laer, L. & Van Loon, L.R. (2015b): Treatment of boundary conditions in through-diffusion: A case study of $^{85}\text{Sr}^{2+}$ diffusion in compacted illite. *Journal of Contaminant Hydrology* 177-178, 239-248. DOI: 10.1016/j.jconhyd.2015.03.010.
- Glaus, M.A., Baeyens, B., Bradbury, M.H., Jakob, A., Van Loon, L.R. & Yaroshchuk, A. (2007): Diffusion of $^{22}\text{Na}^{+}$ and $^{85}\text{Sr}^{2+}$ in montmorillonite: Evidence of interlayer diffusion being the dominant pathway at high compaction. *Environmental Science & Technology* 41/2, 478-485.
- Glaus, M.A., Birgersson, M., Karnland, O. & Van Loon, L.R. (2013): Seeming steady-state uphill diffusion of $^{22}\text{Na}^{+}$ in compacted montmorillonite. *Environmental Science & Technology* 47/20, 11522-11527.
- Glaus, M.A., Frick, S., Rossé, R. & Van Loon, L.R. (2010): Comparative study of tracer diffusion of HTO, $^{22}\text{Na}^{+}$ and $^{36}\text{Cl}^{-}$ in compacted kaolinite, illite and montmorillonite. *Geochimica et Cosmochimica Acta* 74/7, 1999-2010.

- Glaus, M.A., Frick, S. & Van Loon, L.R. (2017): Diffusion of selected cations and anions in compacted montmorillonite and bentonite. PSI Bericht Nr. 17-08. PSI, Villigen.
- Glaus, M.A., Frick, S. & Van Loon, L.R. (2020): A coherent approach for cation surface diffusion in clay minerals and cation sorption models: Diffusion of Cs^+ and Eu^{3+} in compacted illite as case examples. *Geochimica et Cosmochimica Acta* 274, 79-96. DOI: 10.1016/j.gca.2020.01.054.
- Glaus, M.A., Frick, S. & Van Loon, L.R. (2021): Competitive effects of cations on the diffusion properties of strongly sorbing trace cations in compacted illite and Opalinus Clay. *ACS Earth and Space Chemistry*. 5/10, 2621-2625. DOI: 10.1021/acsearthspacechem.1c00250.
- Glaus, M.A., Kulik, D.A., Miron, G.D., Van Loon, L.R., Wüst, R.A.J., Becker, J. & Li, X. (2024a): Diffusion databases for Opalinus Clay, confining geological units, and bentonite: Methods, concepts, and upscaling of data. Nagra Technical Report NTB 23-08.
- Glaus, M.A., Rossé, R., Van Loon, L.R. & Yaroshchuk, A.E. (2008): Tracer diffusion in sintered stainless steel filters: Measurement of effective diffusion coefficients and implications for diffusion studies with compacted clays. *Clays and Clay Minerals* 56/6, 677-685. DOI: 10.1346/CCMN.2008.0560608.
- Glaus, M.A., Van Loon, L.R. & Wüst, R.A.J. (2024b): Diffusion of HTO, ^{36}Cl and ^{22}Na in the Mesozoic rocks of northern Switzerland. II: Data interpretation in terms of an electrical double layer model. *Applied Geochemistry* 162, 105842. DOI: 10.1016/j.apgeochem.2023.105842.
- González Sánchez, F., Jurányi, F., Gimmi, T., Van Loon, L.R., Unruh T. & Diamond, L.W. (2008): Translational diffusion of water and its dependence on temperature in charged and uncharged clays: A neutron scattering study. *Journal of Chemical Physics* 129, 174706 1-11.
- Houben, M.E., Desbois, G. & Urai, J.L. (2014): A comparative study of representative 2D microstructures in shaly and sandy facies of Opalinus Clay (Mont Terri, Switzerland) inferred from BIB-SEM and MIP methods. *Marine and Petroleum Geology* 49, 143-161.
- Hummel, W. & Thoenen, T. (2023): The PSI Chemical Thermodynamic Database 2020. Nagra Technical Report NTB 21-03.
- Jacops, E., Aertsens, M., Maes, N., Bruggeman, C., Swennen, R., Krooss, B., Amann-Hildenbrand, A. & Littke, R. (2017): The dependency of diffusion coefficients and geometric factor on the size of the diffusing molecule: Observations for different clay-based materials. *Geofluids* 2017, 8652560. DOI: 10.1155/2017/8652560.
- Jensen, D.J. & Radke, C.J. (1988): Caesium and strontium diffusion through sodium montmorillonite at elevated temperature. *Journal of Soil Science* 39/1, 53-64. DOI: 10.1111/j.1365-2389.1988.tb01193.x.
- Jordan, P. & Deplazes, G. (2019): Lithostratigraphy of consolidated rocks expected in the Jura Ost, Nördlich Lägern and Zürich Nordost regions. Nagra Arbeitsbericht NAB 19-14.
- Joseph, C., Van Loon, L.R., Jakob, A., Steudtner, R., Schmeide, K., Sachs, S. & Bernhard, G. (2013): Diffusion of U(VI) in Opalinus Clay: Influence of temperature and humic acid. *Geochimica et Cosmochimica Acta* 109, 74-89.

- Kaplan, U. (2013): Speziation von Plutonium bei der Sorption und Diffusion in Opalinuston. PhD Thesis. Johannes Gutenberg-Universität Mainz.
- Krejci, P., Gimmi, T. & Van Loon, L.R. (2021): On the concentration-dependent diffusion of sorbed cesium in Opalinus Clay. *Geochimica et Cosmochimica Acta* 298, 149-166. DOI: 10.1016/j.gca.2021.01.012.
- Lash, G.G. & Blood, D.R. (2004): Origin of shale fabric by mechanical compaction of flocculated clay: Evidence from the Upper Devonian Rhinestreet Shale, Western New York, U.S.A. *Journal of Sedimentary Research* 74/1, 110-116. DOI: 10.1306/060103740110.
- Leupin, O.X., Van Loon, L.R., Gimmi, T., Wersin, P. & Soler, J.M. (2017): Exploring diffusion and sorption processes at the Mont Terri rock laboratory: lessons learned from 20 years of field research. *Swiss Journal of Geosciences, Special Issue: Mont Terri rock laboratory, 20 years of research* 110, 391-403.
- Li, Y.-H. & Gregory, S. (1974): Diffusion of ions in sea water and in deep-sea sediments. *Geochimica et Cosmochimica Acta* 38/5, 703-714.
- Lübke, M. (2015): Migration von Technetium in natürlichem Tongestein. PhD Thesis. Johannes Gutenberg-Universität Mainz.
- Mäder, U., Aschwanden, L., Camesi, L., Gimmi, T., Jenni, A., Kiczka, M., Mazurek, M., Rufer, D., Waber, H.N., Wersin, P., Zwahlen, C. & Traber, D. (2021): TBO Marthalen-1-1: Data Report Dossier VIII Rock properties, porewater characterisation and natural tracer profiles. Nagra Arbeitsbericht NAB 21-20 VIII.
- Maes, N., Glaus, M., Baeyens, B., Marques Fernandes, M., Churakov, S., Dähn, R., Grangeon, S., Tournassat, C., Geckeis, H., Charlet, L., Brandt, F., Poonoosamy, J., Hoving, A., Havlova, V., Scheinost, A., Fischer, C., Noseck, U., Britz, S., Siitari-Kauppi, M. & Missana, T. (2021): State-of-the-Art report on the understanding of radionuclide retention and transport in clay and crystalline rocks. Final version as of 30.04.2021 of deliverable D5.1 of the HORIZON 2020 project EURAD. European Joint Programme on Radioactive Waste Management EURAD.
- Marnat, S. & Becker, J.K. (2021): TBO Trüllikon-1-1: Dossier X Petrophysical Log Analysis. Nagra Arbeitsbericht NAB 20-09 X.
- Mazurek, M., Alt-Epping, P., Bath, A., Gimmi, T., Waber, H.N., Buschaert, S., De Cannière, P., De Craen, M., Gautschi, A., Savoye, S., Vinsot, A., Vermaere, I. & Wouters, L. (2011): Natural tracer profiles across argillaceous formations. *Applied Geochemistry* 26/7, 1035-1064.
- Mazurek, M., Aschwanden, L., Camesi, L., Gimmi, T., Jenni, A., Kiczka, M., Mäder, U., Rufer, D., Waber, H.N., Wanner, P., Wersin, P. & Traber, D. (2021): TBO Bülach-1-1: Data report Dossier VIII Rock Properties, Porewater Characterisation and Natural Tracer Profiles. Nagra Arbeitsbericht NAB 20-08 VIII.

- Mazurek, M., Gimmi, T., Zwahlen, C., Aschwanden, L., Gaucher, E.C., Kiczka, M., Rufer, D., Wersin, P., Marques Fernandes, M., Glaus, M.A., Van Loon, L.R., Traber, D., Schnellmann, M. & Vietor, T. (2023): Swiss deep drilling campaign 2019–2022: Geological overview and rock properties with focus on porosity and pore-space architecture. *Applied Geochemistry* 159, 105839. DOI: 10.1016/j.apgeochem.2023.105839.
- Mazurek, M., Hurford, A.J. & Leu, W. (2006): Unravelling the multi-stage burial history of the Swiss Molasse Basin: integration of apatite fission track, vitrinite reflectance and biomarker isomerisation analysis. *Basin Research* 18, 27-50.
- Molera, M. & Eriksen, T. (2002): Diffusion of $^{22}\text{Na}^+$, $^{85}\text{Sr}^{2+}$, $^{134}\text{Cs}^+$ and $^{57}\text{Co}^{2+}$ in bentonite clay compacted to different densities: experiments and modeling. *Migration '01: Chemistry and migration behaviour of actinides and fission products in the geosphere - Special issue of Radiochimica Acta* 90, 753-760.
- Naef, H., Büchi, M., Bläsi, H.R., Deplazes, G. & Gysi, M. (2019): Lithology Manual: Lithological description of drill cores and cuttings in Northern Switzerland. Nagra Arbeitsbericht NAB 19-11.
- Nagra (2001): Sondierbohrung Benken: Untersuchungsbericht: Textband + Beilagenband. Nagra Technischer Bericht NTB 00-01.
- Nagra (2002): Project Opalinus Clay: Safety report: Demonstration of disposal feasibility for spent fuel, vitrified high-level waste and long-lived intermediate-level waste (Entsorgungsnachweis). Nagra Technical Report NTB 02-05.
- Nagra (2021): The Nagra research, development and demonstration (RD&D) plan for the disposal of radioactive waste in Switzerland. Nagra Technical Report NTB 21-02.
- Nagra (2024): Geosynthesis of Northern Switzerland. Nagra Technischer Bericht NTB 24-17.
- Savoye, S., Goutelard, F., Beaucaire, C., Charles, Y., Fayette, A., Herbette, M., Larabi, Y. & Coelho, D. (2011): Effect of temperature on the containment properties of argillaceous rocks: The case study of Callovo-Oxfordian claystones. *Journal of Contaminant Hydrology* 125, 102-112.
- Soler, J.M., Steefel, C.I., Gimmi, T., Leupin, O.X. & Cloet, V. (2019): Modeling the ionic strength effect on diffusion in clay. The DR-A experiment at Mont Terri. *ACS Earth Space Chemistry* 3/3, 442-451.
- Stumm, W. & Morgan, J.J. (1996): Aquatic chemistry. Chemical equilibria and rates in natural waters. Third edition. John Wiley & Sons Inc, New York.
- Tournassat, C., Steefel, C.I., Bourg, I.C. & Bergaya, F. (2015): Natural and engineered clay barriers. *Developments in Clay Science*, 6, 446 pp.
- van Brakel, J. & Heertjes, P.M. (1974): Analysis of diffusion in macroporous media in terms of a porosity, a tortuosity and a constrictivity factor. *International Journal of Heat and Mass Transfer* 17/9, 1093-1103.

- Van Laer, L., Aertsens, M., Maes, N., Van Loon, L.R., Glaus, M.A. & Wüst, R.A.J. (2024): Diffusion of HTO, ^{36}Cl and ^{22}Na in the Mesozoic rocks of Northern Switzerland: III. Cross-lab comparison of diffusion measurements on argillaceous twin samples. *Applied Geochemistry* 160, 105840. DOI: 10.1016/j.apgeochem.2023.105840.
- Van Loon, L.R. (2014): Effective diffusion coefficients and porosity values for argillaceous rocks and bentonite: Measured and estimated values for the provisional safety analyses for SGT-E2. Nagra Technical Report NTB 12-03.
- Van Loon, L.R., Baeyens, B. & Bradbury, M.H. (2009): The sorption behaviour of caesium on Opalinus Clay: A comparison between intact and crushed material. *Applied Geochemistry* 24/5, 999-1004.
- Van Loon, L.R., Bunic, P., Frick, S., Glaus, M.A. & Wüst, R.A.J. (2023): Diffusion of HTO, ^{36}Cl and ^{22}Na in the Mesozoic rocks of northern Switzerland: I. Effective diffusion coefficients and capacity factors across the heterogeneous sediment sequences. *Applied Geochemistry* 159, 105843. DOI: 10.1016/j.apgeochem.2023.105843.
- Van Loon, L.R., Leupin, O.X. & Cloet, V. (2018): The diffusion of SO_4^{2-} in Opalinus Clay: Measurements of effective diffusion coefficients and evaluation of their importance in view of microbial mediated reactions in the near field of radioactive waste repositories. *Applied Geochemistry* 95, 19-24. DOI: 10.1016/j.apgeochem.2018.05.009.
- Van Loon, L.R. & Mibus, J. (2015): A modified version of Archie's law to estimate effective diffusion coefficients of radionuclides in argillaceous rocks and its application in safety analysis studies. *Applied Geochemistry* 59, 85-94.
- Van Loon, L.R., Müller, W. & Iijima, K. (2005): Activation energies of the self-diffusion of HTO, $^{22}\text{Na}^+$ and $^{36}\text{Cl}^-$ in a highly compacted argillaceous rock (Opalinus Clay). *Applied Geochemistry* 20/5, 961-972.
- Van Loon, L.R. & Soler, J.M. (2003): Diffusion of HTO, $^{36}\text{Cl}^-$, $^{125}\text{I}^-$ and $^{22}\text{Na}^+$ in Opalinus Clay: effect of confining pressure, sample orientation, sample depth and temperature. Nagra Technical Report NTB 03-07.
- Van Loon, L.R., Soler, J.M. & Bradbury, M.H. (2003a): Diffusion of HTO, $^{36}\text{Cl}^-$ and $^{125}\text{I}^-$ in Opalinus Clay samples from Mont Terri: Effect of confining pressure. *Journal of Contaminant Hydrology* 61, 73-83.
- Van Loon, L.R., Soler, J.M., Jakob, A. & Bradbury, M.H. (2003b): Effect of confining pressure on the diffusion of HTO, $^{36}\text{Cl}^-$ and $^{125}\text{I}^-$ in a layered argillaceous rock (Opalinus Clay): diffusion perpendicular to the fabric. *Applied Geochemistry* 18/10, 1653-1662. DOI: 10.1016/S0883-2927(03)00047-7.
- Van Loon, L.R., Soler, J.M., Müller, W. & Bradbury, M.H. (2004): Anisotropic diffusion in layered argillaceous rocks: A case study with Opalinus Clay. *Environmental Science & Technology* 38/21, 5721-5728.
- van Schaik, J.C., Kemper, W.D. & Olsen, S.R. (1966): Contribution of adsorbed cations to diffusion in clay-water systems. *Soil Science Society of America Journal* 30, 17-22, available online at <https://api.semanticscholar.org/CorpusID:128824229>.

- Wenk, H.-R., Voltolini, M., Mazurek, M., Van Loon, L.R. & Vinsot, A. (2008): Preferred orientations and anisotropy in shales: Callovo-Oxfordian shale (France) and Opalinus Clay (Switzerland). *Clays and Clay Minerals* 56/3, 285-306. DOI: 10.1346/CCMN.2008.0560301.
- Wersin, P., Aschwanden, L., Camesi, L., Gaucher, E.C., Gimmi, T., Jenni, A., Kiczka, M., Mäder, U., Mazurek, M., Rufer, D., Waber, H.N., Zwahlen, C. & Traber, D. (2022): TBO Bözberg-1-1: Data Report Dossier VIII Rock properties, porewater characterisation and natural tracer profiles. Nagra Arbeitsbericht NAB 21-21 VIII.
- Wersin, P., Gimmi, T., Ma, J., Mazurek, M., Zwahlen, C., Aschwanden, L., Gaucher, E. & Traber, D. (2023): Porewater profiles of Cl and Br in boreholes penetrating the Mesozoic sequence in northern Switzerland. *Applied Geochemistry* 159, 105845. DOI: 10.1016/j.apgeochem.2023.105845.
- Wersin, P., Gimmi, T., Mazurek, M., Alt-Epping, P., Pekala, M. & Traber, D. (2018): Multi-component diffusion in a 280m thick argillaceous rock sequence. *Applied Geochemistry* 95/8, 110-123.
- Wersin, P., Mazurek, M., Waber, H.N., Mäder, U.K., Gimmi, T., Rufer, D. & de Haller, A. (2013): Rock and porewater characterisation on drillcores from the Schlattingen borehole. Nagra Arbeitsbericht NAB 12-54.
- Wigger, C., Kennell-Morrison, L., Jensen, M., Glaus, M. & Van Loon, L. (2018): A comparative anion diffusion study on different argillaceous, low permeability sedimentary rocks with various pore waters. *Applied Geochemistry* 92/5, 157-165.
- Wigger, C. & Van Loon, L.R. (2017): Importance of interlayer equivalent pores for anion diffusion in clay-rich sedimentary rocks. *Environmental Science & Technology* 51/4, 1998-2006.
- Winhausen, L., Khaledi, K., Jalali, M., Bretthauer, M. & Amann, F. (2023): The anisotropic behavior of a clay shale: strength, hydro-mechanical couplings and failure processes. *Journal of Geophysical Research: Solid Earth* 128/11, e2023JB027382. DOI: 10.1029/2023JB027382.
- Woolf, L.A. (1975): Tracer diffusion of tritiated water (THO) in ordinary water (H₂O) under pressure. *Journal of the Chemical Society, Faraday Transactions 1: Physical Chemistry in Condensed Phases* 71/0, 784-796. DOI: 10.1039/F19757100784.
- Wu, T., Amayri, S., Drebert, J., Van Loon, L.R. & Reich, T. (2009): Neptunium(V) sorption and diffusion in Opalinus Clay. *Environmental Science & Technology* 43/17, 6567-6571.
- Wüst, R.A.J., Tu, S., Nassichuk, B., Bozarth, T., Tucker, J. & Cui, A. (2018): Chemostratigraphy, petrography, and SEM investigations of the Lower Triassic Montney Formation in Alberta: Implications for a new and revised diagenetic and depositional model. *Bulletin of Canadian Petroleum Geology* 66/2, 436-471.
- Xiang, Y., Al, T. & Mazurek, M. (2016): Effect of confining pressure on diffusion coefficients in clay-rich, low-permeability sedimentary rocks. *Journal of Contaminant Hydrology* 195, 1-10.

- Xiang, Y., Al, T., Scott, L. & Loomer, D. (2013): Diffusive anisotropy in low-permeability Ordovician sedimentary rocks from the Michigan Basin in southwest Ontario. *Journal of Contaminant Hydrology* 155, 31-45. DOI: 10.1016/j.jconhyd.2013.09.002.
- Zwahlen, C., Aschwanden, L., Gaucher, E., Gimmi, T., Jenni, A., Kiczka, M., Mäder, U., Mazurek, M., Ross, D., Rufer, D., Waber, H.N., Wersin, P. & Traber, D. (2022): TBO Stadel-2-1: Data report dossier VIII Rock properties, porewater characterisation and natural tracer profiles. Nagra Arbeitsbericht NAB 22-02 VIII.
- Zwahlen, C., Gimmi, T., Jenni, A., Kiczka, M., Mazurek, M., Van Loon, L.R., Mäder, U. & Traber, D. (2024): Chloride accessible porosity fractions across the Jurassic sedimentary rocks of northern Switzerland. *Applied Geochemistry* 162, 105841. DOI: 10.1016/j.apgeochem.2023.105841.

App. A Diffusion data for HTO, $^{36}\text{Cl}^-$ and $^{22}\text{Na}^+$

Tab. A-1: Overview of the effective diffusion coefficients (D_e) and diffusion-accessible porosities (ε) for HTO, $^{36}\text{Cl}^-$ and distribution coefficients (R_d) for $^{22}\text{Na}^+$ for samples from the BOZ1-1 and BOZ2-1 boreholes in the Jura Ost (JO) siting region

Sample	SPW	Formation	Clay	Calcite	Quartz	HTO		$^{36}\text{Cl}^-$		$^{22}\text{Na}^+$	
			[wt.%]	[wt.%]	[wt.%]	D_e [m ² s ⁻¹]	ε [-]	D_e [m ² s ⁻¹]	ε [-]	D_e [m ² s ⁻¹]	R_d [m ³ kg ⁻¹]
BOZ1-1-266.75-DI	SPW-BOZ	Wildeggen Fm.	24.0	58.3	10.4	$(1.40 \pm 0.04) \times 10^{-11}$	0.094 ± 0.007	$(2.58 \pm 0.05) \times 10^{-12}$	0.039 ± 0.001	$(2.56 \pm 0.18) \times 10^{-11}$	$(1.30 \pm 0.13) \times 10^{-4}$
BOZ1-1-290.63-DI	SPW-BOZ	Wildeggen Fm.	19.0	69.6	5.7	$(1.13 \pm 0.03) \times 10^{-11}$	0.071 ± 0.006	$(2.31 \pm 0.08) \times 10^{-12}$	0.031 ± 0.002	$(2.18 \pm 0.14) \times 10^{-11}$	$(1.10 \pm 0.10) \times 10^{-4}$
BOZ1-1-323.16-DI	SPW-BOZ	Wildeggen Fm.	11.4	81.3	3.0	$(8.06 \pm 0.19) \times 10^{-12}$	0.063 ± 0.004	$(9.78 \pm 0.20) \times 10^{-13}$	0.019 ± 0.001	$(1.95 \pm 0.12) \times 10^{-11}$	$(1.17 \pm 0.10) \times 10^{-4}$
BOZ1-1-353.36-DI	SPW-BOZ	Wildeggen Fm.	31.4	55.0	7.9	$(1.27 \pm 0.04) \times 10^{-11}$	0.099 ± 0.007	$(2.11 \pm 0.11) \times 10^{-12}$	0.035 ± 0.003	$(2.45 \pm 0.19) \times 10^{-11}$	$(1.55 \pm 0.17) \times 10^{-4}$
BOZ1-1-391.30-DI	SPW-BOZ	Wildeggen Fm.	24.2	67.0	5.5	$(1.04 \pm 0.03) \times 10^{-11}$	0.080 ± 0.005	$(1.64 \pm 0.04) \times 10^{-12}$	0.030 ± 0.001	$(1.94 \pm 0.13) \times 10^{-11}$	$(1.22 \pm 0.13) \times 10^{-4}$
BOZ1-1-416.35-DI	SPW-BOZ	Hauptrogenstein	15.5	69.9	2.1	$(1.78 \pm 0.07) \times 10^{-11}$	0.116 ± 0.008	$(9.85 \pm 0.24) \times 10^{-12}$	0.101 ± 0.005	$(1.57 \pm 0.07) \times 10^{-11}$	$(4.14 \pm 0.60) \times 10^{-5}$
BOZ1-1-434.29-DI	SPW-BOZ	Klingnau Fm.	15.2	74.3	4.4	$(1.01 \pm 0.03) \times 10^{-11}$	0.074 ± 0.005	$(3.04 \pm 0.05) \times 10^{-12}$	0.040 ± 0.001	$(1.58 \pm 0.08) \times 10^{-11}$	$(8.68 \pm 0.93) \times 10^{-5}$
BOZ1-1-438.20-DI	SPW-BOZ	Klingnau Fm.	27.2	56.2	4.1	$(1.63 \pm 0.07) \times 10^{-11}$	0.119 ± 0.009	$(6.93 \pm 0.08) \times 10^{-12}$	0.095 ± 0.002	$(1.87 \pm 0.10) \times 10^{-11}$	$(7.10 \pm 0.99) \times 10^{-5}$
BOZ1-1-440.55-DI	SPW-BOZ	Klingnau Fm.	28.0	55.3	6.0	$(1.36 \pm 0.04) \times 10^{-11}$	0.113 ± 0.008	$(4.18 \pm 0.09) \times 10^{-12}$	0.075 ± 0.003	$(1.97 \pm 0.13) \times 10^{-11}$	$(1.06 \pm 0.13) \times 10^{-4}$
BOZ1-1-487.01-DI	SPW-BOZ	Passwang Fm.	36.3	33.6	19.9	$(1.24 \pm 0.04) \times 10^{-11}$	0.129 ± 0.007	$(9.71 \pm 0.42) \times 10^{-13}$	0.028 ± 0.002	$(2.66 \pm 0.27) \times 10^{-11}$	$(2.61 \pm 0.34) \times 10^{-4}$
BOZ1-1-493.15-DI	SPW-BOZ	Passwang Fm.	35.2	34.6	21.5	$(1.25 \pm 0.05) \times 10^{-11}$	0.133 ± 0.011	$(1.59 \pm 0.19) \times 10^{-12}$	0.050 ± 0.010	$(3.20 \pm 0.34) \times 10^{-11}$	$(2.64 \pm 0.31) \times 10^{-4}$
BOZ1-1-498.32-DI	SPW-BOZ	Passwang Fm.	44.2	30.0	18.6	$(1.43 \pm 0.05) \times 10^{-11}$	0.118 ± 0.008	$(1.56 \pm 0.03) \times 10^{-12}$	0.035 ± 0.001	$(2.60 \pm 0.23) \times 10^{-11}$	$(1.96 \pm 0.26) \times 10^{-4}$
BOZ1-1-503.87-DI	SPW-BOZ	Passwang Fm.	28.7	49.5	14.9	$(1.23 \pm 0.04) \times 10^{-11}$	0.101 ± 0.007	$(1.88 \pm 0.06) \times 10^{-12}$	0.038 ± 0.002	$(2.72 \pm 0.23) \times 10^{-11}$	$(1.62 \pm 0.19) \times 10^{-4}$
BOZ1-1-512.03-DI	SPW-BOZ	Passwang Fm.	45.2	18.8	27.1	$(1.44 \pm 0.05) \times 10^{-11}$	0.127 ± 0.007	$(1.82 \pm 0.09) \times 10^{-12}$	0.038 ± 0.003	$(2.06 \pm 0.17) \times 10^{-11}$	$(1.63 \pm 0.23) \times 10^{-4}$
BOZ1-1-521.50-DI	SPW-BOZ	Passwang Fm.	41.3	16.2	31.3	$(1.44 \pm 0.05) \times 10^{-11}$	0.128 ± 0.008	$(1.71 \pm 0.04) \times 10^{-12}$	0.041 ± 0.002	$(3.19 \pm 0.29) \times 10^{-11}$	$(2.50 \pm 0.23) \times 10^{-4}$
BOZ1-1-529.56-DI	SPW-BOZ	Passwang Fm.	46.6	3.1	39.7	$(1.59 \pm 0.06) \times 10^{-11}$	0.127 ± 0.009	$(2.89 \pm 0.09) \times 10^{-12}$	0.051 ± 0.003	$(3.14 \pm 0.29) \times 10^{-11}$	$(1.89 \pm 0.21) \times 10^{-4}$
BOZ1-1-550.63-DI	SPW-BOZ	Opalinus Clay	48.5	14.0	27.5	$(8.70 \pm 0.19) \times 10^{-12}$	0.120 ± 0.005	$(9.61 \pm 0.33) \times 10^{-13}$	0.036 ± 0.002	$(1.13 \pm 0.10) \times 10^{-11}$	$(1.62 \pm 0.23) \times 10^{-4}$
BOZ1-1-562.07-DI	SPW-BOZ	Opalinus Clay	58.2	9.7	20.9	$(9.32 \pm 0.24) \times 10^{-12}$	0.119 ± 0.006	$(1.14 \pm 0.04) \times 10^{-12}$	0.036 ± 0.002	$(1.67 \pm 0.13) \times 10^{-11}$	$(1.97 \pm 0.27) \times 10^{-4}$
BOZ1-1-572.43-DI	SPW-BOZ	Opalinus Clay	42.9	14.6	31.1	$(1.10 \pm 0.03) \times 10^{-11}$	0.095 ± 0.006	$(1.84 \pm 0.03) \times 10^{-12}$	0.040 ± 0.001	$(2.35 \pm 0.18) \times 10^{-11}$	$(1.67 \pm 0.17) \times 10^{-4}$
BOZ1-1-580.34-DI	SPW-BOZ	Opalinus Clay	52.2	11.2	25.1	$(9.00 \pm 0.22) \times 10^{-12}$	0.121 ± 0.006	$(9.89 \pm 0.51) \times 10^{-13}$	0.038 ± 0.003	$(3.53 \pm 0.42) \times 10^{-12}$	$(7.04 \pm 3.00) \times 10^{-5}$

Tab. A-1: Cont.

Sample	SPW	Formation	Clay	Calcite	Quartz	HTO		³⁶ Cl		²² Na ⁺	
			[wt.%]	[wt.%]	[wt.%]	D_e [m ² s ⁻¹]	ε [-]	D_e [m ² s ⁻¹]	ε [-]	D_e [m ² s ⁻¹]	R_d [m ³ kg ⁻¹]
BOZ1-1-597.28-DI	SPW-BOZ	Opalinus Clay	61.0	5.7	20.3	$(9.09 \pm 0.19) \times 10^{-12}$	0.137 ± 0.006	$(1.07 \pm 0.04) \times 10^{-12}$	0.041 ± 0.002	$(1.49 \pm 0.13) \times 10^{-11}$	$(2.41 \pm 0.37) \times 10^{-4}$
BOZ1-1-610.79-DI	SPW-BOZ	Opalinus Clay	67.0	4.1	14.7	$(7.95 \pm 0.18) \times 10^{-12}$	0.134 ± 0.006	$(8.42 \pm 0.33) \times 10^{-13}$	0.038 ± 0.002	$(1.11 \pm 0.08) \times 10^{-11}$	$(2.05 \pm 0.30) \times 10^{-4}$
BOZ1-1-619.10-DI	SPW-BOZ	Opalinus Clay	68.4	4.0	16.1	$(8.26 \pm 0.19) \times 10^{-12}$	0.143 ± 0.006	$(8.30 \pm 0.33) \times 10^{-13}$	0.041 ± 0.003	$(1.07 \pm 0.08) \times 10^{-11}$	$(2.08 \pm 0.35) \times 10^{-4}$
BOZ1-1-623.60-DI	SPW-BOZ	Opalinus Clay	69.0	3.1	16.0	$(3.15 \pm 0.13) \times 10^{-12}$	0.081 ± 0.005	$(2.80 \pm 0.15) \times 10^{-13}$	0.020 ± 0.002	$(2.47 \pm 0.20) \times 10^{-12}$	$(5.71 \pm 1.17) \times 10^{-5}$
BOZ1-1-634.90-DI	SPW-BOZ	Opalinus Clay	67.2	4.8	14.5	$(8.54 \pm 0.20) \times 10^{-12}$	0.149 ± 0.007	$(8.54 \pm 0.43) \times 10^{-13}$	0.039 ± 0.003	$(7.88 \pm 0.55) \times 10^{-12}$	$(1.49 \pm 0.27) \times 10^{-4}$
BOZ1-1-646.44-DI	SPW-BOZ	Opalinus Clay	63.3	8.2	15.8	$(7.53 \pm 0.21) \times 10^{-12}$	0.135 ± 0.006	$(7.56 \pm 0.28) \times 10^{-13}$	0.038 ± 0.002	$(1.35 \pm 0.11) \times 10^{-11}$	$(2.68 \pm 0.36) \times 10^{-4}$
BOZ1-1-665.00-DI	SPW-BOZ	Staffelegg Fm.	28.5	9.6	44.3	$(1.63 \pm 0.05) \times 10^{-11}$	0.094 ± 0.007	$(4.09 \pm 0.11) \times 10^{-12}$	0.049 ± 0.002	$(3.15 \pm 0.25) \times 10^{-11}$	$(1.40 \pm 0.16) \times 10^{-4}$
BOZ1-1-670.95-DI	SPW-BOZ	Staffelegg Fm.	36.7	19.2	30.9	$(1.05 \pm 0.03) \times 10^{-11}$	0.096 ± 0.006	$(1.72 \pm 0.05) \times 10^{-12}$	0.038 ± 0.002	$(2.27 \pm 0.17) \times 10^{-11}$	$(1.62 \pm 0.17) \times 10^{-4}$
BOZ1-1-679.67-DI	SPW-BOZ	Staffelegg Fm.	64.1	15.7	13.5	$(9.22 \pm 0.24) \times 10^{-12}$	0.152 ± 0.007	$(7.74 \pm 0.30) \times 10^{-13}$	0.036 ± 0.002	$(1.61 \pm 0.18) \times 10^{-11}$	$(2.93 \pm 0.37) \times 10^{-4}$
BOZ1-1-681.57-DI	SPW-BOZ	Staffelegg Fm.	79.1	8.0	9.1	$(7.83 \pm 0.16) \times 10^{-12}$	0.167 ± 0.006	$(5.87 \pm 0.12) \times 10^{-13}$	0.046 ± 0.001	$(8.42 \pm 0.80) \times 10^{-12}$	$(3.00 \pm 0.42) \times 10^{-4}$
BOZ1-1-686.81-DI	SPW-BOZ	Staffelegg Fm.	61.8	6.4	17.1	$(7.87 \pm 0.25) \times 10^{-12}$	0.176 ± 0.008	$(6.75 \pm 0.37) \times 10^{-13}$	0.039 ± 0.003	$(9.84 \pm 0.46) \times 10^{-12}$	$(3.46 \pm 0.56) \times 10^{-4}$
BOZ1-1-717.96-DI	SPW-BOZ	Klettgau Fm.	39.7	0.0	28.8	$(1.52 \pm 0.06) \times 10^{-11}$	0.125 ± 0.009	$(1.92 \pm 0.20) \times 10^{-12}$	0.093 ± 0.015	$(1.55 \pm 0.13) \times 10^{-11}$	$(1.67 \pm 0.27) \times 10^{-4}$
BOZ1-1-723.97-DI	SPW-BOZ	Bänkerjoch Fm.	44.1	0.0	11.2	$(1.51 \pm 0.05) \times 10^{-11}$	0.115 ± 0.008	$(1.89 \pm 0.05) \times 10^{-12}$	0.042 ± 0.002	$(2.88 \pm 0.26) \times 10^{-11}$	$(1.87 \pm 0.22) \times 10^{-4}$
BOZ1-1-806.41-DI	SPW-BOZ	Bänkerjoch Fm.		3		>90% anhydrite, no breakthrough of tracer during experiment					
BOZ2-1-313.13-DI	SPW-BOZ	Hauptrogenstein	25.2	59.0	1.0	$(2.44 \pm 0.13) \times 10^{-11}$	0.145 ± 0.014	$(1.52 \pm 0.02) \times 10^{-11}$	0.130 ± 0.004	$(1.84 \pm 0.10) \times 10^{-11}$	$(2.69 \pm 0.69) \times 10^{-5}$
BOZ2-1-443.93-DI	SPW-BOZ	Passwang Fm.	38.5	16.0	34.0	$(1.66 \pm 0.05) \times 10^{-11}$	0.109 ± 0.008	$(3.42 \pm 0.05) \times 10^{-12}$	0.049 ± 0.001	$(3.30 \pm 0.33) \times 10^{-11}$	$(1.68 \pm 0.17) \times 10^{-4}$
BOZ2-1-588.64-DI	SPW-BOZ	Staffelegg Fm.	34.2	8.0	48.0	$(1.55 \pm 0.06) \times 10^{-11}$	0.099 ± 0.008	$(3.31 \pm 0.04) \times 10^{-12}$	0.048 ± 0.001	$(2.95 \pm 0.25) \times 10^{-11}$	$(1.34 \pm 0.14) \times 10^{-4}$
BOZ2-1-595.15-DI	SPW-BOZ	Staffelegg Fm.	43.1	11.0	37.0	$(1.74 \pm 0.06) \times 10^{-11}$	0.121 ± 0.010	$(3.70 \pm 0.06) \times 10^{-12}$	0.050 ± 0.002	$(3.17 \pm 0.31) \times 10^{-11}$	$(1.89 \pm 0.17) \times 10^{-4}$
BOZ2-1-606.95-DI	SPW-BOZ	Staffelegg Fm.	70.1	10.0	12.0	$(9.02 \pm 0.19) \times 10^{-12}$	0.159 ± 0.007	$(7.01 \pm 0.16) \times 10^{-13}$	0.035 ± 0.001	$(1.68 \pm 0.12) \times 10^{-11}$	$(3.62 \pm 0.34) \times 10^{-4}$

Tab. A-2: Overview of the effective diffusion coefficients (D_e) and diffusion-accessible porosities (ε) for HTO, $^{36}\text{Cl}^-$ and distribution coefficients (R_d) for $^{22}\text{Na}^+$ for samples from the BUL1-1, STA2-1 and STA3-1 boreholes in the Nördlich Läger (NL) siting region

Sample	SPW	Formation	Clay	Calcite	Quartz	HTO		$^{36}\text{Cl}^-$		$^{22}\text{Na}^+$	
			[wt.%]	[wt.%]	[wt.%]	D_e [m ² s ⁻¹]	ε [-]	D_e [m ² s ⁻¹]	ε [-]	D_e [m ² s ⁻¹]	R_d [m ³ kg ⁻¹]
BUL1-1-734.45-DI	SPW-BUL	Villigen Fm.	10.6	87.5	1.6	$(5.07 \pm 0.09) \times 10^{-12}$	0.038 ± 0.003	$(8.76 \pm 0.45) \times 10^{-13}$	0.014 ± 0.002	$(5.08 \pm 0.10) \times 10^{-12}$	$(2.21 \pm 0.20) \times 10^{-5}$
BUL1-1-752.48-DI	SPW-BUL	Wildegge Fm.	18.4	64.0	7.0	$(9.75 \pm 0.25) \times 10^{-12}$	0.048 ± 0.004	$(2.92 \pm 0.13) \times 10^{-12}$	0.024 ± 0.003	$(1.13 \pm 0.04) \times 10^{-11}$	$(3.28 \pm 0.35) \times 10^{-5}$
BUL1-1-766.65-DI	SPW-BUL	Wildegge Fm.	17.6	73.0	5.0	$(9.30 \pm 0.21) \times 10^{-12}$	0.055 ± 0.004	$(2.36 \pm 0.11) \times 10^{-12}$	0.027 ± 0.004	$(1.14 \pm 0.05) \times 10^{-11}$	$(4.16 \pm 0.49) \times 10^{-5}$
BUL1-1-794.35-DI	SPW-BUL	Wildegge Fm.	22.3	69.0	5.0	$(9.35 \pm 0.20) \times 10^{-12}$	0.064 ± 0.004	$(2.18 \pm 0.09) \times 10^{-12}$	0.030 ± 0.002	$(1.21 \pm 0.04) \times 10^{-11}$	$(5.55 \pm 0.45) \times 10^{-5}$
BUL1-1-808.92-DI	SPW-BUL	'Parkinsoni-Württembergica-Schichten.'	51.5	26.0	15.0	$(1.41 \pm 0.05) \times 10^{-11}$	0.131 ± 0.011	$(3.23 \pm 0.12) \times 10^{-12}$	0.065 ± 0.004	$(1.75 \pm 0.15) \times 10^{-11}$	$(1.62 \pm 0.29) \times 10^{-4}$
BUL1-1-840.45-DI_S1	SPW-BUL	'Herrenwis Unit'	5.3	78.0	10.0	$(4.01 \pm 0.10) \times 10^{-12}$	0.012 ± 0.002	$(3.02 \pm 0.13) \times 10^{-12}$	0.009 ± 0.002	$(3.62 \pm 0.20) \times 10^{-12}$	$(4.94 \pm 1.70) \times 10^{-6}$
BUL1-1-840.45-DI_S2	SPW-BUL	'Herrenwis Unit'	5.3	78.0	10.0	$(6.36 \pm 0.14) \times 10^{-12}$	0.032 ± 0.003	$(2.85 \pm 0.16) \times 10^{-12}$	0.019 ± 0.003	$(6.09 \pm 0.15) \times 10^{-12}$	$(1.87 \pm 0.24) \times 10^{-5}$
BUL1-1-845.05-DI	SPW-BUL	'Herrenwis Unit'	1.0	98.0	1.0	$(2.83 \pm 0.05) \times 10^{-12}$	0.011 ± 0.001	$(2.24 \pm 0.07) \times 10^{-12}$	0.011 ± 0.001	$(1.92 \pm 0.10) \times 10^{-12}$	$(3.81 \pm 0.59) \times 10^{-6}$
BUL1-1-851.50-DI	SPW-BUL	'Herrenwis Unit'	6.8	90.0	3.0	$(6.06 \pm 0.60) \times 10^{-12}$	0.033 ± 0.006	$(3.94 \pm 0.39) \times 10^{-12}$	0.033 ± 0.006	$(6.42 \pm 0.40) \times 10^{-12}$	$(6.34 \pm 2.96) \times 10^{-6}$
BUL1-1-872.35-DI	SPW-BUL	Wedelsandstein Fm.	34.6	16.0	37.0	$(1.61 \pm 0.07) \times 10^{-11}$	0.095 ± 0.008	$(4.93 \pm 0.23) \times 10^{-12}$	0.053 ± 0.005	$(1.89 \pm 0.10) \times 10^{-11}$	$(6.05 \pm 0.62) \times 10^{-5}$
BUL1-1-879.20-DI	SPW-BUL	Wedelsandstein Fm.	39.4	13.0	37.0	$(1.31 \pm 0.05) \times 10^{-11}$	0.082 ± 0.008	$(3.74 \pm 0.11) \times 10^{-12}$	0.045 ± 0.003	$(1.55 \pm 0.07) \times 10^{-11}$	$(5.41 \pm 0.61) \times 10^{-5}$
BUL1-1-908.93-DI	SPW-BUL	Opalinus Clay	57.3	4.0	24.0	$(1.40 \pm 0.05) \times 10^{-11}$	0.113 ± 0.008	$(3.52 \pm 0.14) \times 10^{-12}$	0.058 ± 0.006	$(1.80 \pm 0.09) \times 10^{-11}$	$(9.75 \pm 0.91) \times 10^{-5}$
BUL1-1-914.61-DI	SPW-BUL	Opalinus Clay	54.0	7.0	25.0	$(9.62 \pm 0.26) \times 10^{-12}$	0.115 ± 0.007	$(2.31 \pm 0.10) \times 10^{-12}$	0.056 ± 0.004	$(1.25 \pm 0.05) \times 10^{-11}$	$(1.06 \pm 0.08) \times 10^{-4}$
BUL1-1-921.43-DI_S1	SPW-BUL	Opalinus Clay	48.5	10.0	28.0	$(9.03 \pm 0.27) \times 10^{-12}$	0.111 ± 0.007	$(2.47 \pm 0.09) \times 10^{-12}$	0.058 ± 0.003	$(1.13 \pm 0.04) \times 10^{-11}$	$(1.01 \pm 0.08) \times 10^{-4}$
BUL1-1-921.43-DI_S2	SPW-BUL	Opalinus Clay	48.5	10.0	28.0	$(8.21 \pm 0.23) \times 10^{-12}$	0.088 ± 0.007	$(2.95 \pm 0.08) \times 10^{-12}$	0.048 ± 0.002	$(8.38 \pm 0.25) \times 10^{-12}$	$(5.83 \pm 0.53) \times 10^{-5}$
BUL1-1-932.82-DI	SPW-BUL	Opalinus Clay	58.1	6.0	23.0	$(8.56 \pm 0.20) \times 10^{-12}$	0.120 ± 0.006	$(2.10 \pm 0.08) \times 10^{-12}$	0.055 ± 0.003	$(1.03 \pm 0.05) \times 10^{-11}$	$(1.10 \pm 0.09) \times 10^{-4}$
BUL1-1-945.46-DI	SPW-BUL	Opalinus Clay	61.2	4.0	18.0	$(1.09 \pm 0.03) \times 10^{-11}$	0.131 ± 0.007	$(2.87 \pm 0.16) \times 10^{-12}$	0.069 ± 0.007	$(1.45 \pm 0.06) \times 10^{-11}$	$(1.27 \pm 0.11) \times 10^{-4}$
BUL1-1-955.62-DI_S1	SPW-BUL	Opalinus Clay	69.2	4.0	15.0	$(8.44 \pm 0.21) \times 10^{-12}$	0.133 ± 0.007	$(1.91 \pm 0.08) \times 10^{-12}$	0.064 ± 0.004	$(1.04 \pm 0.05) \times 10^{-11}$	$(1.31 \pm 0.10) \times 10^{-4}$

Tab. A-2: Cont.

Sample	SPW	Formation	Clay	Calcite	Quartz	HTO		³⁶ Cl ⁻		²² Na ⁺	
			[wt. %]	[wt. %]	[wt. %]	D_e [m ² s ⁻¹]	ϵ [-]	D_e [m ² s ⁻¹]	ϵ [-]	D_e [m ² s ⁻¹]	R_d [m ³ kg ⁻¹]
BUL1-1-955.62-DI_S2	SPW-BUL	Opalinus Clay	69.2	4.0	15.0	$(8.29 \pm 0.21) \times 10^{-12}$	0.108 ± 0.007	$(1.97 \pm 0.09) \times 10^{-12}$	0.058 ± 0.004	$(1.05 \pm 0.05) \times 10^{-11}$	$(9.60 \pm 0.70) \times 10^{-5}$
BUL1-1-965.46-DI	SPW-BUL	Opalinus Clay	59.4	8.0	18.0	$(8.96 \pm 0.22) \times 10^{-12}$	0.120 ± 0.007	$(2.24 \pm 0.07) \times 10^{-12}$	0.060 ± 0.003	$(1.13 \pm 0.05) \times 10^{-11}$	$(1.13 \pm 0.09) \times 10^{-4}$
BUL1-1-969.15-DI	SPW-BUL	Opalinus Clay	48.4	13.0	25.0	$(8.25 \pm 0.19) \times 10^{-12}$	0.083 ± 0.005	$(1.90 \pm 0.06) \times 10^{-12}$	0.041 ± 0.002	$(1.10 \pm 0.04) \times 10^{-11}$	$(8.41 \pm 0.58) \times 10^{-5}$
BUL1-1-975.04-DI	SPW-BUL	Opalinus Clay	53.7	7.0	26.0	$(8.46 \pm 0.22) \times 10^{-12}$	0.115 ± 0.006	$(1.85 \pm 0.12) \times 10^{-12}$	0.051 ± 0.005	$(1.05 \pm 0.04) \times 10^{-11}$	$(1.02 \pm 0.08) \times 10^{-4}$
BUL1-1-984.49-DI	SPW-BUL	Opalinus Clay	63.2	6.0	17.0	$(8.53 \pm 0.22) \times 10^{-12}$	0.123 ± 0.007	$(1.98 \pm 0.10) \times 10^{-12}$	0.060 ± 0.006	$(1.07 \pm 0.04) \times 10^{-11}$	$(1.13 \pm 0.10) \times 10^{-4}$
BUL1-1-995.09-DI	SPW-BUL	Opalinus Clay	62.9	9.0	16.0	$(7.94 \pm 0.17) \times 10^{-12}$	0.127 ± 0.006	$(1.65 \pm 0.04) \times 10^{-12}$	0.060 ± 0.002	$(1.01 \pm 0.03) \times 10^{-11}$	$(1.14 \pm 0.09) \times 10^{-4}$
BUL1-1-1002.19-DI	SPW-BUL	Staffelegg Fm.	33.3	47.0	9.0	$(6.37 \pm 0.13) \times 10^{-12}$	0.109 ± 0.006	$(1.61 \pm 0.07) \times 10^{-12}$	0.070 ± 0.004	$(6.71 \pm 0.2) \times 10^{-12}$	$(7.05 \pm 0.53) \times 10^{-5}$
BUL1-1-1008.42-DI	SPW-BUL	Staffelegg Fm.	5.2	93.0	2.0	$(4.03 \pm 0.06) \times 10^{-12}$	0.019 ± 0.001	$(2.12 \pm 0.10) \times 10^{-12}$	0.012 ± 0.002	$(3.62 \pm 0.05) \times 10^{-12}$	$(1.36 \pm 0.10) \times 10^{-5}$
BUL1-1-1014.23-DI	SPW-BUL	Staffelegg Fm.	40.2	6.0	41.0	$(1.34 \pm 0.05) \times 10^{-11}$	0.096 ± 0.008	$(3.92 \pm 0.20) \times 10^{-12}$	0.052 ± 0.004	$(1.57 \pm 0.07) \times 10^{-11}$	$(7.02 \pm 0.73) \times 10^{-5}$
BUL1-1-1017.33-DI	SPW-BUL	Staffelegg Fm.	42.0	7.0	37.0	$(1.11 \pm 0.05) \times 10^{-11}$	0.094 ± 0.008	$(3.08 \pm 0.16) \times 10^{-12}$	0.051 ± 0.004	$(1.27 \pm 0.05) \times 10^{-11}$	$(6.46 \pm 0.60) \times 10^{-5}$
BUL1-1-1025.96-DI	SPW-BUL	Staffelegg Fm.	22.7	61.0	12.0	$(5.32 \pm 0.16) \times 10^{-12}$	0.043 ± 0.003	$(1.37 \pm 0.05) \times 10^{-12}$	0.021 ± 0.002	$(6.14 \pm 0.2) \times 10^{-12}$	$(3.23 \pm 0.23) \times 10^{-5}$
STA2-1-633.23-DI	SPW-STa	Villigen Fm.	10.5	86.0	2.0	$(5.79 \pm 0.86) \times 10^{-12}$	0.042 ± 0.007	$(1.53 \pm 0.08) \times 10^{-12}$	0.019 ± 0.002	$(6.39 \pm 0.34) \times 10^{-12}$	$(2.94 \pm 0.47) \times 10^{-5}$
STA2-1-752.73-DI	SPW-STa	'Parkinsoni-Württembergica-Schichten'	42.6	22.0	22.0	$(1.24 \pm 0.04) \times 10^{-11}$	0.113 ± 0.009	$(2.61 \pm 0.05) \times 10^{-12}$	0.053 ± 0.002	$(1.58 \pm 0.09) \times 10^{-11}$	$(9.55 \pm 1.06) \times 10^{-5}$
STA2-1-758.02-DI	SPW-STa	'Parkinsoni-Württembergica-Schichten'	46.0	23.0	20.0	$(1.25 \pm 0.05) \times 10^{-11}$	0.122 ± 0.009	$(2.35 \pm 0.04) \times 10^{-12}$	0.053 ± 0.002	$(1.64 \pm 0.10) \times 10^{-11}$	$(1.11 \pm 0.12) \times 10^{-4}$
STA2-1-758.23-DI	SPW-STa	'Parkinsoni-Württembergica-Schichten'	45.0	22.0	21.0	$(1.35 \pm 0.05) \times 10^{-11}$	0.131 ± 0.010	$(2.45 \pm 0.05) \times 10^{-12}$	0.054 ± 0.002	$(1.67 \pm 0.11) \times 10^{-11}$	$(1.09 \pm 0.13) \times 10^{-4}$
STA2-1-765.21-DI	SPW-STa	'Parkinsoni-Württembergica-Schichten'	51.0	23.0	15.0	$(1.24 \pm 0.04) \times 10^{-11}$	0.134 ± 0.010	$(2.22 \pm 0.06) \times 10^{-12}$	0.056 ± 0.002	$(1.50 \pm 0.10) \times 10^{-11}$	$(1.13 \pm 0.14) \times 10^{-4}$
STA2-1-779.96-DI	SPW-STa	Wedelsandstein Fm.	61.0	11.0	23.0	$(1.22 \pm 0.04) \times 10^{-11}$	0.141 ± 0.009	$(2.26 \pm 0.06) \times 10^{-12}$	0.058 ± 0.002	$(1.47 \pm 0.10) \times 10^{-11}$	$(1.22 \pm 0.15) \times 10^{-4}$
STA2-1-912.69-DI	SPW-STa	Staffelegg Fm.	36.0	45.0	9.0	$(4.69 \pm 0.10) \times 10^{-12}$	0.111 ± 0.004	$(1.11 \pm 0.03) \times 10^{-12}$	0.070 ± 0.003	$(4.28 \pm 0.09) \times 10^{-12}$	$(6.90 \pm 0.44) \times 10^{-5}$

Tab. A-2: Cont.

Sample	SPW	Formation	Clay	Calcite	Quartz	HTO		³⁶ Cl ⁻		²² Na ⁺	
			[wt.%]	[wt.%]	[wt.%]	D_e [m ² s ⁻¹]	ϵ [-]	D_e [m ² s ⁻¹]	ϵ [-]	D_e [m ² s ⁻¹]	R_d [m ³ kg ⁻¹]
STA2-1-925.51-DI	SPW-STa	Staffelegg Fm.	40.9	9.0	38.0	$(1.00 \pm 0.03) \times 10^{-11}$	0.100 ± 0.006	$(2.33 \pm 0.03) \times 10^{-12}$	0.050 ± 0.001	$(1.28 \pm 0.06) \times 10^{-11}$	$(8.61 \pm 0.79) \times 10^{-5}$
STA2-1-930.53-DI	SPW-STa	Staffelegg Fm.	7.5	91.0	2.0	$(8.24 \pm 0.34) \times 10^{-12}$	0.074 ± 0.004	$(5.23 \pm 0.11) \times 10^{-12}$	0.065 ± 0.003	$(5.86 \pm 0.16) \times 10^{-12}$	$(1.41 \pm 0.27) \times 10^{-5}$
STA2-1-955.40-DI	SPW-STa	Klettgau Fm.	15.7	10.0	16.0	$(2.60 \pm 0.12) \times 10^{-11}$	0.091 ± 0.010	$(1.43 \pm 0.03) \times 10^{-11}$	0.074 ± 0.003	$(2.45 \pm 0.18) \times 10^{-11}$	$(3.36 \pm 0.56) \times 10^{-5}$
STA2-1-957.28-DI	SPW-STa	Klettgau Fm.	13.3	4.0	23.0	$(1.34 \pm 0.04) \times 10^{-11}$	0.067 ± 0.006	$(5.91 \pm 0.08) \times 10^{-12}$	0.050 ± 0.001	$(1.27 \pm 0.06) \times 10^{-11}$	$(2.97 \pm 0.45) \times 10^{-5}$
STA2-1-963.35-DI	SPW-STa	Klettgau Fm.	45.5	0.0	31.0	$(2.36 \pm 0.12) \times 10^{-11}$	0.125 ± 0.013	$(8.22 \pm 0.09) \times 10^{-12}$	0.091 ± 0.002	$(2.68 \pm 0.19) \times 10^{-11}$	$(7.81 \pm 0.11) \times 10^{-5}$
STA3-1-650.49-DI	SPW-STa	Wildegge Fm.	9.0	87.0	2.0	$(4.09 \pm 0.10) \times 10^{-12}$	0.035 ± 0.002	$(1.80 \pm 0.02) \times 10^{-12}$	0.024 ± 0.001	$(4.11 \pm 0.08) \times 10^{-12}$	$(1.75 \pm 0.17) \times 10^{-5}$
STA3-1-686.50-DI	SPW-STa	Wildegge Fm.	16.0	79.0	4.0	Advective flow determined – possible sample damage					
STA3-1-748.38-DI	SPW-STa	Wedelsandstein Fm.	33.2	31.0	25.0	$(9.30 \pm 0.28) \times 10^{-12}$	0.157 ± 0.009	$(1.62 \pm 0.03) \times 10^{-12}$	0.061 ± 0.002	$(1.24 \pm 0.08) \times 10^{-11}$	$(1.71 \pm 0.19) \times 10^{-4}$
STA3-1-758.75-DI	SPW-STa	Wedelsandstein Fm.	58.4	8.0	18.0	$(1.15 \pm 0.04) \times 10^{-11}$	0.150 ± 0.009	$(2.04 \pm 0.05) \times 10^{-12}$	0.059 ± 0.002	$(1.51 \pm 0.10) \times 10^{-11}$	$(1.56 \pm 0.16) \times 10^{-4}$
STA3-1-768.34-DI	SPW-STa	'Murchisonae-Oolith Fm.'	24.0	9.0	62.0	$(2.89 \pm 0.16) \times 10^{-11}$	0.094 ± 0.016	$(1.66 \pm 0.06) \times 10^{-11}$	0.082 ± 0.007	$(2.63 \pm 0.19) \times 10^{-11}$	$(3.51 \pm 0.69) \times 10^{-5}$
STA3-1-896.82-DI	SPW-STa	Staffelegg Fm.	28.2	54.0	8.0	$(6.02 \pm 0.16) \times 10^{-12}$	0.111 ± 0.006	$(1.35 \pm 0.02) \times 10^{-12}$	0.067 ± 0.002	$(5.11 \pm 0.25) \times 10^{-12}$	$(5.45 \pm 0.85) \times 10^{-5}$
STA3-1-919.66-DI	SPW-STa	Staffelegg Fm.	16.4	74.0	7.0	$(1.70 \pm 0.05) \times 10^{-12}$	0.021 ± 0.001	$(5.40 \pm 0.14) \times 10^{-13}$	0.007 ± 0.001	$(1.81 \pm 0.06) \times 10^{-12}$	$(1.56 \pm 0.17) \times 10^{-5}$

Tab. A-3: Overview of the effective diffusion coefficients (D_e) and diffusion-accessible porosities (ε) for HTO, $^{36}\text{Cl}^-$ and distribution coefficients (R_d) for $^{22}\text{Na}^+$ for samples from the TRU1-1 and MAR1-1 boreholes in the Zürich Nordost (ZNO) siting region

Sample	SPW	Formation	Clay	Calcite	Quartz	HTO		$^{36}\text{Cl}^-$		$^{22}\text{Na}^+$	
			[wt.%]	[wt.%]	[wt.%]	D_e [m ² s ⁻¹]	ε [-]	D_e [m ² s ⁻¹]	ε [-]	D_e [m ² s ⁻¹]	R_d [m ³ kg ⁻¹]
TRU1-1-620.33-DI	SPW-TRU	'Felsenkalke' / 'Massenkalk'	2.8	96.0	1.0	$(1.26 \pm 0.09) \times 10^{-12}$	0.017 ± 0.003	$(6.38 \pm 0.54) \times 10^{-13}$	0.012 ± 0.002	$(1.09 \pm 0.03) \times 10^{-12}$	$(5.87 \pm 0.65) \times 10^{-6}$
TRU1-1-660.04-DI	SPW-TRU	Villigen Fm.	2.8	93.0	2.0	$(6.59 \pm 0.10) \times 10^{-13}$	0.014 ± 0.001	$(1.66 \pm 0.05) \times 10^{-13}$	0.008 ± 0.001	$(8.28 \pm 0.10) \times 10^{-13}$	$(1.12 \pm 0.05) \times 10^{-5}$
TRU1-1-710.95-DI	SPW-TRU	Villigen Fm.	6.4	91.9	1.5	$(2.40 \pm 0.28) \times 10^{-12}$	0.025 ± 0.008	$(5.02 \pm 2.36) \times 10^{-13}$	0.008 ± 0.010	$(5.16 \pm 0.53) \times 10^{-12}$	$(1.59 \pm 0.53) \times 10^{-5}$
TRU1-1-728.55-DI	SPW-TRU	Variansmergel Fm.	49.3	17.5	19.2	$(1.62 \pm 0.07) \times 10^{-11}$	0.118 ± 0.009	$(5.00 \pm 0.22) \times 10^{-12}$	0.065 ± 0.005	$(1.99 \pm 0.10) \times 10^{-11}$	$(9.36 \pm 0.85) \times 10^{-5}$
TRU1-1-735.62-DI	SPW-TRU	Variansmergel Fm.	29.7	31.7	28.1	$(1.54 \pm 0.05) \times 10^{-11}$	0.127 ± 0.008	$(3.48 \pm 0.19) \times 10^{-12}$	0.061 ± 0.005	$(1.82 \pm 0.13) \times 10^{-11}$	$(9.34 \pm 0.71) \times 10^{-5}$
TRU1-1-748.98-DI	SPW-TRU	'Parkinsoni-Württembergica-Schichten'	43.5	26.9	16.0	$(1.80 \pm 0.07) \times 10^{-11}$	0.129 ± 0.010	$(4.25 \pm 0.15) \times 10^{-12}$	0.066 ± 0.004	$(1.87 \pm 0.12) \times 10^{-11}$	$(1.11 \pm 0.13) \times 10^{-4}$
TRU1-1-750.88-DI	SPW-TRU	'Parkinsoni-Württembergica-Schichten'	41.1	25.4	21.5	$(1.49 \pm 0.04) \times 10^{-11}$	0.113 ± 0.007	$(3.97 \pm 0.15) \times 10^{-12}$	0.059 ± 0.003	$(1.94 \pm 0.10) \times 10^{-11}$	$(9.38 \pm 0.81) \times 10^{-5}$
TRU1-1-756.39-DI_S1	SPW-TRU	'Parkinsoni-Württembergica-Schichten'	43.9	20.5	21.1	$(1.46 \pm 0.06) \times 10^{-11}$	0.131 ± 0.010	$(3.93 \pm 1.98) \times 10^{-12}$	0.202 ± 0.289	$(1.58 \pm 0.09) \times 10^{-11}$	$(1.13 \pm 0.11) \times 10^{-4}$
TRU1-1-756.39-DI_S2	SPW-TRU	'Parkinsoni-Württembergica-Schichten'	43.9	20.5	21.1	$(1.76 \pm 0.06) \times 10^{-11}$	0.130 ± 0.010	$(4.81 \pm 0.24) \times 10^{-12}$	0.065 ± 0.006	$(2.02 \pm 0.13) \times 10^{-11}$	$(9.75 \pm 1.13) \times 10^{-5}$
TRU1-1-778.32-DI	SPW-TRU	'Humphriesoolith Fm.'	38.7	19.9	31.0	$(1.72 \pm 0.06) \times 10^{-11}$	0.127 ± 0.009	$(4.31 \pm 0.18) \times 10^{-12}$	0.071 ± 0.005	$(2.14 \pm 0.14) \times 10^{-11}$	$(1.18 \pm 0.12) \times 10^{-4}$
TRU1-1-788.76-DI	SPW-TRU	Wedelsandstein Fm.	31.6	16.9	40.3	$(1.77 \pm 0.06) \times 10^{-11}$	0.108 ± 0.008	$(5.97 \pm 0.17) \times 10^{-12}$	0.065 ± 0.003	$(2.16 \pm 0.12) \times 10^{-11}$	$(7.74 \pm 0.79) \times 10^{-5}$
TRU1-1-794.56-DI	SPW-TRU	Wedelsandstein Fm.	48.7	13.8	27.2	$(1.47 \pm 0.05) \times 10^{-11}$	0.121 ± 0.009	$(3.15 \pm 0.11) \times 10^{-12}$	0.056 ± 0.003	$(1.86 \pm 0.11) \times 10^{-11}$	$(1.04 \pm 0.11) \times 10^{-4}$
TRU1-1-802.48-DI	SPW-TRU	Wedelsandstein Fm.	47.7	11.9	29.4	$(1.72 \pm 0.07) \times 10^{-11}$	0.157 ± 0.011	$(3.24 \pm 0.12) \times 10^{-12}$	0.064 ± 0.004	$(2.25 \pm 0.16) \times 10^{-11}$	$(1.62 \pm 0.16) \times 10^{-4}$

Tab. A-3: Cont.

Sample	SPW	Formation	Clay	Calcite	Quartz	HTO		³⁶ Cl ⁻		²² Na ⁺	
			[wt. %]	[wt. %]	[wt. %]	D_e [m ² s ⁻¹]	ϵ [-]	D_e [m ² s ⁻¹]	ϵ [-]	D_e [m ² s ⁻¹]	R_d [m ³ kg ⁻¹]
TRU1-1-807.74-DI	SPW-TRU	Wedelsandstein Fm.	13.5	30.7	46.8	$(4.62 \pm 0.12) \times 10^{-12}$	0.031 ± 0.002	$(1.84 \pm 0.07) \times 10^{-12}$	0.018 ± 0.001	$(5.07 \pm 0.13) \times 10^{-12}$	$(2.21 \pm 0.19) \times 10^{-5}$
TRU1-1-814.82-DI	SPW-TRU	Wedelsandstein Fm.	27.9	17.5	42.9	$(1.58 \pm 0.05) \times 10^{-11}$	0.096 ± 0.007	$(5.90 \pm 0.15) \times 10^{-12}$	0.064 ± 0.003	$(1.82 \pm 0.08) \times 10^{-11}$	$(6.03 \pm 0.60) \times 10^{-5}$
TRU1-1-815.28-DI	SPW-TRU	Wedelsandstein Fm.	23.3	23.3	41.6	$(1.51 \pm 0.05) \times 10^{-11}$	0.090 ± 0.007	$(5.48 \pm 0.17) \times 10^{-12}$	0.056 ± 0.003	$(1.78 \pm 0.08) \times 10^{-11}$	$(5.85 \pm 0.62) \times 10^{-5}$
TRU1-1-831.37-DI	SPW-TRU	Opalinus Clay	63.2	4.7	19.1	$(9.58 \pm 0.32) \times 10^{-12}$	0.143 ± 0.008	$(1.81 \pm 0.09) \times 10^{-12}$	0.061 ± 0.005	$(1.07 \pm 0.05) \times 10^{-11}$	$(1.19 \pm 0.12) \times 10^{-4}$
TRU1-1-839.97-DI	SPW-TRU	Opalinus Clay	51.4	6.1	29.9	$(8.99 \pm 0.28) \times 10^{-12}$	0.123 ± 0.007	$(1.82 \pm 0.10) \times 10^{-12}$	0.053 ± 0.005	$(1.09 \pm 0.05) \times 10^{-11}$	$(1.12 \pm 0.10) \times 10^{-4}$
TRU1-1-870.23-DI	SPW-TRU	Opalinus Clay	50.1	8.7	27.2	$(7.64 \pm 0.24) \times 10^{-12}$	0.121 ± 0.007	$(1.54 \pm 0.07) \times 10^{-12}$	0.053 ± 0.003	$(9.47 \pm 0.37) \times 10^{-12}$	$(1.16 \pm 0.09) \times 10^{-4}$
TRU1-1-882.36-DI	SPW-TRU	Opalinus Clay	56.7	7.2	22.0	$(1.23 \pm 0.05) \times 10^{-11}$	0.134 ± 0.009	$(2.37 \pm 0.10) \times 10^{-12}$	0.060 ± 0.004	$(1.65 \pm 0.09) \times 10^{-11}$	$(1.27 \pm 0.07) \times 10^{-4}$
TRU1-1-890.39-DI	SPW-TRU	Opalinus Clay	57.3	7.0	23.2	$(9.44 \pm 0.31) \times 10^{-12}$	0.129 ± 0.008	$(1.98 \pm 0.07) \times 10^{-12}$	0.058 ± 0.003	$(1.16 \pm 0.05) \times 10^{-11}$	$(1.21 \pm 0.10) \times 10^{-4}$
TRU1-1-902.85-DI	SPW-TRU	Opalinus Clay	55.4	6.8	24.0	$(8.07 \pm 0.26) \times 10^{-12}$	0.134 ± 0.007	$(1.53 \pm 0.04) \times 10^{-12}$	0.058 ± 0.002	$(1.05 \pm 0.04) \times 10^{-11}$	$(1.34 \pm 0.08) \times 10^{-4}$
TRU1-1-913.07-DI	SPW-TRU	Opalinus Clay	61.6	8.5	18.3	$(8.73 \pm 0.25) \times 10^{-12}$	0.121 ± 0.007	$(1.73 \pm 0.07) \times 10^{-12}$	0.053 ± 0.003	$(1.14 \pm 0.05) \times 10^{-11}$	$(1.22 \pm 0.10) \times 10^{-4}$
TRU1-1-923.34-DI	SPW-TRU	Opalinus Clay	66.4	6.1	15.0	$(6.17 \pm 0.30) \times 10^{-12}$	0.129 ± 0.009	$(6.86 \pm 0.6) \times 10^{-13}$	0.043 ± 0.008	$(3.05 \pm 0.12) \times 10^{-12}$	$(3.71 \pm 0.72) \times 10^{-5}$
TRU1-1-930.99-DI	SPW-TRU	Staffelegg Fm.	26.9	58.1	5.9	$(5.75 \pm 0.40) \times 10^{-12}$	0.048 ± 0.006	$(1.85 \pm 0.3) \times 10^{-12}$	0.026 ± 0.008	$(9.89 \pm 0.87) \times 10^{-12}$	$(6.49 \pm 1.27) \times 10^{-5}$
TRU1-1-938.90-DI	SPW-TRU	Staffelegg Fm.	35.6	42.8	10.1	$(2.95 \pm 0.21) \times 10^{-12}$	0.092 ± 0.009	$(4.93 \pm 0.7) \times 10^{-13}$	0.035 ± 0.008	$(2.63 \pm 0.07) \times 10^{-12}$	$(7.40 \pm 0.54) \times 10^{-5}$
TRU1-1-943.21-DI	SPW-TRU	Staffelegg Fm.	43.4	20.6	15.3	$(1.86 \pm 0.12) \times 10^{-12}$	0.099 ± 0.008	$(2.99 \pm 0.12) \times 10^{-13}$	0.047 ± 0.003	$(2.07 \pm 0.05) \times 10^{-12}$	$(1.05 \pm 0.05) \times 10^{-4}$
TRU1-1-955.02-DI	SPW-TRU	Staffelegg Fm.	50.4	10.2	28.60	$(9.53 \pm 0.29) \times 10^{-12}$	0.120 ± 0.007	$(1.68 \pm 0.08) \times 10^{-12}$	0.051 ± 0.004	$(1.40 \pm 0.07) \times 10^{-11}$	$(1.25 \pm 0.10) \times 10^{-4}$
TRU1-1-961.72-DI	SPW-TRU	Staffelegg Fm.	60.2	3.6	23.2	$(9.85 \pm 0.27) \times 10^{-12}$	0.134 ± 0.007	$(1.63 \pm 0.07) \times 10^{-12}$	0.049 ± 0.004	$(1.41 \pm 0.06) \times 10^{-11}$	$(1.48 \pm 0.10) \times 10^{-4}$
TRU1-1-1026.55-DI	SPW-TRU	Klettgau Fm.	56.3	1.3	5.1	$(1.38 \pm 0.06) \times 10^{-11}$	0.154 ± 0.010	$(2.73 \pm 0.12) \times 10^{-12}$	0.077 ± 0.005	$(1.59 \pm 0.09) \times 10^{-11}$	$(1.23 \pm 0.13) \times 10^{-4}$
MAR1-1-494.63-DI	SPW-TRU	Wildegge Fm.	34.0	56.0	7.0	$(1.66 \pm 0.06) \times 10^{-11}$	0.110 ± 0.009	$(3.68 \pm 0.07) \times 10^{-12}$	0.053 ± 0.002	$(2.08 \pm 0.15) \times 10^{-11}$	$(9.24 \pm 1.13) \times 10^{-5}$
MAR1-1-566.33-DI	SPW-TRU	Wedelsandstein Fm.	26.0	14.0	51.0	$(1.97 \pm 0.11) \times 10^{-11}$	0.103 ± 0.011	$(7.14 \pm 0.20) \times 10^{-12}$	0.071 ± 0.004	$(2.33 \pm 0.15) \times 10^{-11}$	$(7.84 \pm 0.91) \times 10^{-5}$

Tab. A-3: Cont.

Sample	SPW	Formation	Clay	Calcite	Quartz	HTO		³⁶ Cr		²² Na ⁺	
			[wt. %]	[wt. %]	[wt. %]	D_e [m ² s ⁻¹]	ϵ [-]	D_e [m ² s ⁻¹]	ϵ [-]	D_e [m ² s ⁻¹]	R_d [m ³ kg ⁻¹]
MAR1-1-570.12-DI	SPW-TRU	Wedelsandstein Fm.	9.0	68.0	19.0	$(2.11 \pm 0.09) \times 10^{-11}$	0.082 ± 0.008	$(1.32 \pm 0.02) \times 10^{-11}$	0.073 ± 0.002	$(1.81 \pm 0.08) \times 10^{-11}$	$(3.17 \pm 0.33) \times 10^{-5}$
MAR1-1-588.40-DI	SPW-TRU	Wedelsandstein Fm.	19.0	28.0	43.0	$(7.60 \pm 0.43) \times 10^{-12}$	0.056 ± 0.007	$(3.65 \pm 0.22) \times 10^{-12}$	0.041 ± 0.005	$(9.55 \pm 0.71) \times 10^{-12}$	$(3.31 \pm 0.76) \times 10^{-5}$
MAR1-1-717.76-DI	SPW-TRU	Staffelegg Fm.	37.0	45.0	9.0	$(6.14 \pm 0.19) \times 10^{-12}$	0.113 ± 0.007	$(1.03 \pm 0.04) \times 10^{-12}$	0.047 ± 0.003	$(5.63 \pm 0.24) \times 10^{-12}$	$(7.65 \pm 0.88) \times 10^{-5}$
MAR1-1-729.30-DI	SPW-TRU	Staffelegg Fm.	56.0	9.0	27.0	$(1.10 \pm 0.04) \times 10^{-11}$	0.120 ± 0.008	$(1.92 \pm 0.04) \times 10^{-12}$	0.048 ± 0.002	$(1.34 \pm 0.09) \times 10^{-11}$	$(1.16 \pm 0.14) \times 10^{-4}$
MAR1-1-738.21-DI	SPW-TRU	Staffelegg Fm.	13.0	85.0	2.0	$(8.70 \pm 0.34) \times 10^{-12}$	0.044 ± 0.005	$(6.10 \pm 0.21) \times 10^{-12}$	0.036 ± 0.003	$(7.19 \pm 0.50) \times 10^{-12}$	$(1.67 \pm 0.48) \times 10^{-5}$
MAR1-1-742.25-DI	SPW-TRU	Staffelegg Fm.	61.0	3.0	34.0	$(9.65 \pm 0.27) \times 10^{-12}$	0.155 ± 0.008	$(1.18 \pm 0.05) \times 10^{-12}$	0.048 ± 0.003	$(1.22 \pm 0.08) \times 10^{-11}$	$(1.75 \pm 0.20) \times 10^{-4}$

Tab. A-4: Overview of the diffusion-accessible porosities (ε) for HTO, and water-loss porosity measured at PSI and the University of Bern for samples from the BOZ1-1 and BOZ2-1 boreholes in the Jura Ost (JO) siting region

Sample	SPW	Formation	Clay	Calcite	Quartz	HTO diffusion @ PSI		H ₂ O loss @ PSI		H ₂ O loss @ University of Bern	
			[wt.%]	[wt.%]	[wt.%]	ε [-]	Uncertainty	ε [-]	Uncertainty	ε [-]	Uncertainty
BOZ1-1-266.75-DI	SPW-BOZ	Wildegge Fm.	24.0	58.1	10.4	0.094	0.007	0.095	0.004	0.104	0.0071
BOZ1-1-290.63-DI	SPW-BOZ	Wildegge Fm.	19.0	69.6	5.7	0.071	0.006	0.069	0.003	0.068	0.0074
BOZ1-1-323.16-DI	SPW-BOZ	Wildegge Fm.	11.4	81.3	3.0	0.063	0.004	0.061	0.002	0.057	0.0075
BOZ1-1-353.36-DI	SPW-BOZ	Wildegge Fm.	31.4	55.0	7.9	0.099	0.007	0.100	0.003	0.101	0.0071
BOZ1-1-391.30-DI	SPW-BOZ	Wildegge Fm.	24.2	67.0	5.5	0.080	0.005	0.080	0.002	0.077	0.0073
BOZ1-1-416.35-DI	SPW-BOZ	Hauptrogenstein	15.5	69.9	2.1	0.116	0.008	0.134	0.004	0.132	0.0071
BOZ1-1-434.29-DI	SPW-BOZ	Klingnau Fm.	15.2	74.3	4.4	0.074	0.005	0.072	0.002	0.083	0.0074
BOZ1-1-438.20-DI	SPW-BOZ	Klingnau Fm.	27.2	56.2	4.1	0.119	0.009	0.131	0.005	0.130	0.0071
BOZ1-1-440.55-DI	SPW-BOZ	Klingnau Fm.	28.0	55.3	6.0	0.113	0.008	0.113	0.004	0.108	0.0072
BOZ1-1-487.01-DI	SPW-BOZ	Passwang Fm.	36.3	33.6	19.9	0.129	0.007	0.123	0.004	0.129	0.0069
BOZ1-1-493.15-DI	SPW-BOZ	Passwang Fm.	35.2	34.6	21.5	0.133	0.011	0.123	0.004	0.113	0.0070
BOZ1-1-498.32-DI	SPW-BOZ	Passwang Fm.	44.2	30.0	18.6	0.118	0.008	0.123	0.004	0.114	0.0070
BOZ1-1-503.87-DI	SPW-BOZ	Passwang Fm.	28.7	49.5	14.9	0.101	0.007	0.095	0.003	0.086	0.0073
BOZ1-1-512.03-DI	SPW-BOZ	Passwang Fm.	45.2	18.8	27.1	0.127	0.007	0.134	0.005	0.128	0.0069
BOZ1-1-521.50-DI	SPW-BOZ	Passwang Fm.	41.3	16.2	31.3	0.128	0.008	0.133	0.004	0.128	0.0068
BOZ1-1-529.56-DI	SPW-BOZ	Passwang Fm.	46.6	3.1	39.7	0.127	0.009	0.132	0.005	0.121	0.0069
BOZ1-1-550.63-DI	SPW-BOZ	Opalinus Clay	48.5	14.0	27.5	0.120	0.005	0.116	0.004	0.115	0.0070
BOZ1-1-562.07-DI	SPW-BOZ	Opalinus Clay	58.2	9.7	20.9	0.119	0.006	0.123	0.005	0.114	0.0070
BOZ1-1-572.43-DI	SPW-BOZ	Opalinus Clay	42.9	14.6	31.1	0.095	0.006	0.096	0.003	0.099	0.0071
BOZ1-1-580.34-DI	SPW-BOZ	Opalinus Clay	52.2	11.2	25.1	0.121	0.006	0.122	0.004	0.118	0.0069
BOZ1-1-597.28-DI	SPW-BOZ	Opalinus Clay	61.0	5.7	20.3	0.137	0.006	0.136	0.005	0.127	0.0069
BOZ1-1-610.79-DI	SPW-BOZ	Opalinus Clay	67.0	4.1	14.7	0.134	0.006	0.129	0.007	0.128	0.0069
BOZ1-1-619.10-DI	SPW-BOZ	Opalinus Clay	68.4	4.0	16.1	0.143	0.006	0.138	0.006	0.130	0.0068

Tab. A-4: Cont.

Sample	SPW	Formation	Clay	Calcite	Quartz	HTO diffusion @ PSI		H ₂ O loss @ PSI		H ₂ O loss @ University of Bern	
			[wt.%]	[wt.%]	[wt.%]	ε [-]	Uncertainty	ε [-]	Uncertainty	ε [-]	Uncertainty
BOZ1-1-623.60-DI	SPW-BOZ	Opalinus Clay	69.0	3.1	16.0	0.081	0.005	0.143	0.005	0.138	0.0067
BOZ1-1-634.90-DI	SPW-BOZ	Opalinus Clay	67.2	4.8	14.5	0.149	0.007	0.150	0.006	0.137	0.0068
BOZ1-1-646.44-DI	SPW-BOZ	Opalinus Clay	63.3	8.2	15.8	0.135	0.006	0.135	0.005	0.123	0.0069
BOZ1-1-665.00-DI	SPW-BOZ	Staffelegg Fm.	28.5	9.6	44.3	0.094	0.007	0.098	0.006	0.095	0.0072
BOZ1-1-670.95-DI	SPW-BOZ	Staffelegg Fm.	36.7	19.2	30.9	0.096	0.006	0.092	0.003	0.097	0.0071
BOZ1-1-679.67-DI	SPW-BOZ	Staffelegg Fm.	64.1	15.7	13.5	0.152	0.007	0.145	0.005	0.133	0.0068
BOZ1-1-681.57-DI	SPW-BOZ	Staffelegg Fm.	79.1	8.0	9.1	0.167	0.006	0.157	0.006	0.154	0.0067
BOZ1-1-686.81-DI	SPW-BOZ	Staffelegg Fm.	61.8	6.4	17.1	0.176	0.008	0.159	0.005	0.146	0.0067
BOZ1-1-717.96-DI	SPW-BOZ	Klettgau Fm.	39.7	0.0	28.8	0.125	0.009	0.128	0.005	0.131	0.0068
BOZ1-1-723.97-DI	SPW-BOZ	Bänkerjoch Fm.	44.1	0.0	11.2	0.115	0.008	0.111	0.003	0.117	0.0071
BOZ2-1-313.13-DI	SPW-BOZ	Hauptrogenstein	25.2	59.0	1.0	0.145	0.014	0.167	0.006	0.166	0.008
BOZ2-1-443.93-DI	SPW-BOZ	Passwang Fm.	38.5	16.0	34.0	0.109	0.008	0.123	0.004	0.110	0.008
BOZ2-1-588.64-DI	SPW-BOZ	Staffelegg Fm.	34.2	8.0	48.0	0.099	0.008	0.112	0.003	0.096	0.008
BOZ2-1-595.15-DI	SPW-BOZ	Staffelegg Fm.	43.1	11.0	37.0	0.121	0.010	0.136	0.005	0.097	0.008
BOZ2-1-606.95-DI	SPW-BOZ	Staffelegg Fm.	70.1	10.0	12.0	0.159	0.007	0.166	0.007	0.146	0.008

Tab. A-5: Overview of the diffusion-accessible porosities (ε) for HTO, and water-loss porosity measured at PSI and the University of Bern for samples from the BUL1-1, STA2-1 and STA3-1 boreholes in the Nördlich Lägern (NL) siting region

Sample	SPW	Formation	Clay	Calcite	Quartz	HTO diffusion @ PSI		H ₂ O loss @ PSI		H ₂ O loss @ University of Bern	
			[wt.%]	[wt.%]	[wt.%]	ε [-]	Uncertainty	ε [-]	Uncertainty	ε [-]	Uncertainty
BUL1-1-734.45-DI	SPW-BUL	Villigen Fm.	10.6	87.5	1.6	0.038	0.003	0.041	0.001	0.034	0.002
BUL1-1-752.48-DI	SPW-BUL	Wildegge Fm.	18.4	64.0	7.0	0.048	0.004	0.056	0.002	0.043	0.003
BUL1-1-766.65-DI	SPW-BUL	Wildegge Fm.	17.6	73.0	5.0	0.055	0.004	0.064	0.003	0.054	0.004
BUL1-1-794.35-DI	SPW-BUL	Wildegge Fm.	22.3	69.0	5.0	0.064	0.004	0.074	0.003	0.056	0.004
BUL1-1-808.92-DI	SPW-BUL	'Parkinsoni-Württembergica-Schichten'	51.5	26.0	15.0	0.131	0.011	0.159	0.009	0.133	0.009
BUL1-1-840.45-DI	SPW-BUL	'Herrenwis Unit'	5.3	78.0	10.0	0.012	0.002	0.040	0.001	0.014	0.001
BUL1-1-840.45-DI	SPW-BUL	'Herrenwis Unit'	5.3	78.0	10.0	0.032	0.003	0.026	0.001	0.015	0.001
BUL1-1-845.05-DI	SPW-BUL	'Herrenwis Unit'	1.0	98.0	1.0	0.011	0.001	0.055	0.002	0.015	0.001
BUL1-1-851.50-DI	SPW-BUL	'Herrenwis Unit'	6.8	90.0	3.0	0.033	0.006	0.021	0.001	0.011	0.001
BUL1-1-872.35-DI	SPW-BUL	Wedelsandstein Fm.	34.6	16.0	37.0	0.095	0.008	0.114	0.008	0.081	0.005
BUL1-1-879.20-DI	SPW-BUL	Wedelsandstein Fm.	39.4	13.0	37.0	0.082	0.008	0.097	0.003	0.092	0.006
BUL1-1-908.93-DI	SPW-BUL	Opalinus Clay	57.3	4.0	24.0	0.113	0.008	0.138	0.011	0.115	0.007
BUL1-1-914.61-DI	SPW-BUL	Opalinus Clay	54.0	7.0	25.0	0.115	0.007	0.133	0.004	0.104	0.007
BUL1-1-921.43-DI	SPW-BUL	Opalinus Clay	48.5	10.0	28.0	0.111	0.007	0.137	0.010	0.092	0.006
BUL1-1-921.43-DI	SPW-BUL	Opalinus Clay	48.5	10.0	28.0	0.088	0.007	0.109	0.003	0.092	0.006
BUL1-1-932.82-DI	SPW-BUL	Opalinus Clay	58.1	6.0	23.0	0.120	0.006	0.137	0.008	0.107	0.007
BUL1-1-945.46-DI	SPW-BUL	Opalinus Clay	61.2	4.0	18.0	0.131	0.007	0.155	0.006	0.130	0.008
BUL1-1-955.62-DI	SPW-BUL	Opalinus Clay	69.2	4.0	15.0	0.133	0.007	0.154	0.010	0.122	0.008
BUL1-1-955.62-DI	SPW-BUL	Opalinus Clay	69.2	4.0	15.0	0.108	0.007	0.127	0.004	0.122	0.008
BUL1-1-965.46-DI	SPW-BUL	Opalinus Clay	59.4	8.0	18.0	0.120	0.007	0.140	0.005	0.115	0.007
BUL1-1-969.15-DI	SPW-BUL	Opalinus Clay	48.4	13.0	25.0	0.083	0.005	0.100	0.004	0.087	0.006

Tab. A-5: Cont.

Sample	SPW	Formation	Clay	Calcite	Quartz	HTO diffusion @ PSI		H ₂ O loss @ PSI		H ₂ O loss @ University of Bern	
			[wt.%]	[wt.%]	[wt.%]	ε [-]	Uncertainty	ε [-]	Uncertainty	ε [-]	Uncertainty
BUL1-1-975.04-DI	SPW-BUL	Opalinus Clay	53.7	7.0	26.0	0.115	0.006	0.133	0.007	0.096	0.006
BUL1-1-984.49-DI	SPW-BUL	Opalinus Clay	63.2	6.0	17.0	0.123	0.007	0.141	0.003	0.114	0.007
BUL1-1-995.09-DI	SPW-BUL	Opalinus Clay	62.9	9.0	16.0	0.127	0.006	0.139	0.007	0.114	0.007
BUL1-1-1002.19-DI	SPW-BUL	Staffelegg Fm.	33.3	47.0	9.0	0.109	0.006	0.116	0.005	0.095	0.006
BUL1-1-1008.42-DI	SPW-BUL	Staffelegg Fm.	5.2	93.0	2.0	0.019	0.001	0.026	0.001	0.013	0.001
BUL1-1-1014.23-DI	SPW-BUL	Staffelegg Fm.	40.2	6.0	41.0	0.096	0.008	0.112	0.004	0.086	0.006
BUL1-1-1017.33-DI	SPW-BUL	Staffelegg Fm.	42.0	7.0	37.0	0.094	0.008	0.101	0.004	0.089	0.006
BUL1-1-1025.96-DI	SPW-BUL	Staffelegg Fm.	22.7	61.0	12.0	0.043	0.003	0.053	0.002	0.050	0.003
STA2-1-633.23-DI	SPW-STA	Villigen Fm.	10.5	86.0	2.0	0.042	0.007	0.046	0.002	0.044	0.008
STA2-1-752.73-DI	SPW-STA	'Parkinsoni-Württembergica-Schichten'	42.6	22.0	22.0	0.113	0.009	0.121	0.004	0.135	0.008
STA2-1-758.02-DI	SPW-STA	'Parkinsoni-Württembergica-Schichten'	46.0	23.0	20.0	0.122	0.009	0.132	0.004	0.128	0.008
STA2-1-758.23-DI	SPW-STA	'Parkinsoni-Württembergica-Schichten'	45.0	22.0	21.0	0.131	0.010	0.155	0.008	0.128	0.008
STA2-1-765.21-DI	SPW-STA	'Parkinsoni-Württembergica-Schichten'	51.0	23.0	15.0	0.134	0.010	0.143	0.005	0.130	0.008
STA2-1-779.96-DI	SPW-STA	Wedelsandstein Fm.	61.0	11.0	23.0	0.141	0.009	0.159	0.006	0.140	0.008
STA2-1-912.69-DI	SPW-STA	Staffelegg Fm.	36.0	45.0	9.0	0.111	0.004	0.106	0.003	0.104	0.007
STA2-1-925.51-DI	SPW-STA	Staffelegg Fm.	40.9	9.0	38.0	0.100	0.006	0.103	0.003	0.094	0.008
STA2-1-930.53-DI	SPW-STA	Staffelegg Fm.	7.5	91.0	2.0	0.074	0.004	0.083	0.003	0.030	0.008

Tab. A-5: Cont.

Sample	SPW	Formation	Clay	Calcite	Quartz	HTO diffusion @ PSI		H ₂ O loss @ PSI		H ₂ O loss @ University of Bern	
			[wt.%]	[wt.%]	[wt.%]	ε [-]	Uncertainty	ε [-]	Uncertainty	ε [-]	Uncertainty
STA2-1-955.40-DI	SPW-STA	Klettgau Fm.	15.7	10.0	16.0	0.091	0.010	0.114	0.004	0.092	0.008
STA2-1-957.28-DI	SPW-STA	Klettgau Fm.	13.3	4.0	23.0	0.067	0.006	0.077	0.003	0.089	0.008
STA2-1-963.35-DI	SPW-STA	Klettgau Fm.	45.5	0.0	31.0	0.125	0.013	0.155	0.005	0.151	0.007
STA3-1-650.49-DI	SPW-STA	Wildegge Fm.	9.0	87.0	2.0	0.035	0.002	0.034	0.001	0.036	0.008
STA3-1-686.50-DI	SPW-STA	Wildegge Fm.	16.0	79.0	4.0	0.099	0.008	0.052	0.002	0.047	0.008
STA3-1-748.38-DI	SPW-STA	Wedelsandstein Fm.	33.2	31.0	25.0	0.157	0.009	0.194	0.107	0.125	0.008
STA3-1-758.75-DI	SPW-STA	Wedelsandstein Fm.	58.4	8.0	18.0	0.150	0.009	0.16	0.008	0.137	0.008
STA3-1-768.34-DI	SPW-STA	'Murchisonae-Oolith Fm.'	24.0	9.0	62.0	0.094	0.016	0.122	0.004	0.113	0.008
STA3-1-896.82-DI	SPW-STA	Staffelegg Fm.	28.2	54.0	8.0	0.111	0.006	0.108	0.004	0.092	0.008
STA3-1-919.66-DI	SPW-STA	Staffelegg Fm.	16.4	74.0	7.0	0.021	0.001	0.021	0.001	0.045	0.008

Tab. A-6: Overview of the diffusion-accessible porosities (ε) for HTO, and water-loss porosity measured at PSI and the University of Bern for samples from the TRU1-1 and MAR1-1 boreholes in the Zürich Nordost (ZNO) siting region

Sample	SPW	Formation	Clay	Calcite	Quartz	HTO diffusion @ PSI		H ₂ O loss @ PSI		H ₂ O loss @ University of Bern	
			[wt.%]	[wt.%]	[wt.%]	ε [-]	Uncertainty	ε [-]	Uncertainty	ε [-]	Uncertainty
TRU1-1-620.33-DI	SPW-TRU	'Felsenkalke' / 'Massenkalk'	2.8	96.0	1.0	0.017	0.003	0.019	0.001	0.025	0.002
TRU1-1-660.04-DI	SPW-TRU	Villigen Fm.	2.8	93.0	2.0	0.014	0.001	0.013	0.001	0.013	0.001
TRU1-1-710.95-DI	SPW-TRU	Villigen Fm.	6.4	91.9	1.5	0.025	0.008	0.024	0.001	0.027	0.002
TRU1-1-728.55-DI	SPW-TRU	Variansmergel Fm.	49.3	17.5	19.2	0.118	0.009	0.124	0.005	0.124	0.008
TRU1-1-735.62-DI	SPW-TRU	Variansmergel Fm.	29.7	31.7	28.1	0.127	0.008	0.129	0.004	0.090	0.006
TRU1-1-748.98-DI	SPW-TRU	'Parkinsoni-Württembergica-Schichten'	43.5	26.9	16.0	0.129	0.010	0.136	0.005	0.129	0.009
TRU1-1-750.88-DI	SPW-TRU	'Parkinsoni-Württembergica-Schichten'	41.1	25.4	21.5	0.113	0.007	0.118	0.004	0.114	0.008
TRU1-1-756.39-DI_S1	SPW-TRU	'Parkinsoni-Württembergica-Schichten'	43.9	20.5	21.1	0.131	0.010	0.128	0.005	0.115	0.008
TRU1-1-756.39-DI_S2	SPW-TRU	'Parkinsoni-Württembergica-Schichten'	43.9	20.5	21.1	0.130	0.010	0.133	0.006	0.115	0.008
TRU1-1-778.32-DI	SPW-TRU	'Humphriesoolith Fm.'	38.7	19.9	31.0	0.127	0.009	0.131	0.005	0.130	0.009
TRU1-1-788.76-DI	SPW-TRU	Wedelsandstein Fm.	31.6	16.9	40.3	0.108	0.008	0.111	0.004	0.099	0.007
TRU1-1-794.56-DI	SPW-TRU	Wedelsandstein Fm.	48.7	13.8	27.2	0.121	0.009	0.122	0.008	0.116	0.008
TRU1-1-802.48-DI	SPW-TRU	Wedelsandstein Fm.	47.7	11.9	29.4	0.157	0.011	0.159	0.009	0.132	0.009
TRU1-1-807.74-DI	SPW-TRU	Wedelsandstein Fm.	13.5	30.7	46.8	0.031	0.002	0.029	0.002	0.057	0.004

Tab. A-6: Cont.

Sample	SPW	Formation	Clay	Calcite	Quartz	HTO diffusion @ PSI		H ₂ O loss @ PSI		H ₂ O loss @ University of Bern	
			[wt.%]	[wt.%]	[wt.%]	ε [-]	Uncertainty	ε [-]	Uncertainty	ε [-]	Uncertainty
TRU1-1-814.82-DI	SPW-TRU	Wedelsandstein Fm.	27.9	17.5	42.9	0.096	0.007	0.100	0.003	0.088	0.006
TRU1-1-815.28-DI	SPW-TRU	Wedelsandstein Fm.	23.3	23.3	41.6	0.090	0.007	0.093	0.003	0.086	0.006
TRU1-1-831.37-DI	SPW-TRU	Opalinus Clay	63.2	4.7	19.1	0.143	0.008	0.148	0.005	0.116	0.008
TRU1-1-839.97-DI	SPW-TRU	Opalinus Clay	51.4	6.1	29.9	0.123	0.007	0.121	0.006	0.110	0.007
TRU1-1-870.23-DI	SPW-TRU	Opalinus Clay	50.1	8.7	27.2	0.121	0.007	0.113	0.004	0.092	0.006
TRU1-1-882.36-DI	SPW-TRU	Opalinus Clay	56.7	7.2	22.0	0.134	0.009	0.133	0.004	0.117	0.008
TRU1-1-890.39-DI	SPW-TRU	Opalinus Clay	57.3	7.0	23.2	0.129	0.008	0.123	0.006	0.097	0.006
TRU1-1-902.85-DI	SPW-TRU	Opalinus Clay	55.4	6.8	24.0	0.134	0.007	0.127	0.006	0.102	0.007
TRU1-1-913.07-DI	SPW-TRU	Opalinus Clay	61.6	8.5	18.3	0.121	0.007	0.123	0.006	0.115	0.008
TRU1-1-923.34-DI	SPW-TRU	Opalinus Clay	66.4	6.1	15.0	0.129	0.009	0.139	0.005	0.115	0.008
TRU1-1-930.99-DI	SPW-TRU	Staffelegg Fm.	26.9	58.1	5.9	0.048	0.006	0.048	0.002	0.078	0.005
TRU1-1-938.90-DI	SPW-TRU	Staffelegg Fm.	35.6	42.8	10.1	0.092	0.009	0.097	0.002	0.103	0.007
TRU1-1-943.21-DI	SPW-TRU	Staffelegg Fm.	43.4	20.6	15.3	0.099	0.008	0.100	0.003	0.105	0.007
TRU1-1-955.02-DI	SPW-TRU	Staffelegg Fm.	50.4	10.2	28.6	0.120	0.007	0.112	0.005	0.099	0.007
TRU1-1-961.72-DI	SPW-TRU	Staffelegg Fm.	60.2	3.6	23.2	0.134	0.007	0.124	0.005	0.109	0.007
TRU1-1-1026.55-DI	SPW-TRU	Klettgau Fm.	56.3	1.3	5.1	0.154	0.010	0.148	0.007	0.130	0.009
MAR1-1-494.63-DI	SPW-TRU	Wildeggen Fm.	33.6	56.0	7.0	0.110	0.009	0.126	0.004	0.103	0.008
MAR1-1-566.33-DI	SPW-TRU	Wedelsandstein Fm.	26.0	14.0	51.0	0.103	0.011	0.121	0.004	0.113	0.007
MAR1-1-570.12-DI	SPW-TRU	Wedelsandstein Fm.	8.5	68.0	19.0	0.082	0.008	0.104	0.004	0.028	0.008
MAR1-1-588.40-DI	SPW-TRU	Wedelsandstein Fm.	18.9	28.0	43.0	0.056	0.007	0.053	0.002	0.070	0.008
MAR1-1-717.76-DI	SPW-TRU	Staffelegg Fm.	37.3	45.0	9.0	0.113	0.007	0.143	0.005	0.098	0.007

Tab. A-6: Cont.

Sample	SPW	Formation	Clay	Calcite	Quartz	HTO diffusion @ PSI		H ₂ O loss @ PSI		H ₂ O loss @ University of Bern	
			[wt.%]	[wt.%]	[wt.%]	ε [-]	Uncertainty	ε [-]	Uncertainty	ε [-]	Uncertainty
MAR1-1-729.30-DI	SPW-TRU	Staffelegg Fm.	56.5	9.0	27.0	0.120	0.008	0.129	0.005	0.113	0.008
MAR1-1-738.21-DI	SPW-TRU	Staffelegg Fm.	12.8	85.0	2.0	0.044	0.005	0.061	0.002	0.017	0.008
MAR1-1-742.25-DI	SPW-TRU	Staffelegg Fm.	61.4	3.0	34.0	0.155	0.008	0.159	0.006	0.140	0.008

App. B Auxiliary rock properties

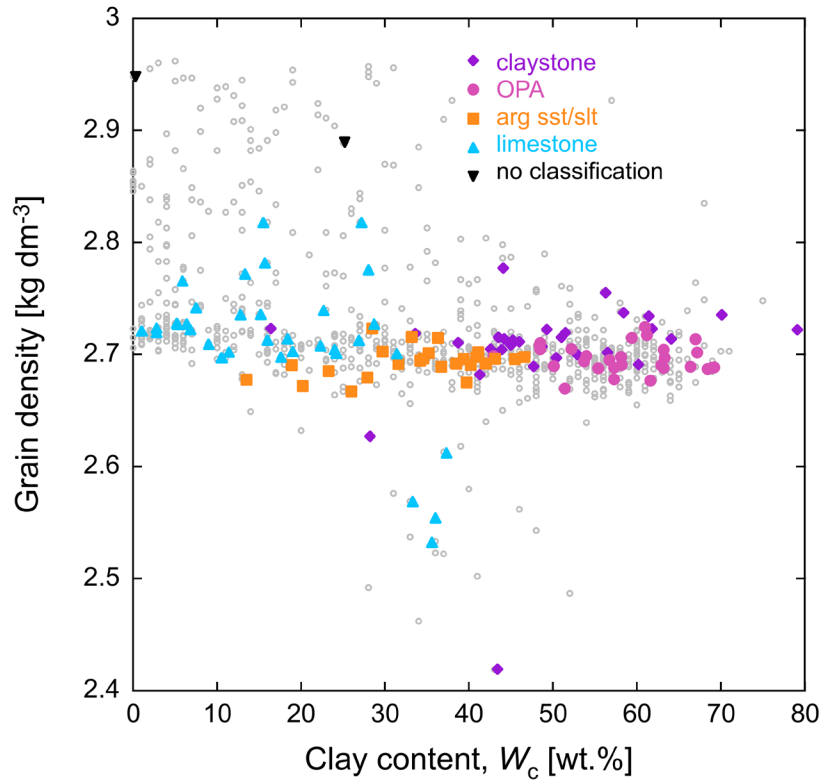


Fig. B-1: Grain density of rock samples as a function of clay content

The data shown in colours represent the rock samples used for the diffusion experiment. Small grey circles represent the entirety of all rock samples investigated (Mazurek et al. 2023). The grain density of the majority of samples is around 2.7 kg dm⁻³. Larger values are generally observed for samples containing large amounts of dolomite, anhydrite and some Fe-oolites. Lower values are related to samples containing organic carbon > 2%. All samples studied in the present work with grain density < 2.6 kg dm⁻³ belong to the Rietheim member.

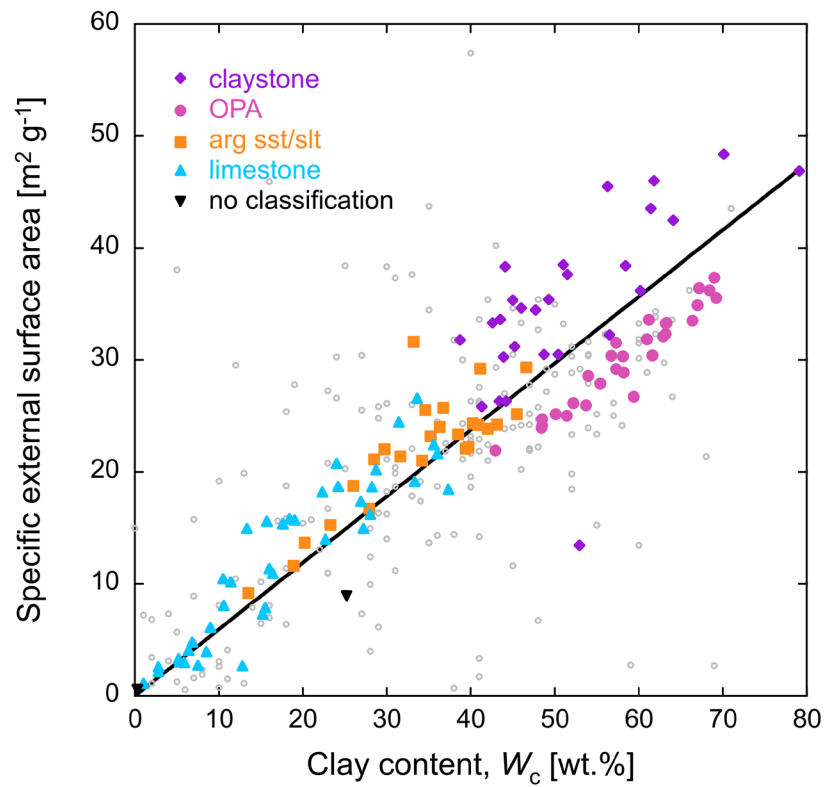


Fig. B-2: Specific external surface area of rock samples as a function of clay content

The data shown in colours represent the rock samples used for the diffusion experiment, small grey circles represent the entirety of all rock samples investigated (Mazurek et al. 2023).

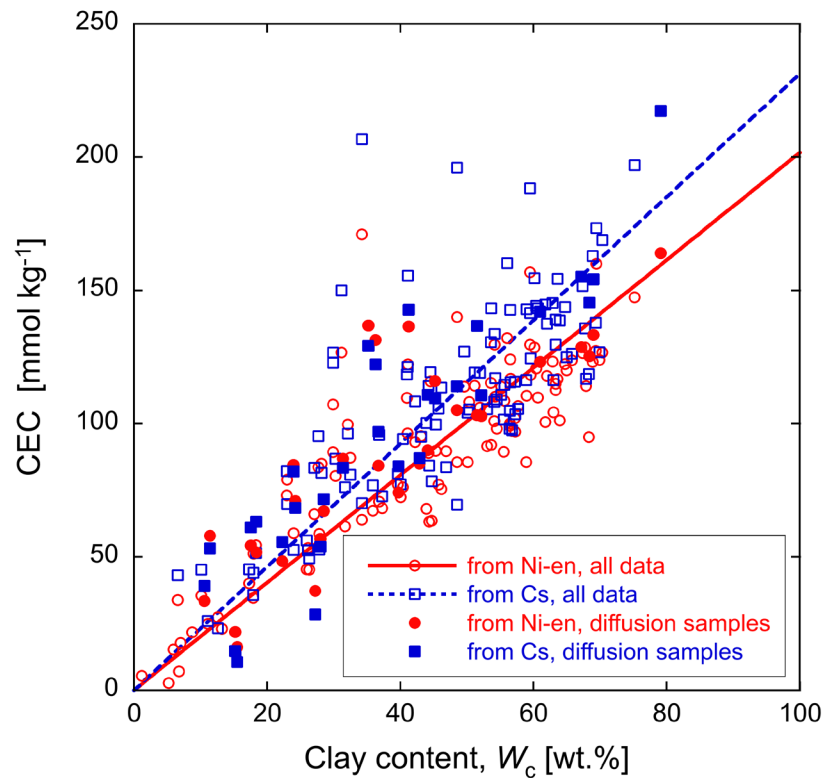


Fig. B-3: Cation-exchange capacity (CEC) of rock samples measured by two different methods (Ni-ethylenediamine and Cs isotope exchange) as a function of clay content

As such data are not available for all the rock samples used for the diffusion experiments, the fit curves were obtained from regression of all rock samples investigated.

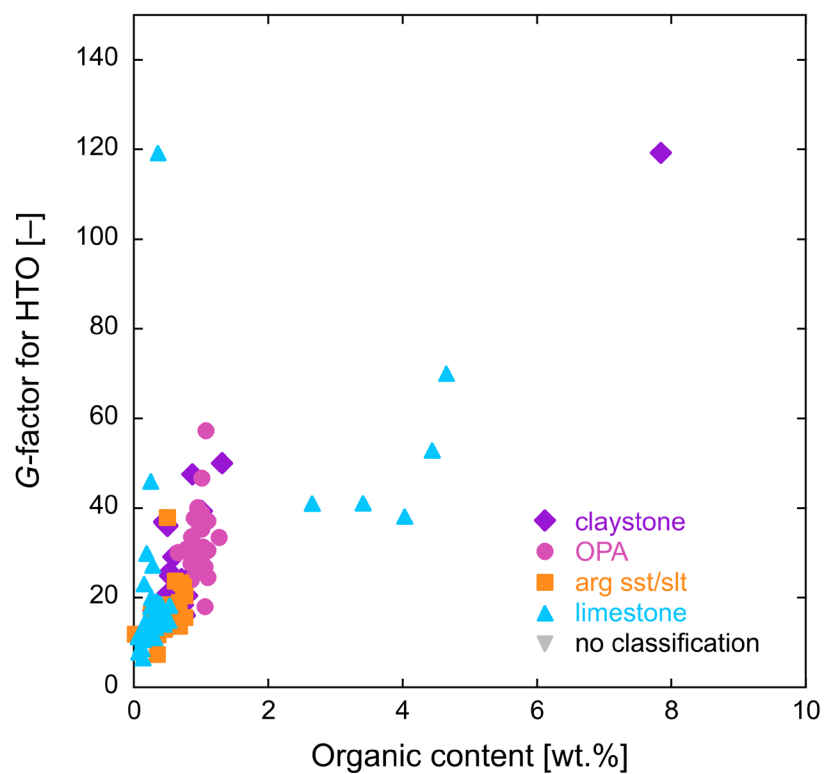


Fig. B-4: G -factor of HTO as a function of the content of organic material

These values are only available for the rock samples used for diffusion measurements.

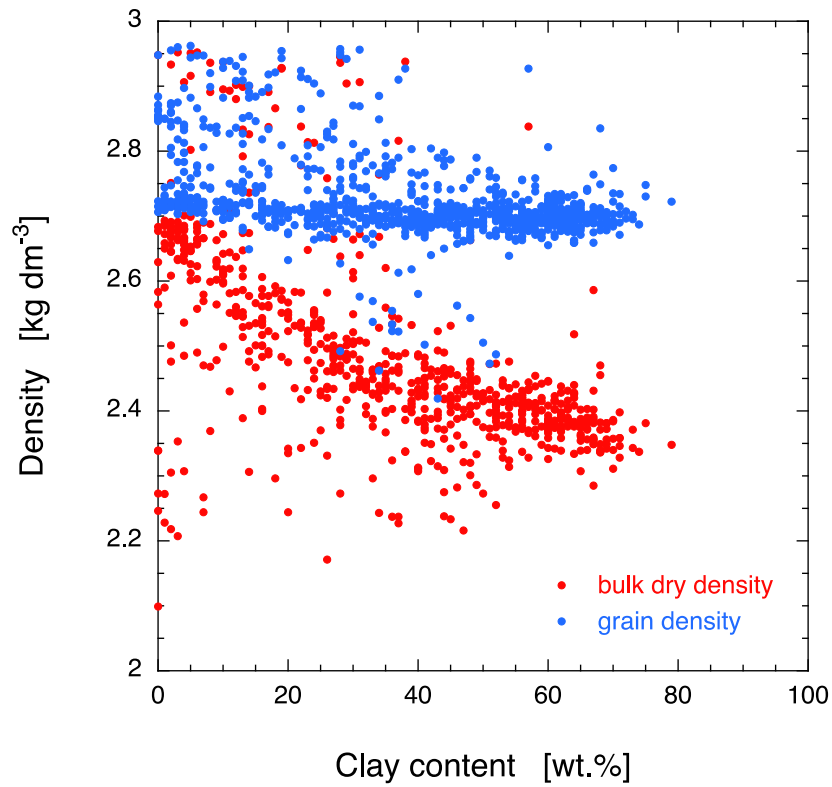


Fig. B-5: Grain and bulk dry density of rock samples as a function of clay content
All data were taken from Mazurek et al. (2023).

Tab. B-1: Overview of constants, parameters and variables used for the EDL simulations

Quantity	Symbol	Units	Value	Function of W_c
Constants				
Faraday constant	F	C mol ⁻¹	96,485	
Molar gas constant	R	J K ⁻¹ mol ⁻¹	8.314	
Diffusion coefficient in free water (25 °C)	D_w	m ² s ⁻¹	^a	
Ratio between Donnan thickness and Debye length	n_{DL}	None	2	
Specific surface area	A_{sp}	m ² g ⁻¹	100	
Grain density of the total clay rock	ρ_{gr}	kg m ⁻³	2,700	
Cation-exchange capacity (100% clay)	CEC ₁₀₀	mol kg ⁻¹	0.21	
Empirical parameters				
-	m_1	None	0.1444	
-	m_2	wt.% ⁻¹	0.0398	
-	a_1	None	10	
-	a_2	wt.% ⁻¹	0.0515	
Variables				
Weight fraction of the clay phase	f_{clay}	None		Yes; Sect. 4.2.3.1
Total clay content in rock	W_c	wt.%		
(Diffusion) accessible porosity, total porosity	ε	None		Yes; Eq. (4-2)
Geometrical factor	G	None		Yes; Eq. (4-3)
Solid:liquid ratio	r_{SL}	kg m ⁻³		Yes; Eq. (4-19)
Bulk dry density of the total rock	ρ_{br}	kg m ⁻³		Yes; Eq. (4-19)
Pore width	d_p	m		Yes; Eq. (4-16)
Pore volume fraction of free porewater	f_{free}	None		Yes; Eq. (4-18)
Pore volume fraction of Donnan layer	f_{DL}	None		Yes; Eq. (4-18)
Cation exchange capacity on a volumetric basis	CEC _{vol}	mol m ⁻³		Yes; Eq. (4-20)
Donnan thickness	d_{DL}	m		No; Eq. (4-17)

^a Value depending on species (see Sect. 4.2.3.4)

App. C Evolution of porewater composition during saturation

The through-diffusion experiments are generally preceded by a saturation phase, during which the porous test medium is brought into repeated contact with a fresh electrolyte solution of defined composition. The purpose of this step in the context of the present through-diffusion experiments with TBO samples is to avoid concentration gradients between the external solutions in the up- and downstream reservoirs (the synthetic porewaters) and the internal pore solution. Owing to the large variability of the mineralogical composition of the rock samples used for the diffusion experiments and the depths below surface from which the samples were taken, the synthetic porewaters applied cannot be expected to be identical with the in-situ porewater composition. Contrarily, the purpose of the saturation phase is to recreate the same conditions in the pore solution as in the external synthetic porewaters as well as possible. The composition of the external porewaters after exchange against fresh porewaters was monitored by various analytical methods to measure the concentration of the main constituents of the synthetic porewaters.

For each ion analysed (i), the results are expressed as concentration differences (Δc) between the concentrations in the equilibrated solutions (c_{eq}) and in the respective synthetic porewaters (c_0). The concentration differences are expressed as equivalents instead of moles by multiplication with the charge number (z [eq mol⁻¹]):

$$\Delta c_i = z_i(c_{i,q} - c_{i,0}) \quad (C-1)$$

The concentration differences are handled on an absolute scale, and the sign of the charge number is taken into account in Eq. (C-1), with regard to positive charge signs for cations and negative charge signs for anions. For this reason, the sign of the resulting Δc has a different meaning for cations and anions with respect to diffusion from or into the solution reservoir: for cations, a positive difference is equivalent to a larger concentration after equilibration and denotes a mass flux from the porous rock sample into the solution reservoir. For anions, the contrary is the case for positive values of Δc : the concentration after equilibration is smaller than the initial concentration, denoting a mass flux from the solution reservoir into the porous rock sample. This approach is used here in order to optically separate the behaviour of cations and anions. Besides the evolution of the individual concentrations, the sum of equivalents of all analysed species is also calculated to confirm the comprehensiveness and accuracy of the analytical results. Ideally, the sum should be zero. However, deviations occurred in all cases because of the uncertainties involved in the analytical measurements.

Aside from interpreting the Δc sign, the numbering of the solution exchanges also requires an explanation. In most cases, numbers 1 – 4 refer to the solution exchanges during the saturation phase. They are representative of the adaptation of the pore solutions to the concentrations of the external solutions. If both concentrations are equal, no mass transfer occurs and the resulting Δc values approach zero. The last two indices (normally 5 and 6) refer to measurements following the through-diffusion phase of the HTO/³⁶Cl⁻ tracers and after through-diffusion of the ²²Na⁺ tracer, respectively. However, these measurements do not actually refer to solution exchanges in the true sense because the electrolyte solutions were left unchanged upon tracer addition. The purpose of these measurements was simply to document the potential changes of the concentrations during the experimental phase of diffusion. Here, deviations in Δc can, for example, indicate water loss from the solutions to the atmosphere of the glove box. The same colour codes for the summarising rock classification are used here as in the main text of this report, i.e. claystones are a bright violet, OPA are dark violet, sand- and siltstones are ochre, limestones are bright blue and non-classified rock formations are grey.

Generally, it can be noted that the regime of saturation involving four weekly changes of solution was adequate to achieve the goal of equilibration of the internal pore solutions with the synthetic porewaters. This finding is supported by the general trend of Δc values approaching zero. Notable exceptions are probably caused by analytical deficiencies, e.g. in the case of Na^+ concentrations in the BUL1-1 samples. However, it has to be kept in mind that the accuracy of the main electrolyte ions, Na^+ and Cl^- , is affected most strongly by the analytical uncertainties because the induced relative concentration changes are lowest for these ions. Furthermore, some effects of water loss can be seen from the increasing Δc values for cations and decreasing Δc values for anions in a comparison of the last saturation solution with the analysis of the diffusion samples.

Otherwise, no detailed further interpretation of the results is performed here as this would go beyond the scope of the present report. A detailed modelling of the results would certainly be interesting but would require a fully coupled multispecies diffusion model involving concentration and electrochemical potential gradients as the driving forces. As a general observation, it should be simply noted that the concentration evolution between argillaceous rock and limestone formations is different, reflecting different mineralogical compositions and thus probably cation loadings of the clay minerals in these formations. Most indicative for this trend is the behaviour of Mg^{2+} and K^+ , which are obviously not in equilibrium with the synthetic porewaters.

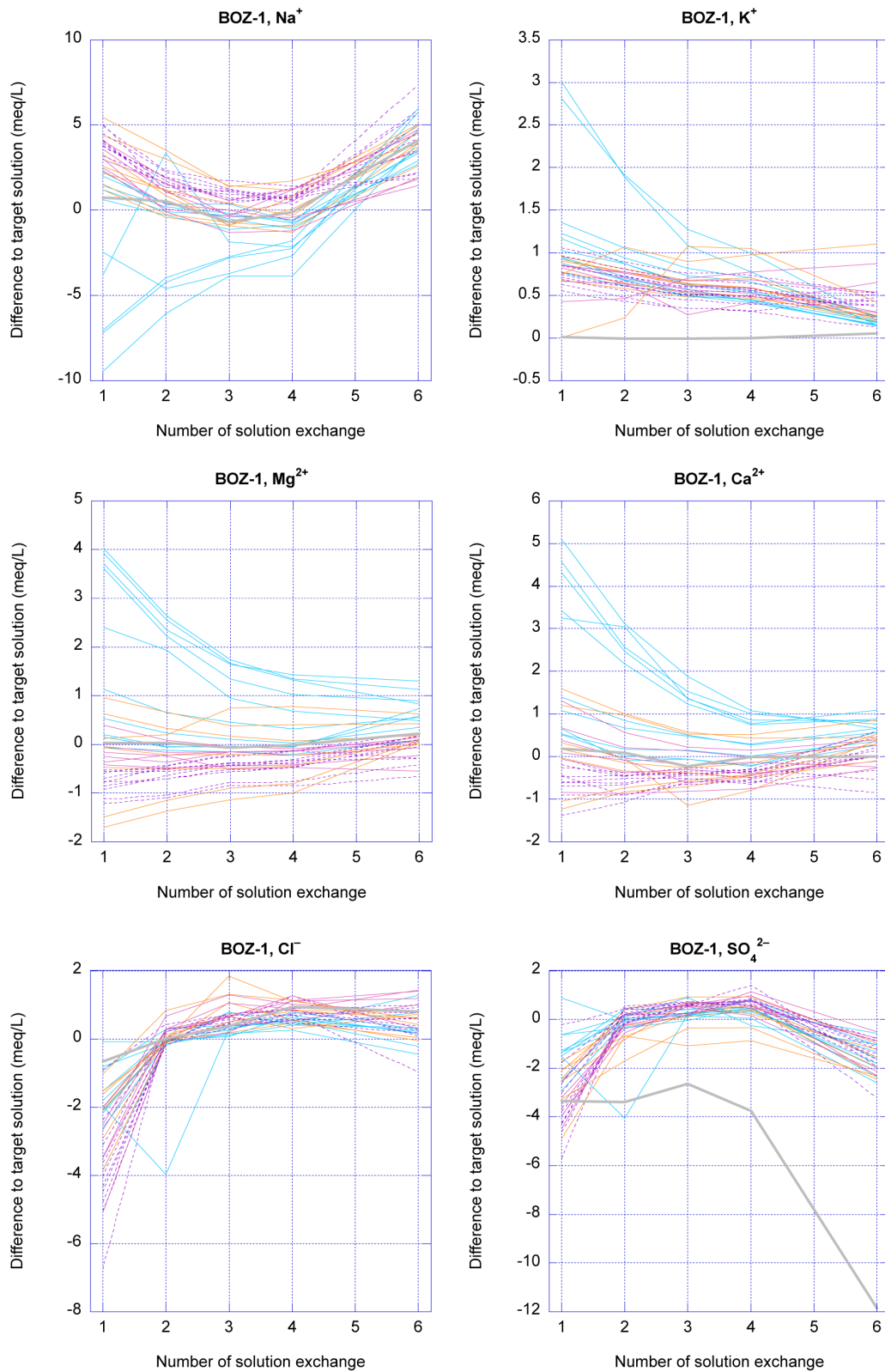


Fig. C-1: Evolution of the concentration differences (Δc) for the main electrolytes in the experiments with BOZ1-1 samples

The indices 1–4 refer to the saturation phase, 5 to the measurements after through-diffusion of HTO and $^{36}\text{Cl}^-$, and 6 after $^{22}\text{Na}^+$.

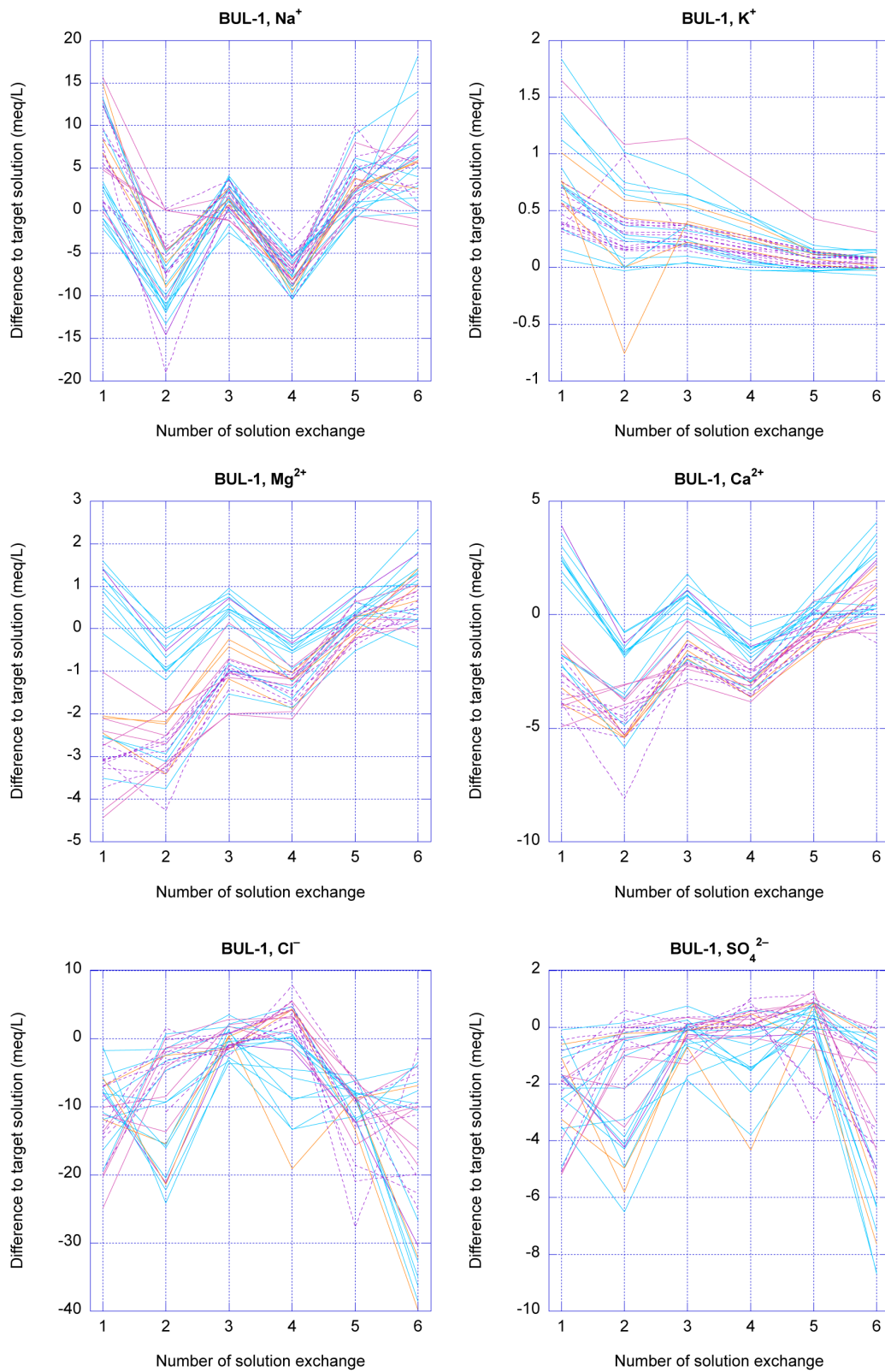


Fig. C-2: Evolution of the concentration differences (Δc) for the main electrolytes in the experiments with BUL1-1 samples

The indices 1–4 refer to the saturation phase, 5 to the measurements after through-diffusion of HTO and $^{36}\text{Cl}^-$, and 6 after $^{22}\text{Na}^+$.

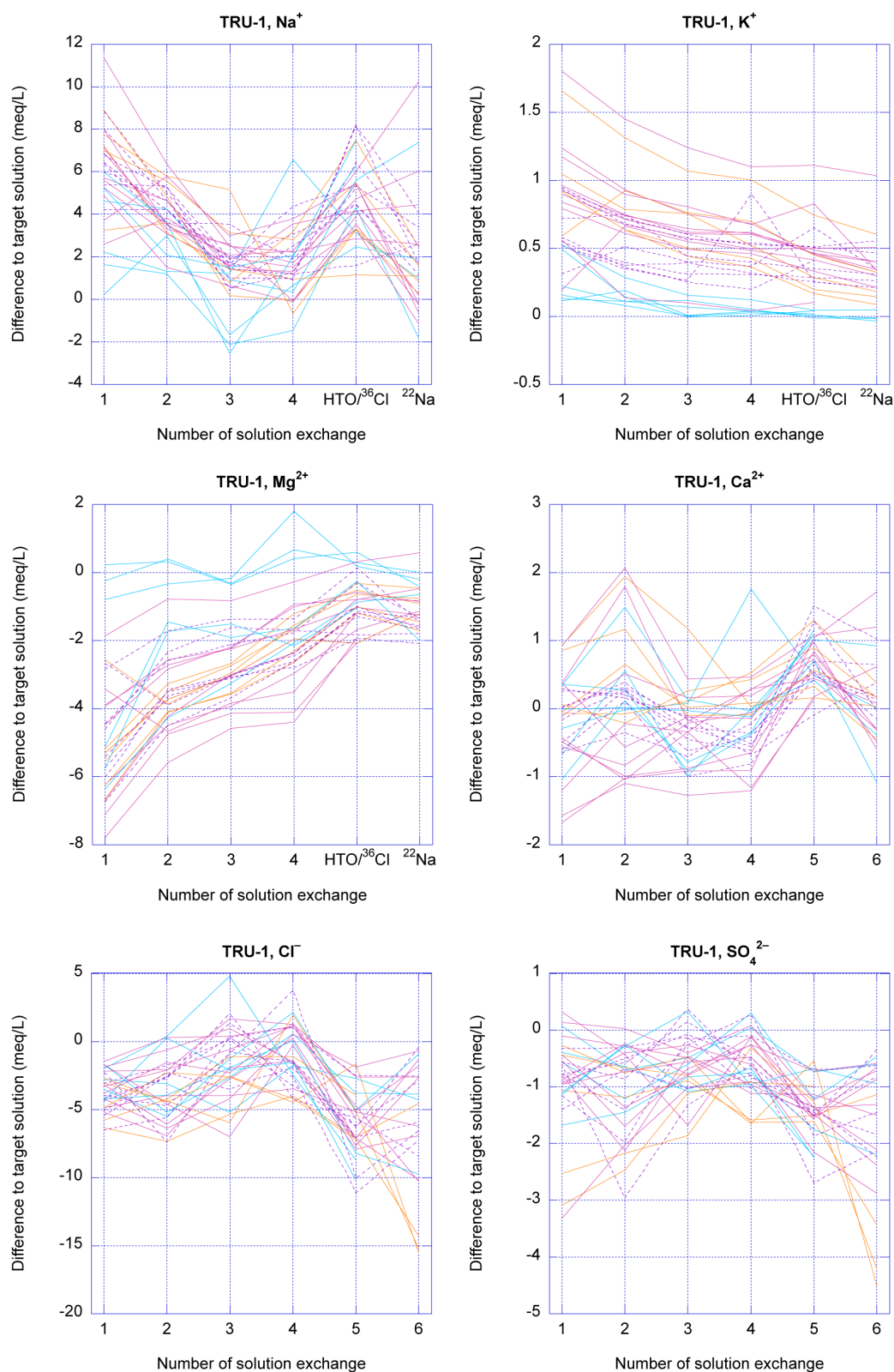


Fig. C-3: Evolution of the concentration differences (Δc) for the main electrolytes in the experiments with TRU1-1 samples

The indices 1 – 4 refer to the saturation phase, 5 to the measurements after through-diffusion of HTO and $^{36}\text{Cl}^-$, and 6 after $^{22}\text{Na}^+$.

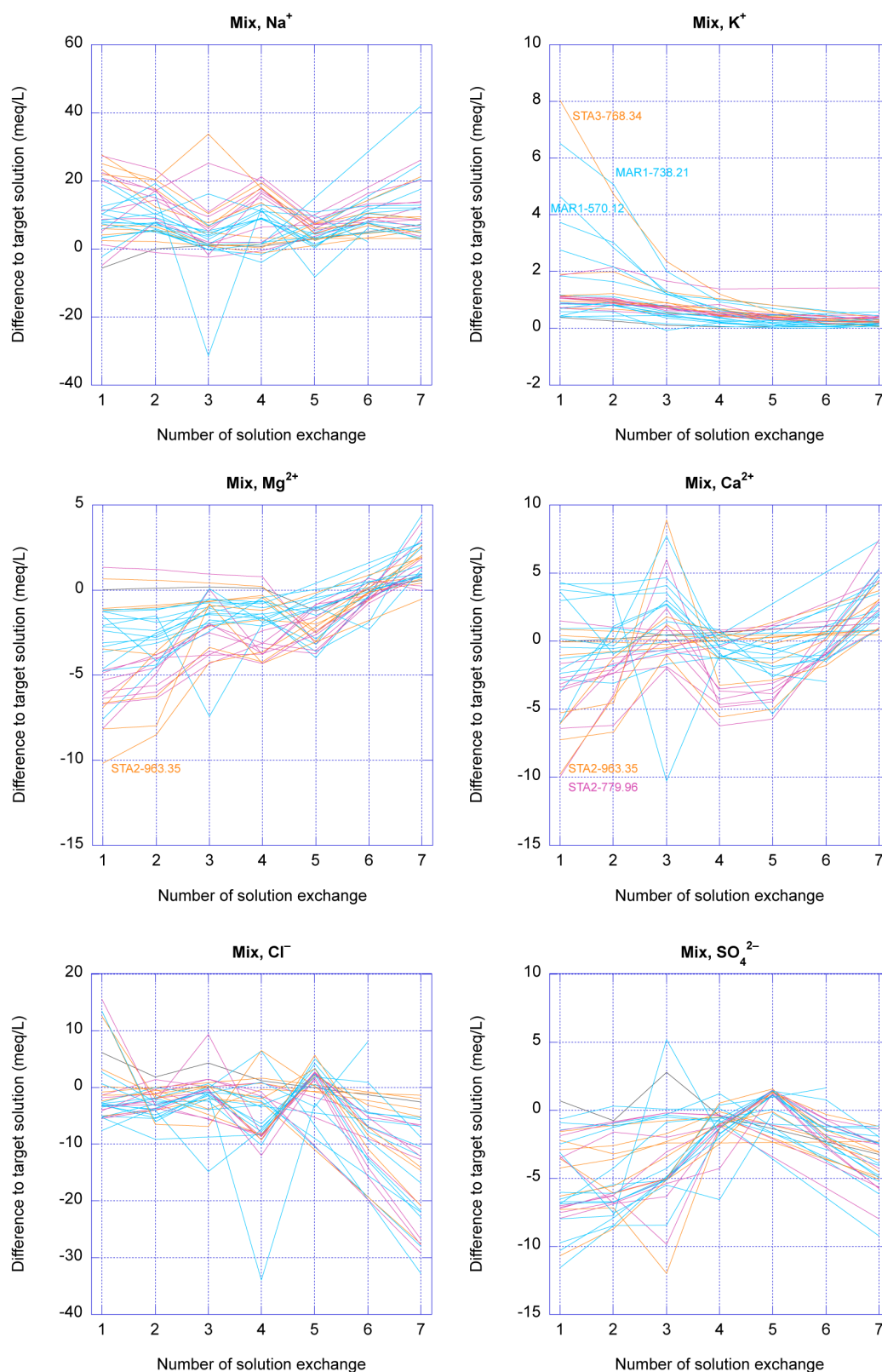


Fig. C-4: Evolution of the concentration differences (Δc) for the main electrolytes in the experiments with a “mixed series”

The indices 1 – 6 refer to the saturation phase, and 7 after $^{22}\text{Na}^+$.

App. D Impact of dimensionality on modelling

Through-diffusion experiments in the context of the measurements on drill core samples (for the experiments in this report) from the deep exploratory boreholes in Northern Switzerland were carried out using cylindrical clay rock samples confined by metallic filter plates (cf. Fig. D-1).

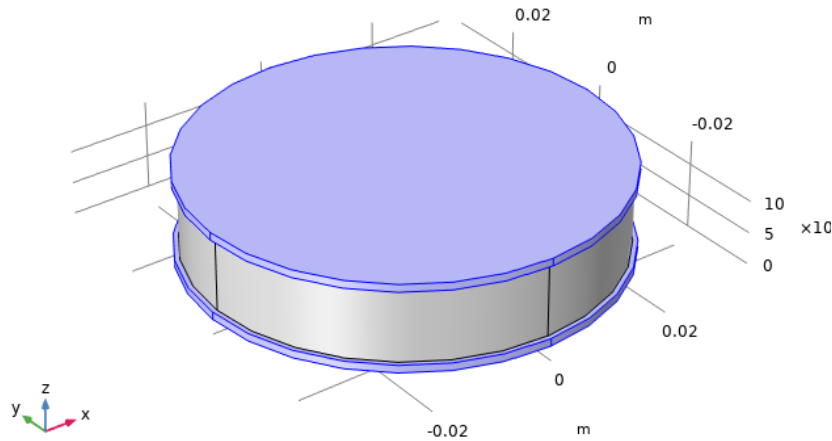


Fig. D-1: 3-dimensional representation of the clay sample (grey) sandwiched between the two confining filters (blue)

The encasing of the clay sample in epoxy resin and the components of the diffusion cell are not shown. The direction of diffusion is perpendicular to the faces of the cylindric shapes.

Since the filter diameter is larger than the diameter of the clay, a 3-dimensional or a 2-dimensional (2D) axisymmetric geometry would be required for an exact representation of the sample in numerical simulations. However, to save computational time, the overlap of the filters was ignored, allowing for the simplification of the geometry to a 1-dimensional (1D) linear representation in which the geometry was identical with the axis of diffusion. Thereby, the diffusive fluxes were calculated with respect to the cross-sectional area of the clay sample instead of the cross-sectional area of the filters, which would have been the appropriate mode according to the geometry shown in Fig. D-1.

It was tested whether the simplifications taken are justified with respect to the small differences of the diameters of clay and filters. To illustrate these circumstances, the diffusion results of an arbitrarily chosen sample from the Bülach-1 borehole (sample BUL1-1-808.92-DI) in 1D-linear geometry were chosen and compared to a more elaborate simulation using a 2D-axisymmetric geometry.

Results in 1D geometry. Fig. D-2 and Fig. D-3 show the results for the flux data and the accumulated mass, respectively, measured at the downstream boundary. Note that the experimental data were evaluated on the basis of the cross-sectional area of the clay. The simulation uses the same cross-sectional area.

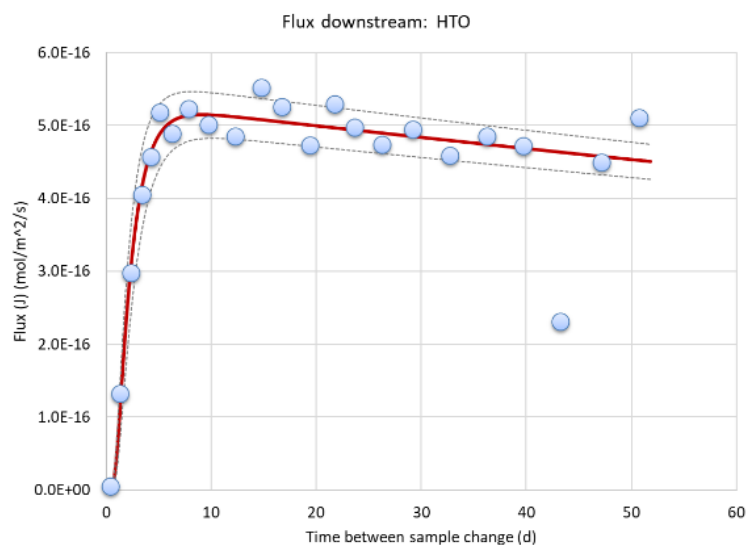


Fig. D-2: Downstream flux of HTO for sample BUL1-1-808.92-DI
Measured data (dots) and 1D-simulations based on the best-fit parameter values (red) and upper and lower confidence intervals (dotted).

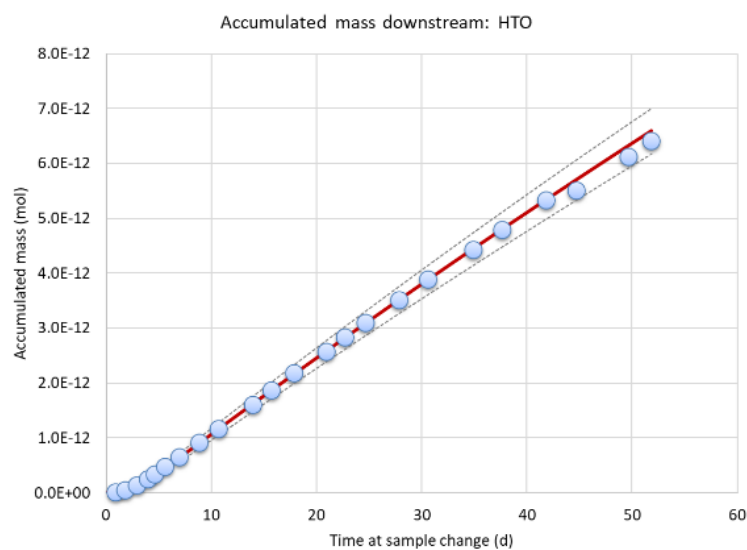


Fig. D-3: Accumulated mass of HTO for sample BUL1-1-808.92-DI
Measured data (dots) and 1D-simulations based on the best-fit parameter values (red) and upper and lower confidence intervals (dotted).

Realisation of a 2D-axisymmetric geometry in Comsol Multiphysics®. The geometrical objects are shown in the 2D-axisymmetric model in Fig. D-4. Note that a comprehensive explanation of the model features would go beyond the scope of the present report, which focuses only on the specific aspects of the issues related to the geometric differences of filters and clay. A particular difficulty in such a model is the calculation of the flux in z-direction at the interface between downstream filter and the solution, which is not represented by a particular geometric entity, but taken into account as a calculatory concentration boundary condition based on an ordinary differential equation (ODE) involving flux integration (not explained in detail for the following purpose). The flux magnitude in z-direction is inhomogeneous as can be seen from Fig. D-5, owing to the existence of rectangular bulges in the geometry. Consequently, the representation of the experimentally observed values in the simulation needs to be based on averaging the flux over r , adequately taking into account the correct weighting in the revolved geometry (revolving a radius to a circular area). Several options are available in Comsol Multiphysics® to perform such averaging calculations, involving averaging operators or linear projection operators. Alternatively, the flux may be derived from calculating the time derivative of the mass diffused across the downstream boundary.

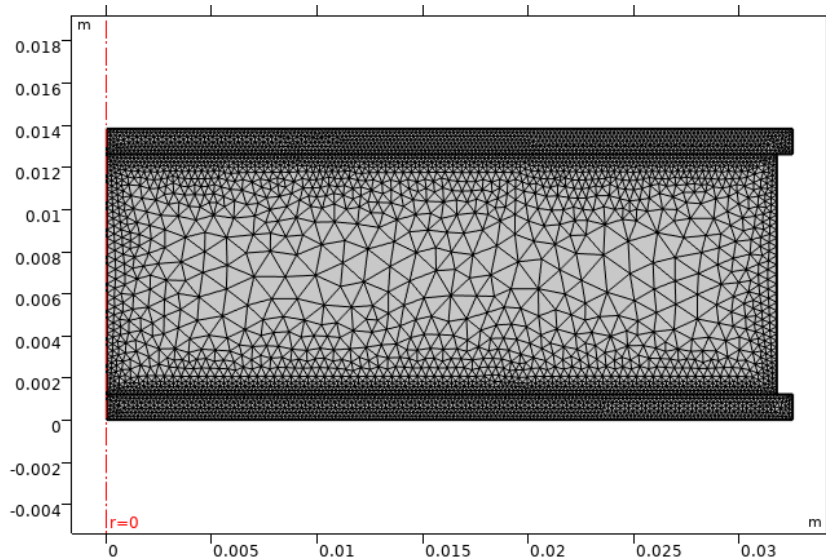


Fig. D-4: 2D axisymmetric representation of the clay sample sandwiched between the two confining filters (cf. Fig. D-1), including the meshing of the geometric entities

The horizontal axis is termed the r -axis, the vertical axis (the direction of diffusion) the z -axis.

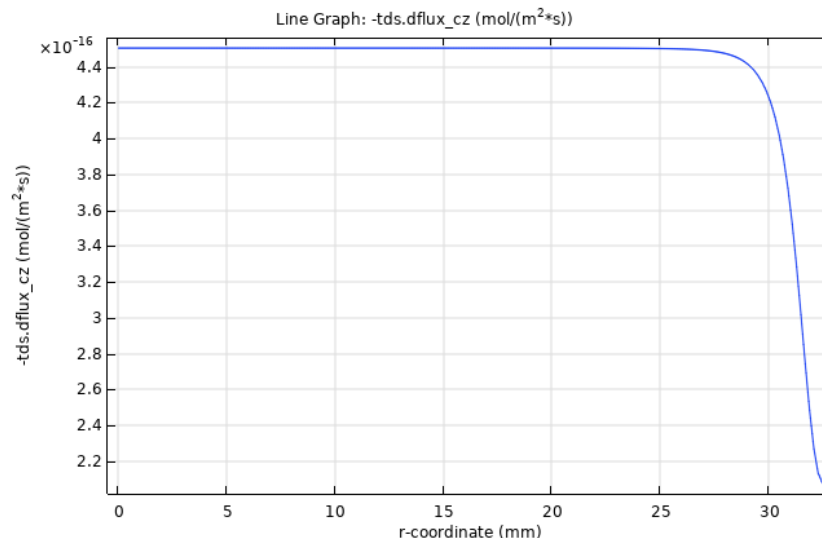


Fig. D-5: 2D axisymmetric representation of the clay sample sandwiched between the two confining filters (cf. Fig. D-1) including the meshing of the geometric entities

The horizontal axis is termed the r-axis, the vertical axis (the direction of diffusion) the z-axis.

Tab. D-1 shows the syntax for the various options chosen. “Intop” denotes an integration operator (definition not shown in Tab. D-1). Note that the integration is not performed directly on the flux, but on so-called Lagrange multipliers (denoted as `comp1.c_lm`), which are invoked in the definition of weak boundary concentration conditions. This procedure leads to a more accurate determination of the boundary fluxes compared to the integration of flux expressions, because the calculation of boundary fluxes may be ill-defined in the finite element numerical scheme. “Jav” is simply a name for the averaged flux. The linear projection operator “linproj1” produces a projection of the model values on the radius *r* to a single point on the z-axis.

Tab. D-1: Syntax used for the various options of flux calculation

The `Cusb` variable is needed to calculate the concentration boundary condition at the upstream boundary and is not related to the present discussion of downstream flux calculations.

Name	Expression	Unit
Flow_usb	<code>intop_usb(comp1.c_lm[mol/m/s])</code>	mol/s
Flow_dsb	<code>intop_dsb(-comp1.c_lm[mol/m/s])</code>	mol/s
Cusb	<code>(c0-Menge_usb/Vusb(t))*step1(t[1/s])</code>	mol/m ³
Jav_aveop	<code>aveop1(-tds.dflux_cz)</code>	mol/(m ² ·s)
Jav_linproj	<code>linproj1(-tds.dflux_cz*r)/linproj1(r)</code>	mol/(m ² ·s)
Jav_SV	<code>Flow_dsb/intop_dsb(2*pi*r)</code>	mol/(m ² ·s)

Note that the “aveop1” requires selecting a checkbox to be integrated in revolved geometry, while the integration operators used to integrate the Lagrange multipliers do not require this definition.

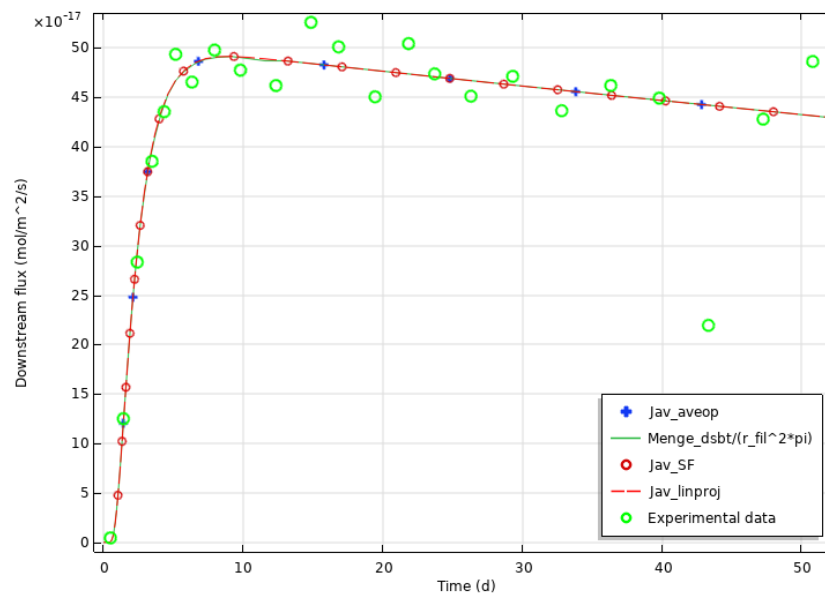


Fig. D-6: Average downstream flux calculated for the 2D-axisymmetric model using the various options to calculate the diffusive flux, according to the definitions of the related variables as shown in Tab. D-1

Note that the experimental data are normalised with respect to the cross-sectional area of the filters. Otherwise, a direct comparability would not be given.

Fig. D-6 shows the results of the various options in the calculation of the averaged downstream flux. It can readily be seen that all options produce the same result and that this result is in good agreement with the experimental data. It can also be seen that both simulation and experimental data are proportionally lower than the data shown in Fig. D-2. This circumstance is clearly related to the fact that the present simulation used the cross-sectional area of the confining filters for flux calculation, which is formally the correct approach. For this reason, the experimental data in Fig. D-6 were renormalised to this cross-sectional area. All these renormalisation issues are not relevant for the calculation of the accumulated mass, because this quantity does not depend on the cross-sectional area.

To provide a direct comparison of the 1D-linear and the 2D-axisymmetric simulations, the expression of the time derivative of the accumulated mass arrived at the downstream boundary (“Menge_dsb”) was renormalised to the cross-sectional area of the clay in Fig. D-7. It can be seen that no differences between the simulations in the two geometries are visible. Only a slight shift in breakthrough behaviour can be recognised in a logarithmic representation of Fig. D-7 (not shown). It can thus be safely postulated that, for the conditions encountered in the TBO experiments, which cover roughly one order of magnitude of variability in effective diffusion coefficients and accessible porosity, the simplifications taken in a 1D-linear geometry can be justified. It is, however, important that the normalisation of the flux data in the experimental calculation and the simulations are made on the same basis (cross-sectional area). Otherwise, a systematic bias proportional to the ratio of cross-sectional areas in the calculation of the effective diffusion coefficients will arise.

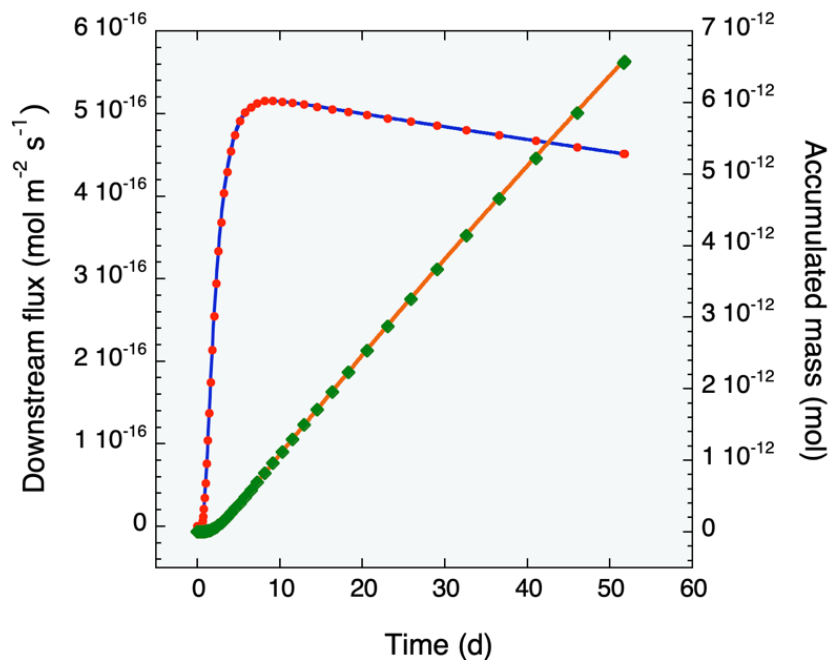


Fig. D-7: Comparison of the 1D-linear (solid lines) and the 2D-axisymmetric (dots) simulations of downstream flux (blue line, red points) and accumulated mass (red line, green points)

For comparison purposes, the flux data (calculated as time derivatives of accumulated mass) were normalised to the cross-sectional area of the clay. The experimental data are not shown; they can be derived from Fig. D-2 and Fig. D-3.

App. E Impact of confining resin on diffusion modelling

Diffusion tests with HTO in small Opalinus Clay (OPA) samples embedded in resin clearly show the signature of the resin being available as a porous medium for diffusion. The potential impact of the resins on the results of the measurements on TBO samples (this work) was tested for one case: HTO diffusion in the BOZ1-1-416.35-DI sample using the diffusion parameters obtained with the epoxy resin from the small cylindrical OPA samples and the computational model applied in Comsol Multiphysics®. Whether these properties are relevant for the resin used for the sample preparation at the University of Bern remains an open question and will not be answered in this report. Measurements on representative resin samples will be required for this purpose.

The Comsol model uses an existing template (described in PN-MIG-2020_01) using a 2D-axi-symmetric geometry, involving Lagrange multipliers for boundary flux calculation as a starting point. To this geometry, the resin is simply added as a further porous domain (no sorption properties). Fig. E-1 shows a 3D view of this geometry.

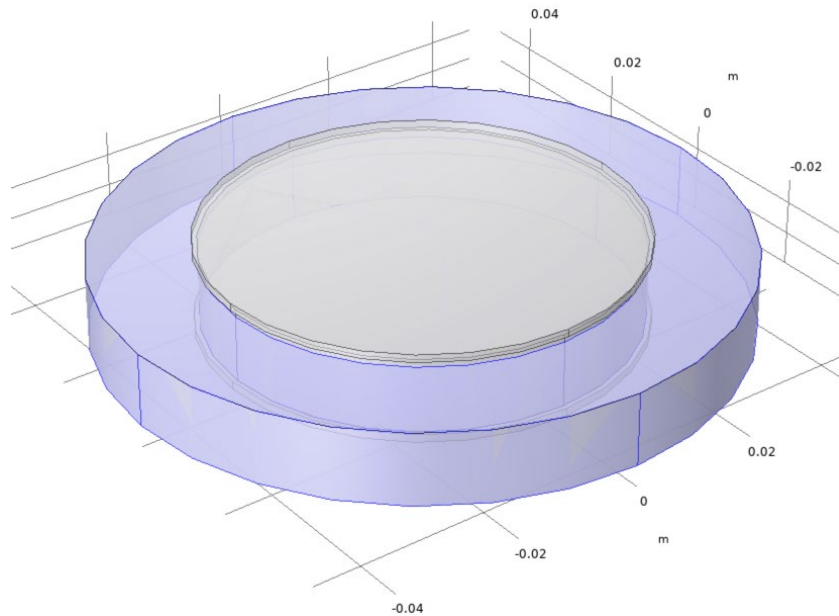


Fig. E-1: 3D representation of the clay sample (transparent) sandwiched between the two confining filters (grey)

The encasing of the clay sample in epoxy resin is shown as transparent blue. The direction of diffusion is parallel to the cylinder axis.

As discussed in App. D, the definition of the cross-sectional area for normalising the diffusive flux is a critical step. Commonly, the clay cross-sectional area is used. Here, the cross-sectional area of the filters is chosen because this is more convenient in view of the boundaries defined in the model. For this reason, the experimental reference data were renormalised with respect to the cross-sectional area of the filters (filter diameter = 65.0 mm, and clay diameter 63.55 mm for sample BOZ1-1-416.35-DI).

A particular difficulty in such a model is the calculation of the flux in z-direction at the interface between downstream filter and the solution, which is not represented by a particular geometric entity, but taken into account as a calculatory concentration boundary condition based on an ordinary differential equation (ODE) involving flux integration (not explained in detail here). The flux magnitude in z-direction is inhomogeneous as can be seen from Fig. E-3, owing to the existence of the rectangular bulges in the geometry. Consequently, the representation of the experimentally observed values in the simulation has to be based on averaging the flux over r , adequately taking into account the correct weighting in the revolved geometry (revolving a radius to a circular area). Several options are available in Comsol Multiphysics® to perform such averaging calculations, involving averaging operators or linear projection operators. Alternatively, the flux can be derived from calculating the time derivative of the mass diffused across the downstream boundary.

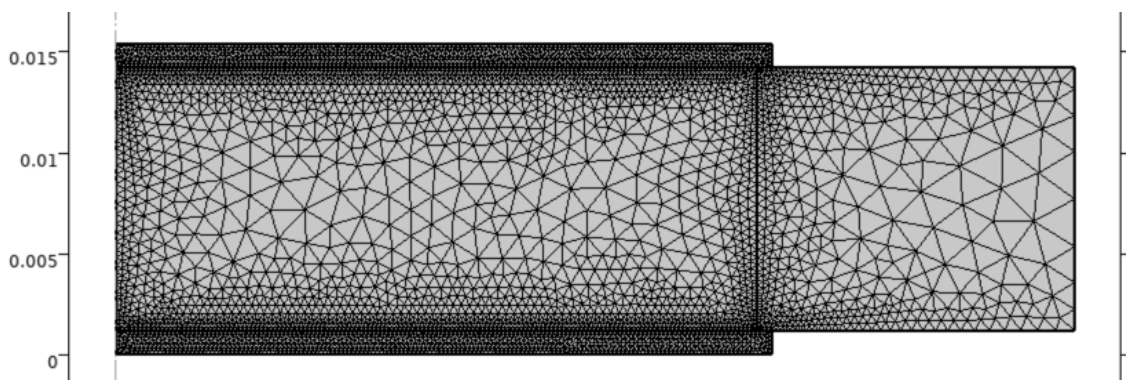


Fig. E-2: 2D axisymmetric representation of the clay sample sandwiched between the two confining filters (cf. E-1), including the meshing of the geometric entities

The horizontal axis is termed the r -axis, the vertical axis (the direction of diffusion) the z -axis.

Tab. E-1 shows the syntax for the various options chosen. “Intop” denotes an integration operator (definition not shown in Tab. E-1). Note that the integration is not performed directly on the flux, but on so-called Lagrange multipliers (denoted as `comp1.c_lm`), which are invoked in the definition of weak boundary concentration conditions. This procedure leads to a more accurate determination of the boundary fluxes compared to the integration of flux expressions, because the calculation of boundary fluxes can be ill-defined in the finite element numerical scheme. “Jav” is simply a name for the averaged flux. The linear projection operator “linproj1” produces a projection of the model values on the radius, r , to a single point on the z -axis.

Tab. E-1: Syntax used for the various options of flux calculation

The Cusb variable is needed to calculate the concentration boundary condition at the upstream boundary and is not related to the present discussion of downstream flux calculations.

Name	Expression	Unit
Flow_usb	intop_usb(comp1.c_Im[mol/m/s])	mol/s
Flow_dsb	intop_dsb(-comp1.c_Im[mol/m/s])	mol/s
Cusb	(c0-Menge_usb/Vusb(t))*step1(t[1/s])	mol/m ³
Jav_aveop	aveop1(-tds.dflux_cz)	mol/(m ² ·s)
Jav_linproj	linproj1(-tds.dflux_cz*r)/linproj1(r)	mol/(m ² ·s)
Jav_SV	Flow_dsb/intop_dsb(2*pi*r)	mol/(m ² ·s)

Note that the “aveop1” requires selecting a checkbox to be integrated in revolved geometry, while the integration operators used to integrate the Lagrange multipliers do not require this definition.

Fig. E-3 shows a comparison of the results obtained using the 1D geometry (neglecting the difference in diameter of clay and filter, which has proven to be justified, see App. D) with the results obtained in 2D geometry, including the resin. The diffusion parameters chosen for the resin are: $D_{e,resin} = 2 \times 10^{-12} \text{ m}^2 \text{ s}^{-1}$ and $\epsilon_{resin} = 0.03$.

Although the difference between the two models is rather insignificant, the comparison shows that knowledge of the pertinent diffusion properties of the resin is important. In particular, whether the diffusion properties could exert a biasing effect on the calculated accessible porosity of the clay has not been clarified in this report.

Diffusion in the resin can be seen as a potential explanation for the slight discrepancies observed between diffusion-accessible porosity and water-loss porosity. In particular, diffusion in the resin could further explain the slight increase of the “steady-state” flux in the case of diffusion of the ³⁶Cl⁻ tracer observed in many samples.

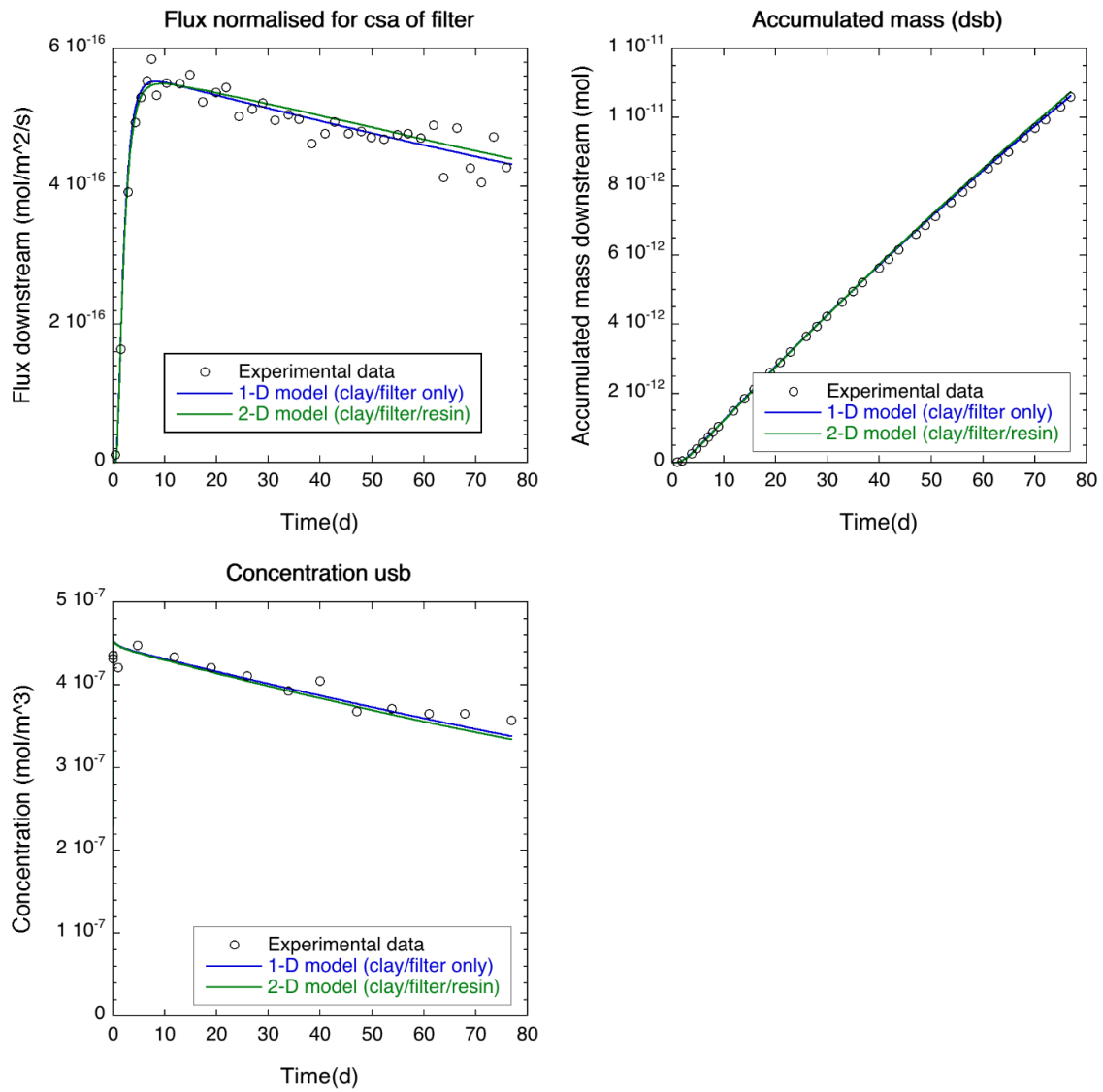


Fig. E-3: Average downstream flux calculated for the 2D-axisymmetric model using the various options shown in Tab. E-1

Note that the experimental data are normalised with respect to the cross-sectional area of the filters. Otherwise, a direct comparison would not be possible.

App. F Diffusion data as a function of sample depth

This section contains all data (effective diffusion coefficients and accessible porosities for HTO and ^{36}Cl , and effective diffusion coefficients and distribution ratios for $^{22}\text{Na}^+$) as a function of depth. The data were grouped per borehole.

Note that for the STA2-1, STA3-1 and MAR2-1 boreholes, only samples of the overlying and underlying units were used in the experiments. No samples of OPA were used. The composition of the used porewaters is given in Tab. 2-1.

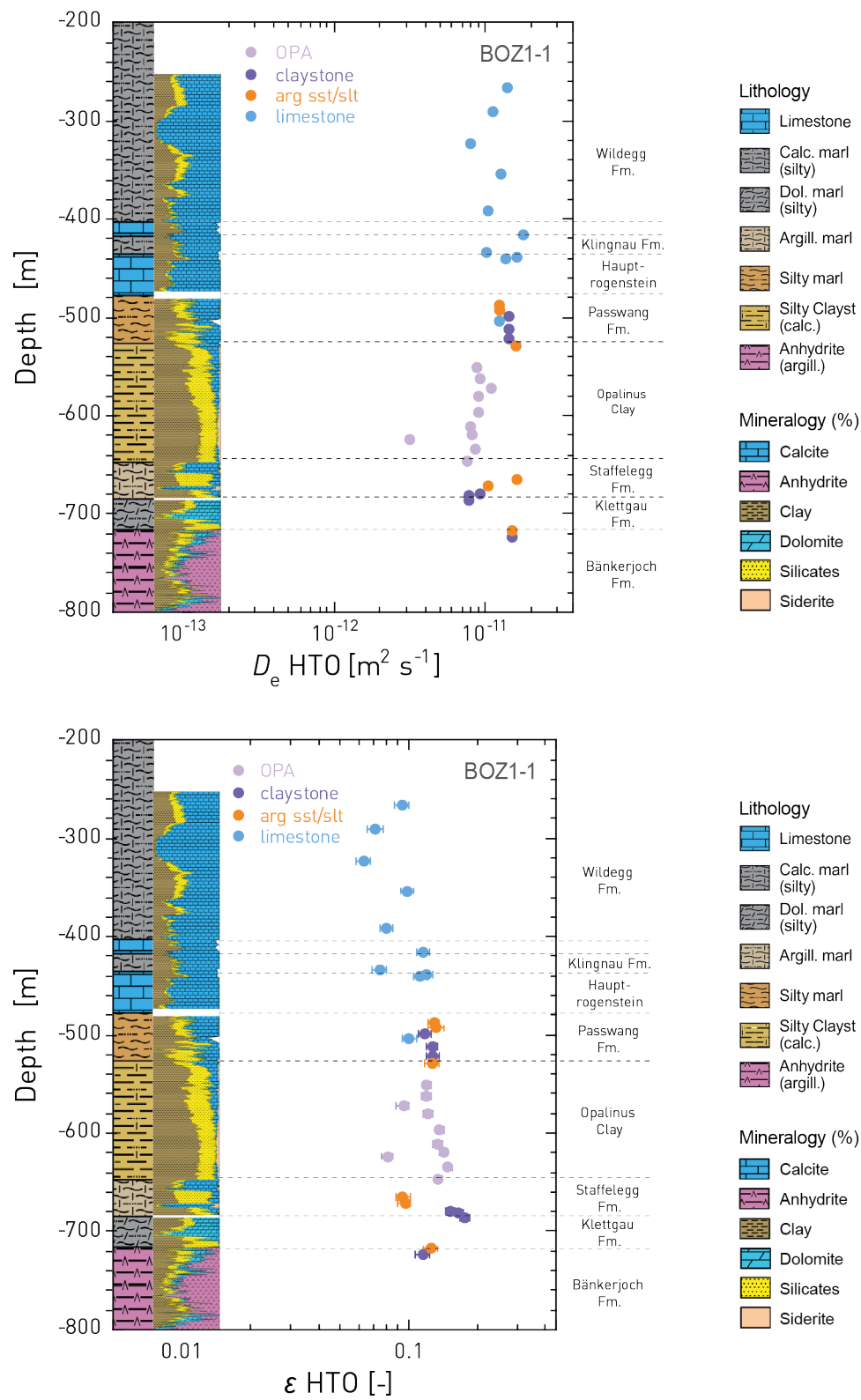


Fig. F-1: Effective diffusion coefficients and accessible porosity values for HTO as a function of sample depth for the BOZ1-1 borehole (JO siting region)

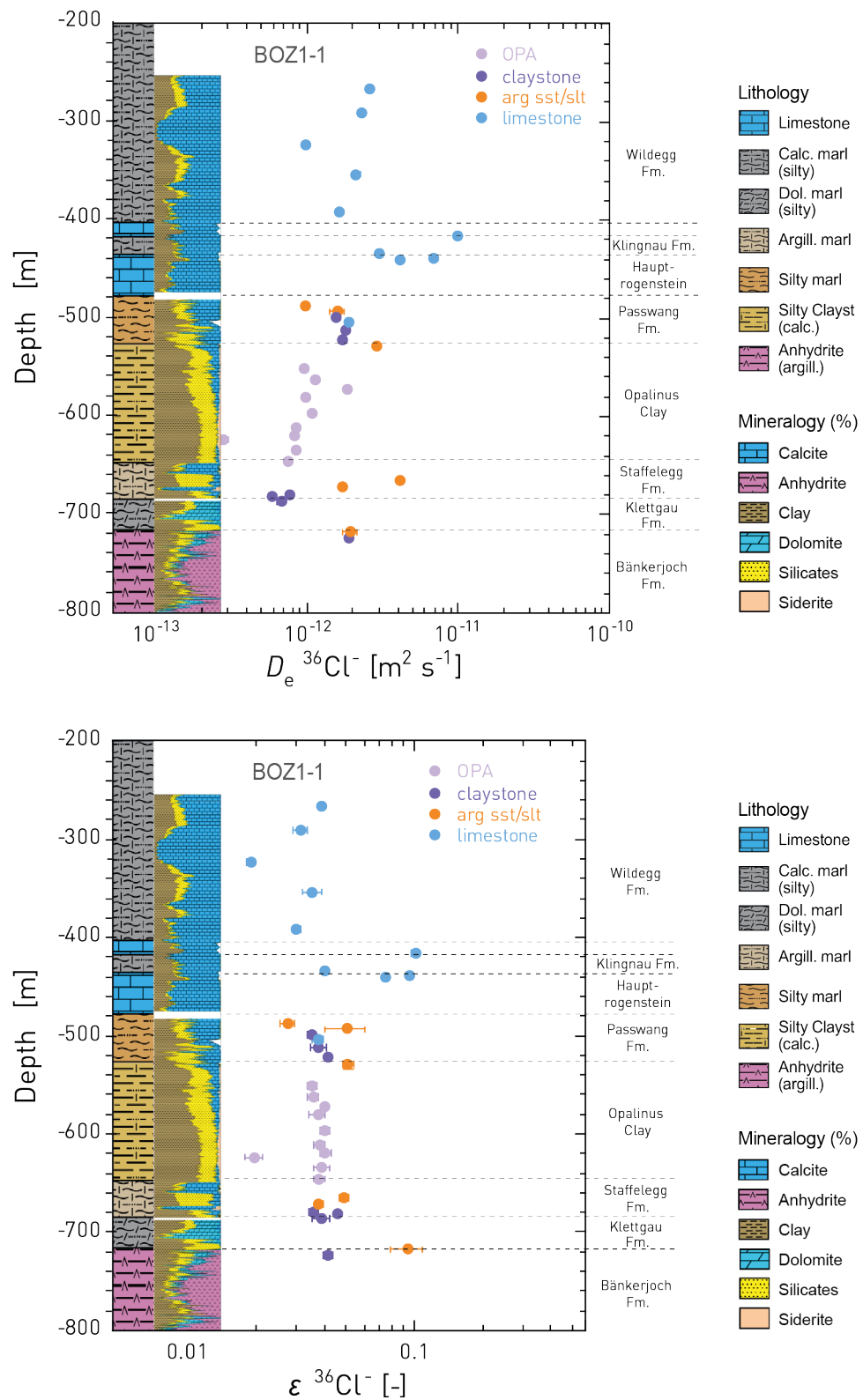


Fig. F-2: Effective diffusion coefficients and accessible porosity values for $^{36}\text{Cl}^-$ as a function of sample depth for the BOZ1-1 borehole (JO siting region)

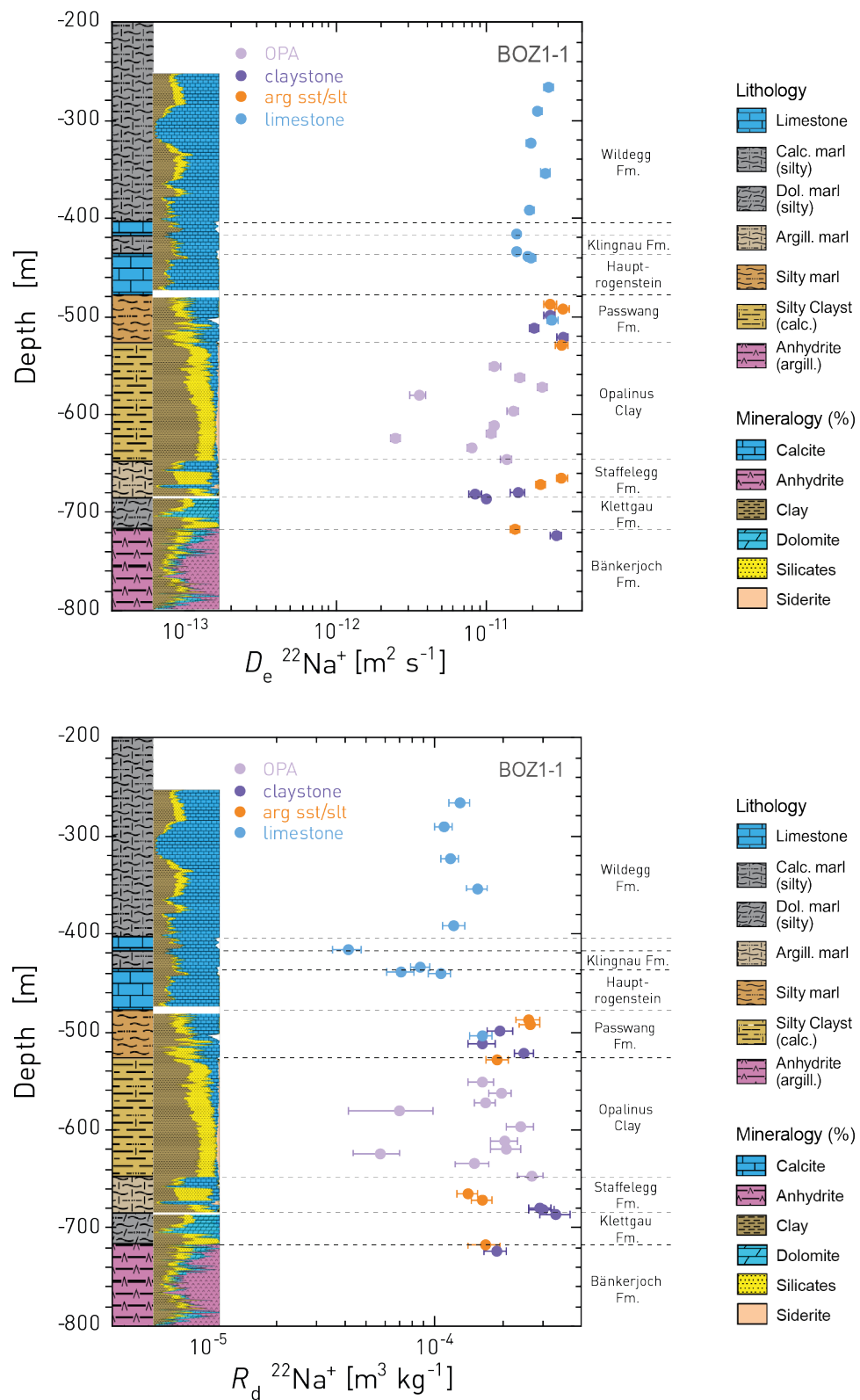


Fig. F-3: Effective diffusion coefficients and R_d values for $^{22}\text{Na}^+$ as a function of sample depth for the BOZ1-1 borehole (JO siting region)

The porosity values selected for $^{22}\text{Na}^+$ are identical to those for HTO.

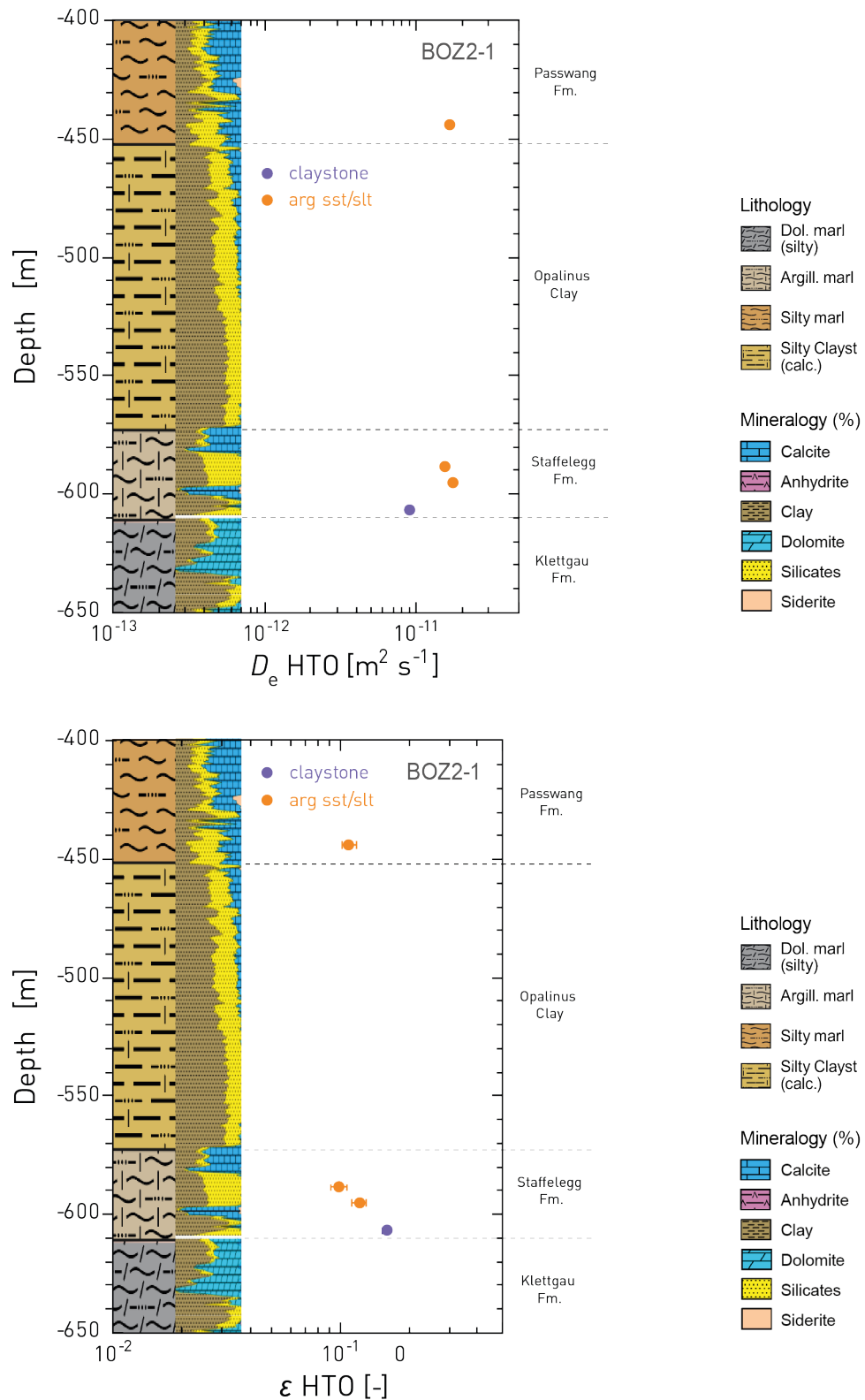


Fig. F-4: Effective diffusion coefficients and accessible porosity values for HTO as a function of sample depth for the BOZ2-1 borehole (JO siting region)

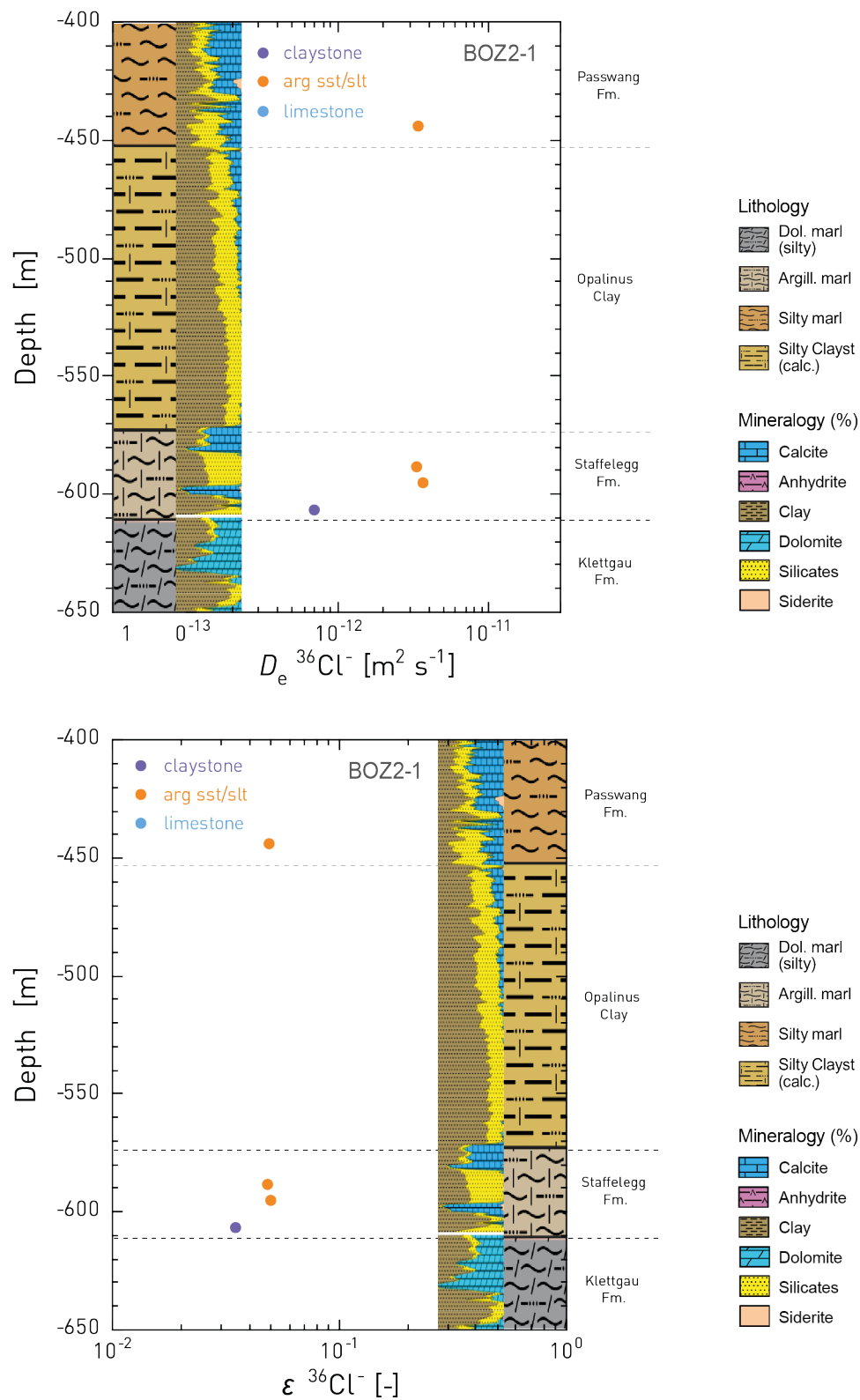


Fig. F-5: Effective diffusion coefficients and accessible porosity values for $^{36}\text{Cl}^-$ as a function of sample depth for the BOZ2-1 borehole (JO siting region)

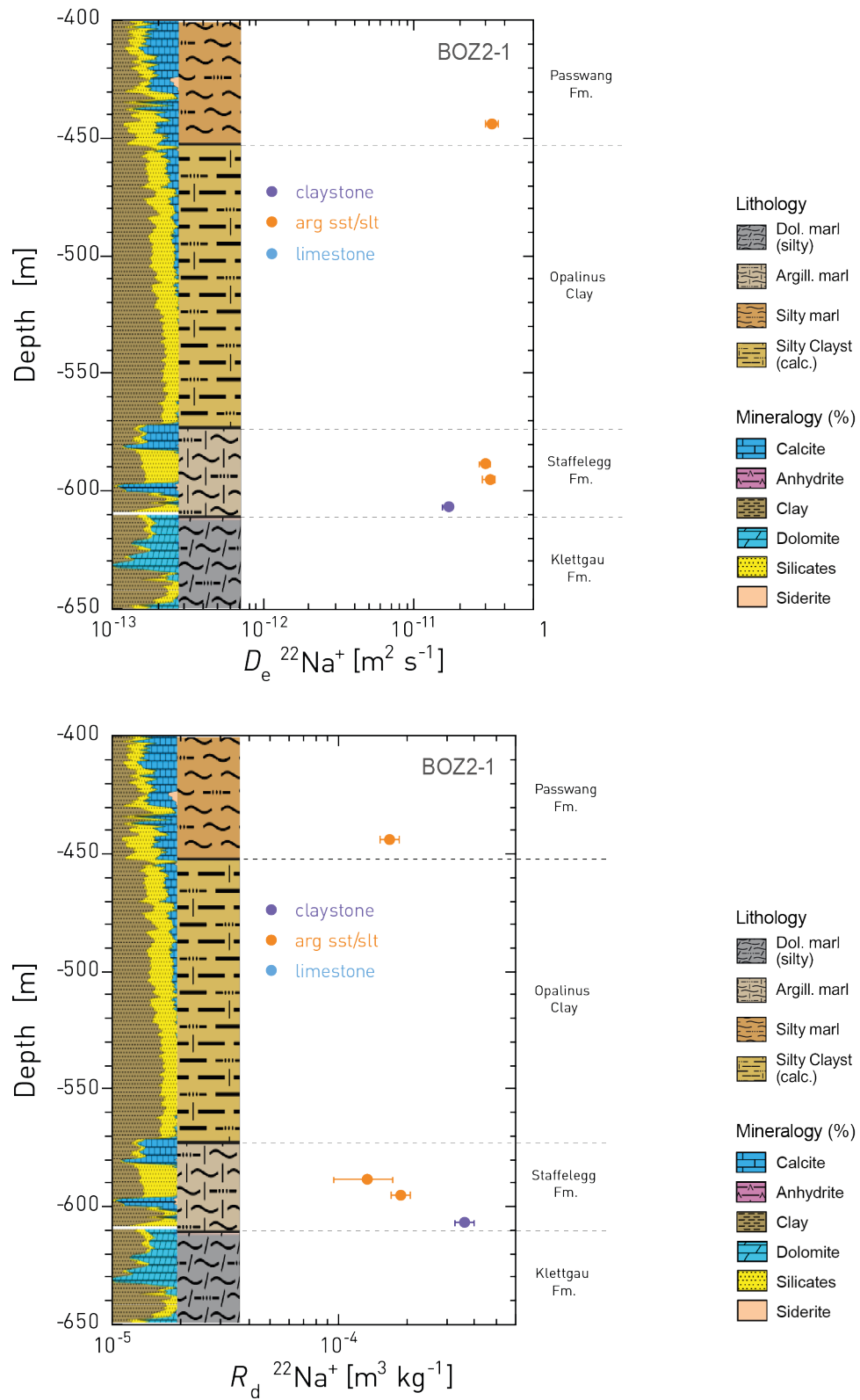


Fig. F-6: Effective diffusion coefficients and R_d values for $^{22}\text{Na}^+$ as a function of sample depth for the BOZ2-1 borehole (JO siting region)

The porosity values selected for $^{22}\text{Na}^+$ are identical to those for HTO.

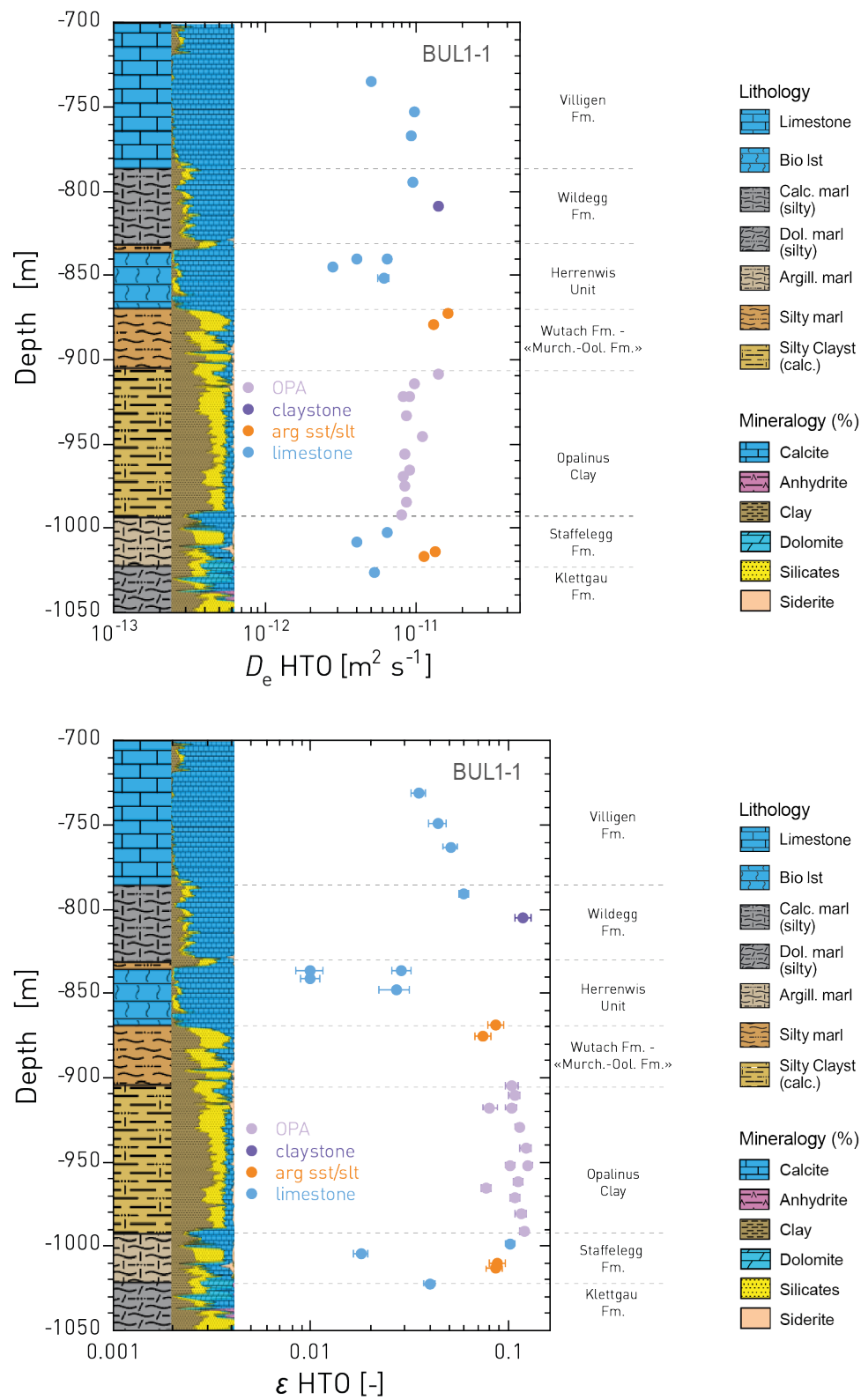


Fig. F-7: Effective diffusion coefficients and accessible porosity values for HTO as a function of sample depth for the BUL1-1 borehole (NL siting region)

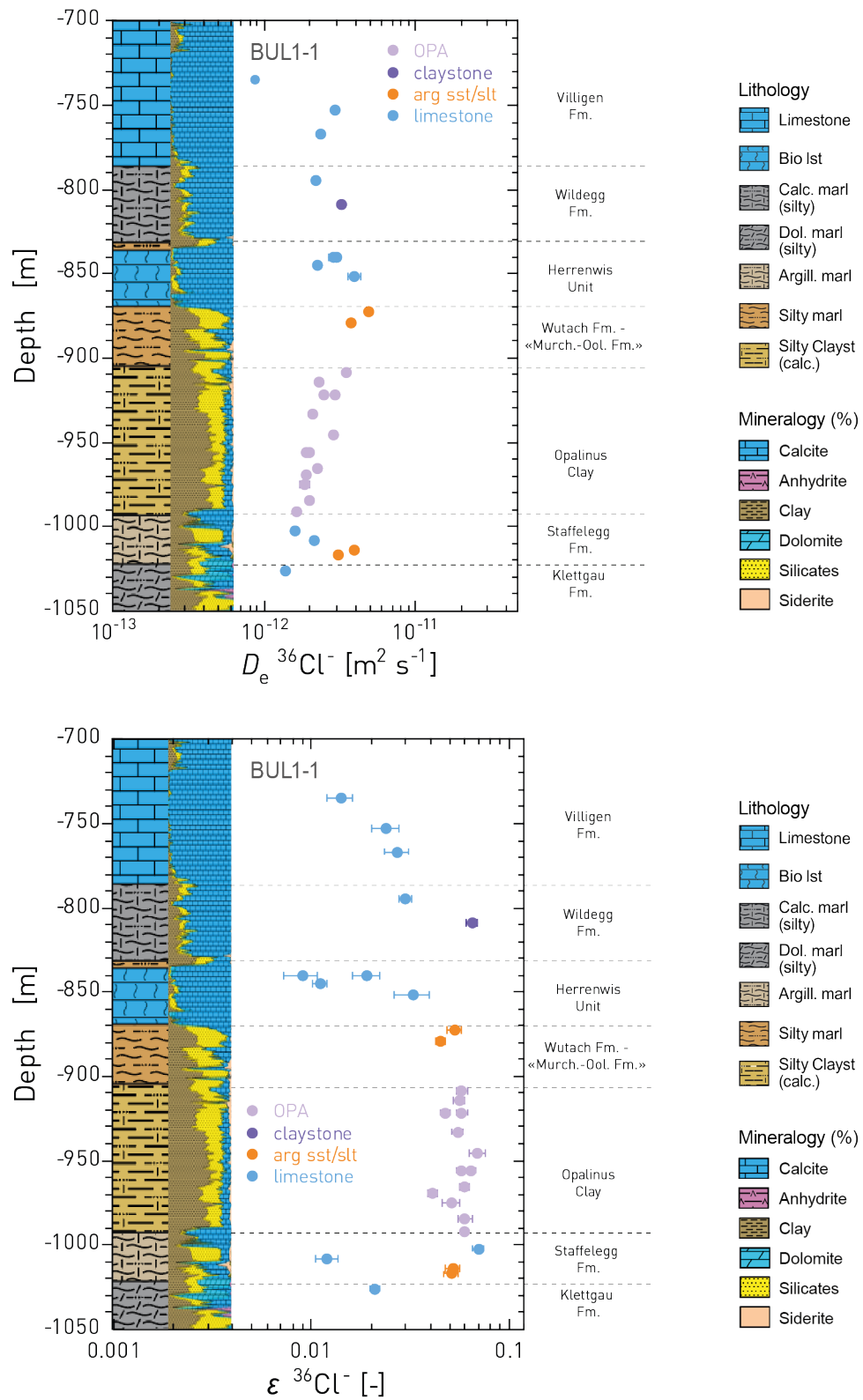


Fig. F-8: Effective diffusion coefficients and accessible porosity values for $^{36}\text{Cl}^-$ as a function of sample depth for the BUL1-1 borehole (NL siting region)

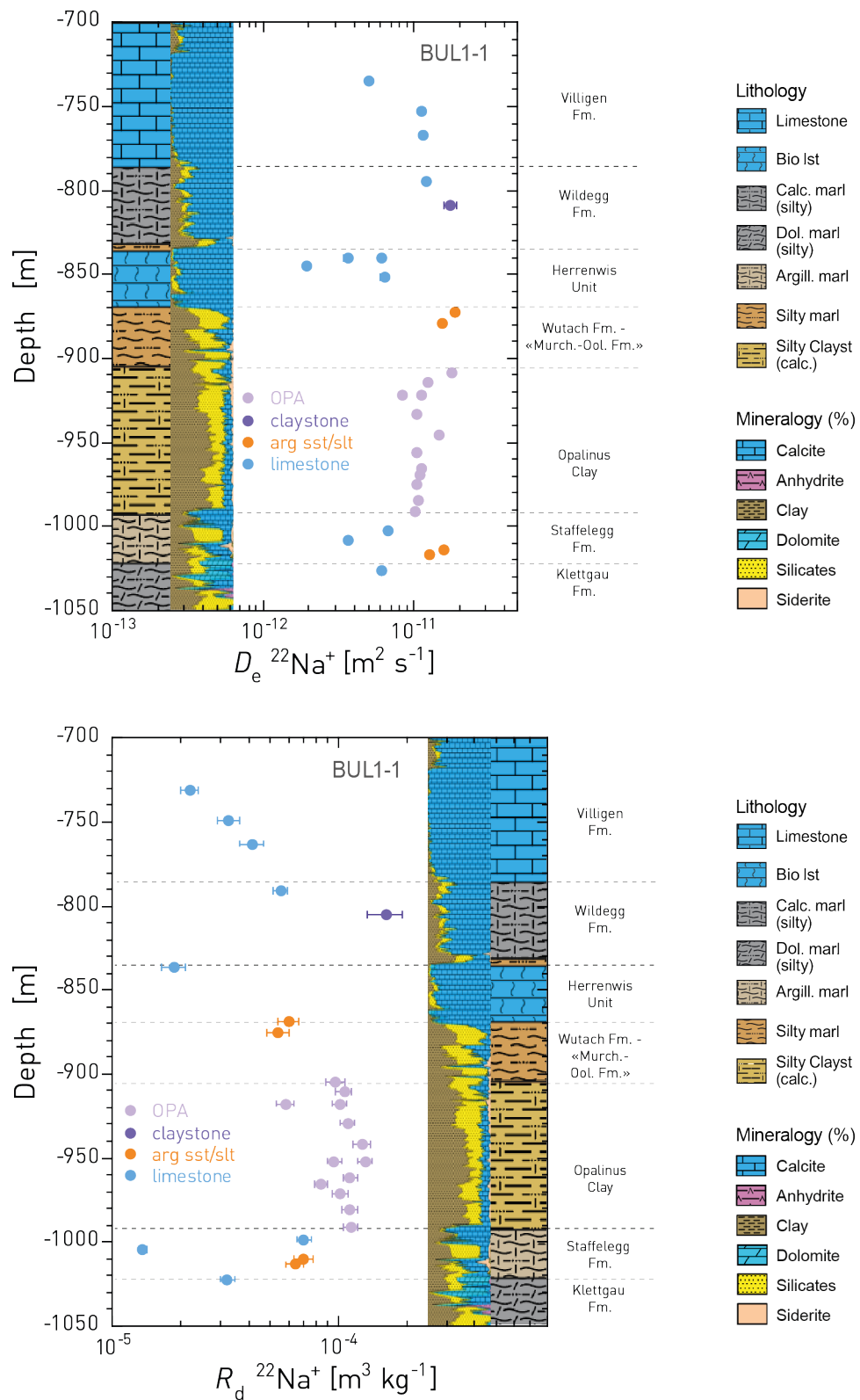


Fig. F-9: Effective diffusion coefficients and R_d values for $^{22}\text{Na}^+$ as a function of sample depth for the BUL1-1 borehole (NL siting region)

The porosity values selected for $^{22}\text{Na}^+$ are identical to those for HTO.

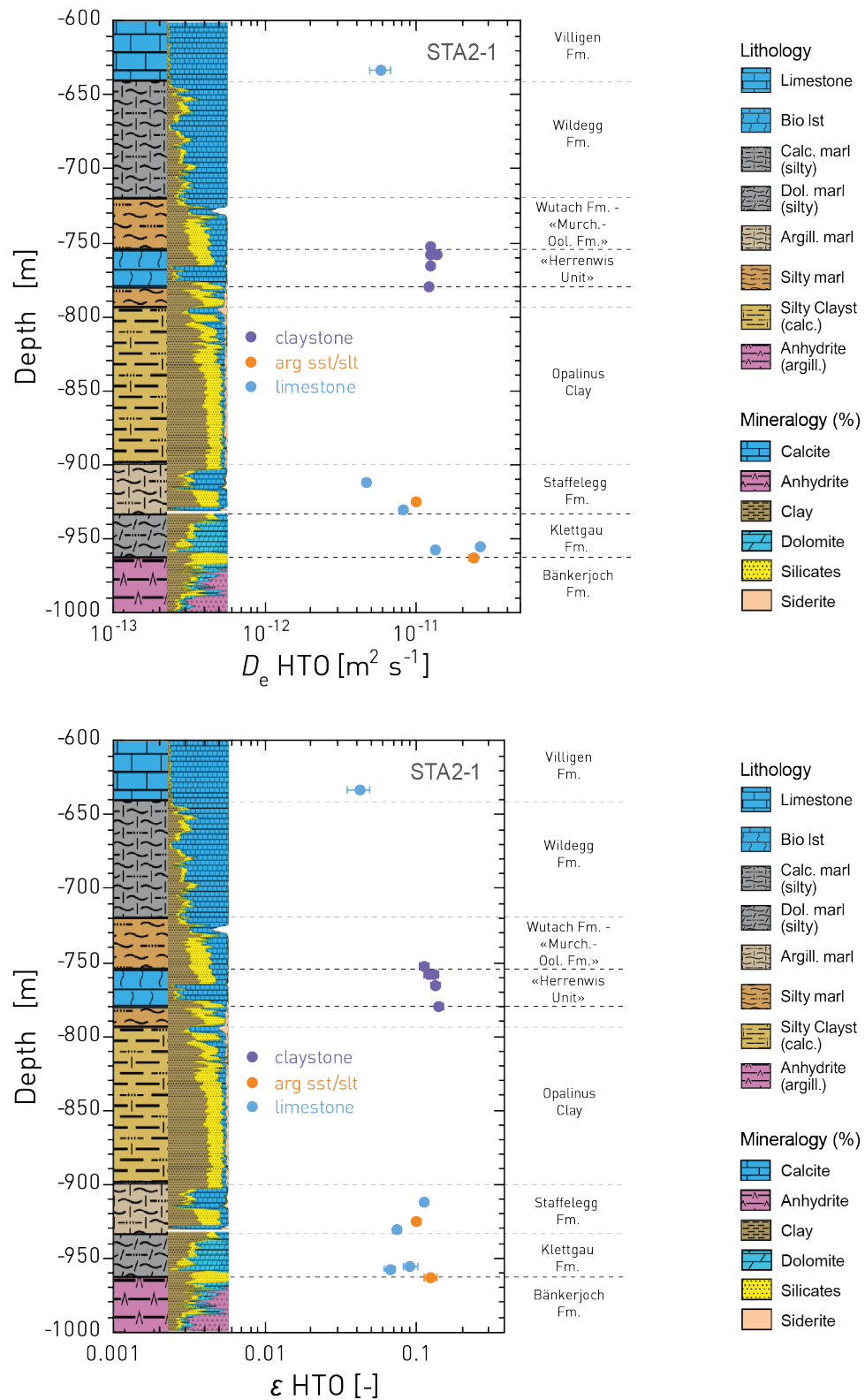


Fig. F-10: Effective diffusion coefficients and accessible porosity values for HTO as a function of sample depth for the STA2-1 borehole (NL siting region)

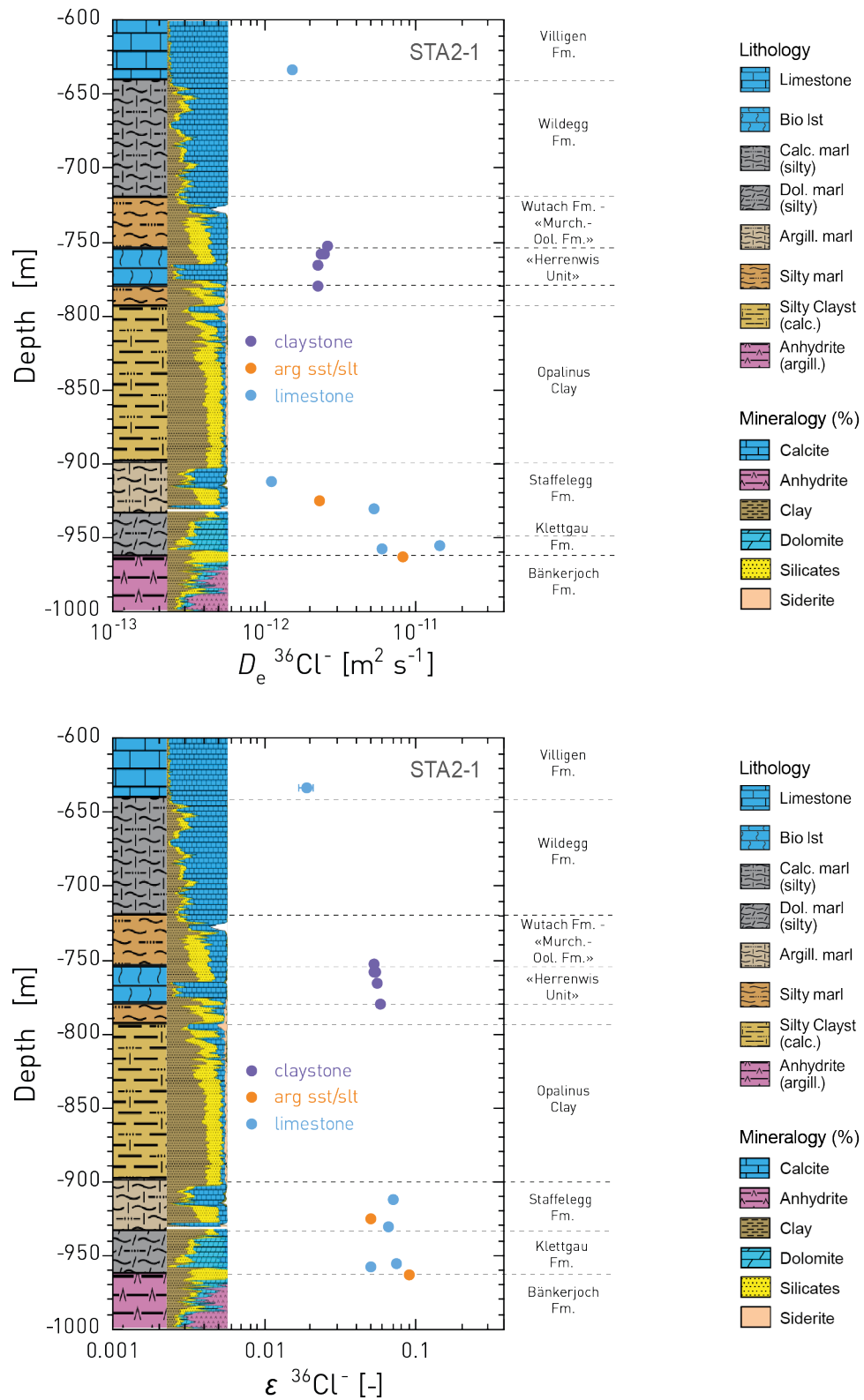


Fig. F-11: Effective diffusion coefficients and accessible porosity values for $^{36}\text{Cl}^-$ as a function of sample depth for the STA2-1 borehole (NL siting region)

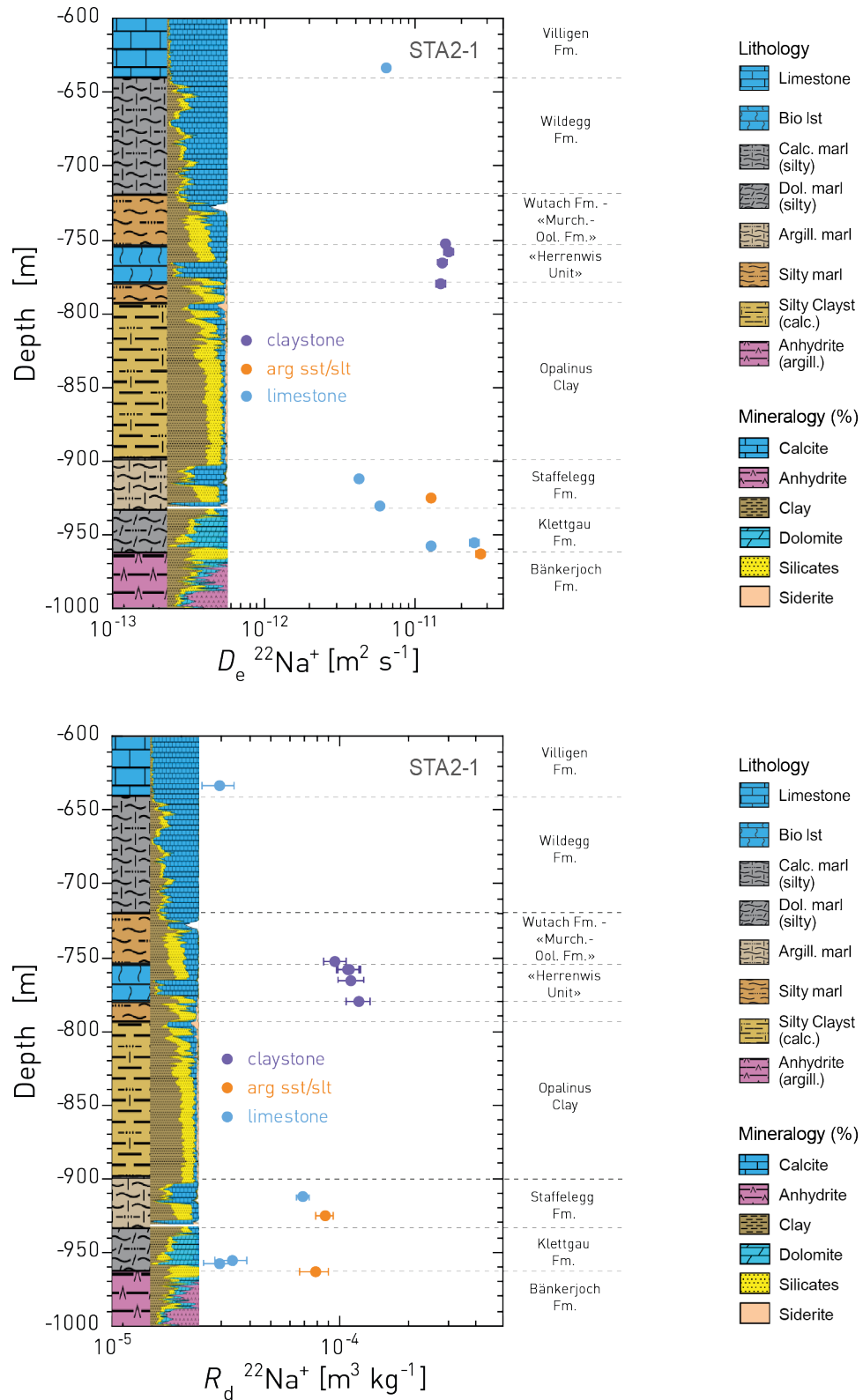


Fig. F-12: Effective diffusion coefficients and R_d values for $^{22}\text{Na}^+$ as a function of sample depth for the STA2-1 borehole (NL siting region)

The porosity values selected for $^{22}\text{Na}^+$ are identical to those for HTO.

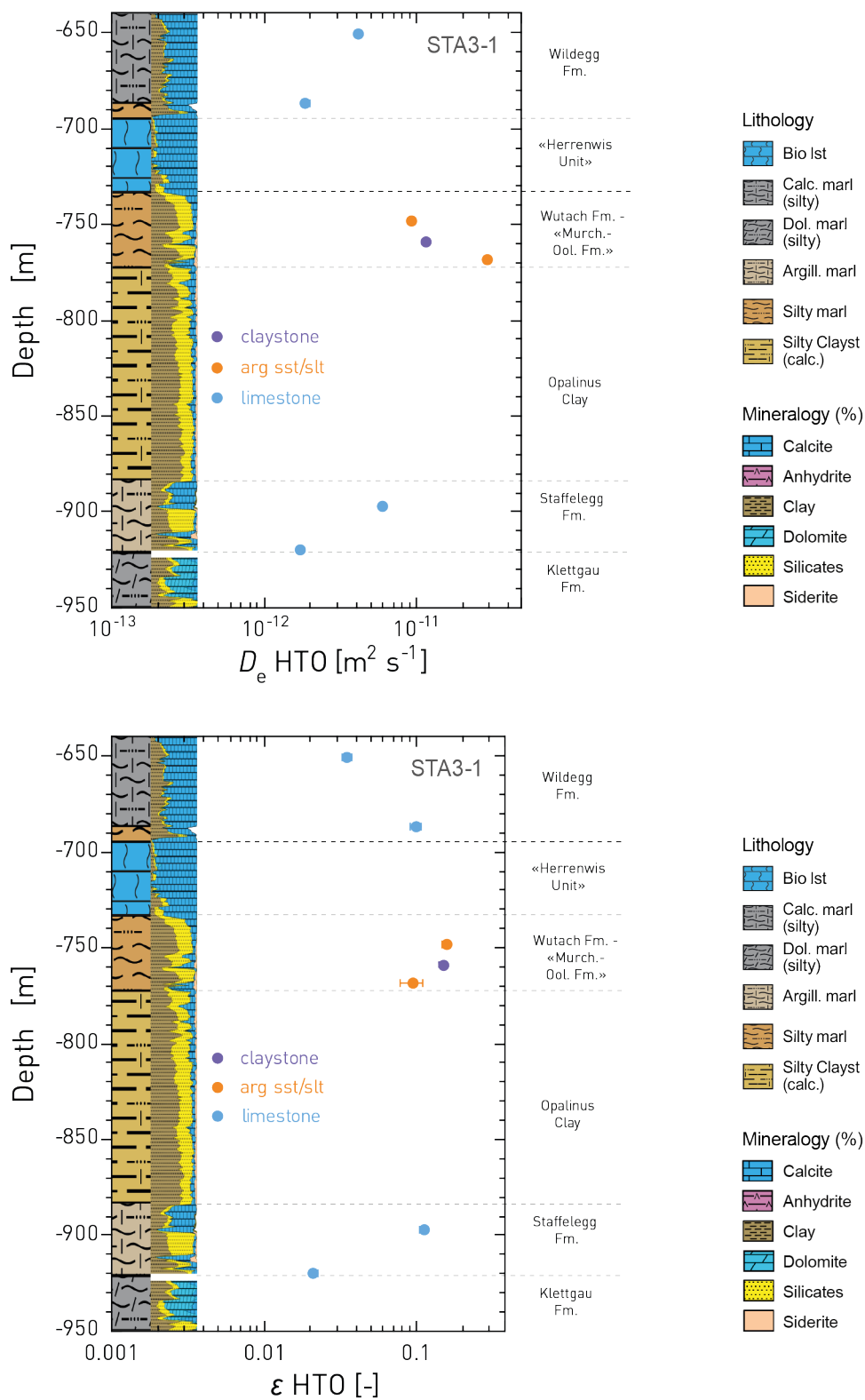


Fig. F-13: Effective diffusion coefficients and accessible porosity values for HTO as a function of sample depth for the STA3-1 borehole (NL siting region)

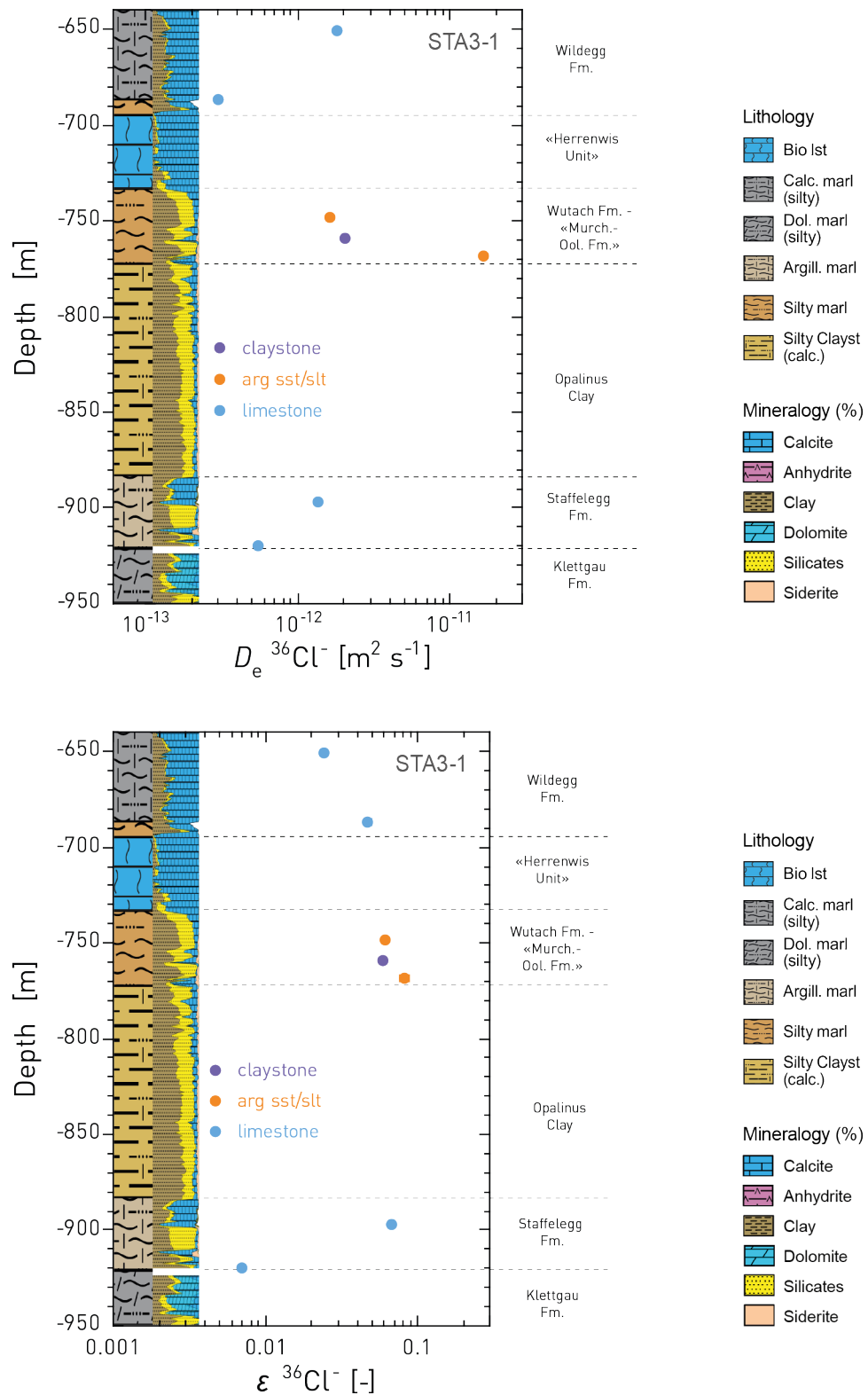


Fig. F-14: Effective diffusion coefficients and accessible porosity values for $^{36}\text{Cl}^-$ as a function of sample depth for the STA3-1 borehole (NL siting region)

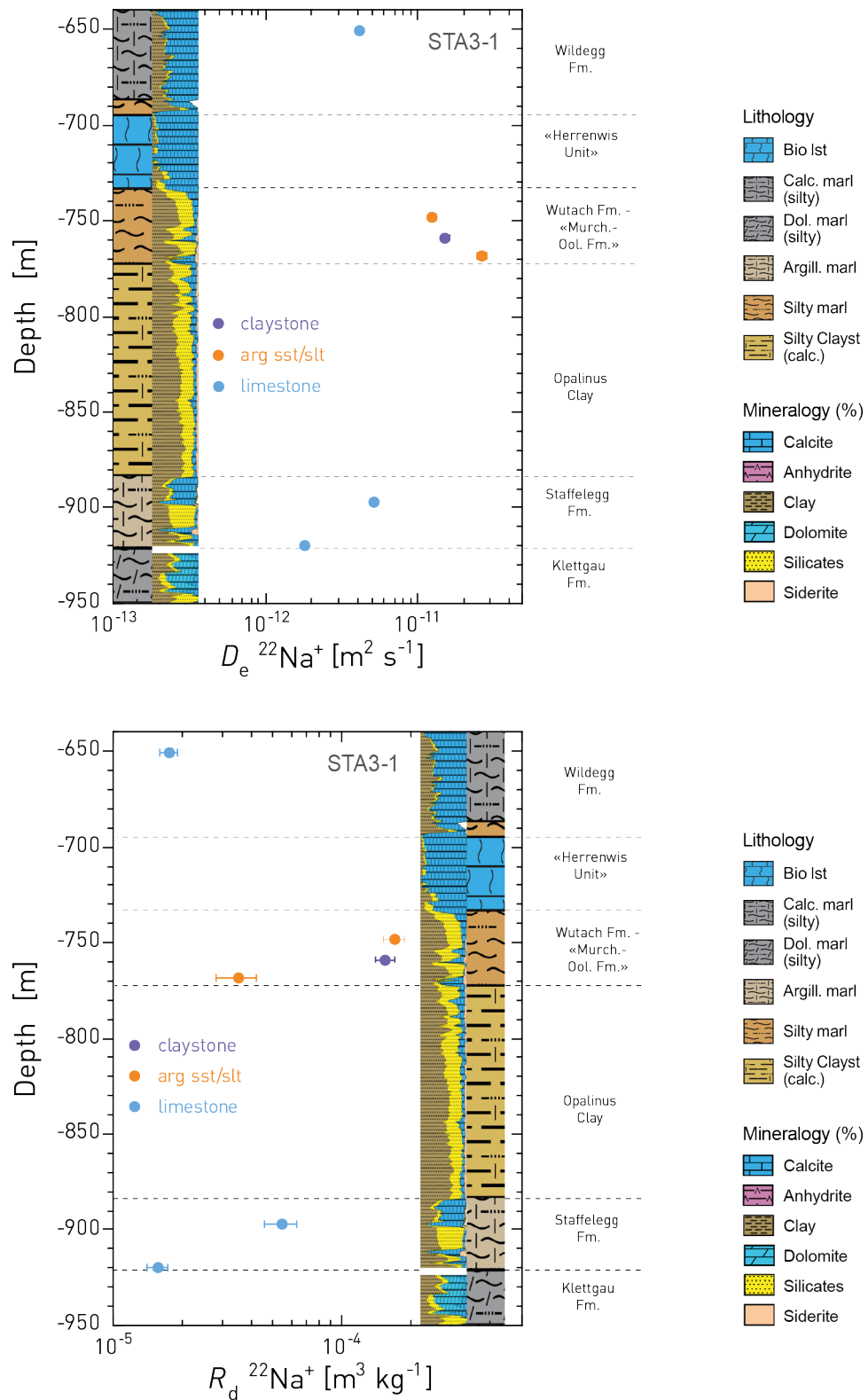


Fig. F-15: Effective diffusion coefficients and R_d values for $^{22}\text{Na}^+$ as a function of sample depth for the STA3-1 borehole (NL siting region)

The porosity values selected for $^{22}\text{Na}^+$ are identical to those for HTO.

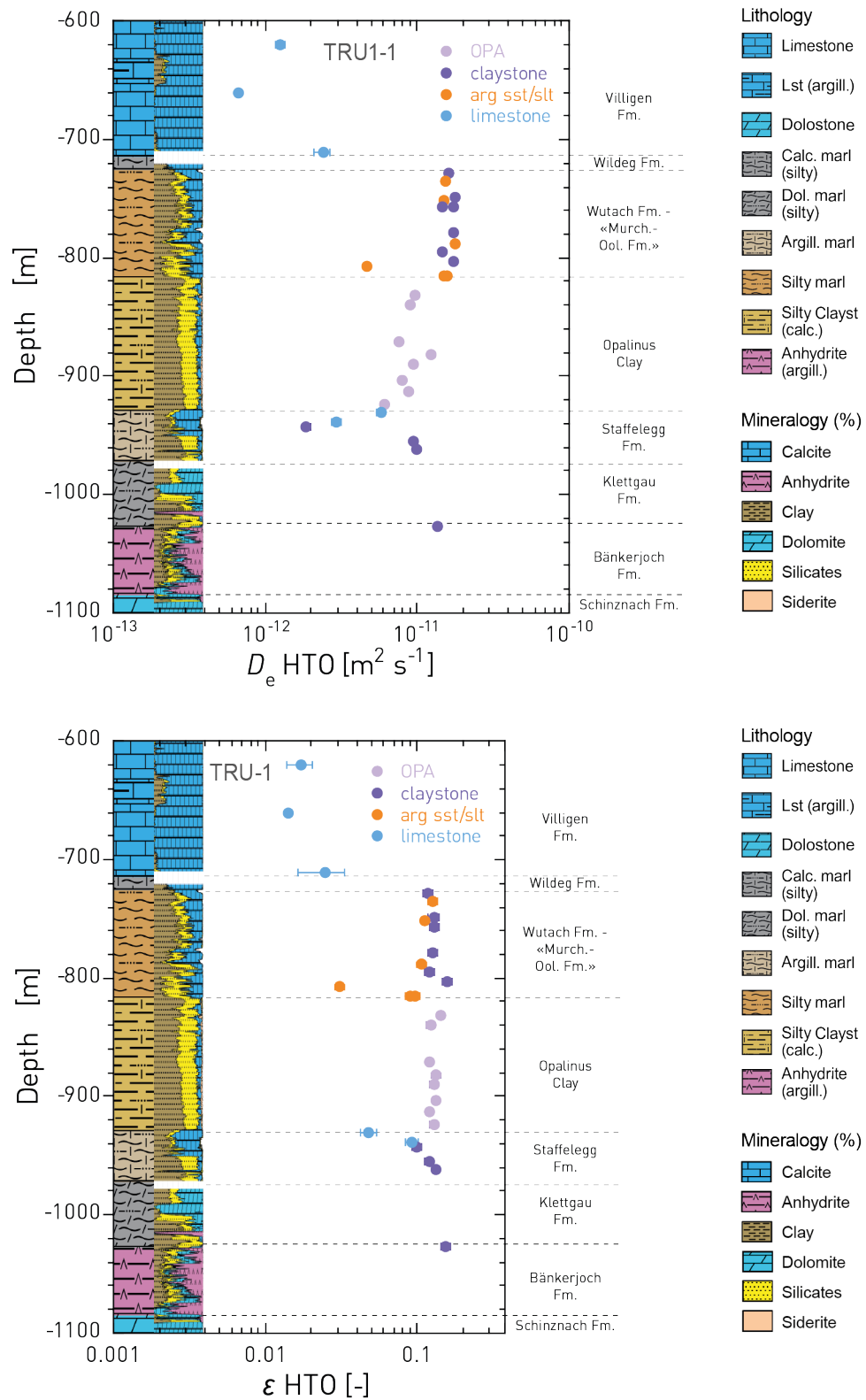


Fig. F-16: Effective diffusion coefficients and accessible porosity values for HTO as a function of sample depth for the TRU1-1 borehole (ZNO siting region)

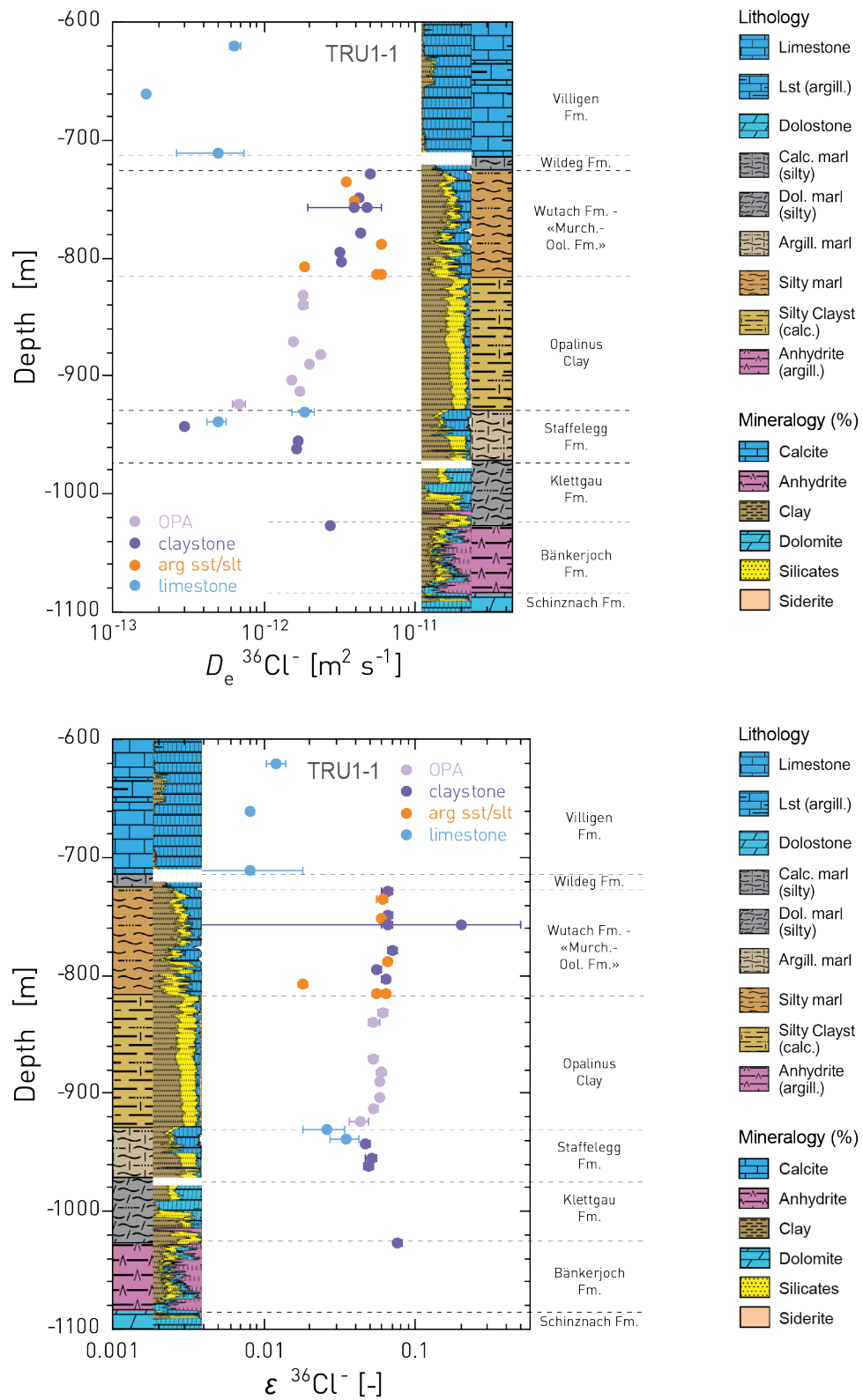


Fig. F-17: Effective diffusion coefficients and accessible porosity values for ${}^{36}\text{Cl}^-$ as a function of sample depth for the TRU1-1 borehole (ZNO siting region)

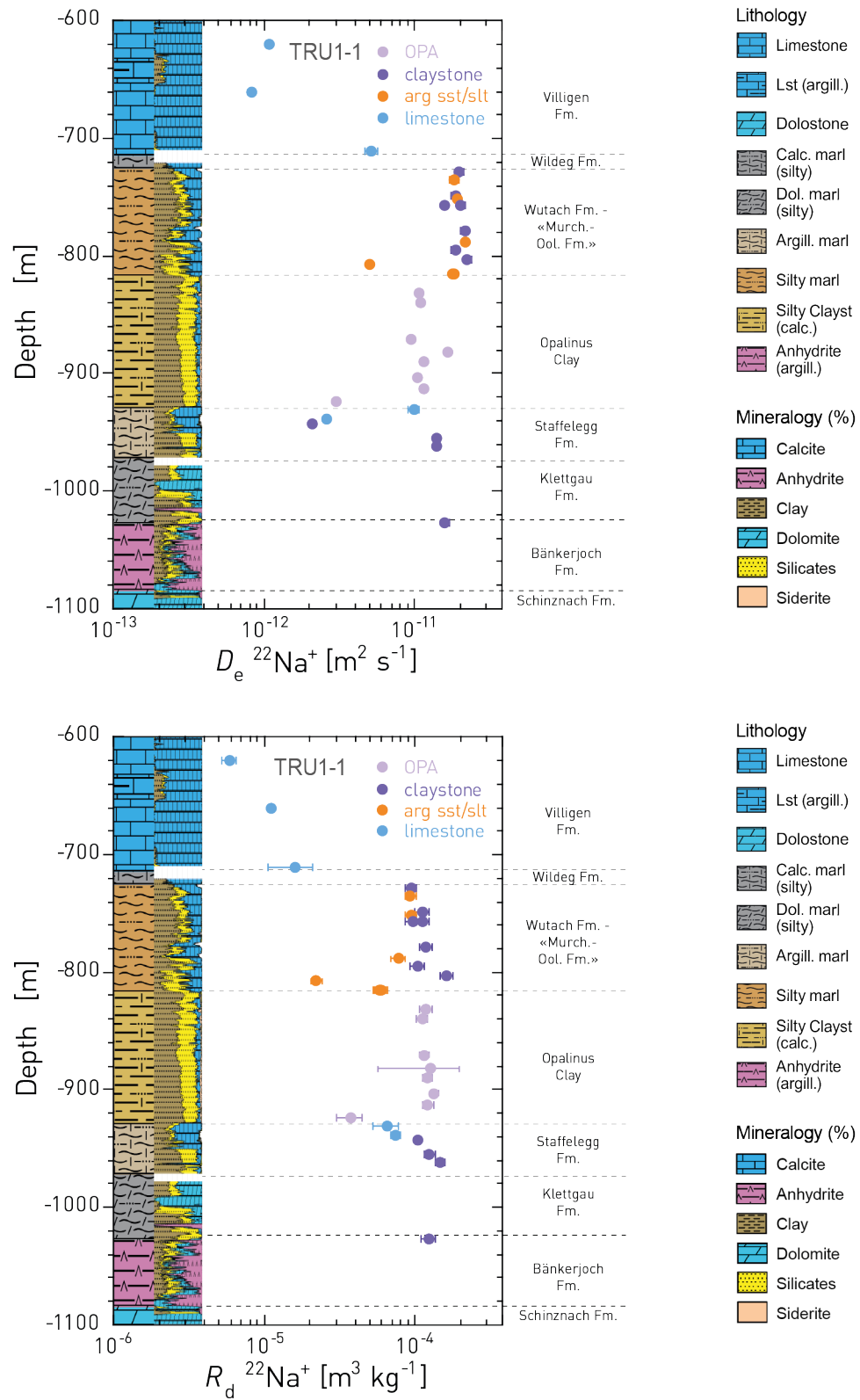


Fig. F-18: Effective diffusion coefficients and R_d values for $^{22}\text{Na}^+$ as a function of sample depth for the TRU1-1 borehole (ZNO siting region)

The porosity values selected for $^{22}\text{Na}^+$ are identical to those for HTO.

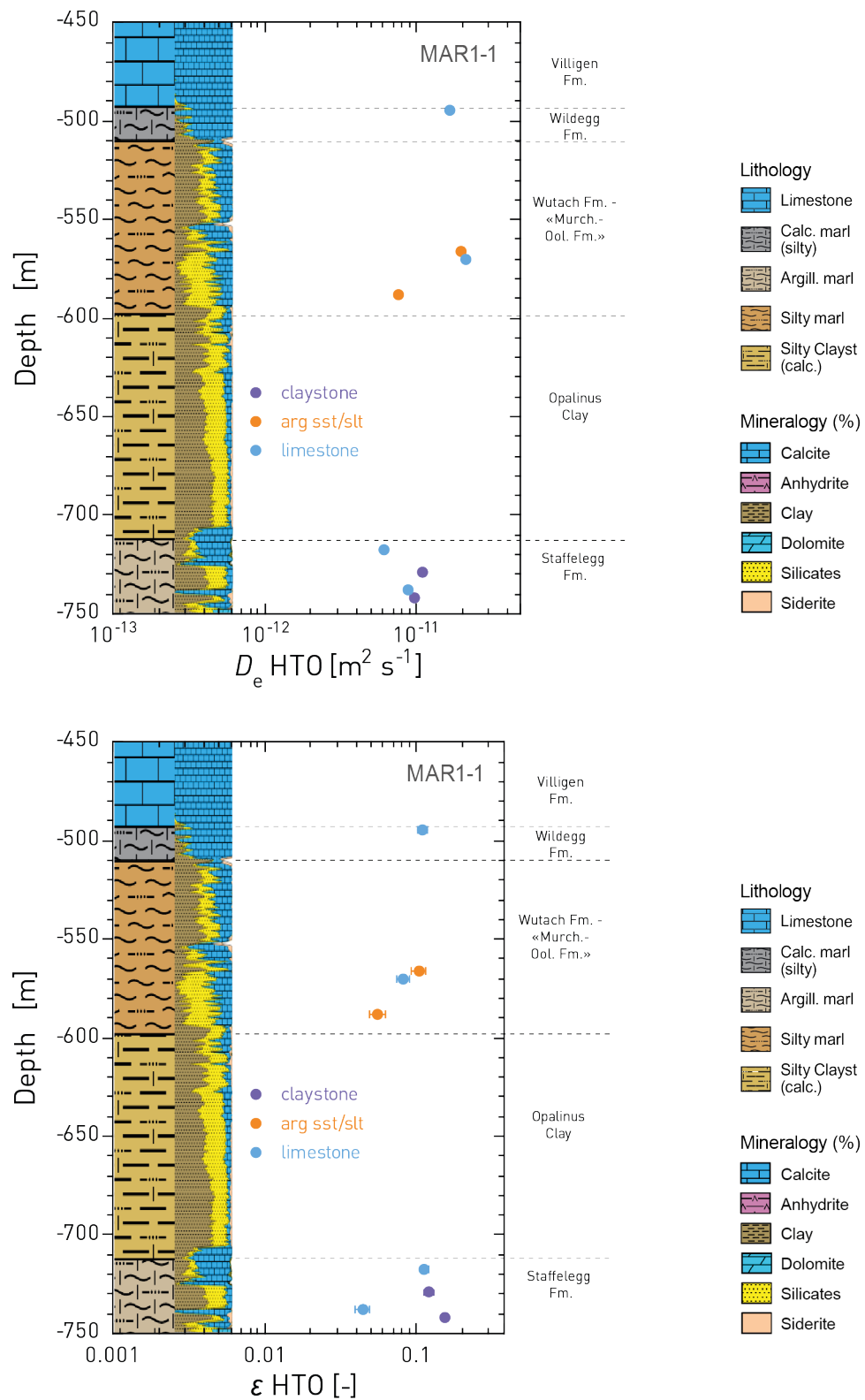


Fig. F-19: Effective diffusion coefficients and accessible porosity values for HTO as a function of sample depth for the MAR1-1 borehole (ZNO siting region)

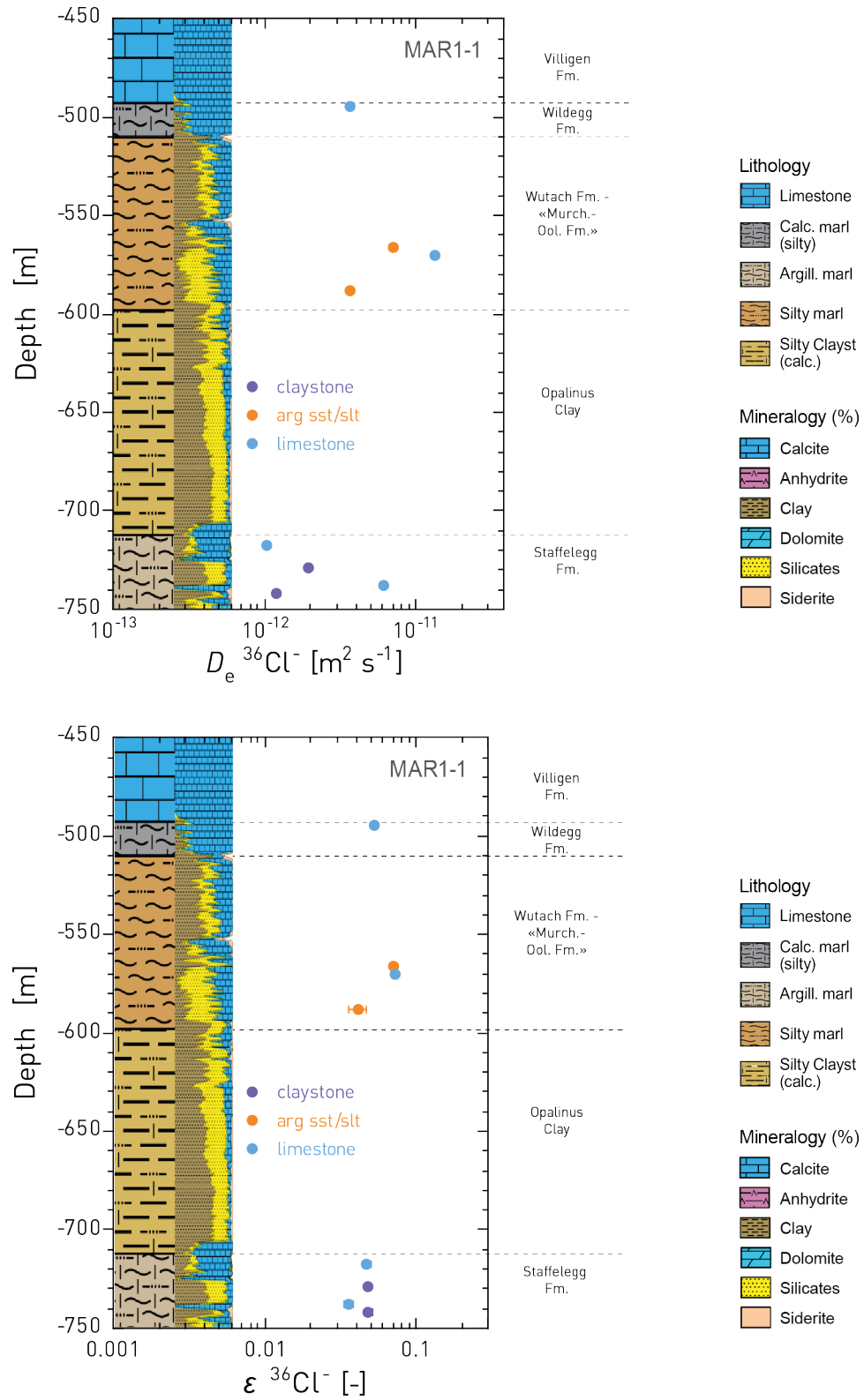


Fig. F-20: Effective diffusion coefficients and accessible porosity values for $^{36}\text{Cl}^-$ as a function of sample depth for the MAR1-1 borehole (ZNO siting region)

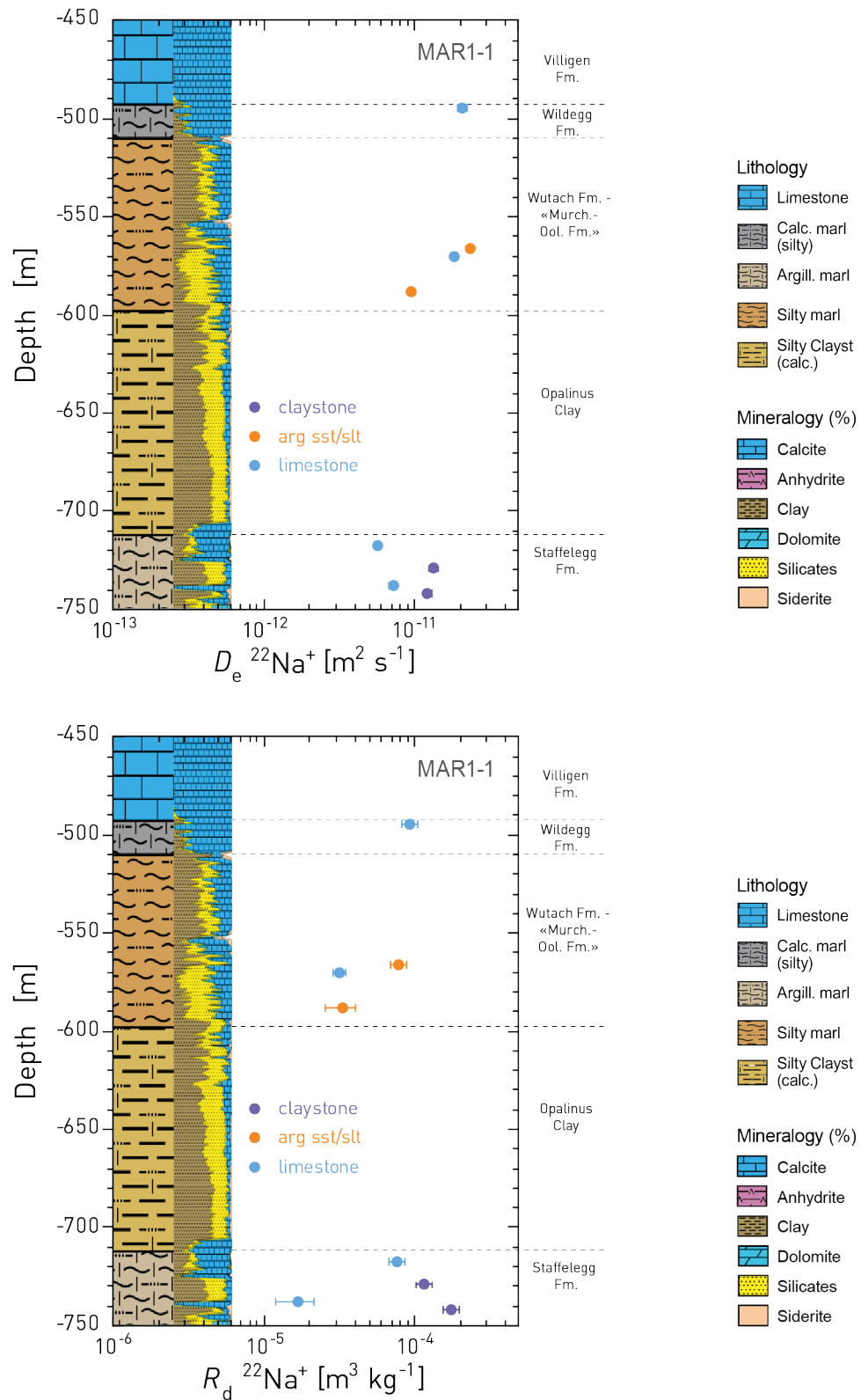


Fig. F-21: Effective diffusion coefficients and R_d values for $^{22}\text{Na}^+$ as a function of sample depth for the MAR1-1 borehole (ZNO siting region)

The porosity values selected for $^{22}\text{Na}^+$ are identical to those for HTO.

App. G Comparison of diffusion data measured at PSI (CH) and SCK-CEN (BE)

SCK-CEN (Belgium) and PSI (Switzerland) measured diffusion parameters for HTO, $^{36}\text{Cl}^-$ and $^{22}\text{Na}^+$ on a few twin samples from the TRU1-1 borehole. The results of the measurements are summarised in Tab. G-1. Besides this exercise, PSI also analysed three sets of twin samples from the BUL1-1 borehole and one set of twin samples of the TRU1-1 borehole. The results are also summarised in Tab. G-1. More details can be found in Van Laer et al. (2024).

Tab. G-1: Overview of the effective diffusion coefficients (D_e) and diffusion-accessible porosities (ε) for HTO, $^{36}\text{Cl}^-$ and rock capacity factors (α) for $^{22}\text{Na}^+$ for selected twin samples from the BUL1-1 and TRU1-1 boreholes

Sample	Exp. data	HTO		$^{36}\text{Cl}^-$		$^{22}\text{Na}^+$	
		ε [-]	D_e [$\times 10^{-12} \text{ m}^2 \text{ s}^{-1}$]	ε [-]	D_e [$\times 10^{-12} \text{ m}^2 \text{ s}^{-1}$]	α [-]	D_e [$\times 10^{-12} \text{ m}^2 \text{ s}^{-1}$]
TRU1-1-870.23-DI	PSI	0.121 ± 0.007	7.6 ± 0.2	0.053 ± 0.003	1.5 ± 0.07	0.41 ± 0.02	9.5 ± 0.4
TRU1-1-882.36-DI	PSI	0.134 ± 0.009	12.3 ± 0.5	0.060 ± 0.004	2.4 ± 0.1	0.44 ± 0.03	16.6 ± 0.9
	SCK (FF)	0.111 ± 0.005	8.46 ± 0.4	0.050 ± 0.002	1.4 ± 0.06	0.720 ± 0.049	13.0 ± 0.9
	SCK (FC)	0.101 ± 0.006	10.9 ± 0.8	0.041 ± 0.002	1.8 ± 0.09	0.576 ± 0.046	16.0 ± 1.3
TRU-1-890.39-DI	PSI	0.129 ± 0.008	9.4 ± 0.3	0.058 ± 0.003	2.0 ± 0.07	0.42 ± 0.02	11.6 ± 0.5
	SCK (FF)	0.117 ± 0.015	8.3 ± 1.1	0.059 ± 0.014	4.0 ± 1.0	0.436 ± 0.043	10.8 ± 1.1
	SCK (FC)	0.110 ± 0.016	11.4 ± 1.8	0.047 ± 0.003	5.0 ± 1.4	0.346 ± 0.040	13.7 ± 1.7
TRU1-1-902.85-DI	PSI	0.134 ± 0.007	8.1 ± 0.3	0.058 ± 0.002	1.5 ± 0.04	0.46 ± 0.02	10.5 ± 0.4
	SCK (FF)	0.099 ± 0.006	6.6 ± 0.4	0.026 ± 0.001	1.4 ± 0.1	0.383 ± 0.013	6.9 ± 0.2
	SCK (FC)	0.083 ± 0.005	8.5 ± 0.6	0.020 ± 0.003	1.8 ± 0.3	0.306 ± 0.012	9.0 ± 0.4
TRU1-1-938.90-DI	PSI	0.092 ± 0.009	3.0 ± 0.2	0.035 ± 0.008	0.49 ± 0.07	0.28 ± 0.01	2.6 ± 0.07
	SCK (FF)	0.084 ± 0.007	2.5 ± 0.2	0.033 ± 0.003	0.49 ± 0.04	0.185 ± 0.010	2.0 ± 0.1
	SCK (FC)	0.070 ± 0.007	3.3 ± 0.4	0.029 ± 0.003	0.68 ± 0.07	0.174 ± 0.010	2.4 ± 0.2
TRU1-1-756.39-DI_S1	PSI	0.131 ± 0.010	14.6 ± 0.6	0.202 ± 0.289	3.93 ± 1.98	0.40 ± 0.04	15.8 ± 0.9
TRU1-1-756.39-DI_S2	PSI	0.130 ± 0.010	17.6 ± 0.6	0.065 ± 0.006	4.81 ± 0.24	0.37 ± 0.04	20.2 ± 1.3
BUL1-1-840.45-DI_S1	PSI	0.012 ± 0.002	4.01 ± 0.10	0.009 ± 0.002	3.02 ± 0.13	0.03 ± 0.01	3.62 ± 0.20
BUL1-1-840.45-DI_S2	PSI	0.032 ± 0.003	6.36 ± 0.14	0.019 ± 0.003	2.85 ± 0.16	0.08 ± 0.01	6.09 ± 0.15
BUL1-1-921.43-DI_S1	PSI	0.111 ± 0.007	9.03 ± 0.27	0.058 ± 0.003	2.47 ± 0.09	0.37 ± 0.03	11.3 ± 0.4
BUL1-1-921.43-DI_S2	PSI	0.088 ± 0.007	8.21 ± 0.23	0.048 ± 0.002	2.95 ± 0.08	0.24 ± 0.02	8.38 ± 0.25
BUL1-1-955.62-DI_S1	PSI	0.133 ± 0.007	8.44 ± 0.21	0.064 ± 0.004	1.04 ± 0.05	0.45 ± 0.03	10.4 ± 0.5
BUL1-1-955.62-DI_S2	PSI	0.108 ± 0.007	8.29 ± 0.21	0.058 ± 0.004	1.05 ± 0.05	0.35 ± 0.03	10.5 ± 0.5

FF: fixed filter parameter values ($\varepsilon_{\text{filter}} = 0.4$; $D_e = 1.4 \times 10^{-10} \text{ m}^2 \text{ s}^{-1}$) were used for modelling.
FC: The same diffusion parameter values were used for filter and clay (F = C).

App. H Image collection of the diffusion samples

H.1 Samples from the Jura Ost siting region

This section documents the final step of the sampling campaign for diffusion experiments (DI) of the Bözberg boreholes (BOZ1-1 and BOZ2-1). In detail, it documents the positioning and selection of the final core slices – which were used for the experiments at PSI – from the subcored and epoxy-embedded samples. The axis of the subcores is parallel to that of the original full core, i.e. normal to bedding. The positioning, selection and description of the slices is mainly based on X-ray CT scans performed at the Institute of Forensic Medicine at the University of Bern. While scans of the whole DI samples were already available, new scans were acquired on the subcored samples and served as a basis for the positioning of the DI slices. On the one hand, these scans allow the assessment of sample homogeneity (or heterogeneity) and, thus, aid the selection of slices such that they are as representative as possible. Moreover, these scans make it possible to identify features such as cracks or fractures (either natural or artificially induced), large pyrite concretions, siderite lenses, macropores (i.e. in calcareous lithologies), stylolites or burrows, which could affect the experiments. Such features were avoided.

In general, 1 to 2 DI slices were collected per sample, of which 1 slice was selected for the PSI experiments. The slice selection is documented in various CT images that are generally arranged as image pairs, one on the left side and one on the right side. Such an image pair illustrates two sections of the subcores parallel to the core axis that are oriented orthogonally with respect to each other. The position of the DI slices within the subcored samples (i.e. top and bottom of slices) is shown in the CT image on the left-hand side, whereas special features relevant to sampling can be illustrated in the second CT image on the right-hand side.

Tab. H-1: Depth information for all samples from Bözberg used for diffusion experiments. The values in grey text are samples for which no diffusion data are available.

BOZ1-1	BOZ2-1
Mean depth [m]	Mean depth [m]
266.75	313.13
281.72	367.56
290.63	402.37
296.08	443.93
301.54	588.64
323.16	595.15
329.10	602.23
353.36	606.95
391.30	
416.35	
434.29	
438.20	
440.55	
452.61	
487.01	
493.15	
498.32	
503.87	
512.03	
521.50	

BOZ1-1	BOZ2-1
529.56	
550.63	
562.07	
572.43	
580.34	
597.28	
610.79	
619.10	
623.60	
634.90	
643.80	
646.44	
665.00	
670.95	
679.67	
681.57	
686.81	
717.96	
723.97	
743.10	
790.16	
808.61	

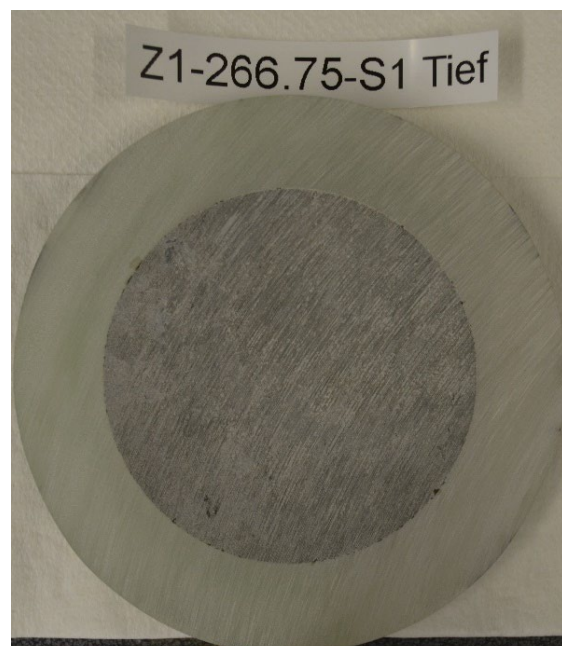
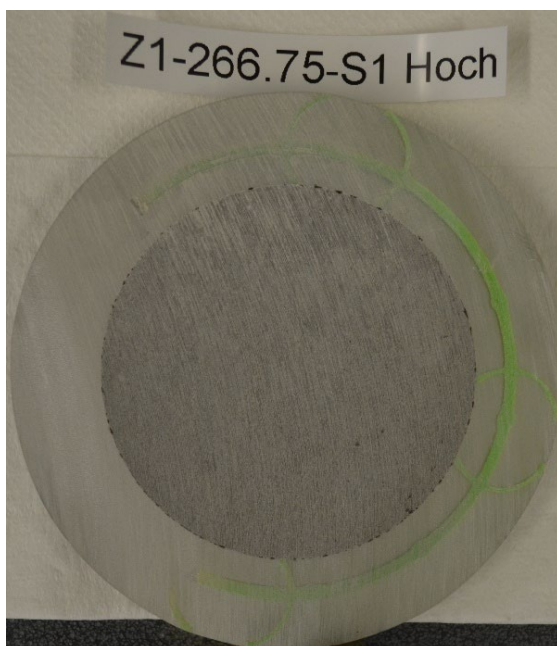
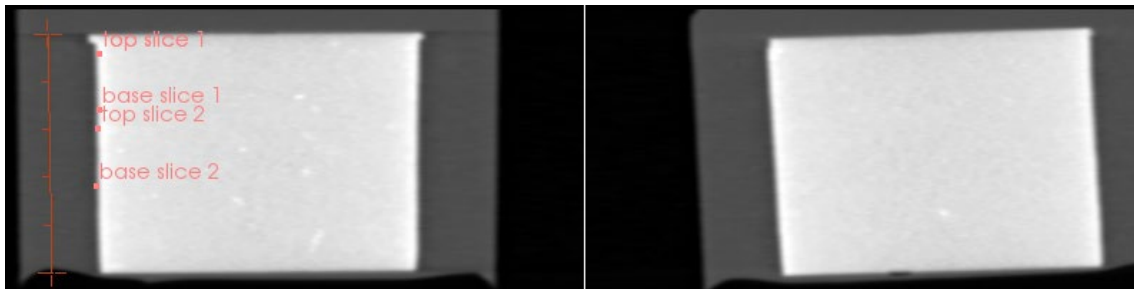
H.1.1 Bözberg 1-1**BOZ1-1-266.75-DI**

Mean depth [m]: 266.75

Stratigraphic position: Wildegg (Malm)

CT-based slice selection: There are bioturbation and concretions but less so in Slice 1.

To be analysed: Slice 1



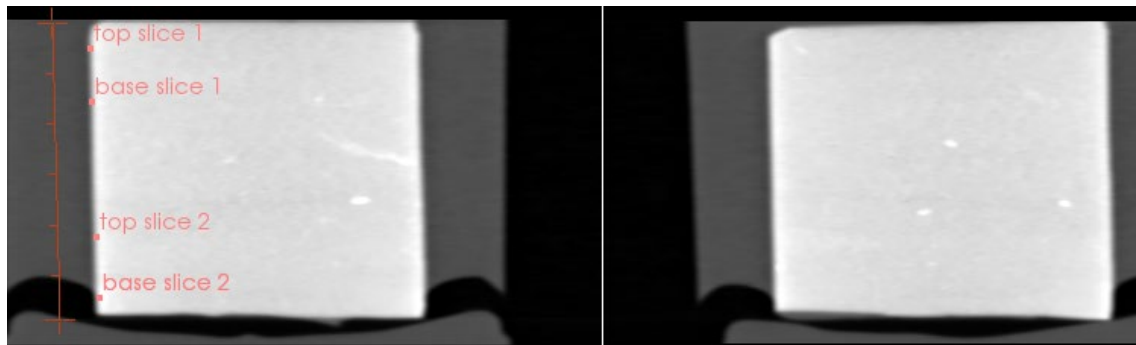
BOZ1-1-281.72-DI

Mean depth [m]: 281.72

Stratigraphic position: Wildegg (Malm)

CT-based slice selection: Slice 1 is less affected by bioturbation.

To be analysed: Slice 1



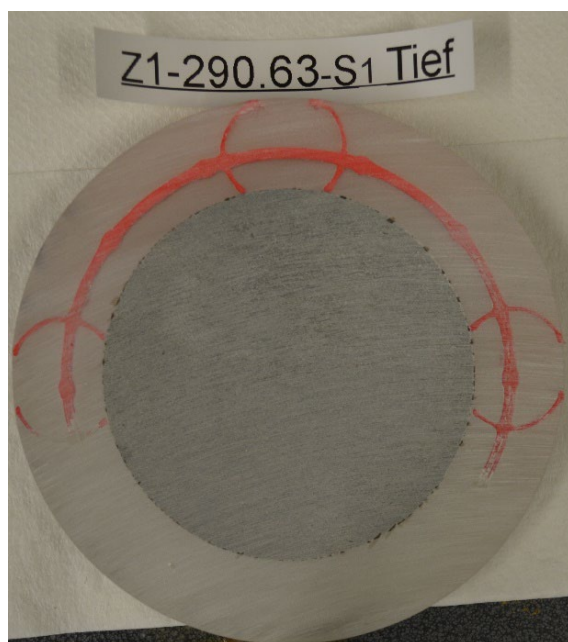
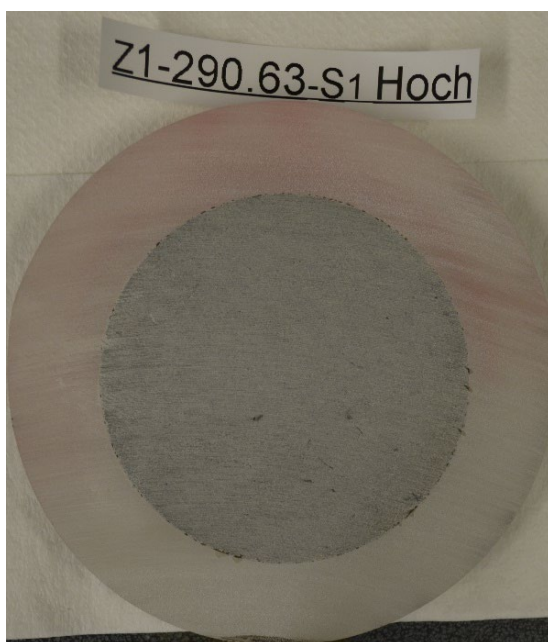
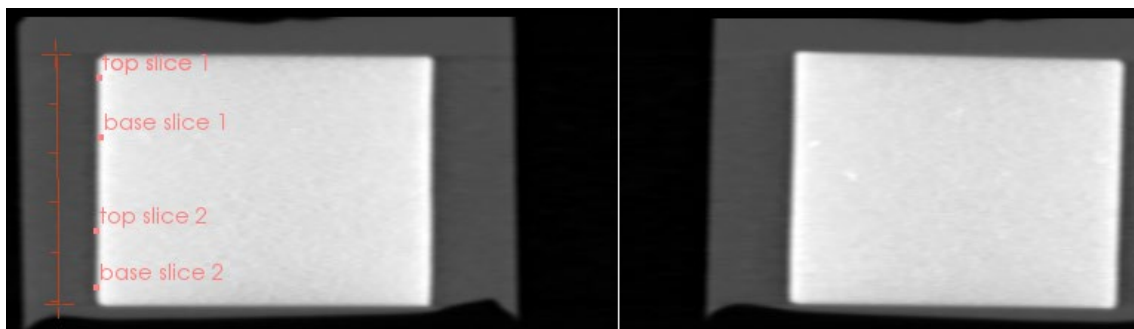
BOZ1-1-290.63-DI

Mean depth [m]: 290.63

Stratigraphic position: Wildegg (Malm)

CT-based slice selection: Slice 1 is less disturbed by bioturbation than Slice 2.

To be analysed: Slice 1



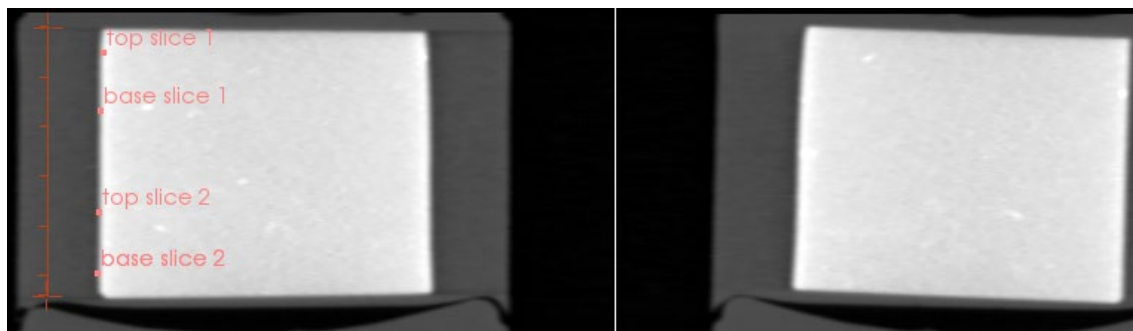
BOZ1-1-296.08-DI

Mean depth [m]: 296.08

Stratigraphic position: Wildegg (Malm)

CT-based slice selection: Slice 1 is less disturbed by pyrite/siderite nodules and vertically oriented bioturbation.

To be analysed: Slice 1

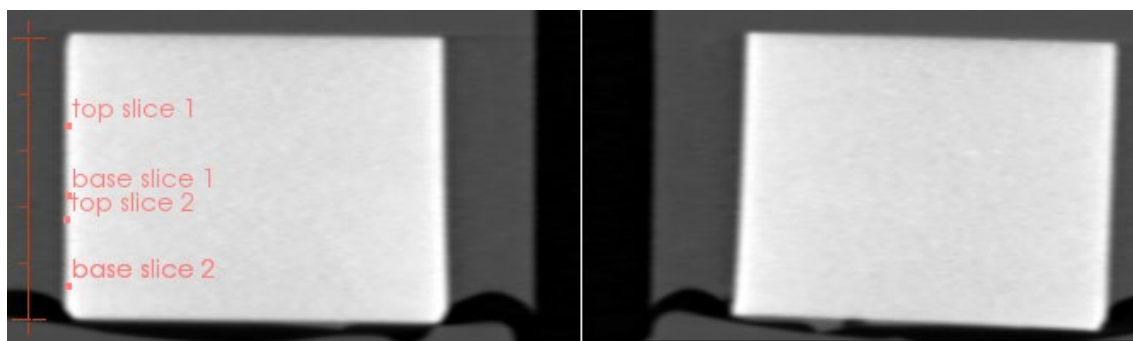
**BOZ1-1-301.54-DI**

Mean depth [m]: 301.54

Stratigraphic position: Wildegg (Malm)

CT-based slice selection: There is no visible difference between Slices 1 and 2.

To be analysed: Slice 1



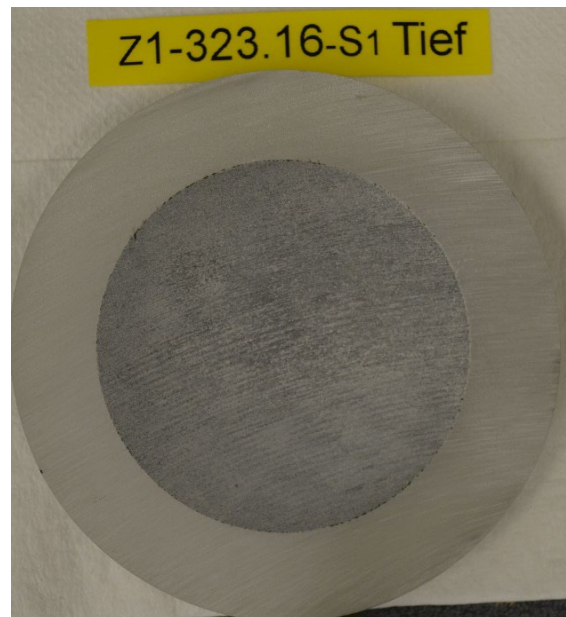
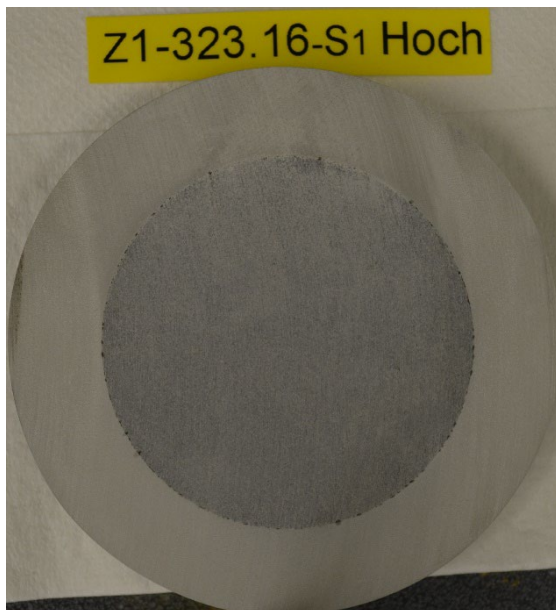
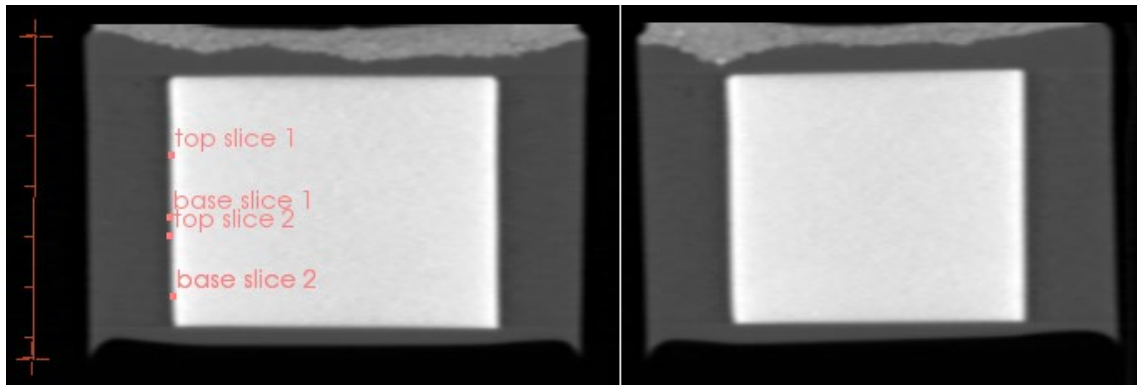
BOZ1-1-323.16-DI

Mean depth [m]: 323.16

Stratigraphic position: Wildegg (Malm)

CT-based slice selection: There is no visible difference between Slices 1 and 2.

To be analysed: Slice 1



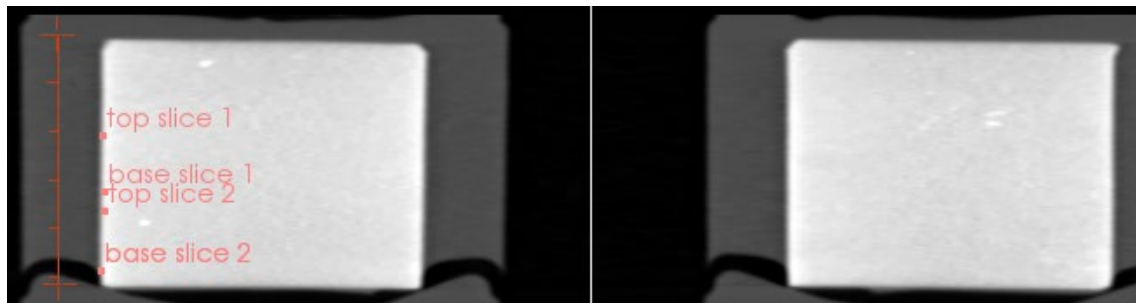
BOZ1-1-329.10-DI

Mean depth [m]: 329.10

Stratigraphic position: Wildegg (Malm)

CT-based slice selection: Slice 2 is less affected by pyrite concretions.

To be analysed: Slice 2



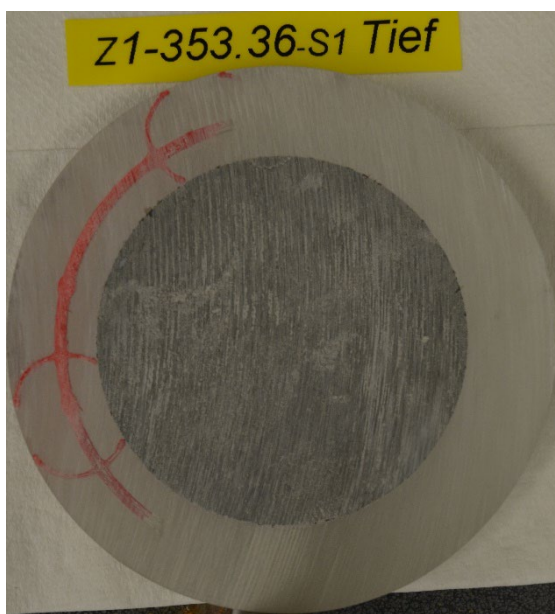
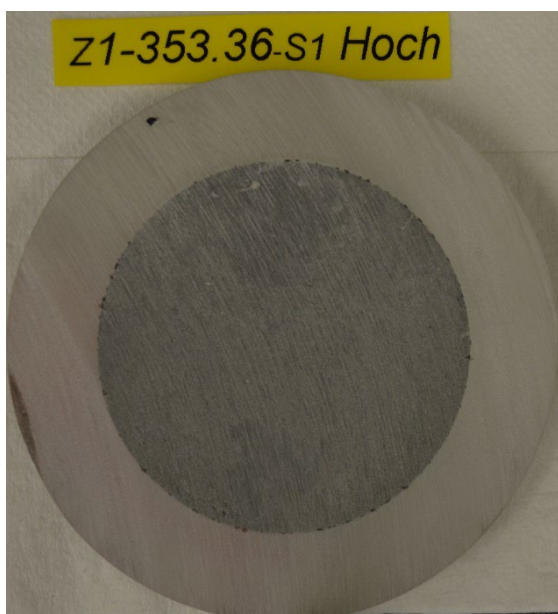
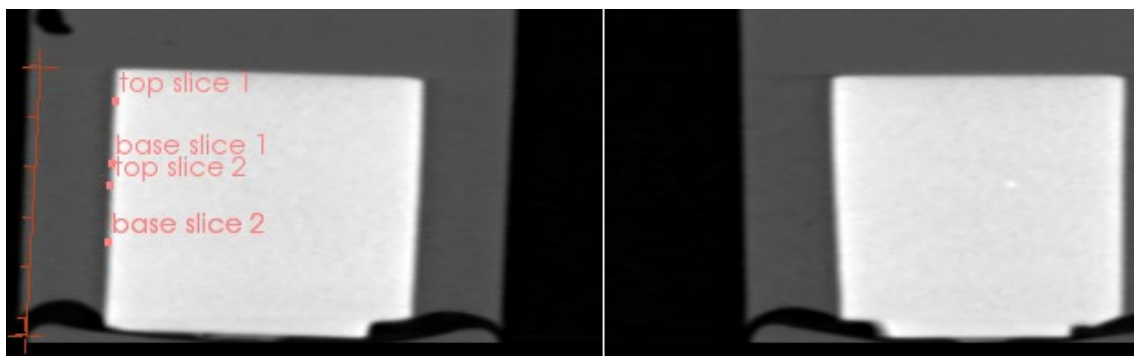
BOZ1-1-353.36-DI

Mean depth [m]: 353.36

Stratigraphic position: Wildegg (Malm)

CT-based slice selection: Slice 1 is less disturbed by concretions and bioturbation.

To be analysed: Slice 1



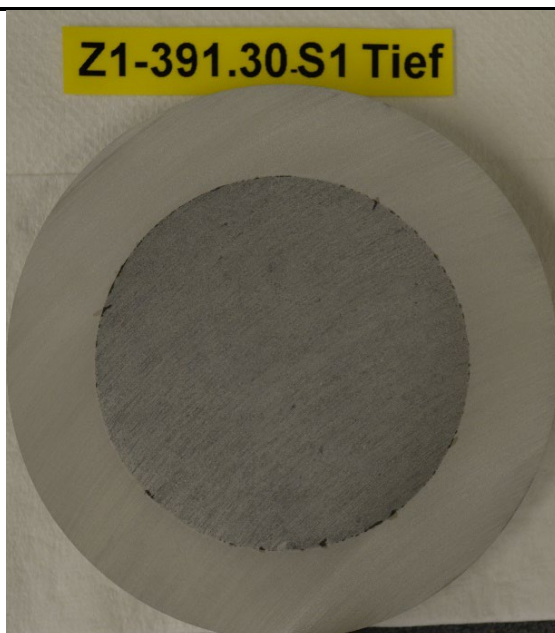
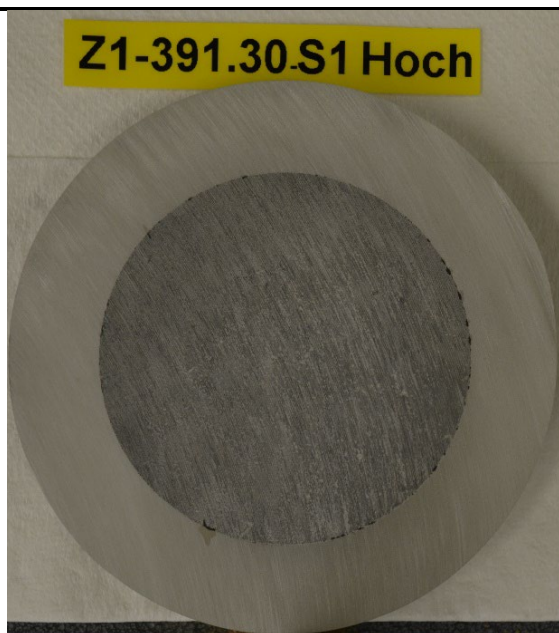
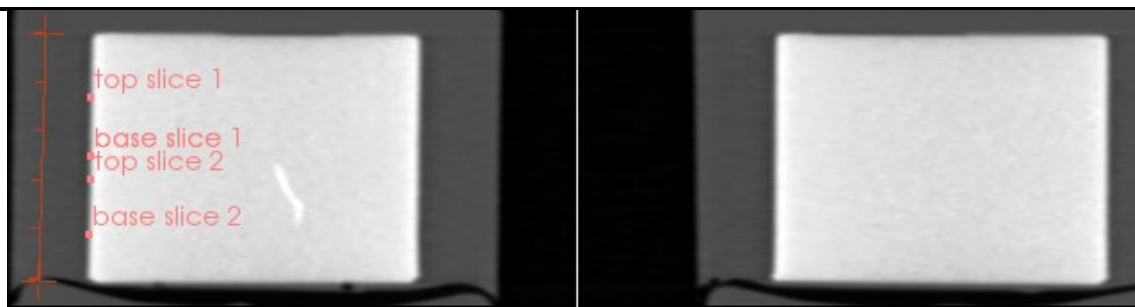
BOZ1-1-391.30-DI

Mean depth [m]: 391.30

Stratigraphic position: Wildegg (Malm)

CT-based slice selection: Slice 2 has vertically oriented bioturbation cutting through and was therefore not chosen.

To be analysed: Slice 1



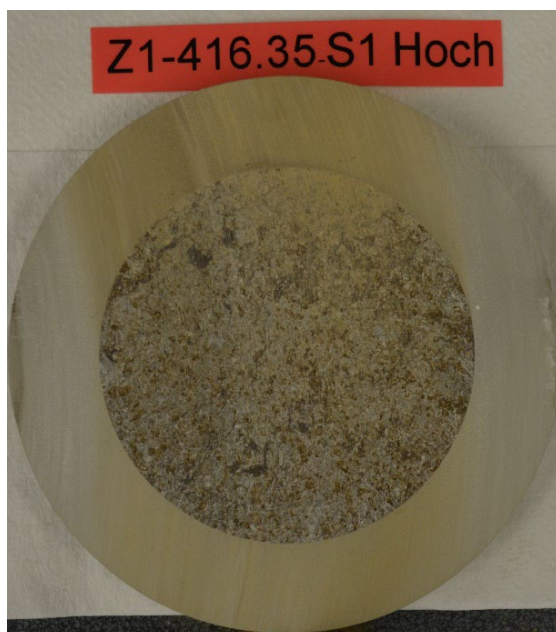
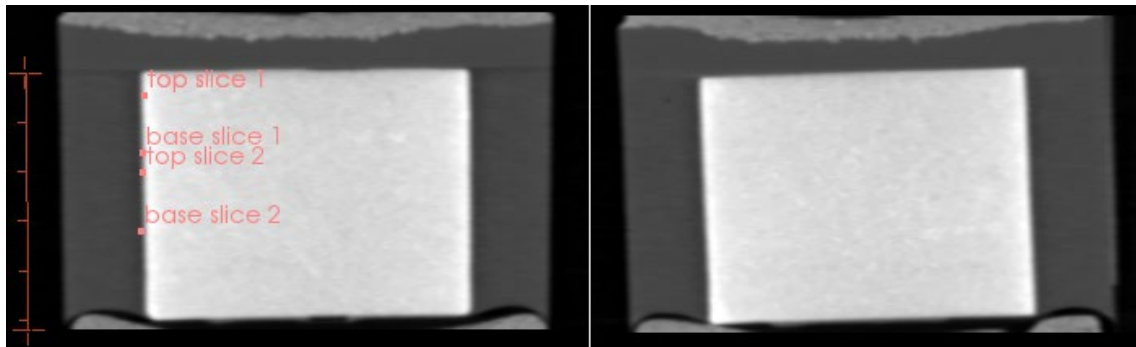
BOZ1-1-416.35-DI

Mean depth [m]: 416.35

Stratigraphic position: Ifenthal (Dogger)

CT-based slice selection: There is no visible difference between Slices 1 and 2.

To be analysed: Slice 1



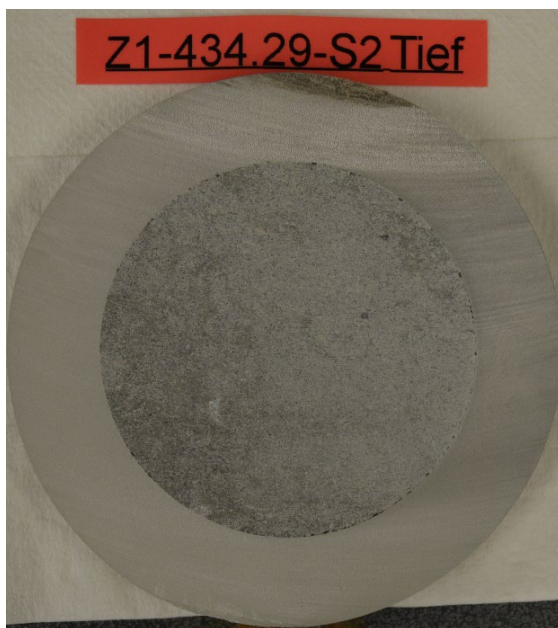
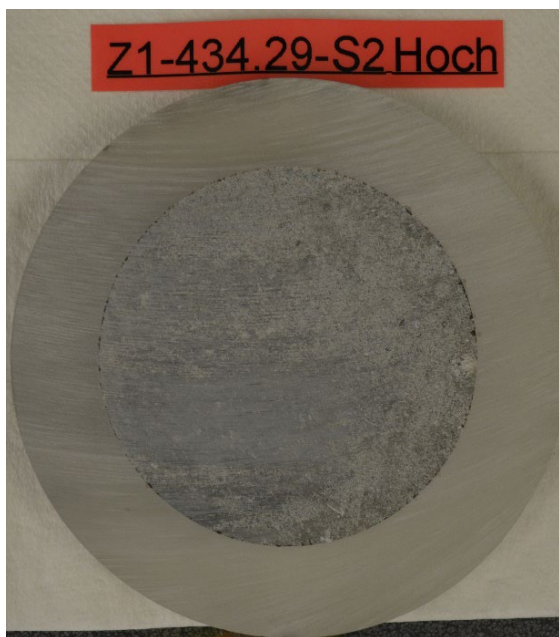
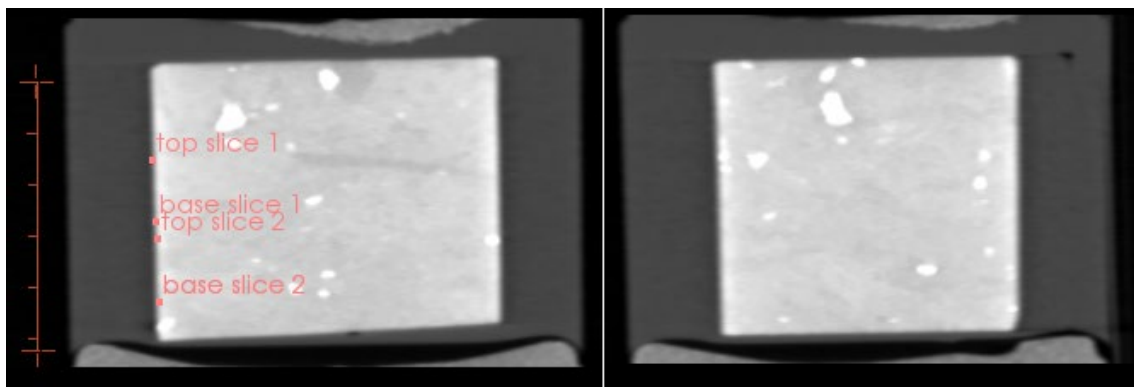
BOZ1-1-434.29-DI

Mean depth [m]: 434.29

Stratigraphic position: Hauptrogenstein (Dogger)

CT-based slice selection: Many concretions and heterogeneities although less in Slice 2.

To be analysed: Slice 2



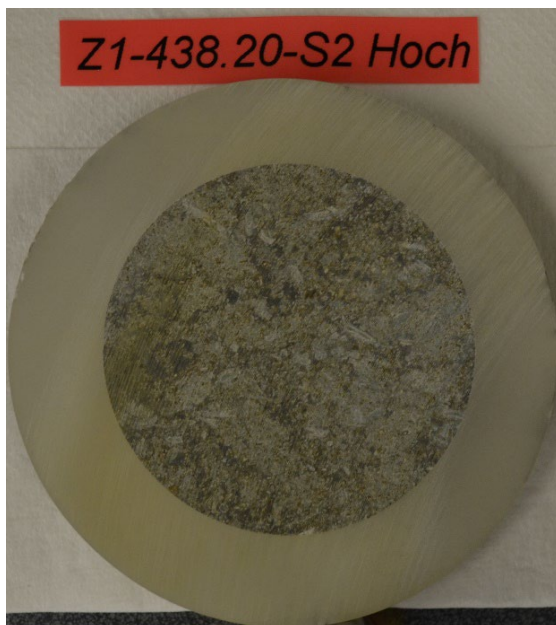
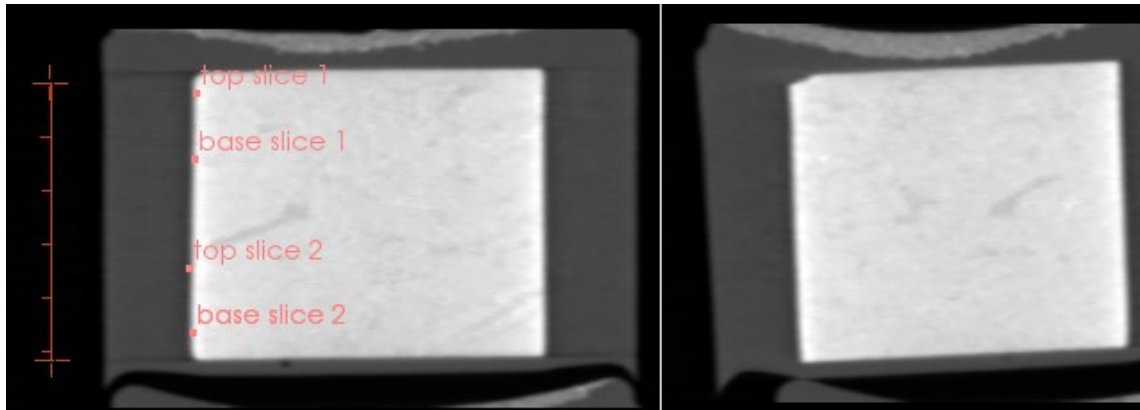
BOZ1-1-438.20-DI

Mean depth [m]: 438.20

Stratigraphic position: Hauptrogenstein (Dogger)

CT-based slice selection: Lot of heterogeneous structures. Slice 2 is a bit more homogeneous.

To be analysed: Slice 2



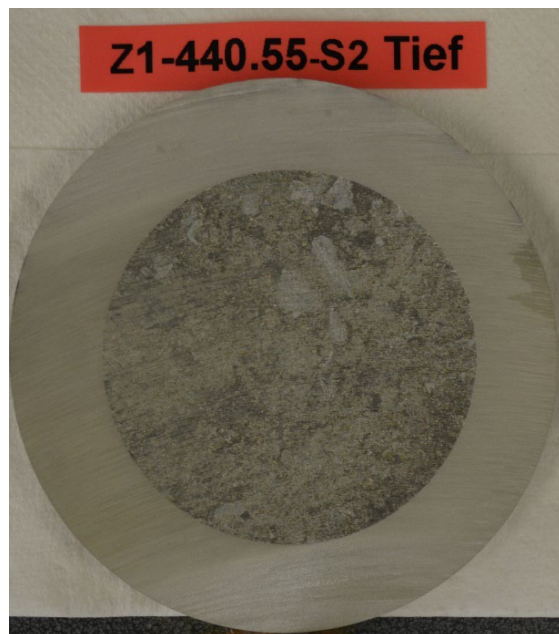
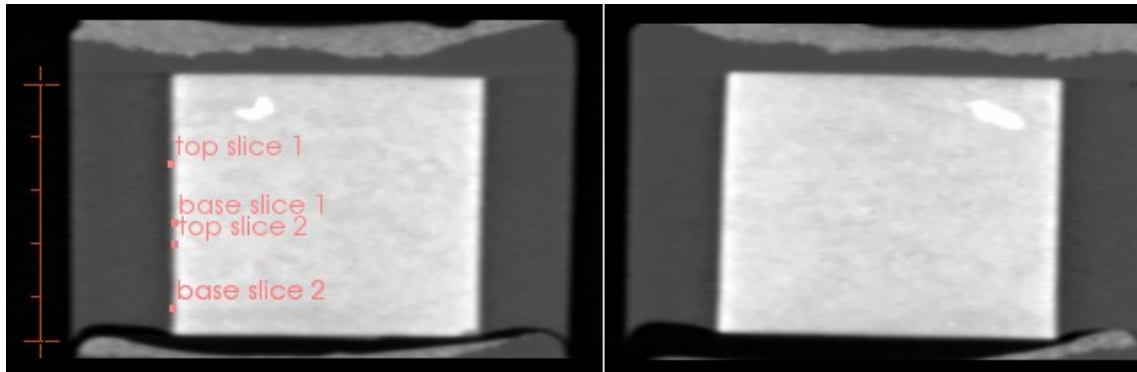
BOZ1-1-440.55-DI

Mean depth [m]: 440.55

Stratigraphic position: Hauptrogenstein (Dogger)

CT-based slice selection: Some heterogeneities on top of the segment. Therefore, Slice 2 was chosen.

To be analysed: Slice 2



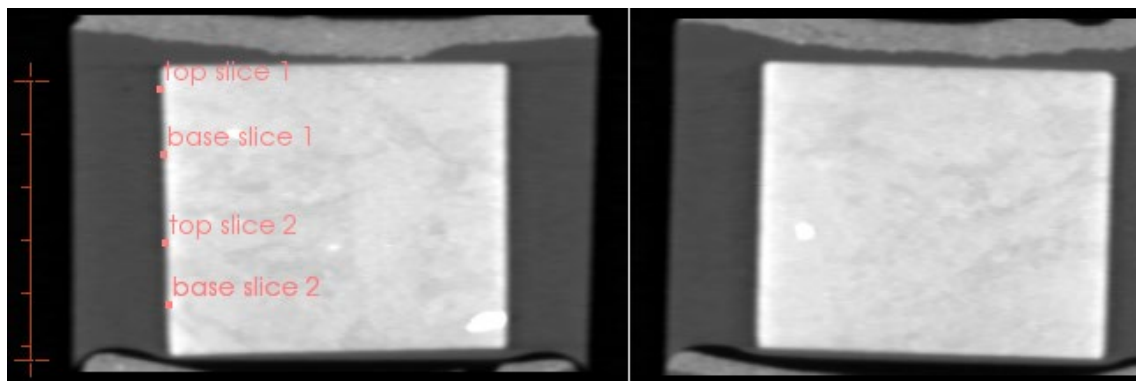
BOZ1-1-452.61-DI

Mean depth [m]: 452.61

Stratigraphic position: Hauptrogenstein (Dogger)

CT-based slice selection: Some bioturbation and stylolites but less in Slice 1.

To be analysed: Slice 1



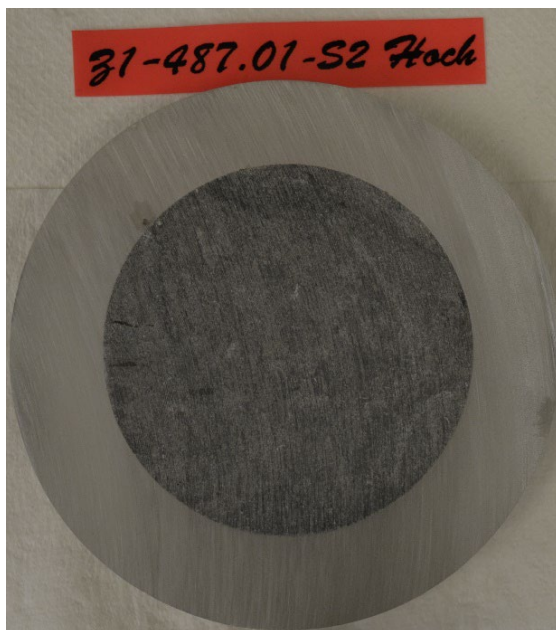
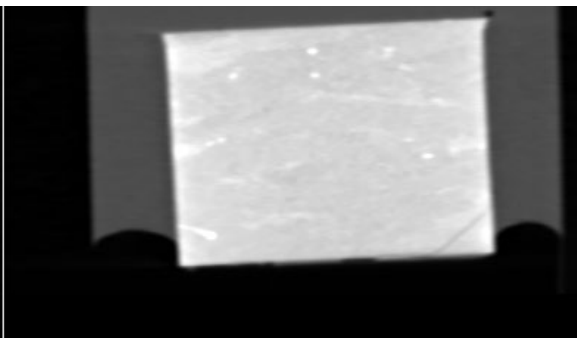
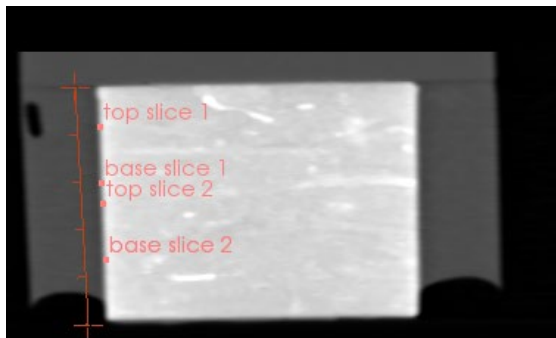
BOZ1-1-487.01-DI

Mean depth [m]: 487.01

Stratigraphic position: Passwang (Dogger)

CT-based slice selection: Slice 1 contains a stylolite and vertically oriented bioturbation. Therefore, Slice 2 was selected.

To be analysed: Slice 2



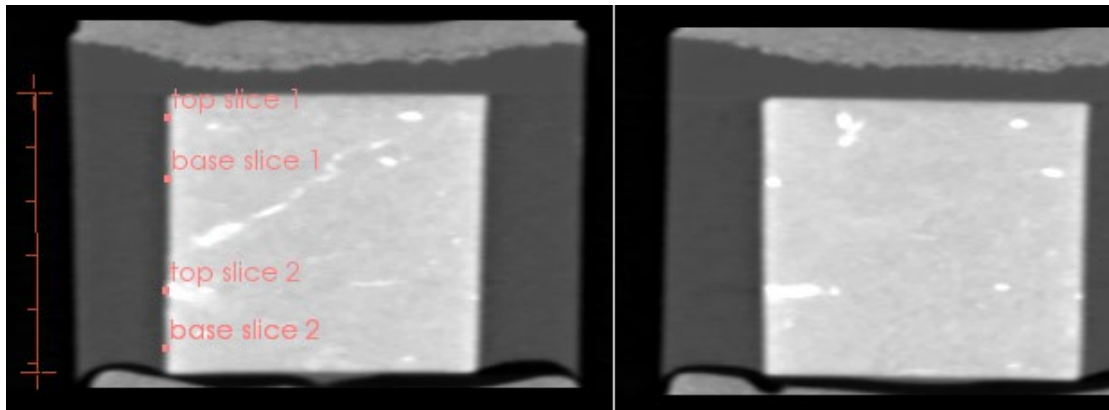
BOZ1-1-493.15-DI

Mean depth [m]: 493.15

Stratigraphic position: Passwang (Dogger)

CT-based slice selection: A lot of bioturbation but somewhat less in Slice 2.

To be analysed: Slice 2



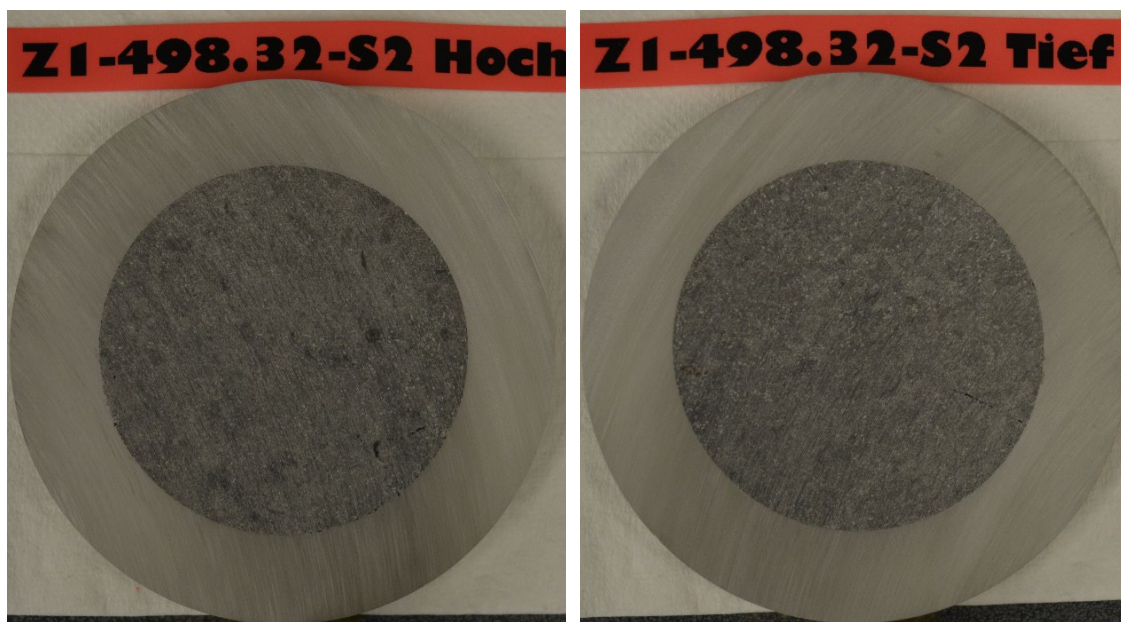
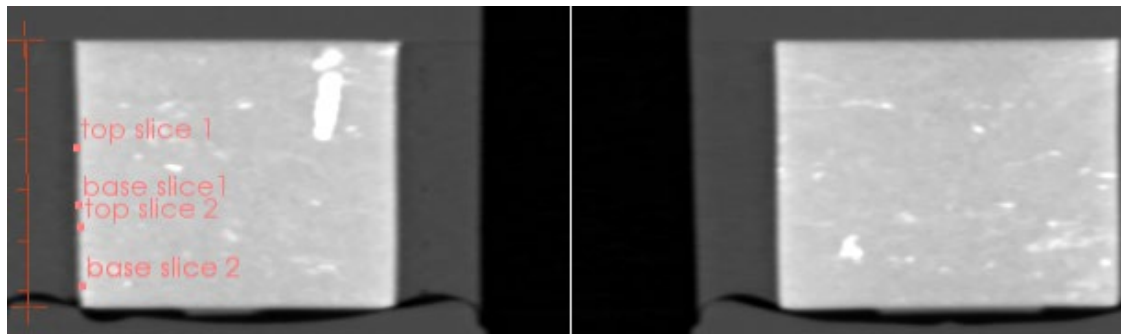
BOZ1-1-498.32-DI

Mean depth [m]: 498.32

Stratigraphic position: Passwang (Dogger)

CT-based slice selection: Slice 2 is less affected by large vertically oriented bioturbation.

To be analysed: Slice 2



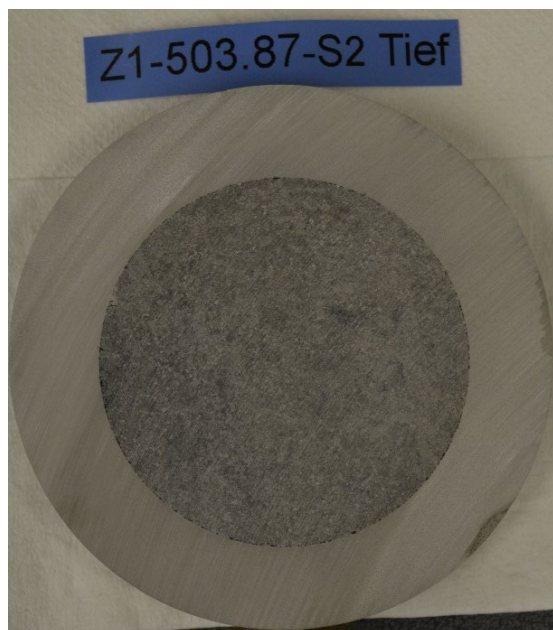
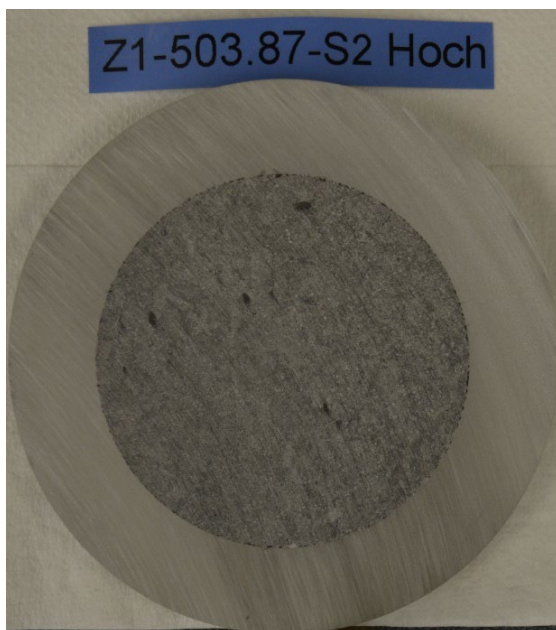
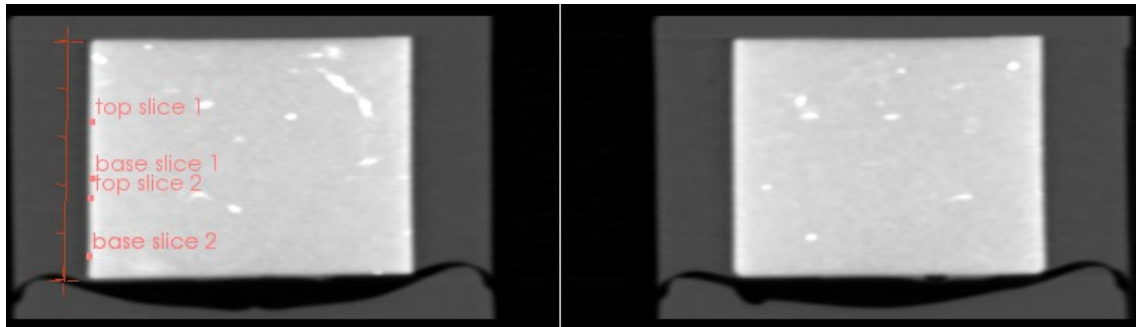
BOZ1-1-503.87-DI

Mean depth [m]: 503.87

Stratigraphic position: Passwang (Dogger)

CT-based slice selection: Slice 1 contains more vertically oriented bioturbation. Therefore, Slice 2 was selected.

To be analysed: Slice 2



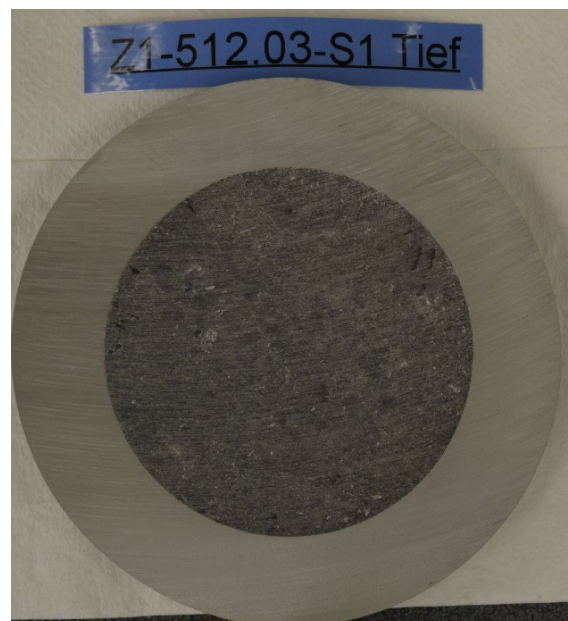
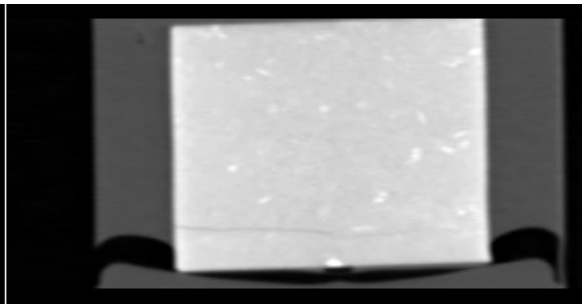
BOZ1-1-512.03-DI

Mean depth [m]: 512.03

Stratigraphic position: Passwang (Dogger)

CT-based slice selection: Slice 1 was selected as it is less disturbed by bioturbation and stylolite.

To be analysed: Slice 1



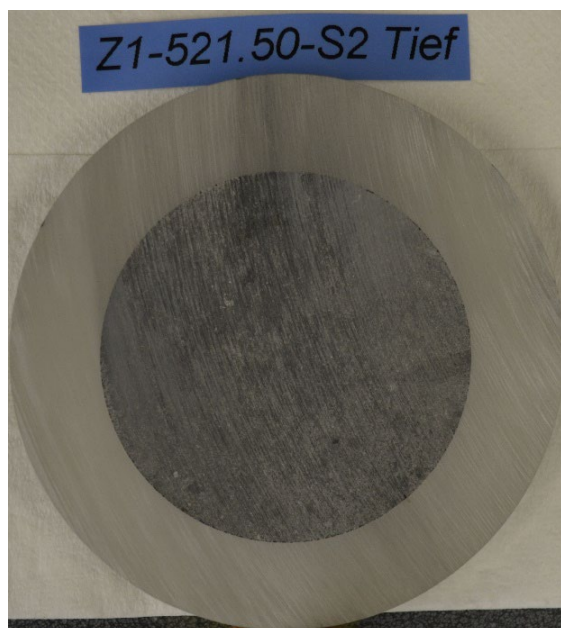
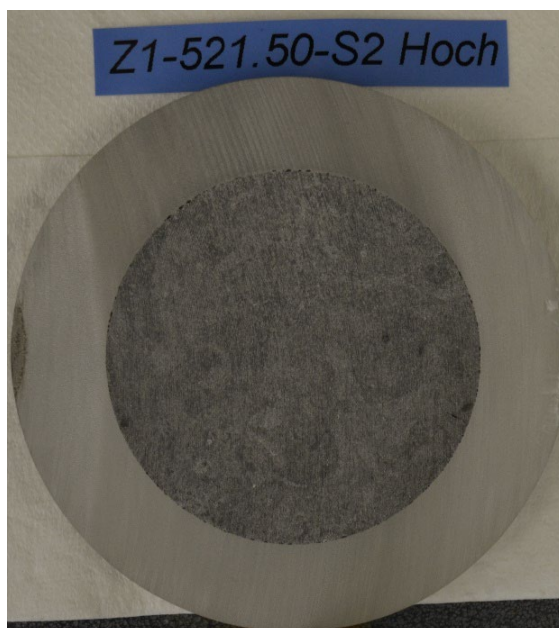
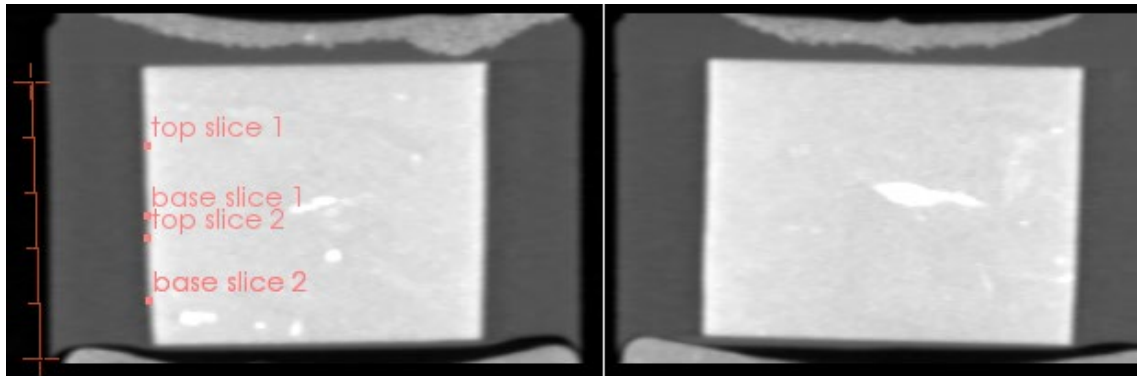
BOZ1-1-521.50-DI

Mean depth [m]: 521.50

Stratigraphic position: Passwang (Dogger)

CT-based slice selection: There are stylolites and many pyrite/siderite concretions. Slice 2 is a bit more homogeneous.

To be analysed: Slice 2



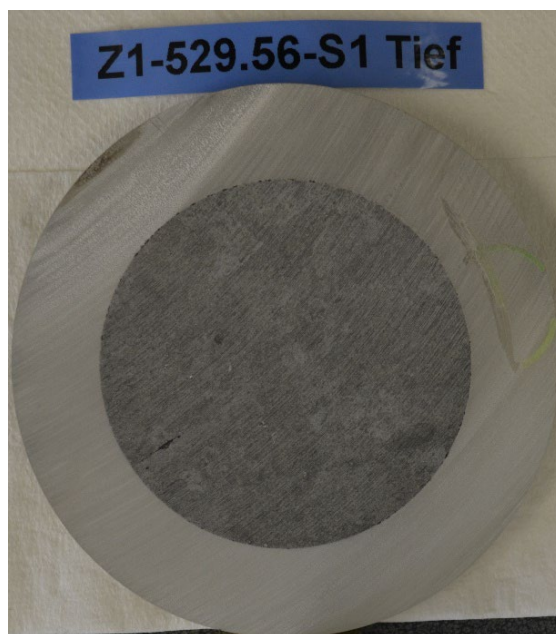
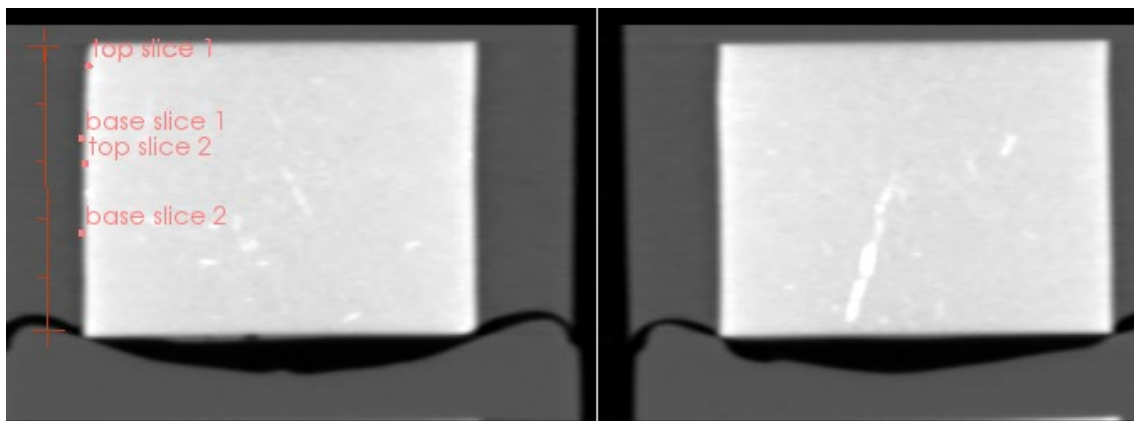
BOZ1-1-529.56-DI

Mean depth [m]: 529.56

Stratigraphic position: Passwang (Dogger)

CT-based slice selection: Slice 1 is less affected by vertically oriented bioturbation.

To be analysed: Slice 1



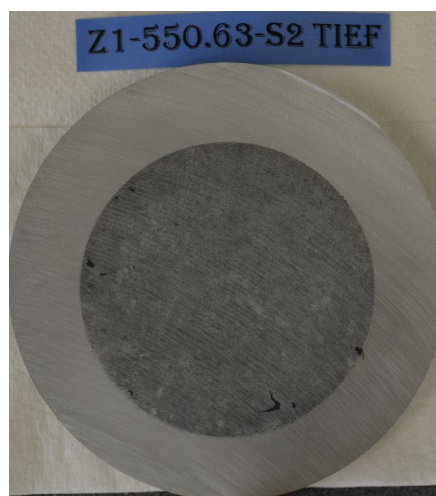
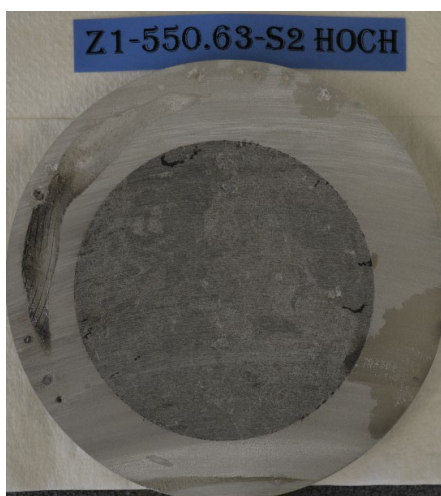
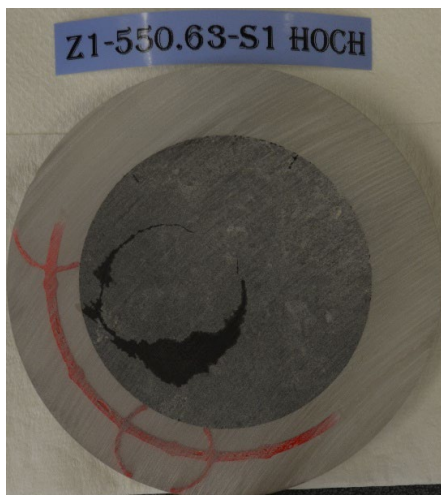
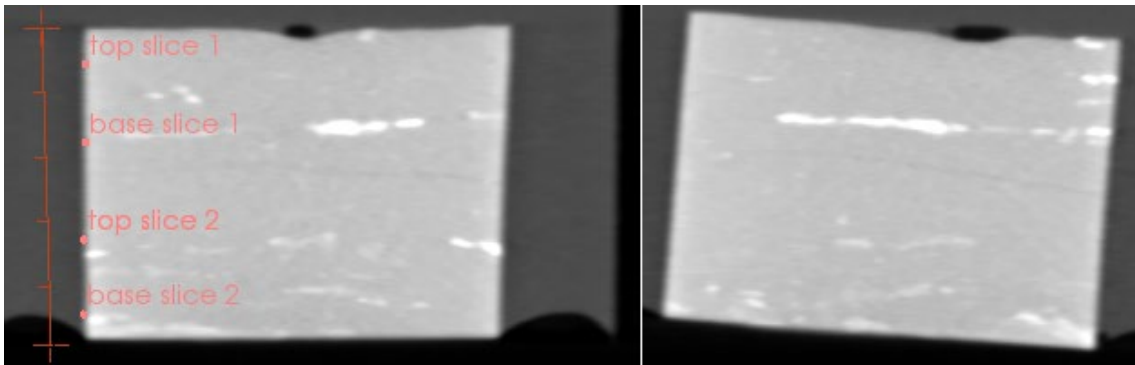
BOZ1-1-550.63-DI

Mean depth [m]: 550.63

Stratigraphic position: Opalinus Clay (Dogger)

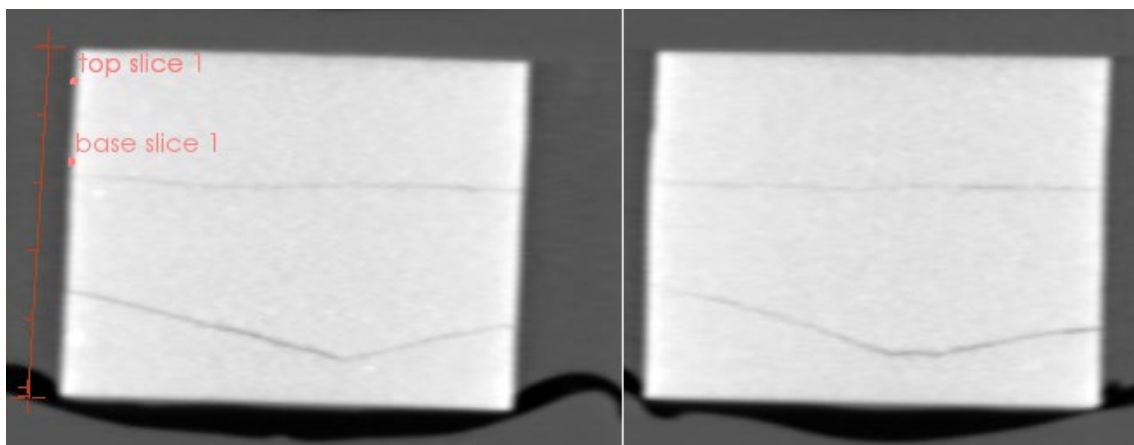
CT-based slice selection: Slice 2 has less vertically oriented bioturbation.

To be analysed: Slice 2



BOZ1-1-562.07-DI

Mean depth [m]: 562.07
Stratigraphic position: Opalinus Clay (Dogger)
CT-based slice selection: Only one slice taken due to fractures.
To be analysed: Slice 1



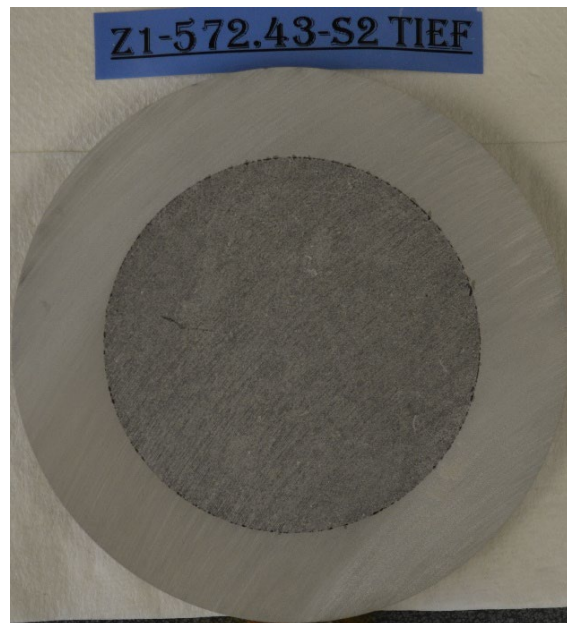
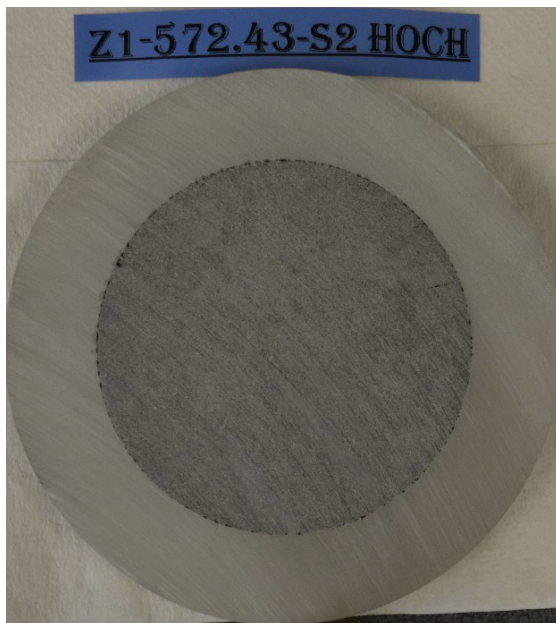
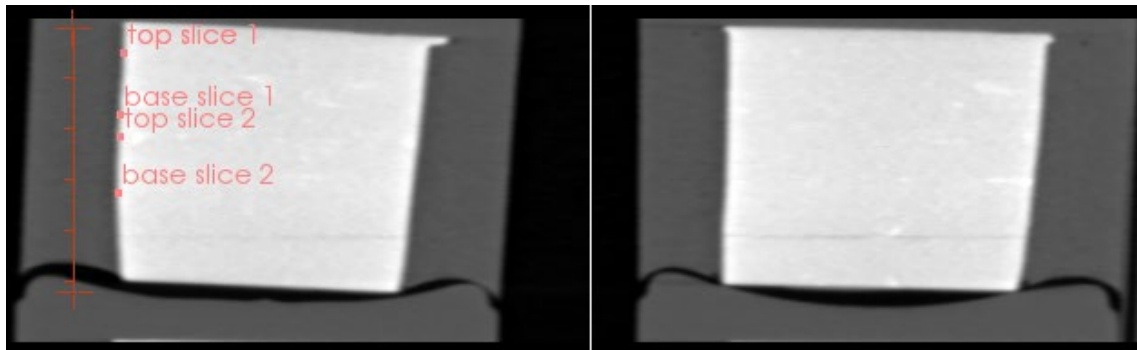
BOZ1-1-572.43-DI

Mean depth [m]: 572.43

Stratigraphic position: Opalinus Clay (Dogger)

CT-based slice selection: There are some small concretions. Slice 2 is more homogeneous.

To be analysed: Slice 2



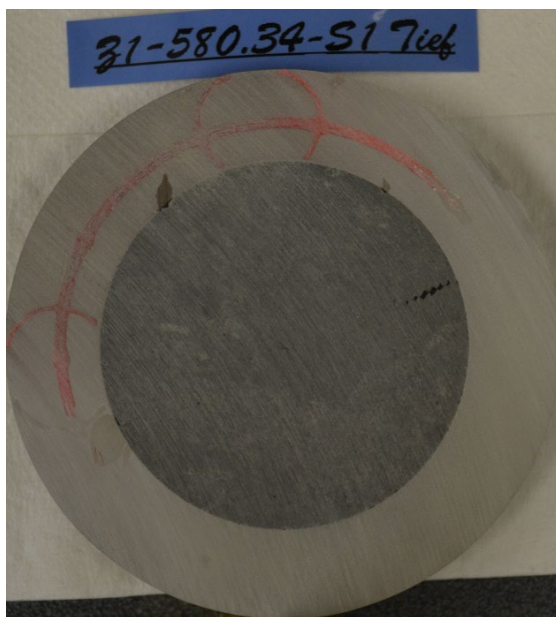
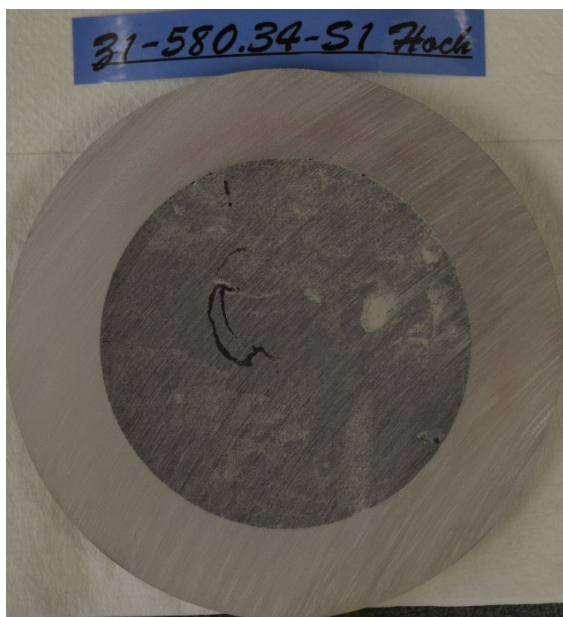
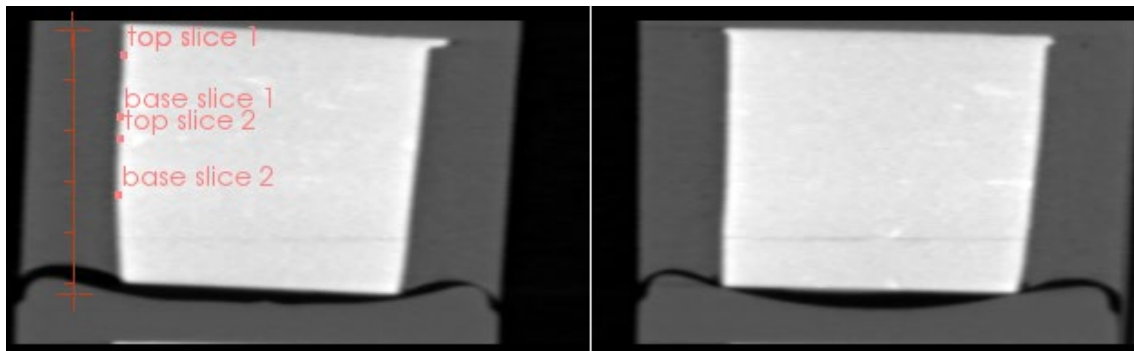
BOZ1-1-580.34-DI

Mean depth [m]: 580.34

Stratigraphic position: Opalinus Clay (Dogger)

CT-based slice selection: Slice 1 contains less pyrite lenses and is not affected by bioturbation.

To be analysed: Slice 1



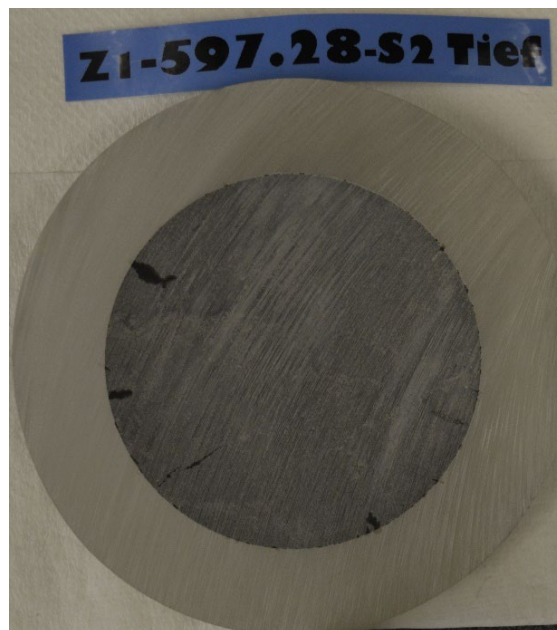
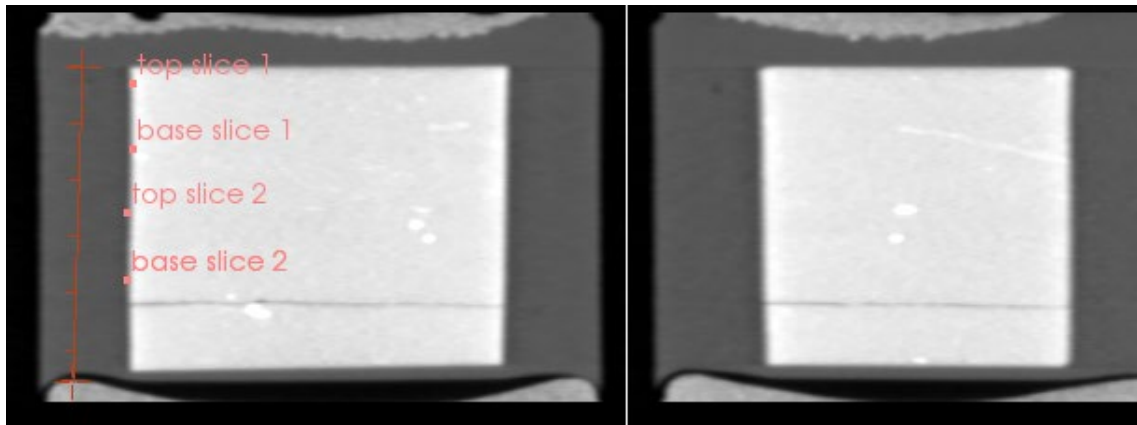
BOZ1-1-597.28-DI

Mean depth [m]: 597.28

Stratigraphic position: Opalinus Clay (Dogger)

CT-based slice selection: There are concretions. Slice 1 is affected by diagonal oriented bioturbation.

To be analysed: Slice 2



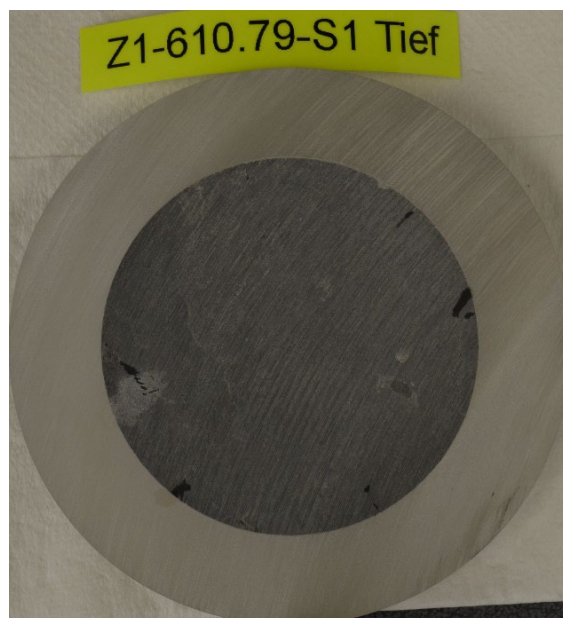
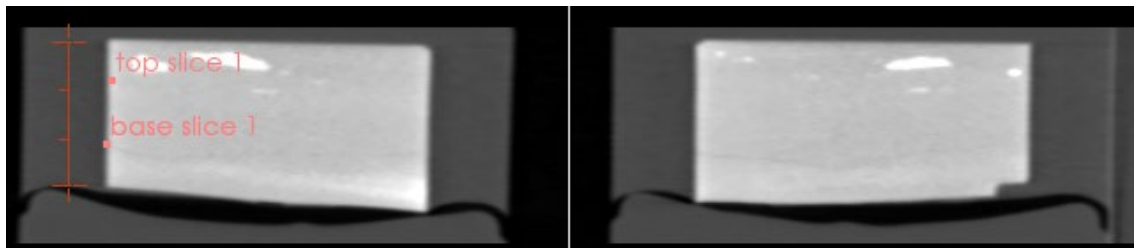
BOZ1-1-610.79-DI

Mean depth [m]: 610.79

Stratigraphic position: Opalinus Clay (Dogger)

CT-based slice selection: Only one slice taken due to short segment. No pyrite lenses or bioturbation visible.

To be analysed: Slice 1



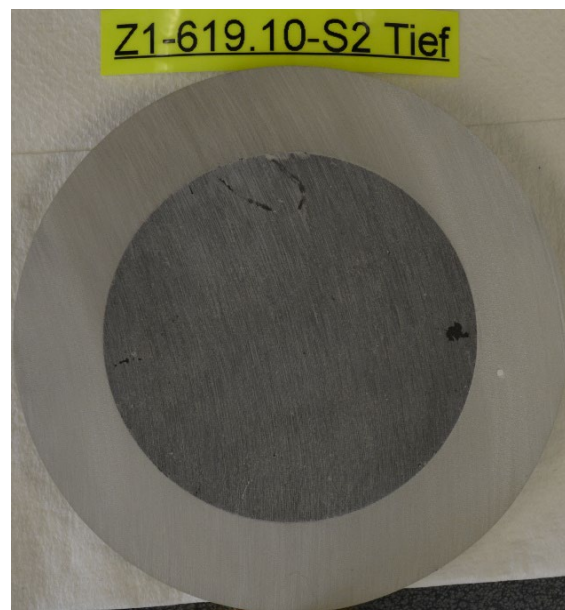
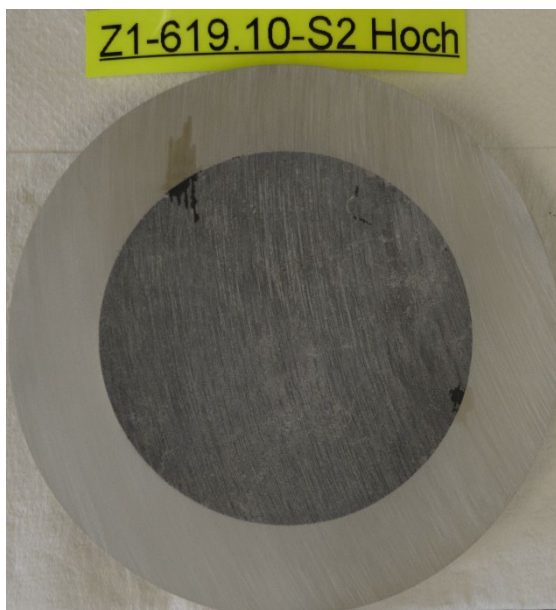
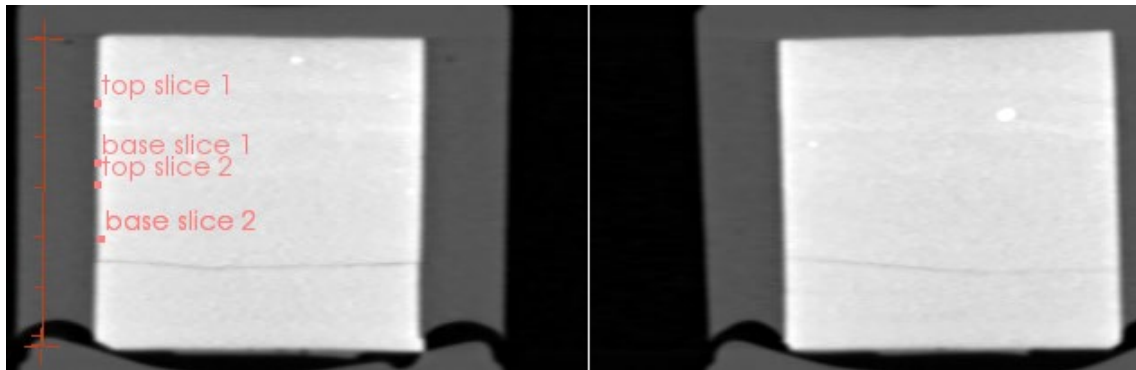
BOZ1-1-619.10-DI

Mean depth [m]: 619.10

Stratigraphic position: Opalinus Clay (Dogger)

CT-based slice selection: Slice 2 is more homogenous and contains less concretions.

To be analysed: Slice 2



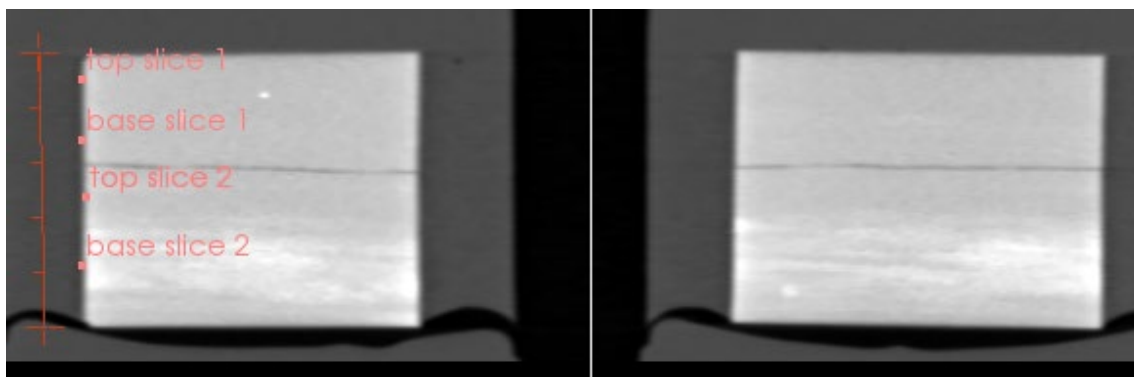
BOZ1-1-623.60-DI

Mean depth [m]: 623.60

Stratigraphic position: Opalinus Clay (Dogger)

CT-based slice selection: Slice 2 is affected by some heterogeneity, therefore Slice 1 was chosen.

To be analysed: Slice 1



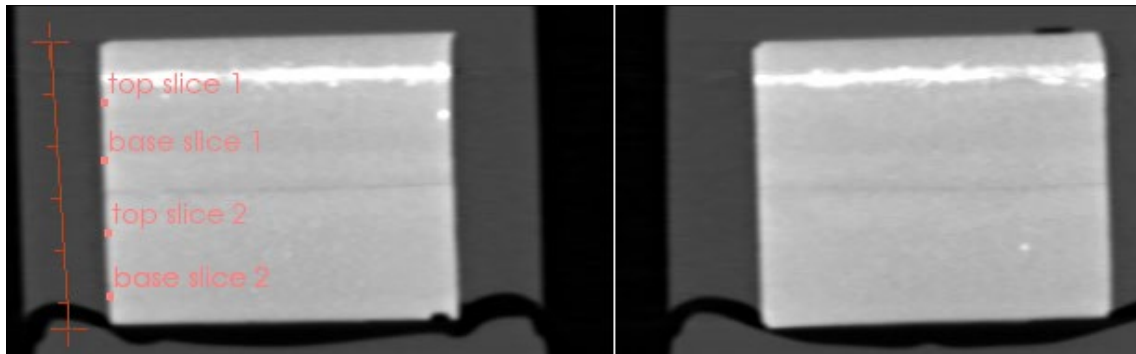
BOZ1-1-634.90-DI

Mean depth [m]: 634.90

Stratigraphic position: Opalinus Clay (Dogger)

CT-based slice selection: Slice 2 is not affected by pyrite concretions.

To be analysed: Slice 2



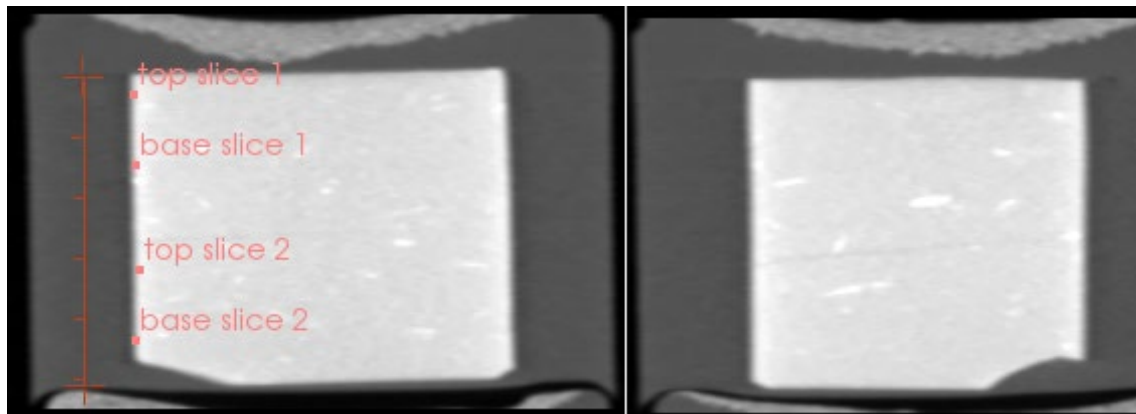
BOZ1-1-643.80-DI

Mean depth [m]: 643.80

Stratigraphic position: Opalinus Clay (Dogger)

CT-based slice selection: Much vertically oriented bioturbation. Slice 1 is slightly more homogeneous.

To be analysed: Slice 1



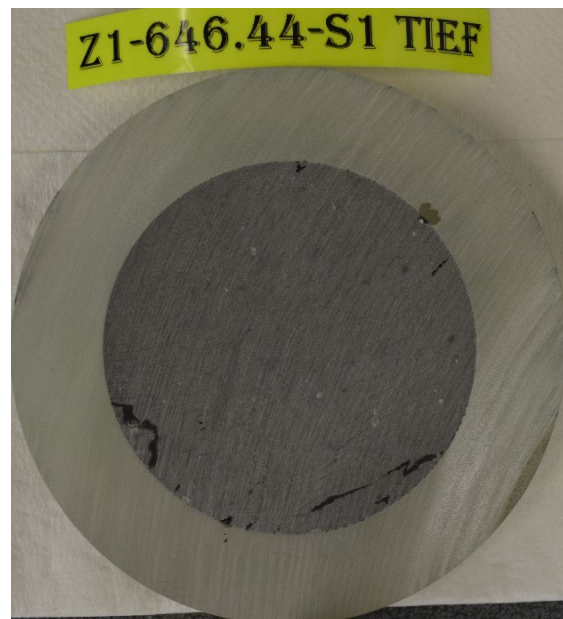
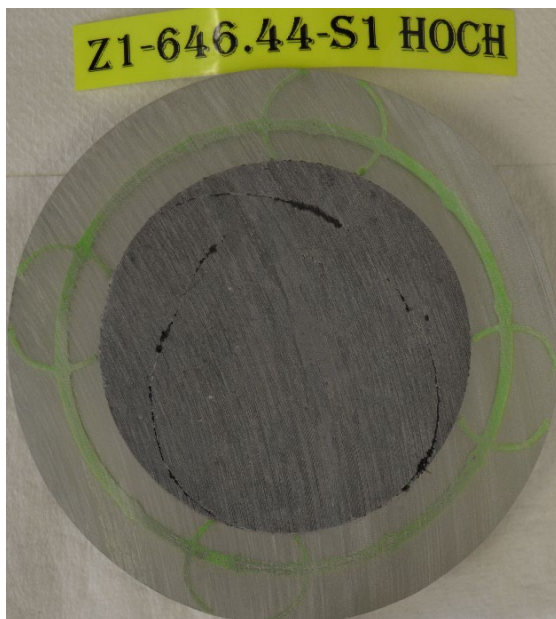
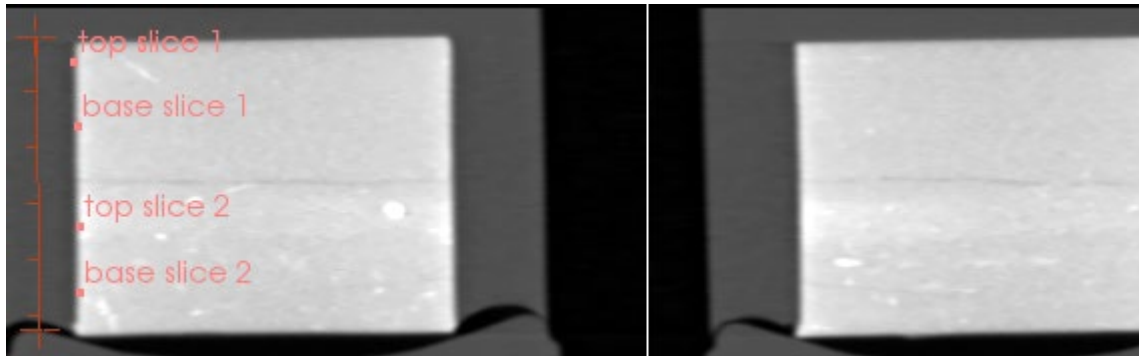
BOZ1-1-646.44-DI

Mean depth [m]: 646.44

Stratigraphic position: Opalinus Clay (Dogger)

CT-based slice selection: Slice 2 contains more pyrite concretions and signatures of bio-turbation.

To be analysed: Slice 1



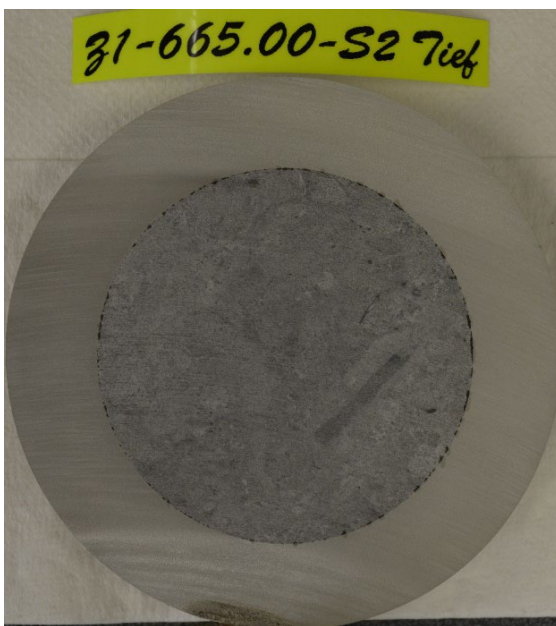
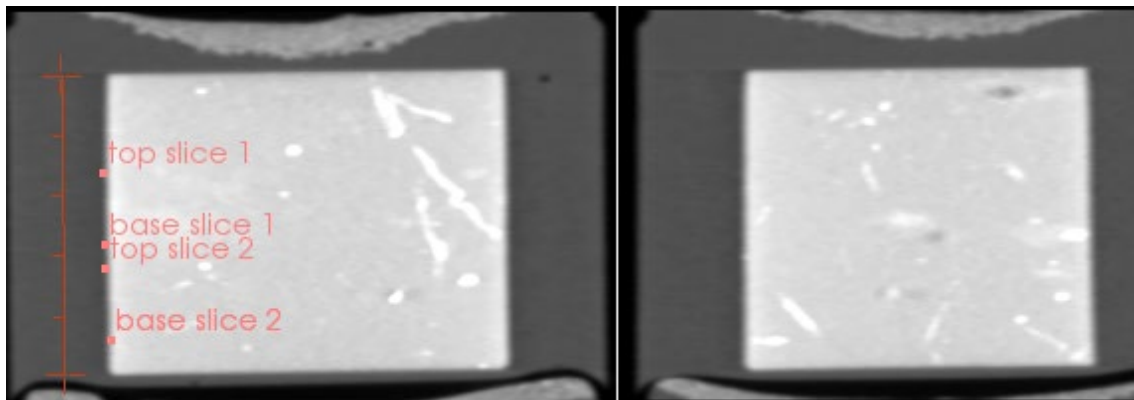
BOZ1-1-665.00-DI

Mean depth [m]: 665.00

Stratigraphic position: Staffelegg (Lias)

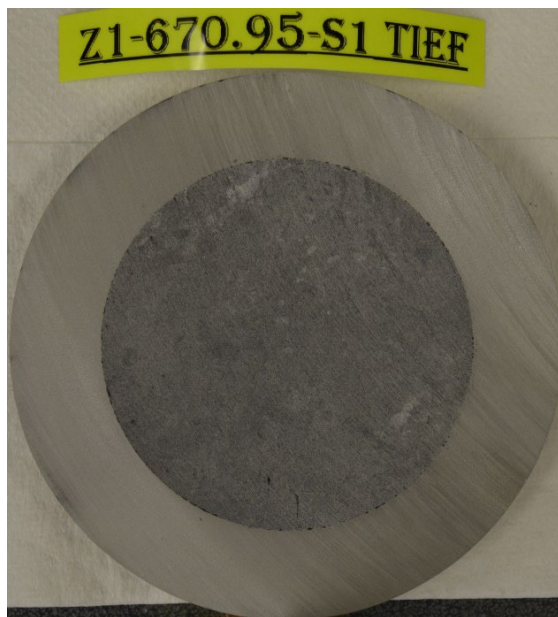
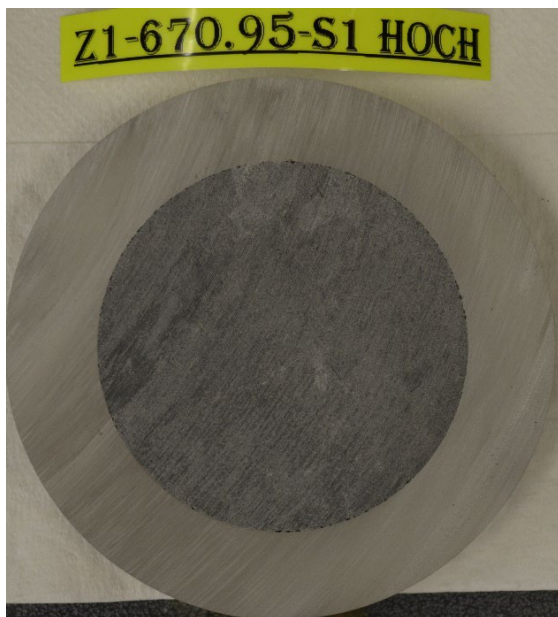
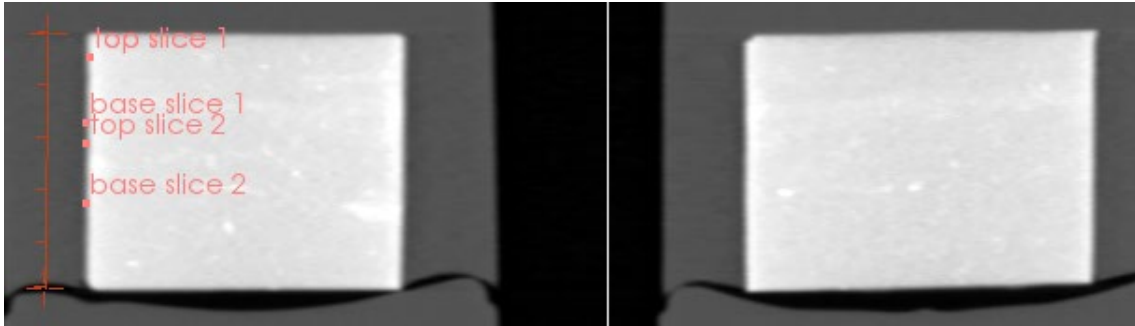
CT-based slice selection: There is a lot of vertically oriented bioturbation. Slice 2 is a bit more homogeneous.

To be analysed: Slice 2



BOZ1-1-670.95-DI

Mean depth [m]: 670.95
Stratigraphic position: Staffelegg (Lias)
CT-based slice selection: Both contain pyrite lenses but Slice 1 is more homogeneous.
To be analysed: Slice 1



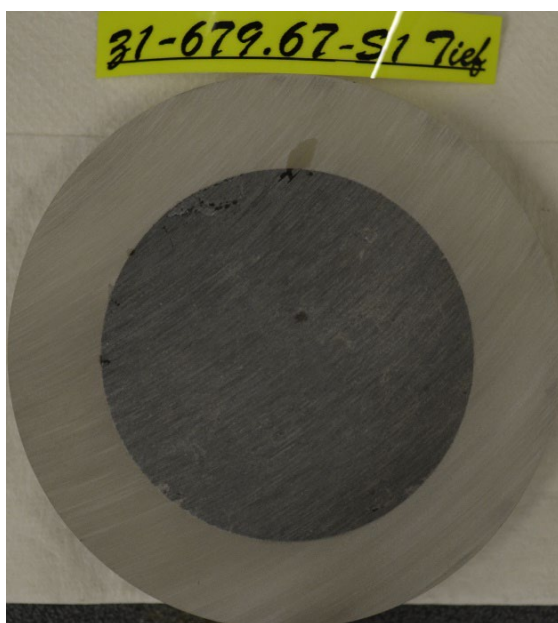
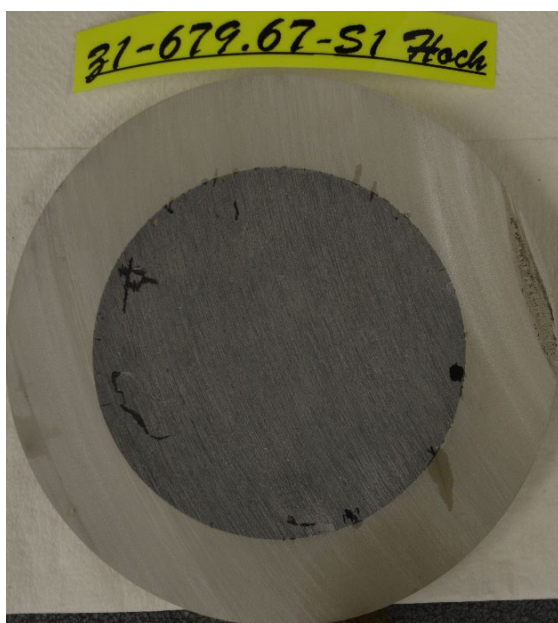
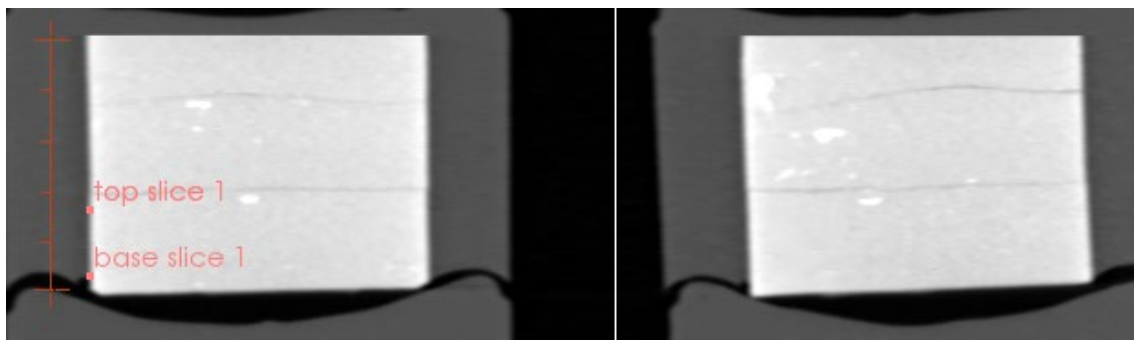
BOZ1-1-679.67-DI

Mean depth [m]: 679.67

Stratigraphic position: Staffelegg (Lias)

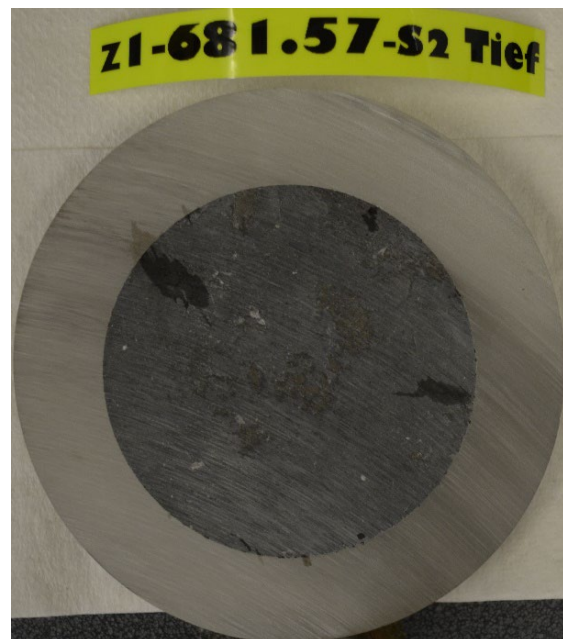
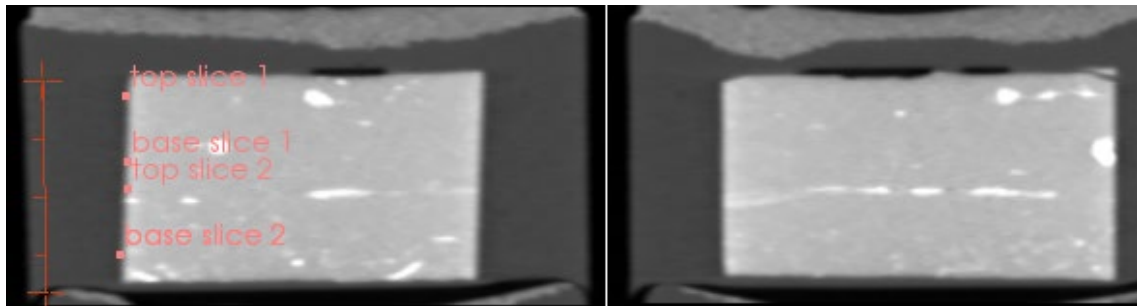
CT-based slice selection: Only one slice was taken due to fragmentation of the other.

To be analysed: Slice 1



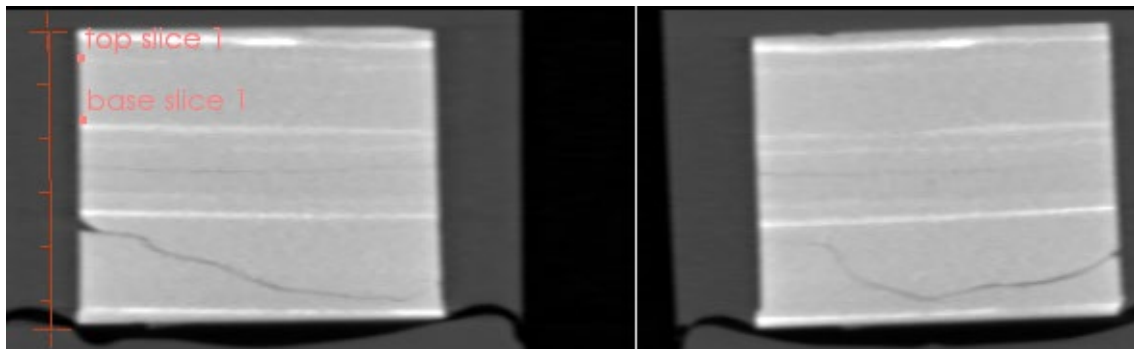
BOZ1-1-681.57-DI

Mean depth [m]: 681.57
Stratigraphic position: Staffelegg (Lias)
CT-based slice selection: Quite a lot of concretions. Slice 2 is a bit less affected.
To be analysed: Slice 2



BOZ1-1-686.81-DI

Mean depth [m]: 686.81
Stratigraphic position: Staffelegg (Lias)
CT-based slice selection: Only one slice taken due to fractures.
To be analysed: Slice 1



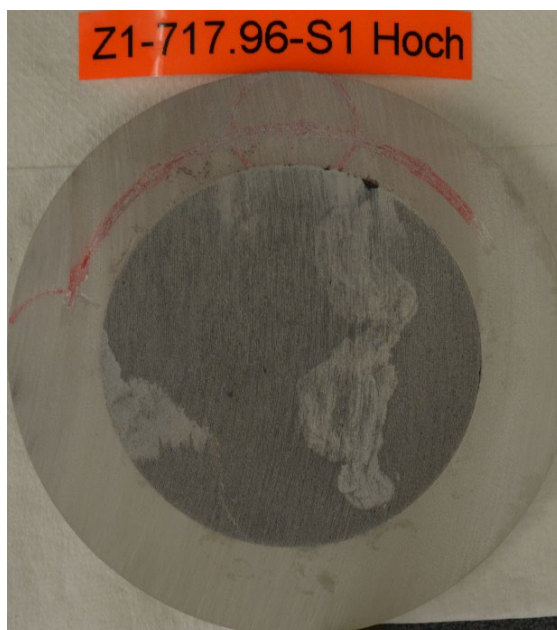
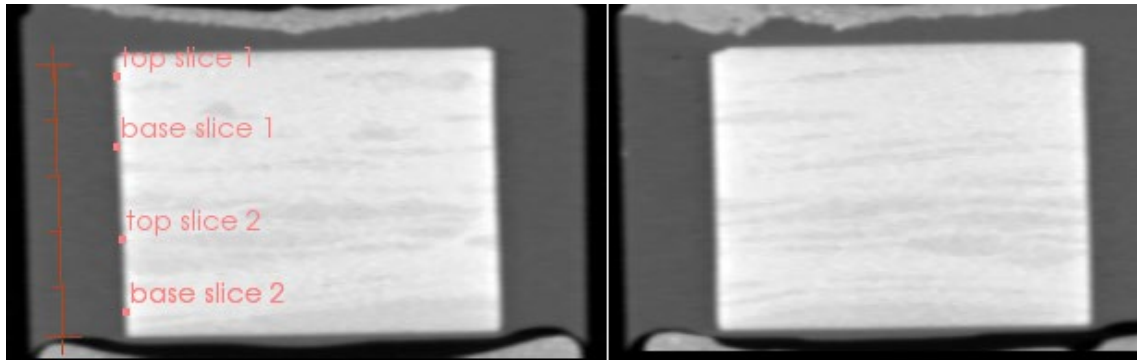
BOZ1-1-717.96-DI

Mean depth [m]: 717.96

Stratigraphic position: Klettgau (Keuper)

CT-based slice selection: Some heterogeneities (silt pockets). Slice 1 is more homogeneous.

To be analysed: Slice 1



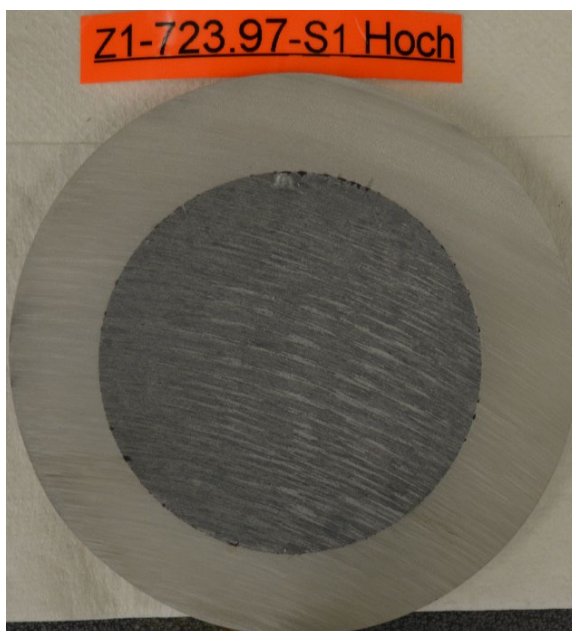
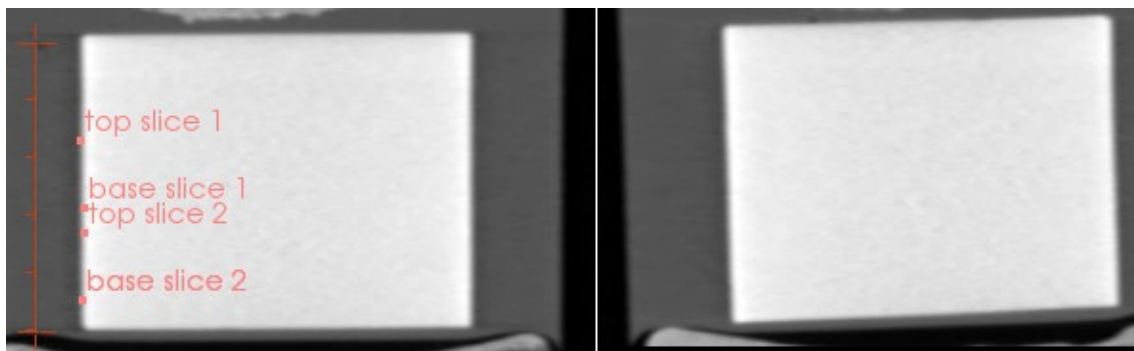
BOZ1-1-723.97-DI

Mean depth [m]: 723.97

Stratigraphic position: Bänkerjoch (Keuper)

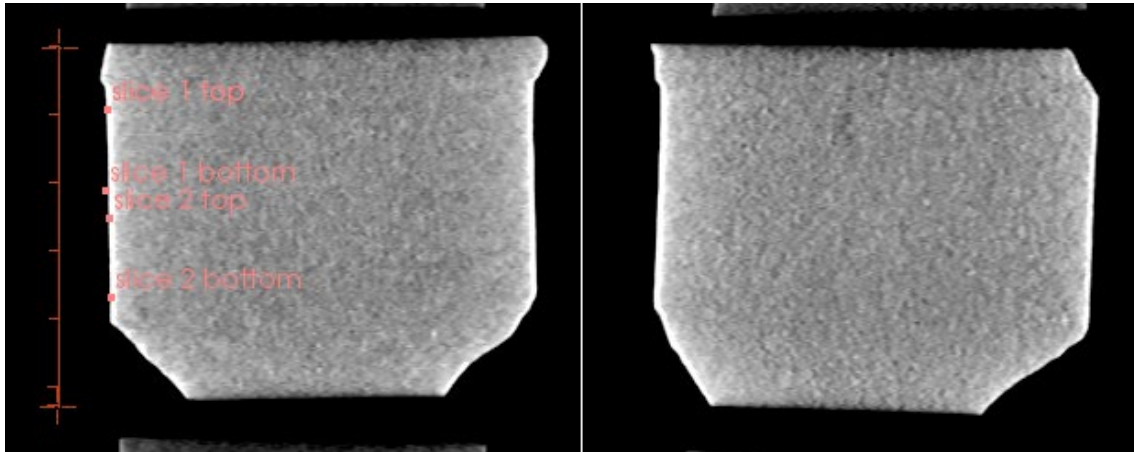
CT-based slice selection: Very homogeneous and no difference between Slices 1 and 2.

To be analysed: Slice 1

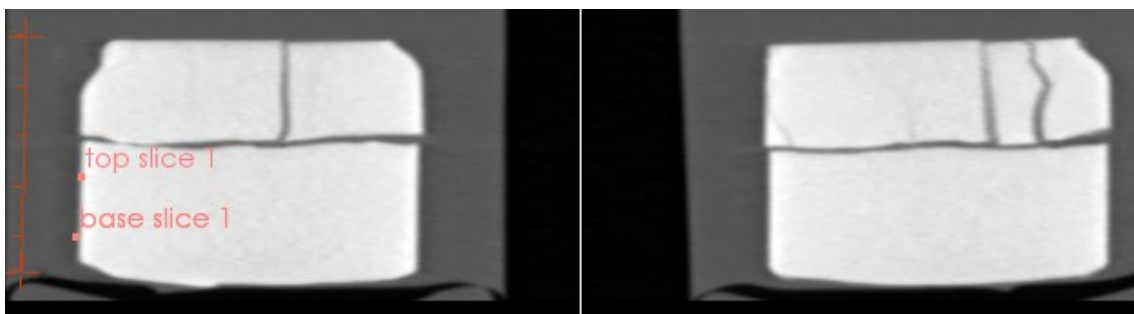


BOZ1-1-743.10-DI

Mean depth [m]: 743.10
Stratigraphic position: Bänkerjoch Fm. (Keuper)
CT-based slice selection: Pure anhydrite.
To be analysed: Both slices are good and homogeneous.

**BOZ1-1-790.16-DI**

Mean depth [m]: 790.16
Stratigraphic position: Bänkerjoch (Keuper)
CT-based slice selection: Only one slice taken due to fractures.
To be analysed: Slice 1



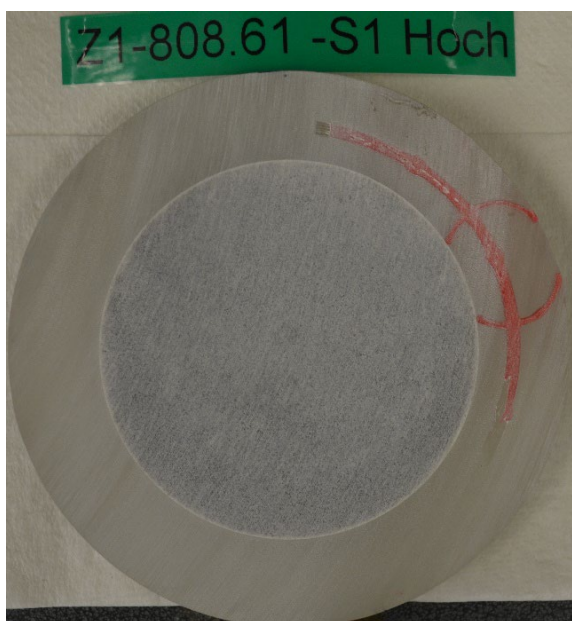
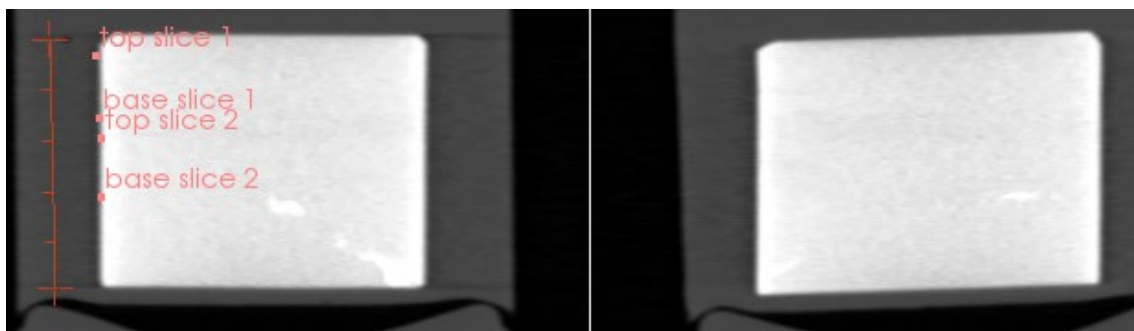
BOZ1-1-808.61-DI

Mean depth [m]: 808.61

Stratigraphic position: Bänkerjoch (Keuper)

CT-based slice selection: Slice 1 is further away from an unusual feature.

To be analysed: Slice 1



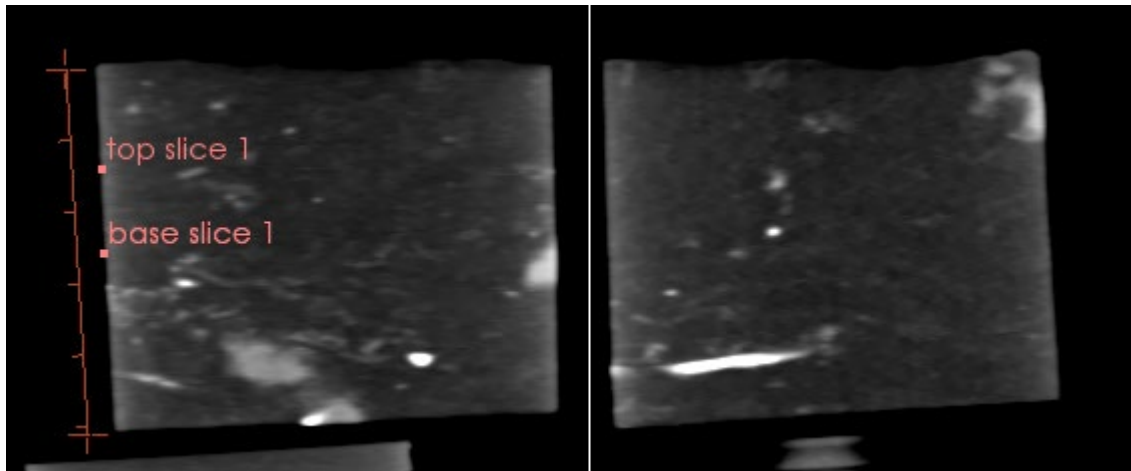
H.1.2 Bözberg 2-1**BOZ2-1-313.13-DI**

Mean depth [m]: 313.13

Stratigraphic position: Hauptrogenstein (Dogger)

CT-based slice selection: Only one slice was selected due to large nodules in the remaining section.

To be analysed: Slice 1



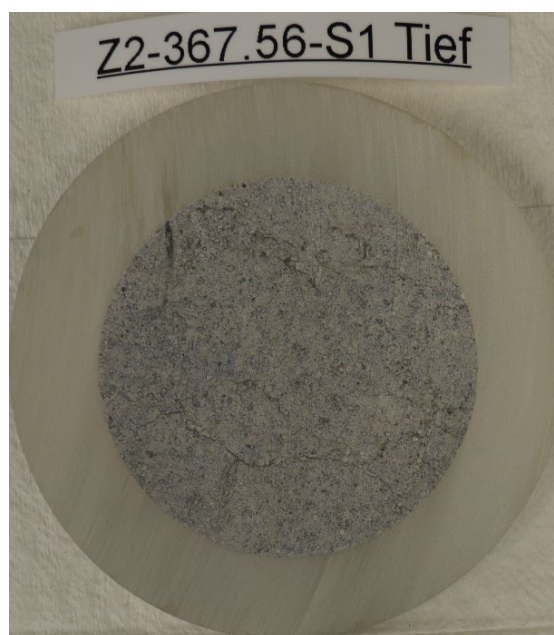
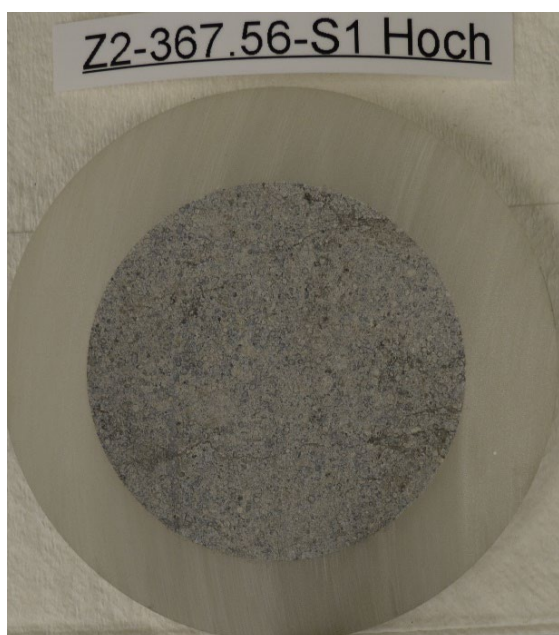
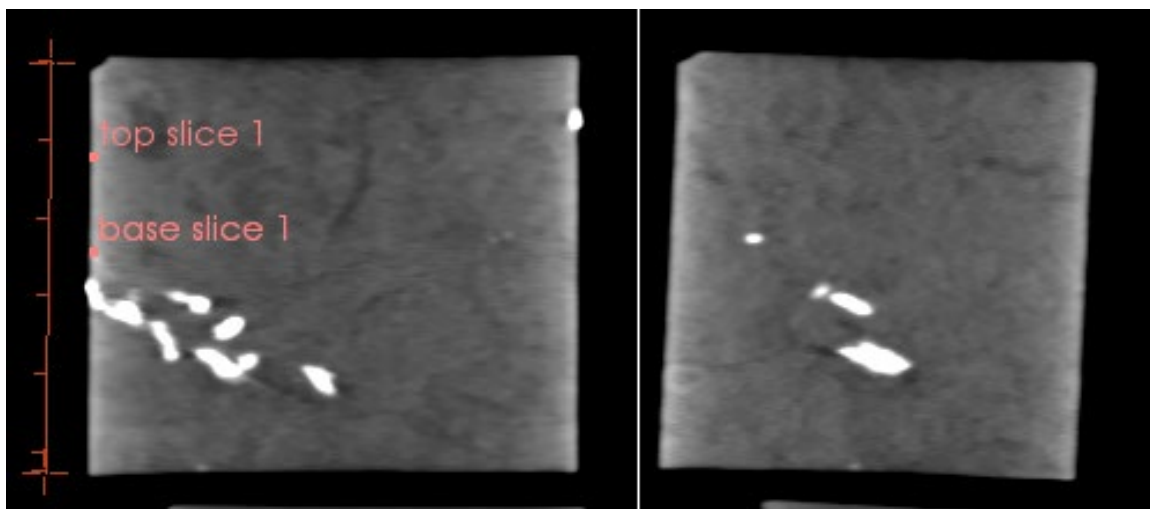
BOZ2-1-367.56-DI

Mean depth [m]: 367.56

Stratigraphic position: Hauptrogenstein (Dogger)

CT-based slice selection: Only one slice was selected due to large lenses in the remaining section.

To be analysed: Slice 1



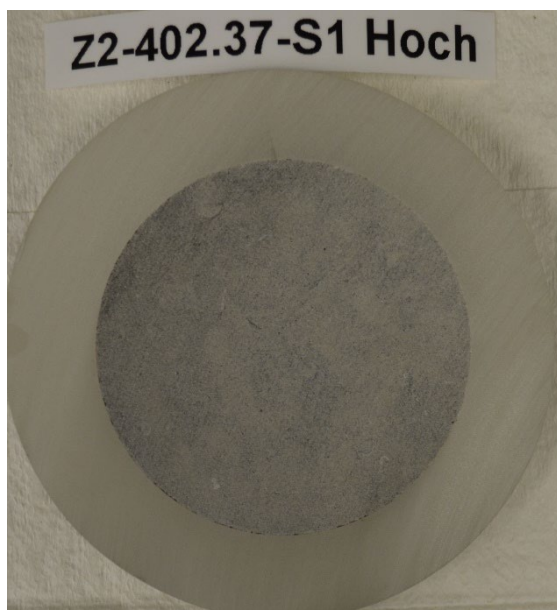
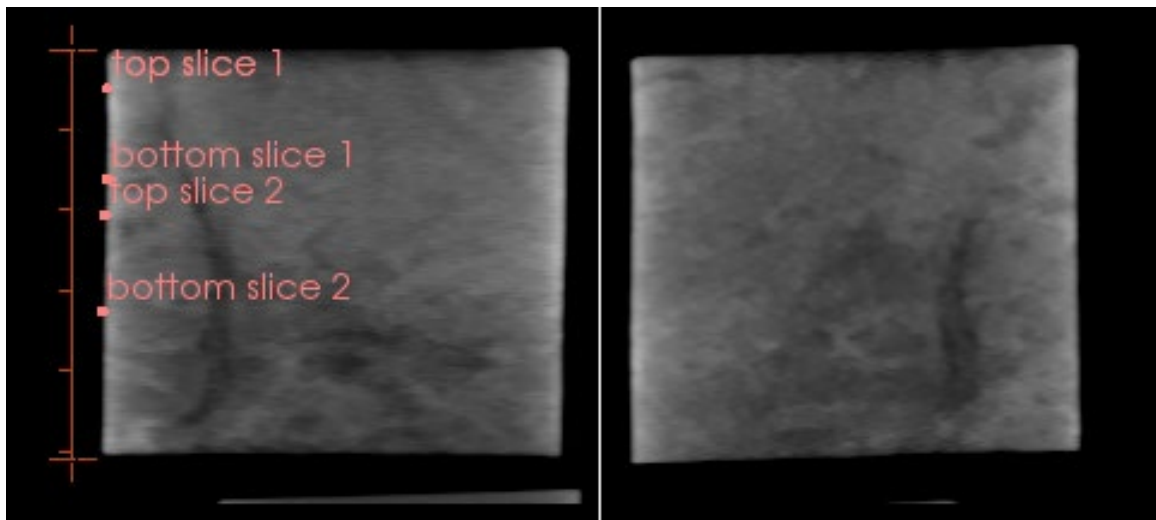
BOZ2-1-402.37-DI

Mean depth [m]: 402.37

Stratigraphic position: Passwang (Dogger)

CT-based slice selection: Slice 1 was chosen due to bioturbation in the remaining section.

To be analysed: Slice 1



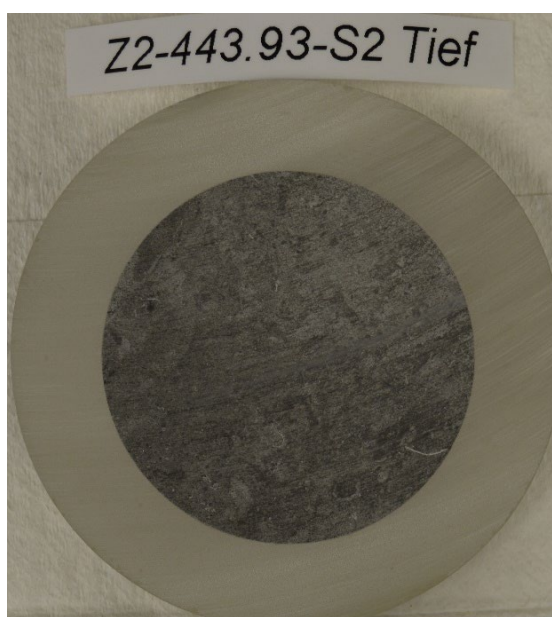
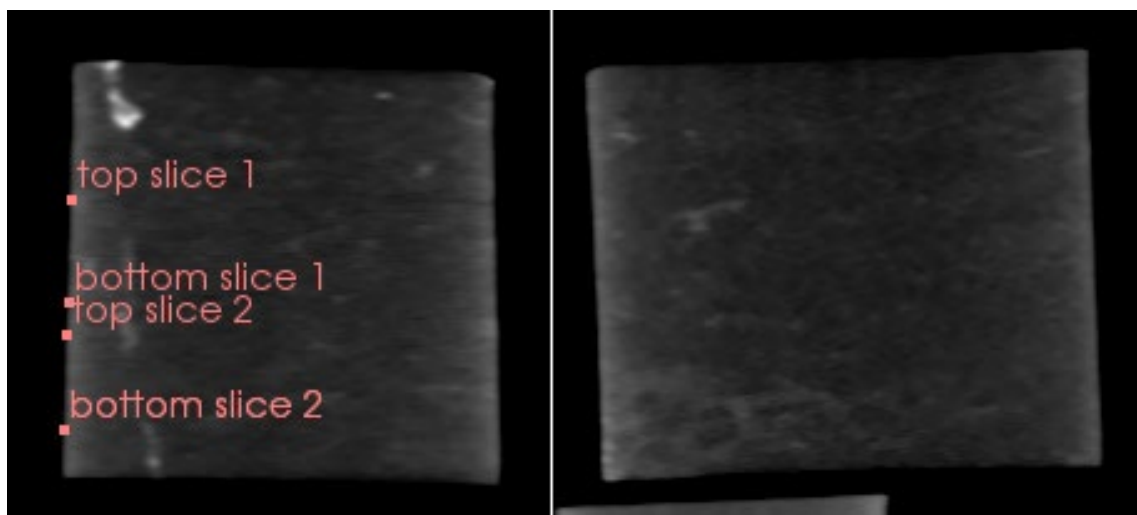
BOZ2-1-443.93-DI

Mean depth [m]: 443.93

Stratigraphic position: Passwang (Dogger)

CT-based slice selection: Slice 2 was chosen because it is more homogeneous.

To be analysed: Slice 2



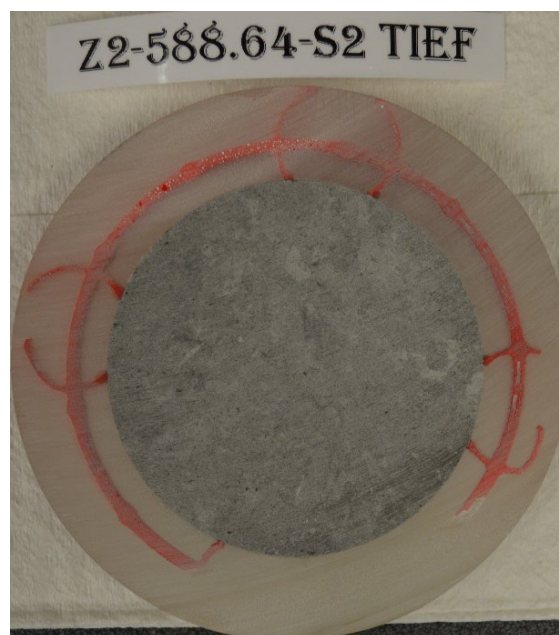
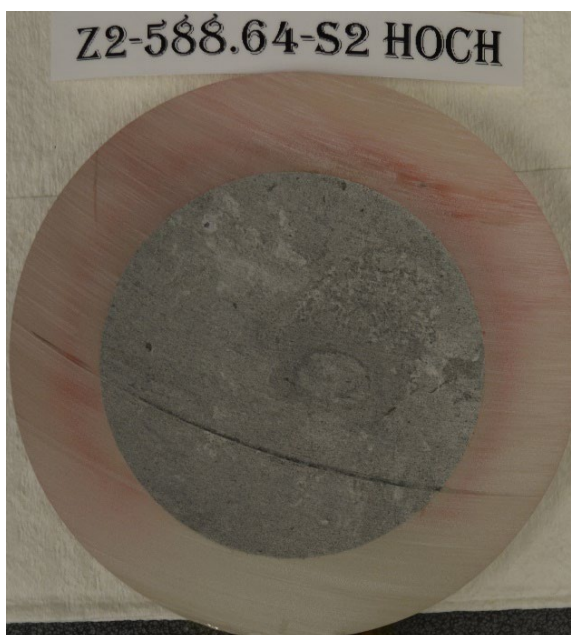
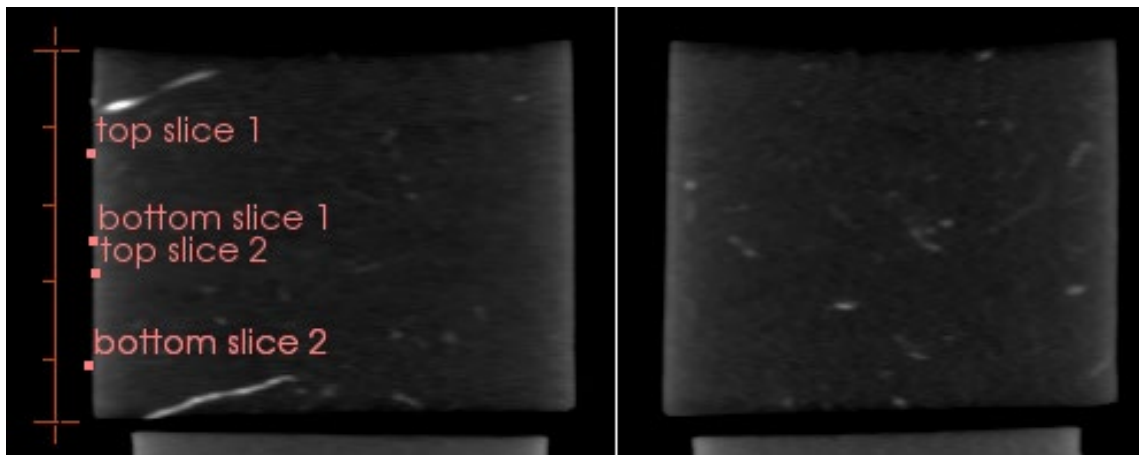
BOZ2-1-588.64-DI

Mean depth [m]: 588.64

Stratigraphic position: Staffelegg Fm. (Lias)

CT-based slice selection: Slice 2 was chosen because it is more homogeneous.

To be analysed: Slice 2



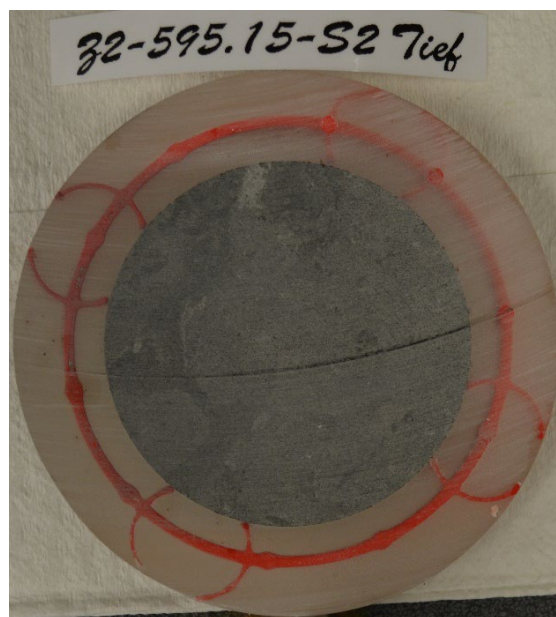
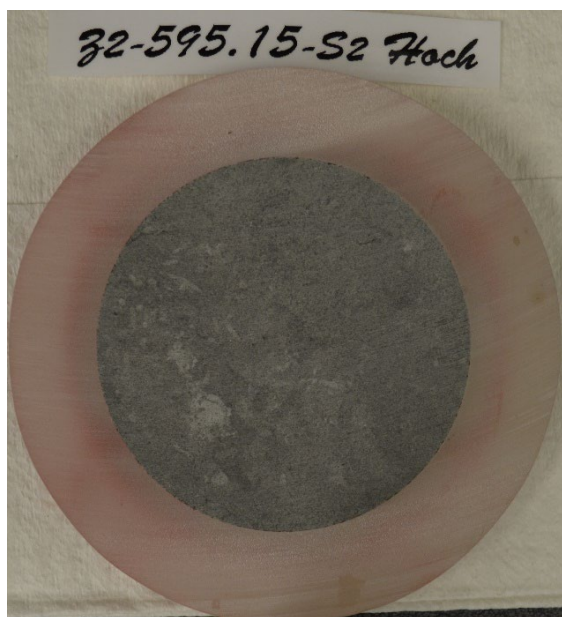
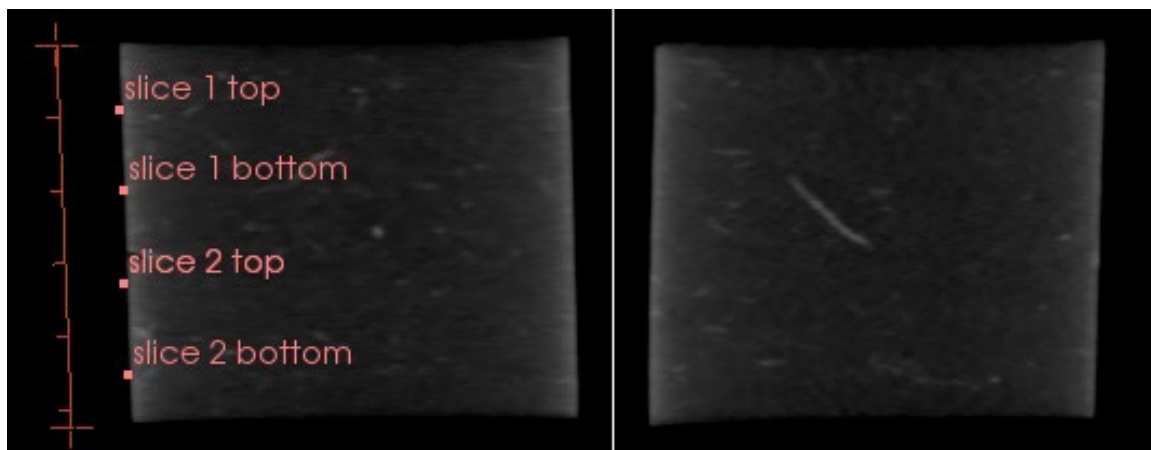
BOZ2-1-595.15-DI

Mean depth [m]: 595.15

Stratigraphic position: Staffelegg Fm. (Lias)

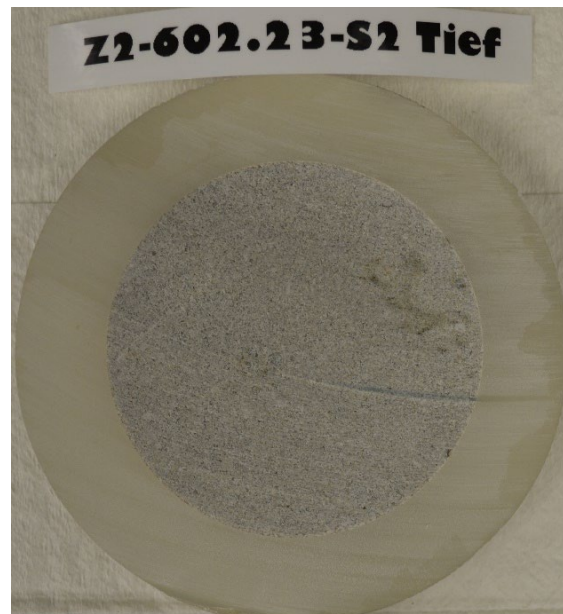
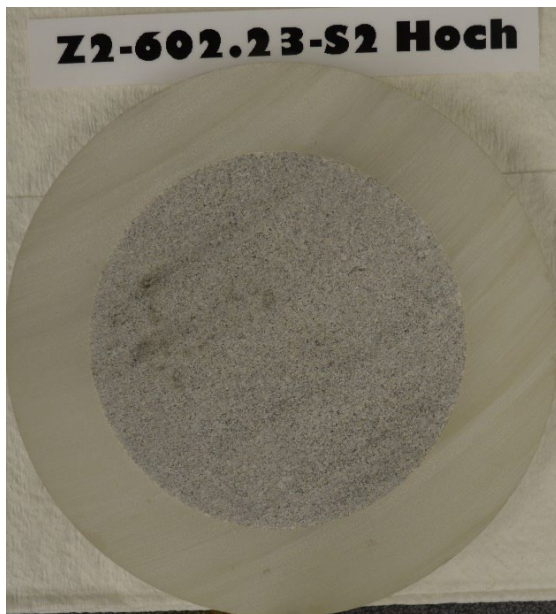
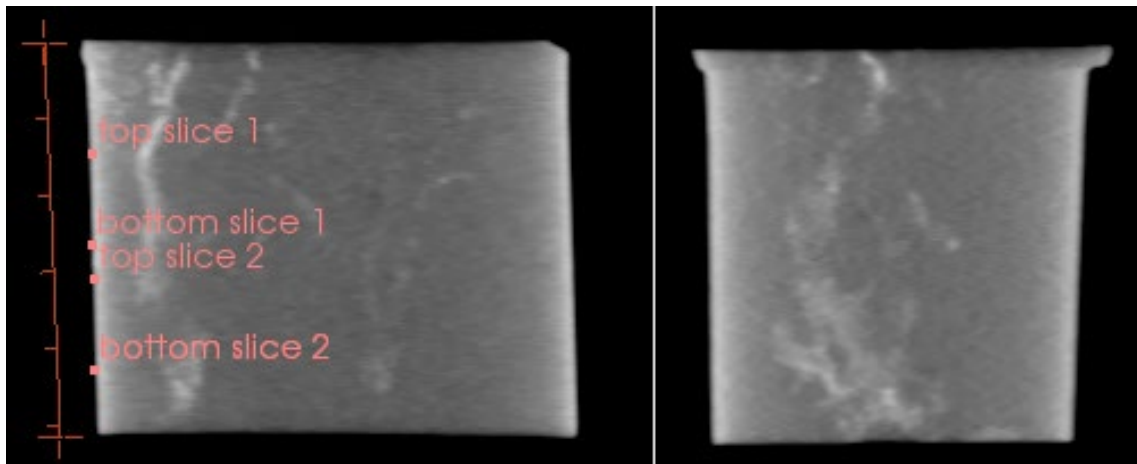
CT-based slice selection: Slice 2 was chosen because it is more homogeneous.

To be analysed: Slice 2



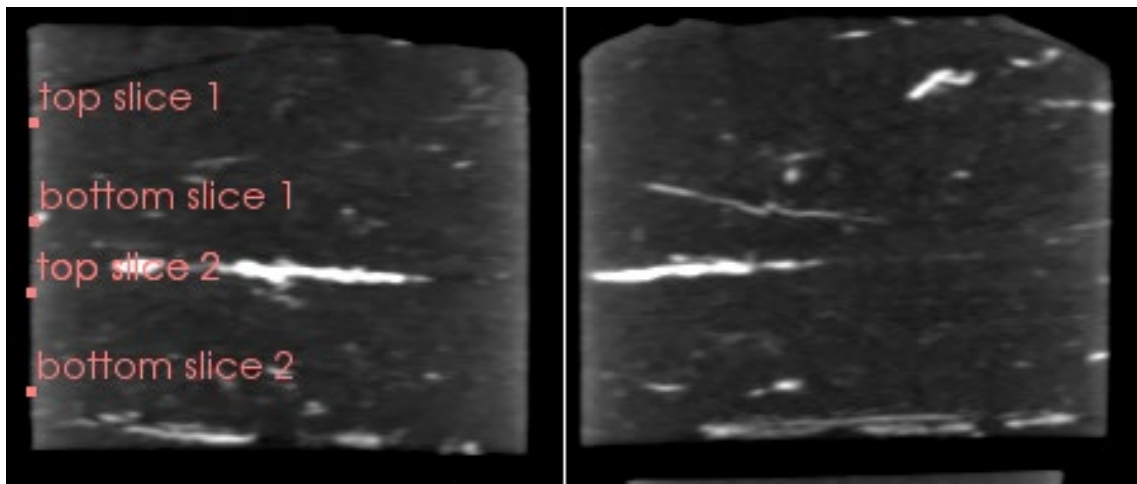
BOZ2-1-602.23-DI

Mean depth [m]: 602.23
Stratigraphic position: Staffelegg Fm. (Lias)
CT-based slice selection: Slice 2 is more homogeneous.
To be analysed: Slice 2



BOZ2-1-606.95-DI

Mean depth [m]: 606.95
Stratigraphic position: Staffelegg Fm. (Lias)
CT-based slice selection: Slice 2 is more homogeneous.
To be analysed: Slice 2



H.2 Samples from the Nördlich Lägern siting region

This section documents the final step of the sampling campaign for diffusion experiments (DI) of the Bachs, Bülach and Stadel boreholes (or BAC, BUL, STA, respectively). In detail, it documents the positioning and selection of the final core slices, which were used for the experiments at PSI, from the subcored and epoxy-embedded samples. The axis of the subcores is parallel to that of the original full core, i.e. normal to bedding. The positioning, selection and description of the slices is mainly based on CT scans performed at the Institute of Forensic Medicine at the University of Bern. While scans of the whole DI samples were already available, new scans were acquired on the subcored samples and served as a basis for the positioning of the DI slices. On the one hand, these scans allow the assessment of sample homogeneity (or heterogeneity) and, thus, aid the selection of slices such that they are as representative as possible. Moreover, these scans make it possible to identify features such as cracks or fractures (either natural or artificially induced), large pyrite concretions, siderite lenses, macropores (i.e. in calcareous lithologies), stylolites or burrows, which could affect the experiments. Such features were avoided.

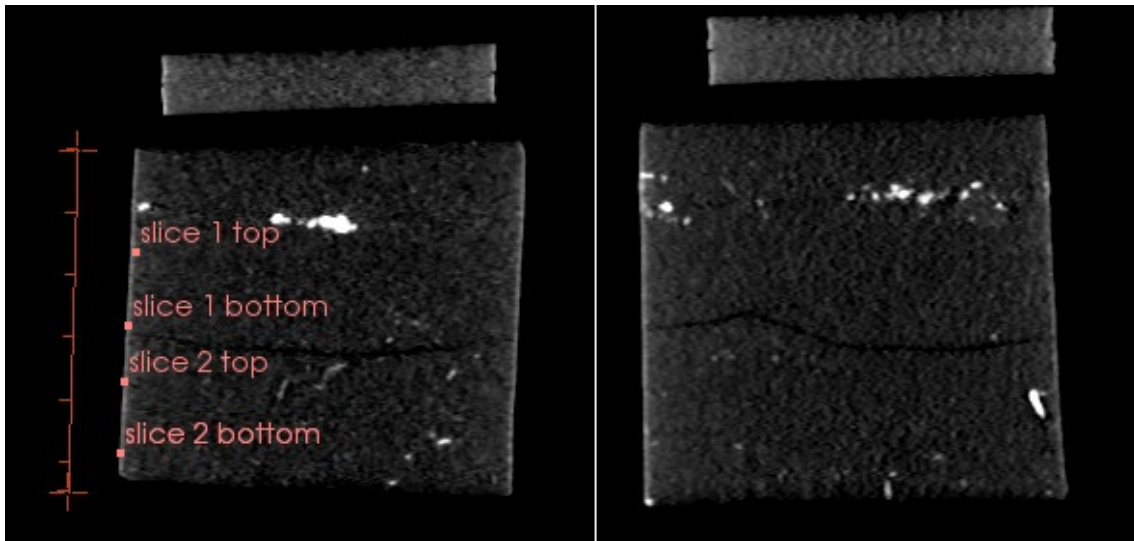
In general, 1 to 2 DI slices were collected per sample, of which one slice is selected for the experiments at PSI. The slice selection is documented in various CT images and generally arranged as image pairs; one on the left side and one on the right side. Such an image pair illustrates two sections of the subcores parallel to the core axis, which are oriented orthogonally with respect to each other. The position of the DI slices within the subcored samples (i.e. top and bottom of slices) is shown in the CT image on the left-hand side, whereas special features relevant to sampling may be illustrated in the second CT image on the right-hand side.

Tab. H-2: Depth information for all core samples from the Bachs, Bülach and Stadel boreholes used for diffusion experiments. The values in grey text are samples for which no diffusion data are available.

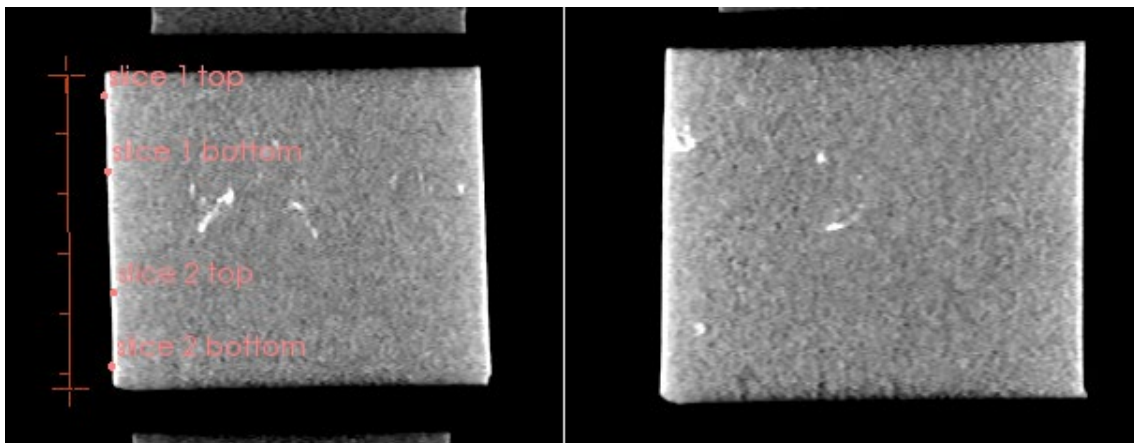
BAC1-1	BUL1-1	STA2-1	STA3-1
Sample mean depth [m]			
933.25	734.45	633.23	650.49
1,033.70	752.48	752.73	686.50
	766.65	758.02	748.38
	794.35	758.23	758.75
	808.92	765.21	768.34
	815.90	779.96	896.82
	840.45	912.69	919.66
	845.05	925.51	
	851.50	930.53	
	861.00	955.40	
	872.35	957.28	
	879.20	963.35	
	908.93	1,126.66	
	914.61		
	921.43		
	932.82		
	945.46		
	955.62		
	965.46		
	969.15		
	975.04		
	984.49		
	995.09		
	1,002.19		
	1,008.42		
	1,014.23		
	1,017.33		
	1,025.96		

H.2.1 Bachs 1-1**BAC1-5-AD-933.25m**

Mean depth [m]: 933.25
Stratigraphic position: Staffelegg (Frick Mb.)
CT-based slice selection: Slice 2 is less affected by pyrite band and fracture.
To be analysed: Slice 2

**BAC1-1-1033.70-DI**

Mean depth [m]: 1,033.70
Stratigraphic position: Bänkerjoch Fm. (Keuper)
CT-based slice selection: Anhydrite with fine clay layers (1mm) which are parallel to subparallel.
To be analysed: Slice 1



H.2.2 Bülach 1-1**BUL1-1-734.45-DI**

Mean depth [m]: 734.45

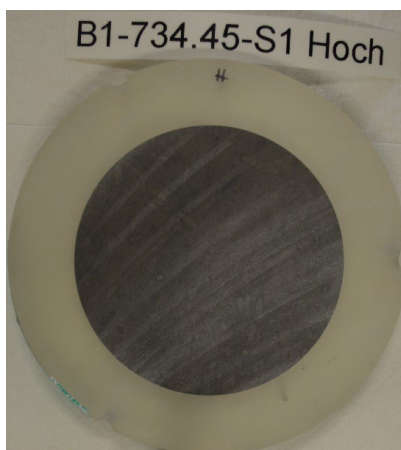
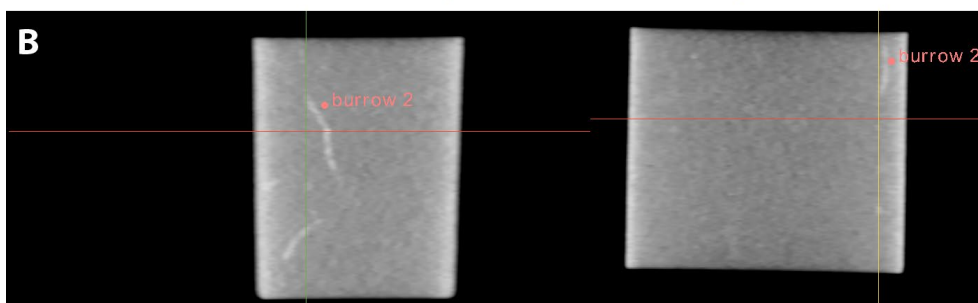
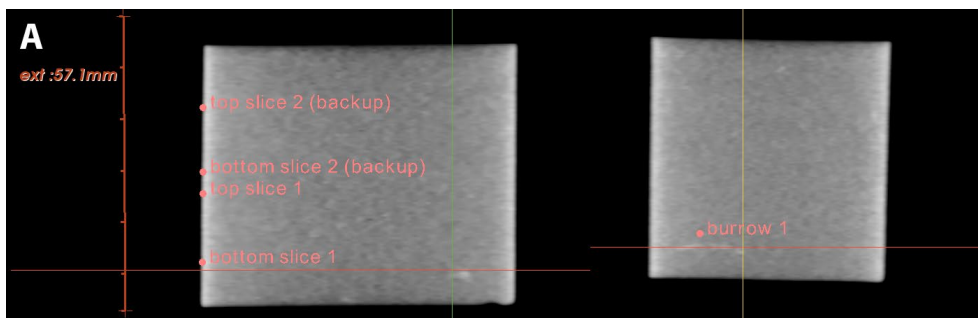
Stratigraphic position: Villigen Fm.

Lithological description: Limestone, marly

CT-based slice selection: Sample is slightly bioturbated, otherwise homogeneous (A). Two slices were sampled, however, small, oblique burrows (filled by pyrite or siderite) cut across both slices (B).

To be analysed: Slice 1

General comments regarding sample preparation: Cut surfaces of DI slices may not be entirely parallel.



BUL1-1-752.48-DI

Mean depth [m]: 752.48

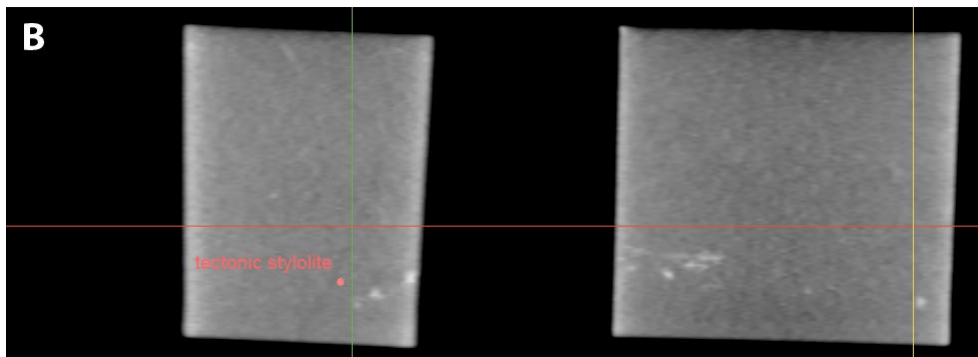
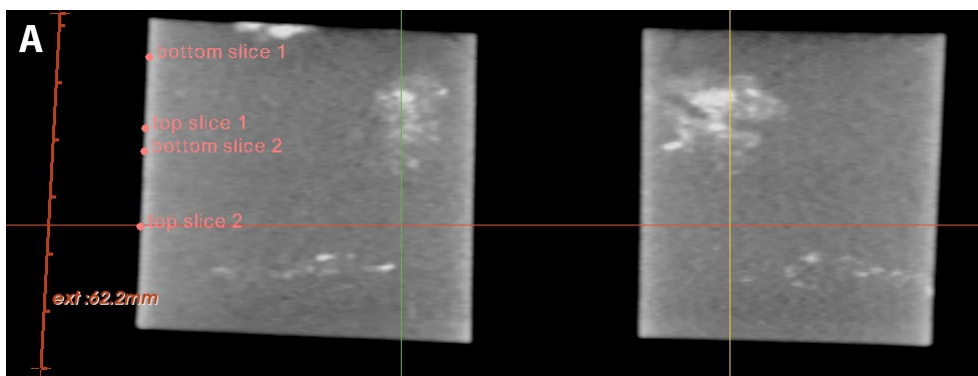
Stratigraphic position: Wildegg Fm.

Lithological description: Limestone, marly

CT-based slice selection: Two slices were sampled: Slice 1 contains a nodular feature of higher density material (A). Slice 2 appears entirely homogeneous. Both slices are crosscut by a tectonic stylolite (B).

To be analysed: Slice 2

General comments regarding sample preparation: Cut surfaces of DI slices may not be entirely parallel.



BUL1-1-766.65-DI

Mean depth [m]: 766.65

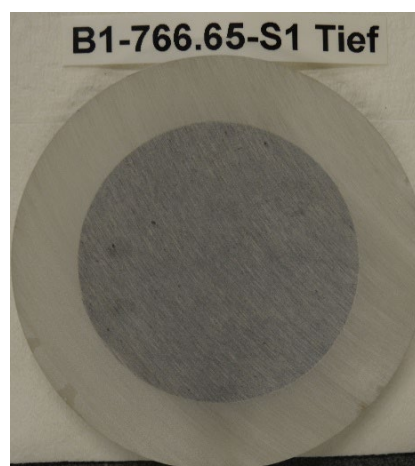
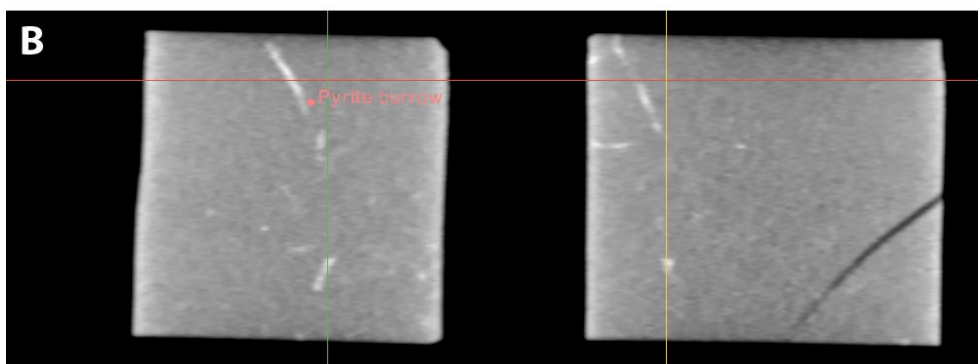
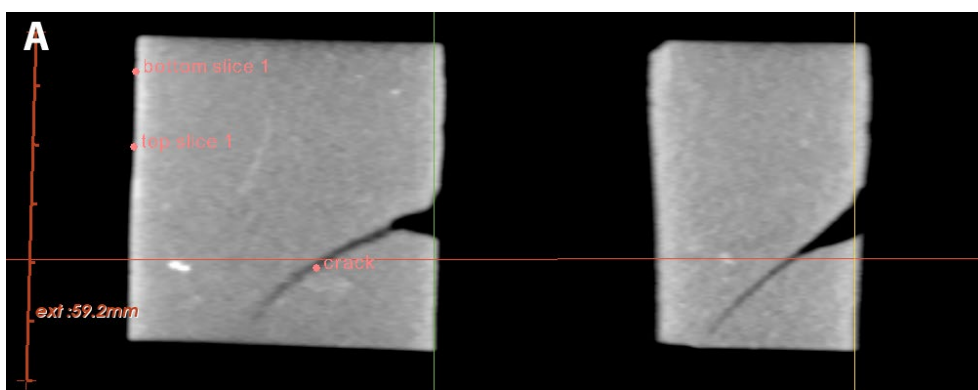
Stratigraphic position: Wildegg Fm.

Lithological description: Marl, calcareous

CT-based slice selection: Sample contains a massive drilling-induced crack (A), thus only one slice was sampled. This slice contains a thin, oblique pyrite (or siderite?) burrow that cuts across the slice (B).

To be analysed: Slice 1

General comments regarding sample preparation: Cut surfaces of DI slice may not be entirely parallel.



BUL1-1-794.35-DI

Mean depth [m]: 794.35

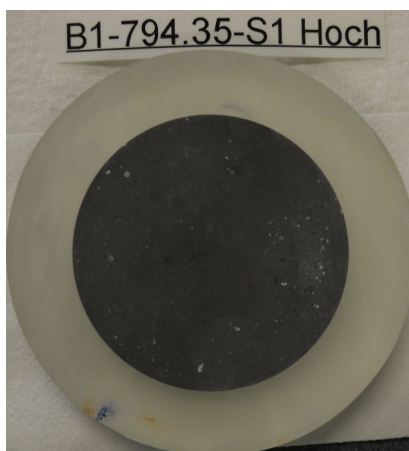
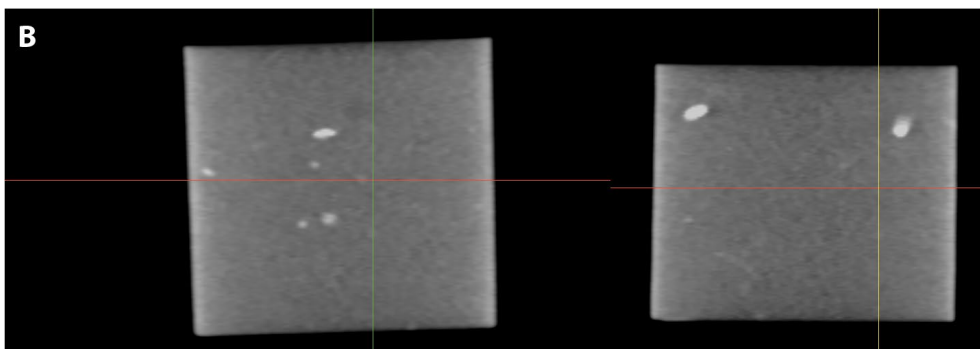
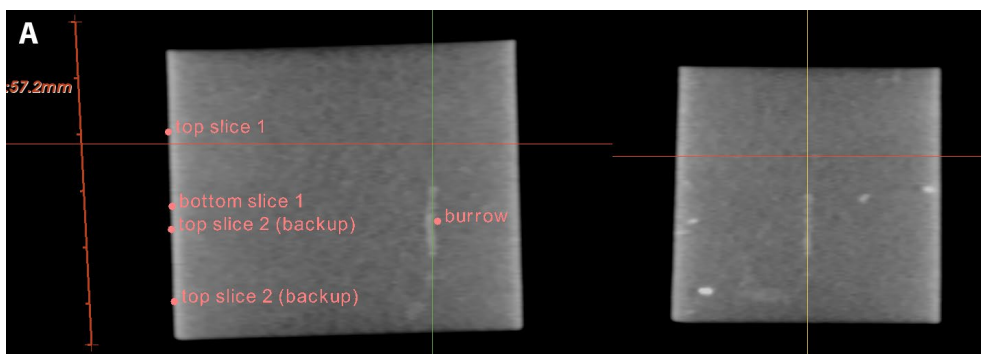
Stratigraphic position: Wildegg Fm.

Lithological description: Marl, argillaceous

CT-based slice selection: The sample contains pyrite concretions of considerable size (< 0.5 cm (A, B)). Two slices were sampled: Slice 1 may contain less and smaller concretions than Slice 2.

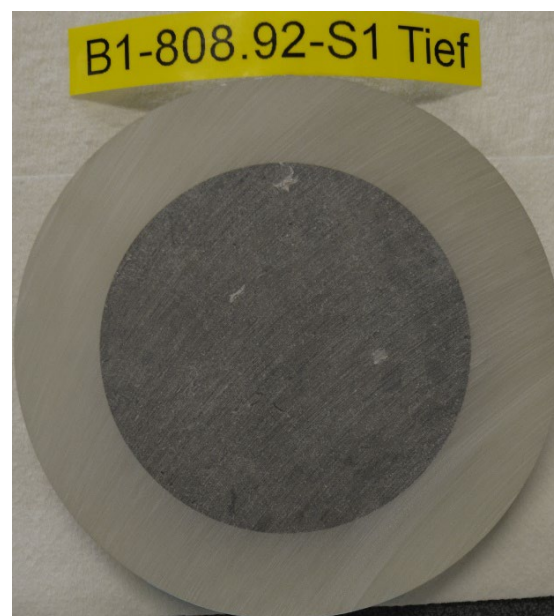
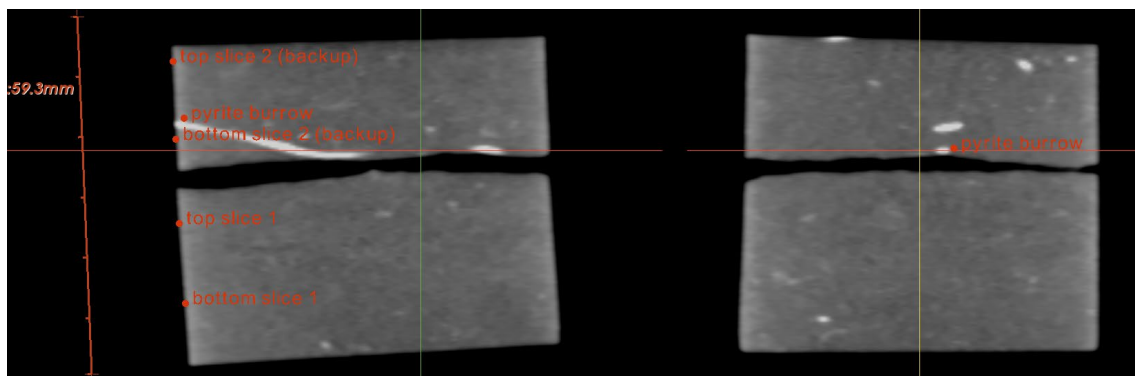
To be analysed: Slice 1

General comments regarding sample preparation: Cut surfaces of DI slices may not be entirely parallel.



BUL1-1-808.92-DI

Mean depth [m]:	808.92
Stratigraphic position:	Variansmergel Fm.
Lithological description:	Claystone
CT-based slice selection:	The sample contains minor amounts of small pyrite concretions and is slightly bioturbated. Two slices were sampled: Slice 2 contains a subhorizontal pyrite (or siderite?) burrow and slightly more pyrite concretions than Slice 1.
To be analysed:	Slice 1
General comments regarding sample preparation:	Cut surfaces of DI slices may not be entirely parallel.



BUL1-1-815.90-DI

Mean depth [m]: 815.90

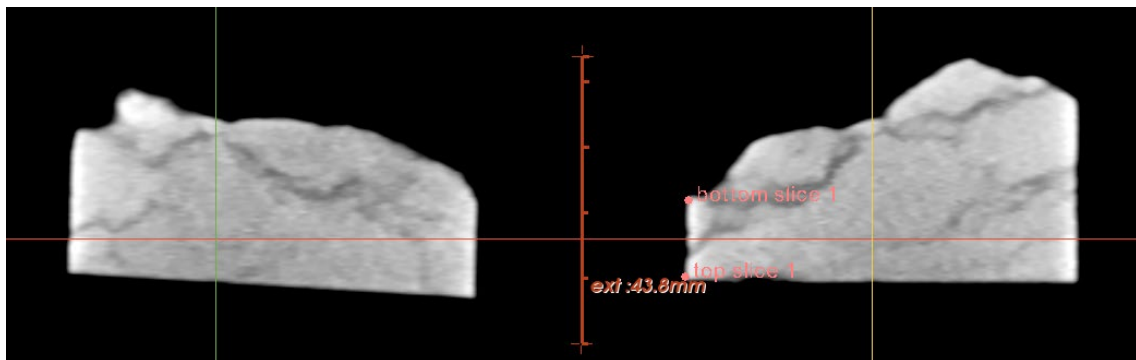
Stratigraphic position: Reef facies

Lithological description: Heterogeneous reef limestone with clay-rich seams (partly argillaceous)

CT-based slice selection: Only one slice was sampled.

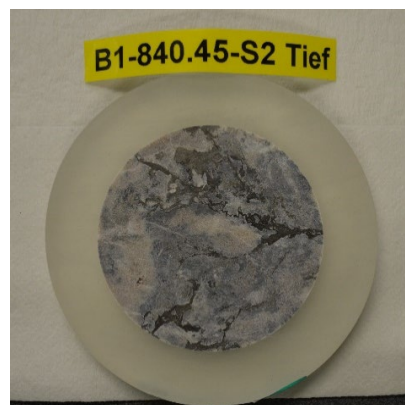
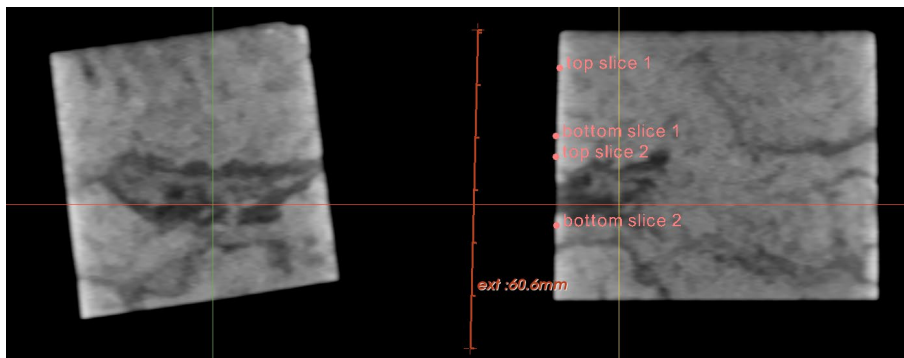
To be analysed: Slice 1

General comments regarding sample preparation: Sample broke apart during drilling. Leftover material is rather thin and thus the slice may be slightly thinner than required. Cut surfaces of DI slice may not be entirely parallel.



BUL1-1-840.45-DI

Mean depth [m]:	840.45
Stratigraphic position:	Reef facies
Lithological description:	Heterogeneous reef limestone with clay-rich seams (partly marly)
CT-based slice selection:	Sample is composed of calcareous material and clay seams. Two slices were sampled: Slice 1 is carbonate-rich and clay-poor, whereas Slice 2 is clay-rich and carbonate-poor.
To be analysed:	Both slices should be analysed.
General comments regarding sample preparation:	Cut surfaces of DI slices may not be entirely parallel.



BUL1-1-845.05-DI

Mean depth [m]: 845.05

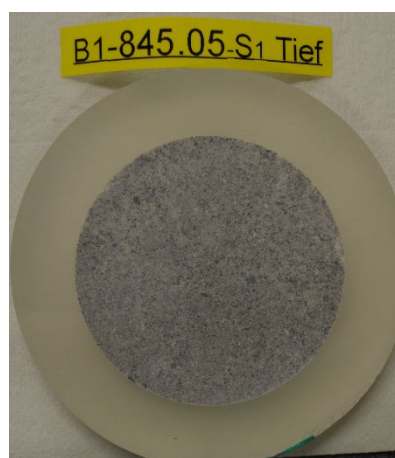
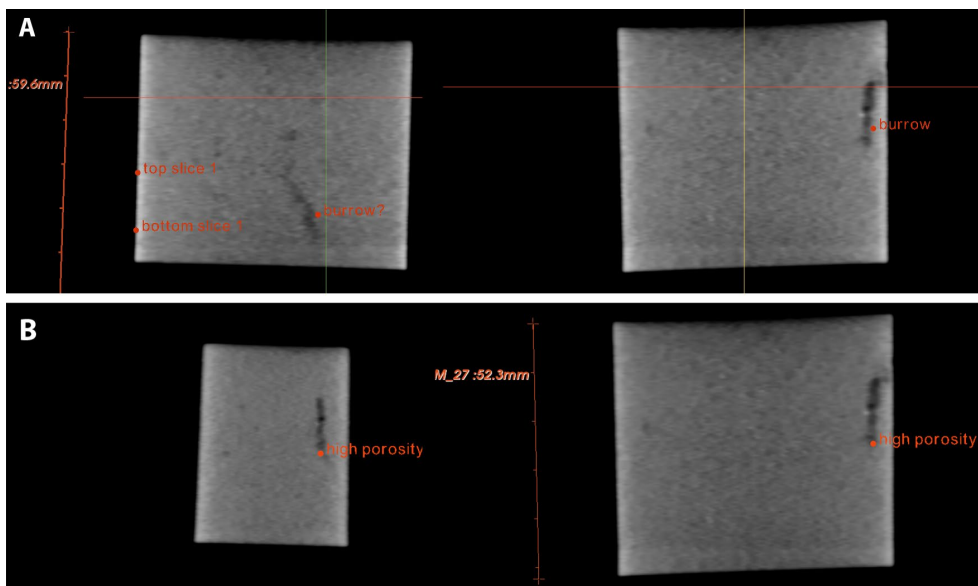
Stratigraphic position: Reef facies

Lithological description: Limestone-marl

CT-based slice selection: Sample is slightly bioturbated. Upper part of sample contains a bedding-orthogonal burrow that contains distinctly less dense material compared to the surrounding rock matrix (porosity?). Lower part of sample also contains a burrow, however, it is more weakly developed and the density contrast between matrix and burrow is not as high as above. Thus, only the lower part is sampled.

To be analysed: Slice 1

General comments regarding sample preparation: Cut surfaces of DI slice may not be entirely parallel.



BUL1-1-851.50-DI

Mean depth [m]: 851.50

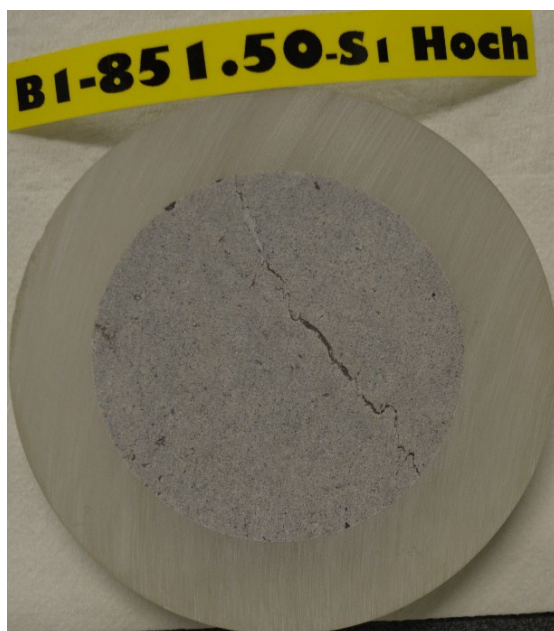
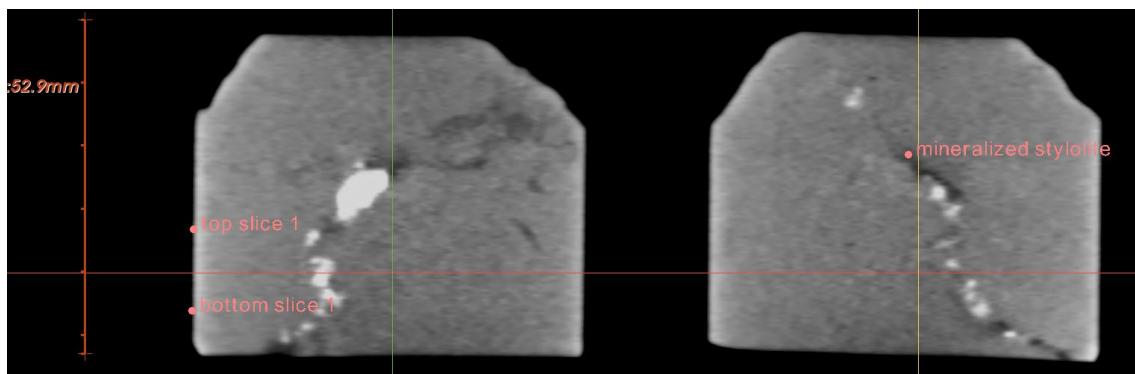
Stratigraphic position: Reef facies

Lithological description: Limestone-marl with one oblique clay seam

CT-based slice selection: Sample contains a mineralised tectonic stylolite which crosscuts the entire sample. The mineralisation shows fibrous radiating textures and may be sulphate. Apart from these features, the sample appears rather homogeneous. One slice was sampled.

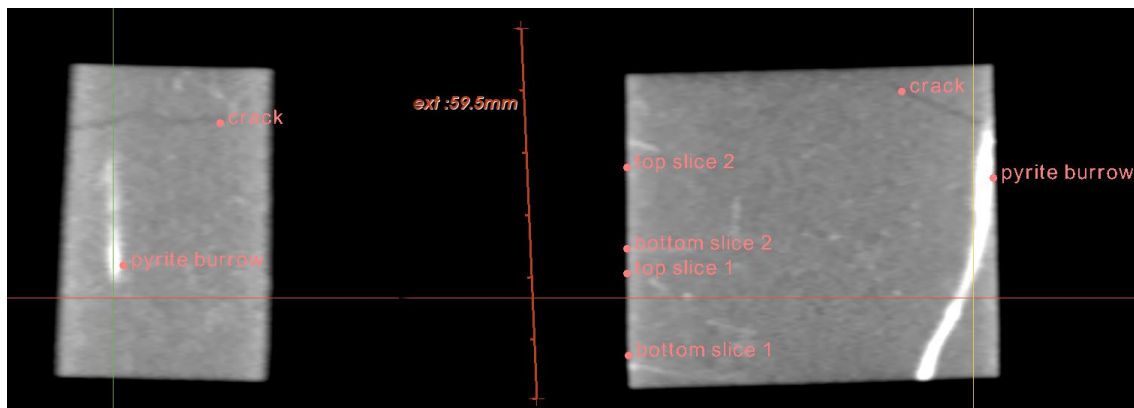
To be analysed: Slice 1

General comments regarding sample preparation: Cut surfaces of DI slice may not be entirely parallel.



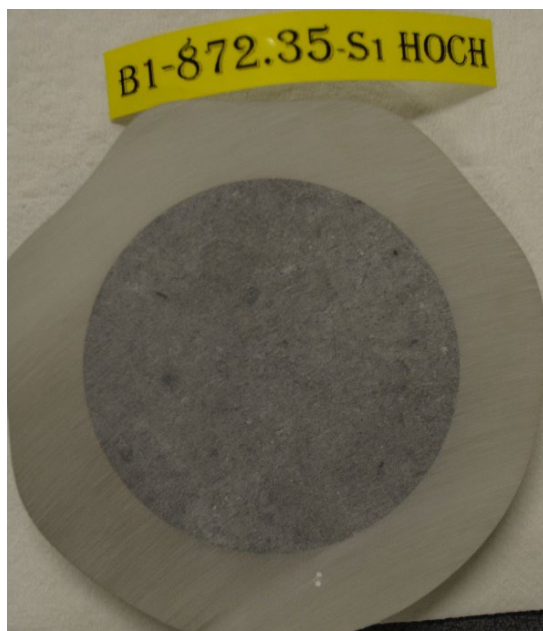
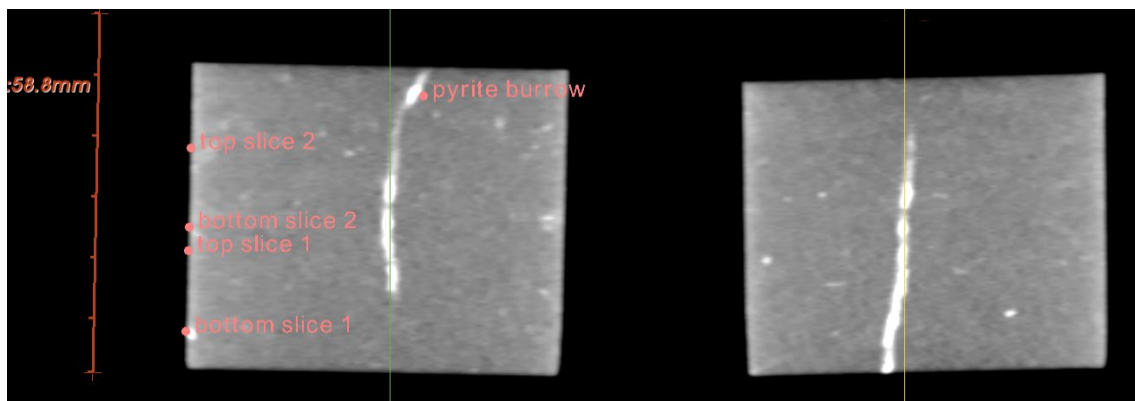
BUL1-1-861.00-DI

Mean depth [m]:	861.00
Stratigraphic position:	Opalinus Clay?
Lithological description:	Claystone
CT-based slice selection:	Bioturbated sample containing a massive, bedding-orthogonal pyrite (or siderite?) burrow that cuts across the entire sample.
To be analysed:	No slices planned for analysis as sample shows same texture and features as BUL1-1-872.35-DI.
General comments regarding sample preparation:	Cut surfaces of DI slices may not be entirely parallel.



BUL1-1-872.35-DI

Mean depth [m]:	872.35
Stratigraphic position:	Opalinus Clay?
Lithological description:	Claystone
CT-based slice selection:	Bioturbated sample containing a massive, bedding-orthogonal pyrite (or siderite?) burrow that cuts across the entire sample. Two slices were sampled.
To be analysed:	Slice 1 (arbitrary selection)
General comments regarding sample preparation:	Sample experienced 80 – 100 °C during epoxy embedding. Cut surfaces of DI slices may not be entirely parallel.



BUL1-1-879.20-DI

Mean depth [m]: 879.20

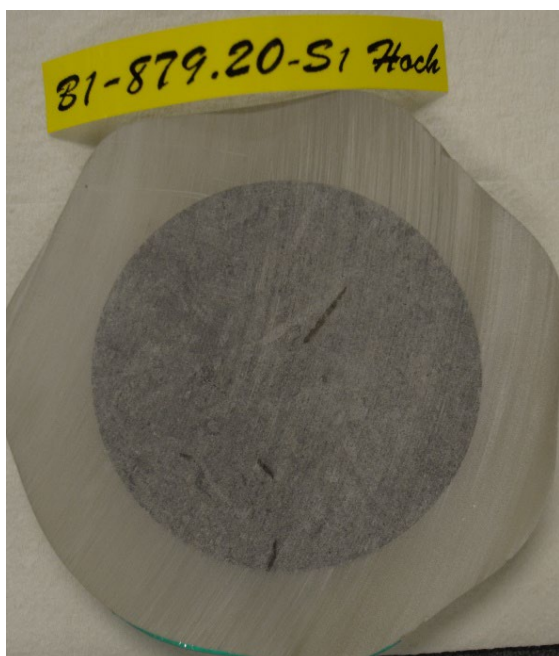
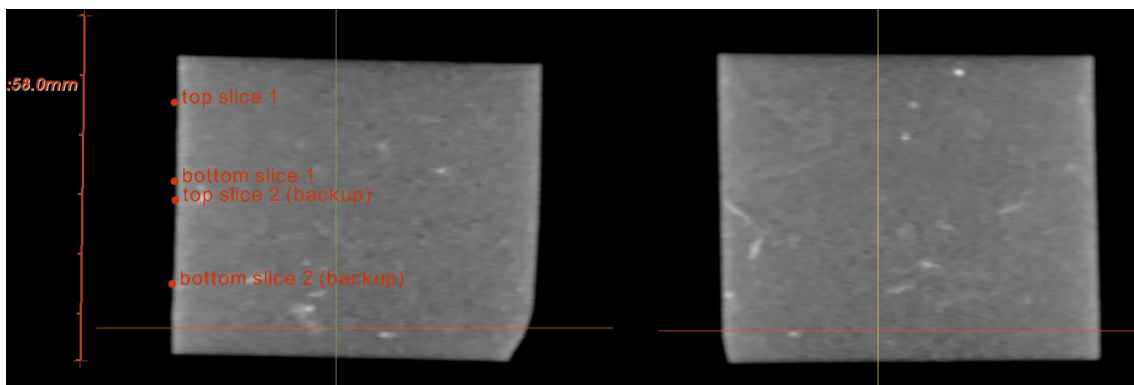
Stratigraphic position: Opalinus Clay?

Lithological description: Claystone

CT-based slice selection: Sample is bioturbated and contains some pyrite concretions and burrows. Two slices were sampled, which may show slight differences in the amount of burrows and pyrite concretions.

To be analysed: Slice 1 (arbitrary selection)

General comments regarding sample preparation: Sample experienced 80 – 100 °C during epoxy embedding. Cut surfaces of DI slices may not be entirely parallel.



BUL1-1-908.93-DI

Mean depth [m]: 908.93

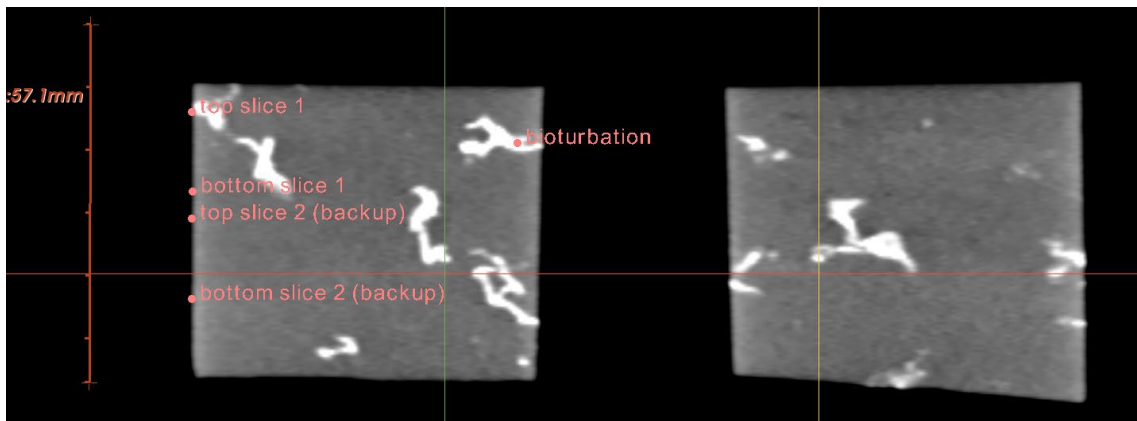
Stratigraphic position: Opalinus Clay

Lithological description: Claystone

CT-based slice selection: Entire sample is strongly bioturbated and contains considerable amounts of massive pyrite (or siderite?) burrows. Two slices were sampled, which may show slight differences in the amount of burrows.

To be analysed: Slice 1 (arbitrary selection)

General comments regarding sample preparation: Sample experienced 80–100 °C during epoxy embedding. Cut surfaces of DI slices may not be entirely parallel.



BUL1-1-914.61-DI

Mean depth [m]: 914.61

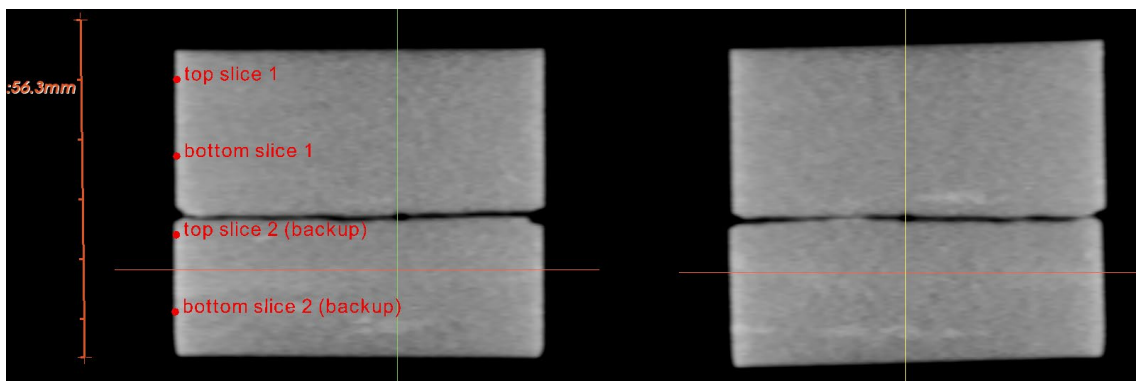
Stratigraphic position: Opalinus Clay

Lithological description: Claystone

CT-based slice selection: Sample contains some silty lenses. Two slices were sampled: From a quantitative point of view, Slice 2 contains slightly more silty lenses than Slice 1. Otherwise, they appear similar.

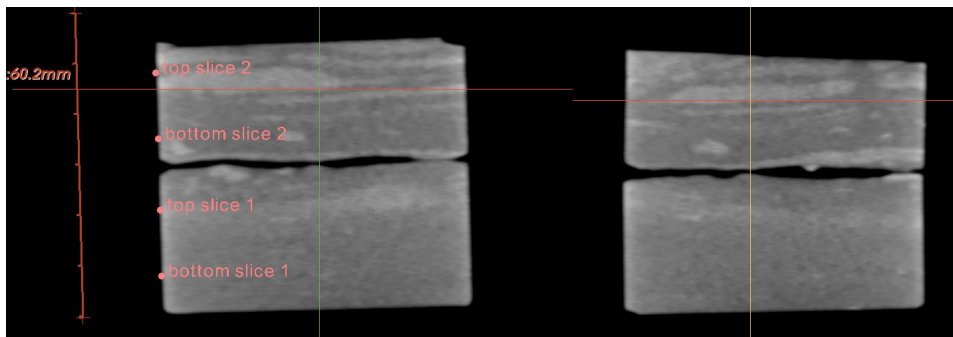
To be analysed: Slice 1

General comments regarding sample preparation: Cut surfaces of DI slices may not be entirely parallel.



BUL1-1-921.43-DI

Mean depth [m]:	921.43
Stratigraphic position:	Opalinus Clay
Lithological description:	Claystone with many silty lenses
CT-based slice selection:	Sample contains many silty lenses. Two slices were sampled: From a quantitative point of view, Slice 2 contains distinctly more of these lenses than Slice 1. In comparison, Slice 1 appears more homogeneous.
To be analysed:	Both slices should be analysed
General comments regarding sample preparation:	Cut surfaces of DI slices may not be entirely parallel.



BUL1-1-932.82-DI

Mean depth [m]: 932.82

Stratigraphic position: Opalinus Clay

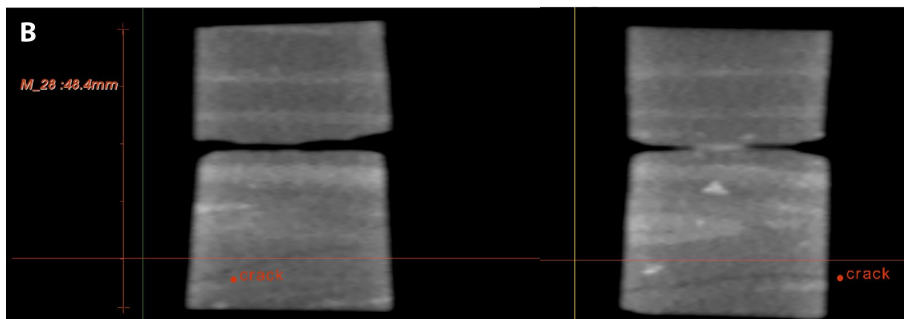
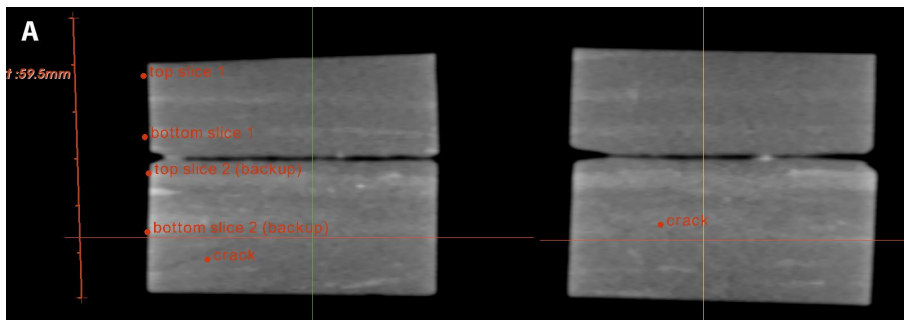
Lithological description: Claystone with silty lenses

CT-based slice selection: Sample contains silty lenses. Two slices were sampled: From a quantitative point of view, Slice 2 contains more and thicker silty lenses than Slice 1 (A). Slice 2 may contain a crack that propagates from below into the slice. However, it may also be a stylolite (B).

To be analysed: Slice 1

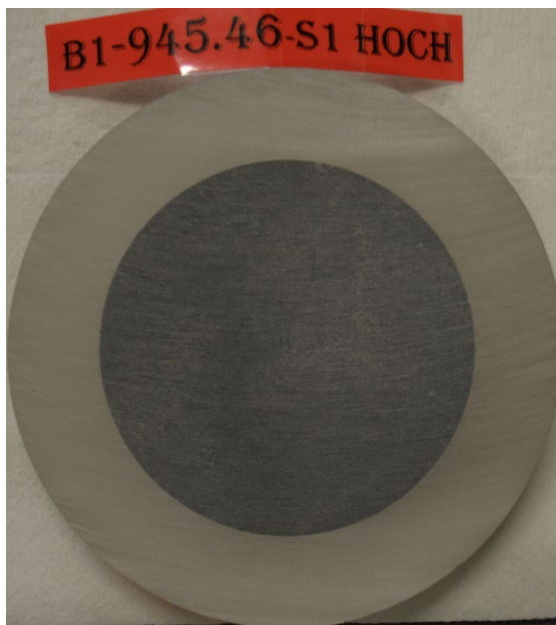
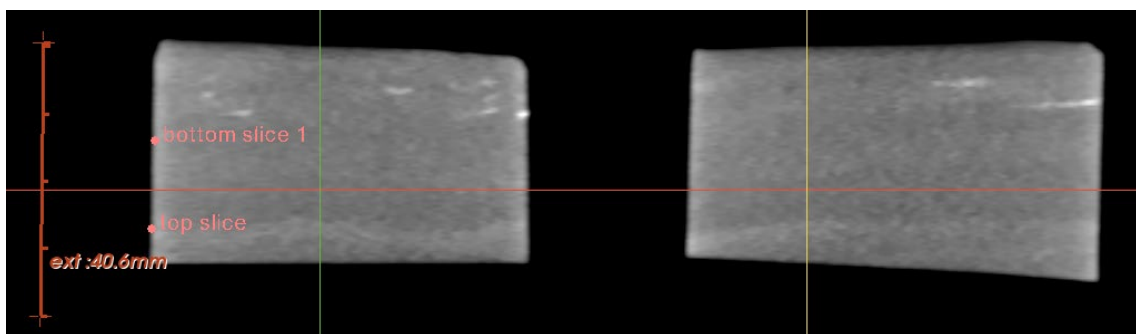
General comments regarding sample preparation:

Sample experienced 80 – 100 °C during epoxy embedding. Cut surfaces of DI slices may not be entirely parallel.



BUL1-1-945.46-DI

Mean depth [m]: 945.46
Stratigraphic position: Opalinus Clay
Lithological description: Claystone
CT-based slice selection: Short sample due to dinking, thus only one slice was sampled. Apart from some small pyrite concretions, this slice appears rather homogeneous.
To be analysed: Slice 1
General comments regarding sample preparation: Cut surfaces of DI slice may not be entirely parallel.



BUL1-1-955.62-DI

Mean depth [m]: 955.62

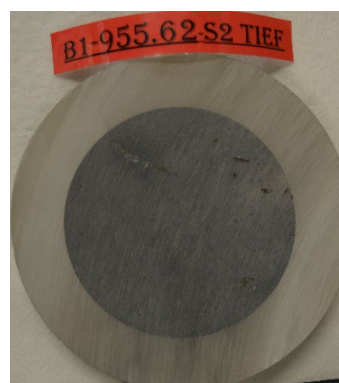
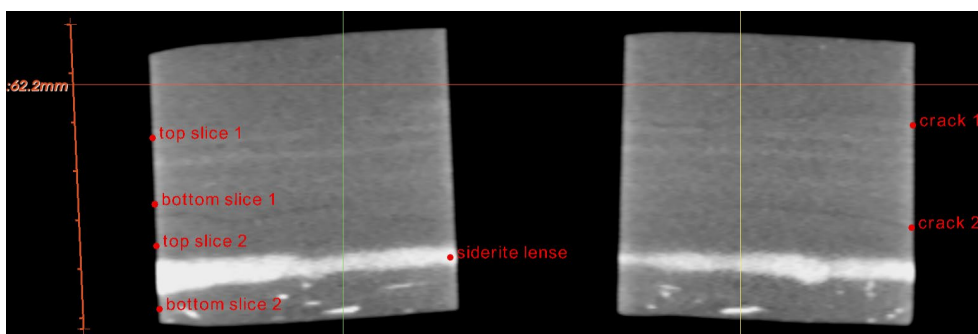
Stratigraphic position: Opalinus Clay

Lithological description: Claystone

CT-based slice selection: Apart from a siderite lens rather homogeneous sample. Two slices were sampled: Slice 1 within the homogeneous clay matrix where it had to be located within 2 narrowly spaced bedding-parallel cracks or stylolites. Slice 2 includes the siderite lens.

To be analysed: Both slices.

General comments regarding sample preparation: Cut surfaces of DI slices may not be entirely parallel.



BUL1-1-965.46-DI

Mean depth [m]: 965.46

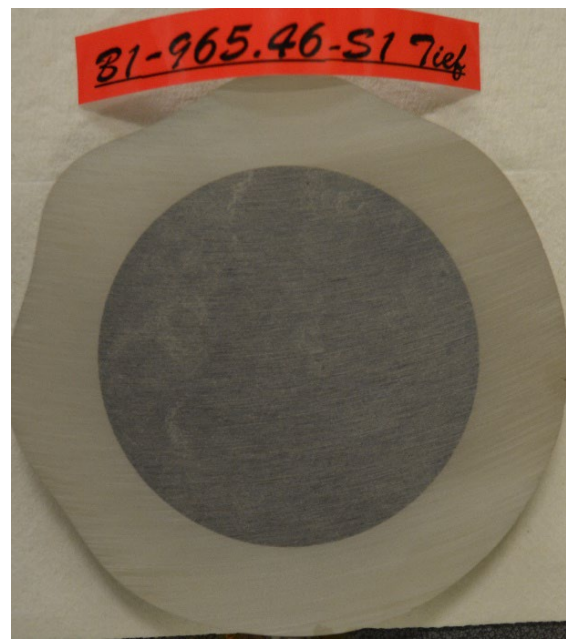
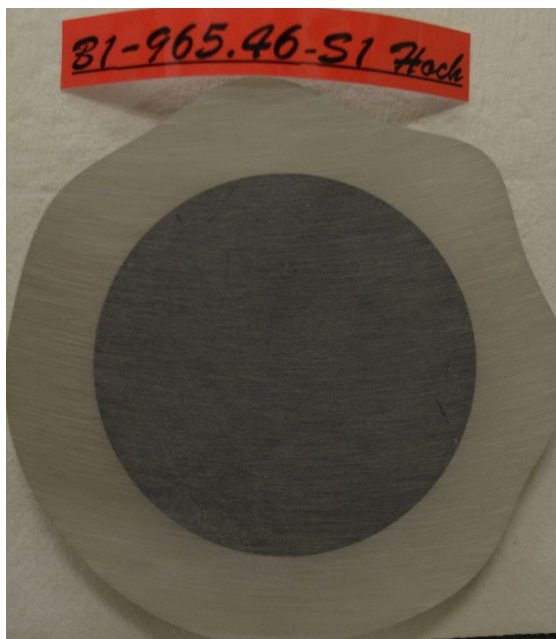
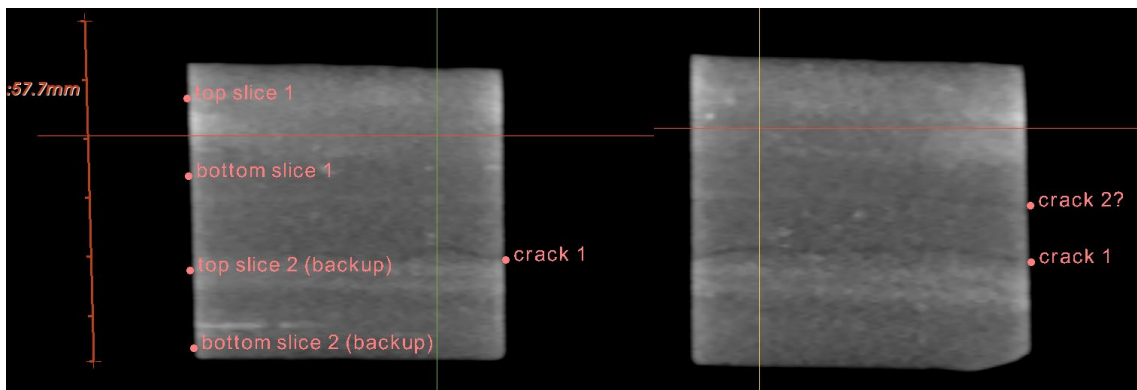
Stratigraphic position: Opalinus Clay

Lithological description: Claystone

CT-based slice selection: Sample shows alternating high and low density layers (possibly marly or silty vs. clay-rich), some small pyrite concretions, and it contains 2 bedding-parallel cracks or stylolites. Two slices were sampled (avoiding the cracks or stylolites): Slice 1 may contain slightly more of the high-density material than Slice 2, whereas Slice 2 may contain slightly more of the low-density material.

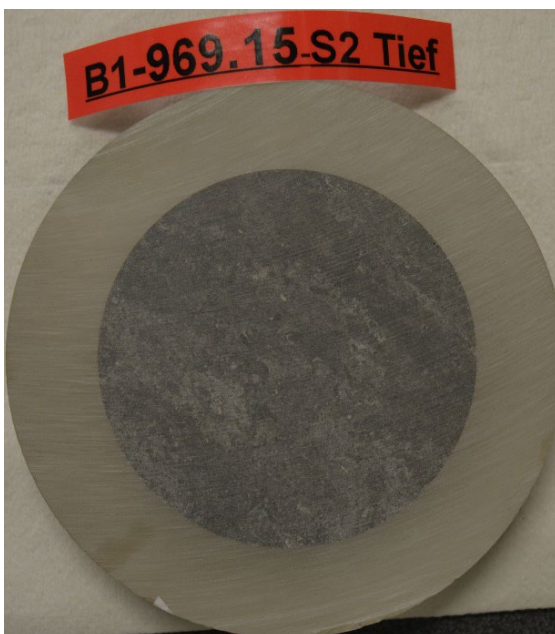
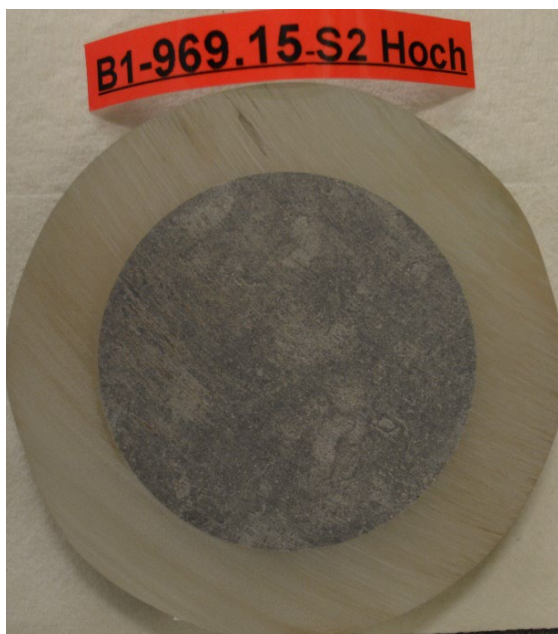
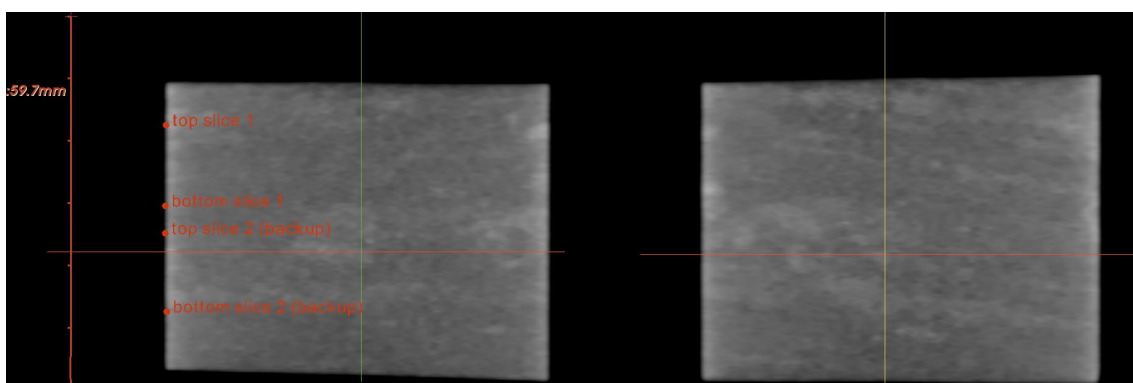
To be analysed: Slice 1

General comments regarding sample preparation: Sample exposed to temperatures of 80 – 100 °C during epoxy embedding. Cut surfaces of DI slices may not be entirely parallel.



BUL1-1-969.15-DI

Mean depth [m]:	969.15
Stratigraphic position:	Opalinus Clay
Lithological description:	Claystone
CT-based slice selection:	Sample contains some homogeneously distributed silty lenses. Only very little pyrite is present (on macro scale). Two slices were planned to be sampled, however, only one slice was successfully recovered (Slice 1 broke apart while cutting).
To be analysed:	Slice 2
General comments regarding sample preparation:	Sample exposed to temperatures of 80 – 100 °C during epoxy embedding. Cut surfaces of DI slices may not be entirely parallel.



BUL1-1-975.04-DI

Mean depth [m]: 975.04

Stratigraphic position: Opalinus Clay

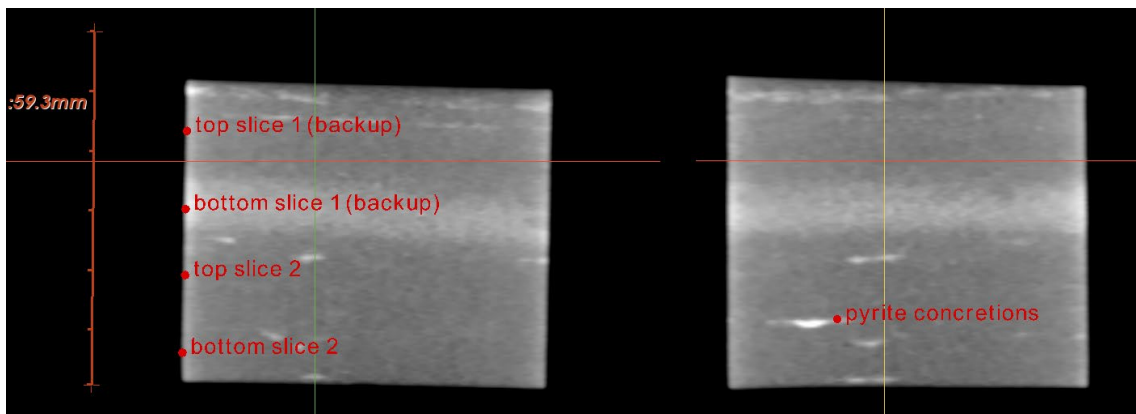
Lithological description: Claystone

CT-based slice selection: The sample contains a cm-thick silty lens in an otherwise clay-rich rock, which contains some pyrite concretions (or siderite?). Two slices were sampled: Slice 1 partly contains the silty lens, whereas Slice 2 is more representative of the clay-rich bulk of the sample.

To be analysed: Slice 2

General comments regarding sample preparation:

Sample exposed to temperatures of 80 – 100 °C during epoxy embedding. Cut surfaces of DI slices may not be entirely parallel.



BUL1-1-984.49-DI

Mean depth [m]: 984.49

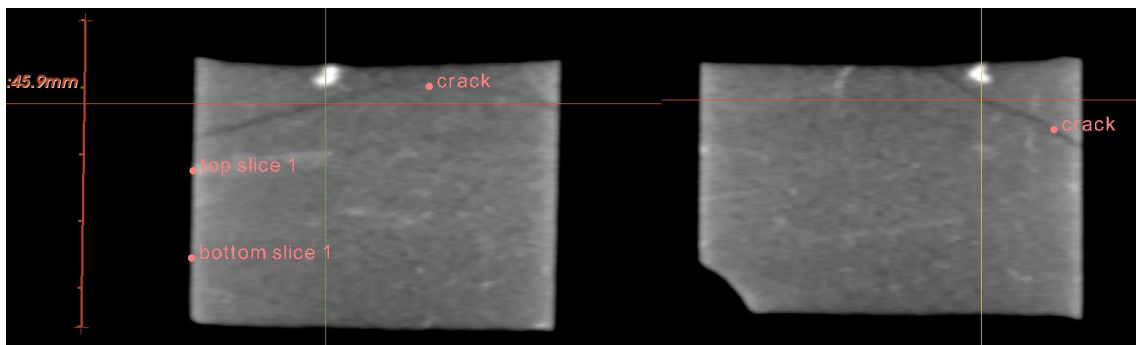
Stratigraphic position: Opalinus Clay

Lithological description: Claystone

CT-based slice selection: Sample contains a crack, thus, only one slice was sampled. This slice shows no major heterogeneities and contains only little pyrite.

To be analysed: Slice 1

General comments regarding sample preparation: Cut surfaces of DI slice may not be entirely parallel.



BUL1-1-995.09-DI

Mean depth [m]: 995.09

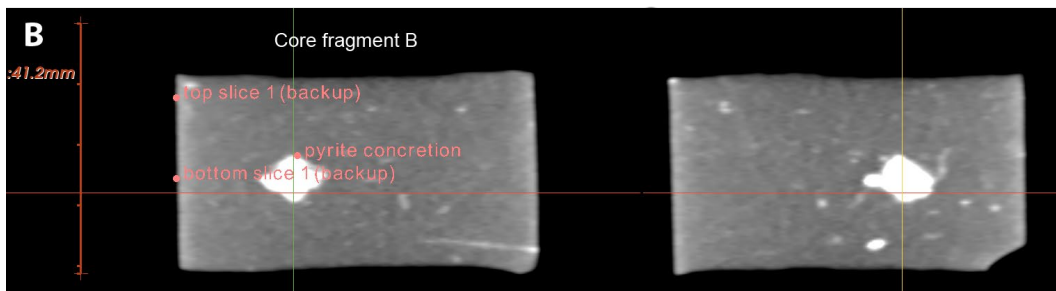
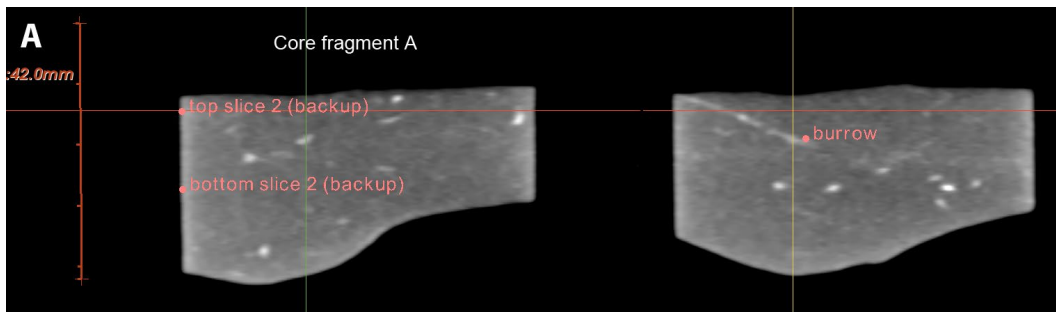
Stratigraphic position: Opalinus Clay

Lithological description: Claystone

CT-based slice selection: Bioturbated sample composed of two core fragments. One slice per fragment was sampled: Slice 2 is crosscut by a thin, oblique pyrite (or siderite?) burrow. Slice 1 partly contains a massive pyrite concretion.

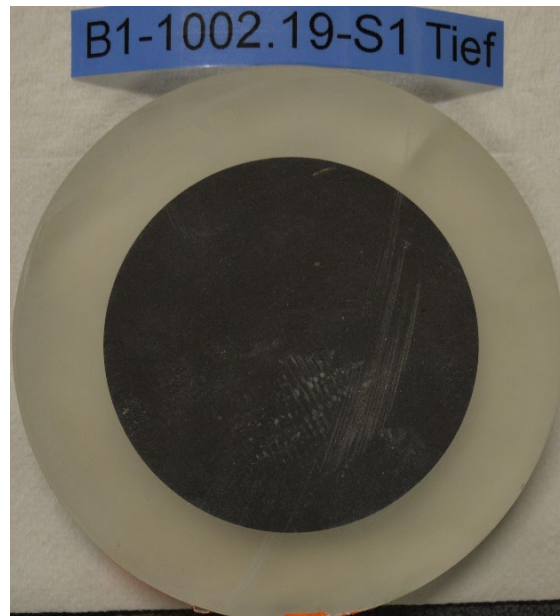
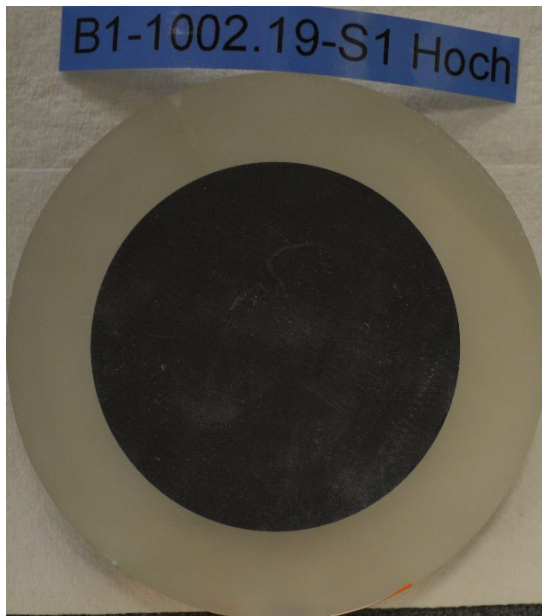
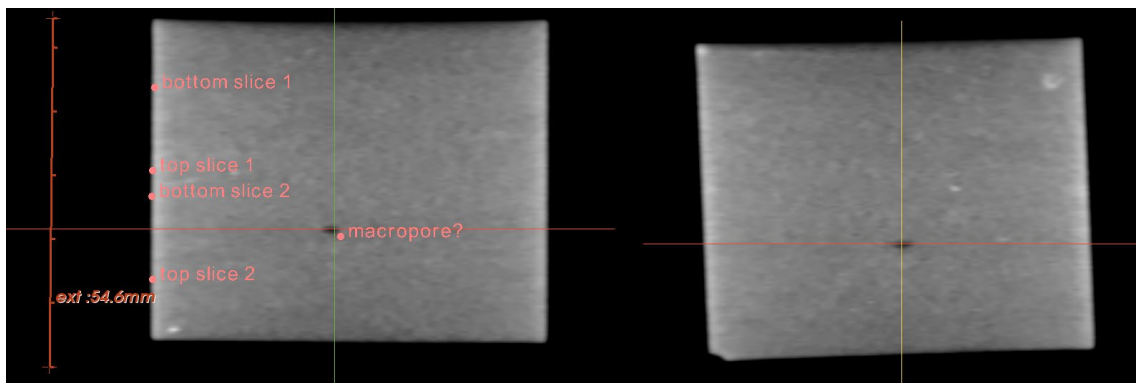
To be analysed: Slice 2

General comments regarding sample preparation: Cut surfaces of DI slices may not be entirely parallel.



BUL1-1-1,002.19-DI

Mean depth [m]: 1,002.19
Stratigraphic position: Staffelegg Fm.
Lithological description: Claystone
CT-based slice selection: Rather homogeneous sample containing only little pyrite. Two slices were sampled: Slice 2 contains a macropore (ca. 0.2 cm). Apart from that, Slices 1 and 2 appear similar.
To be analysed: Slice 1
General comments regarding sample preparation: Cut surfaces of DI slices may not be entirely parallel.



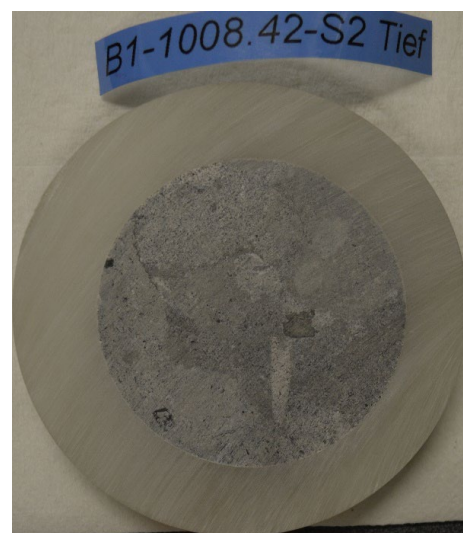
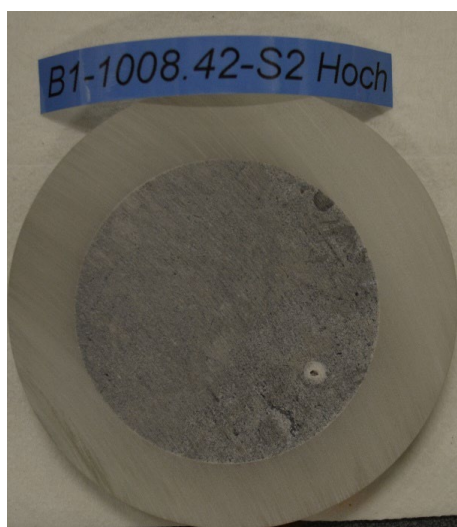
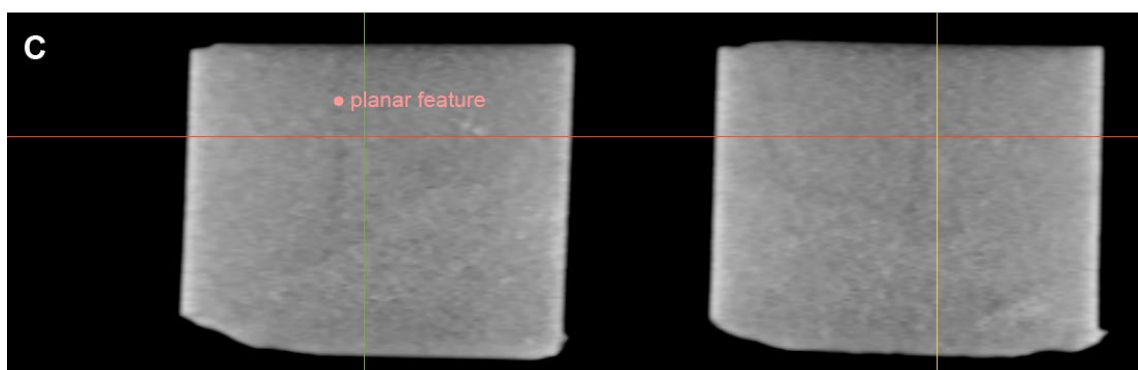
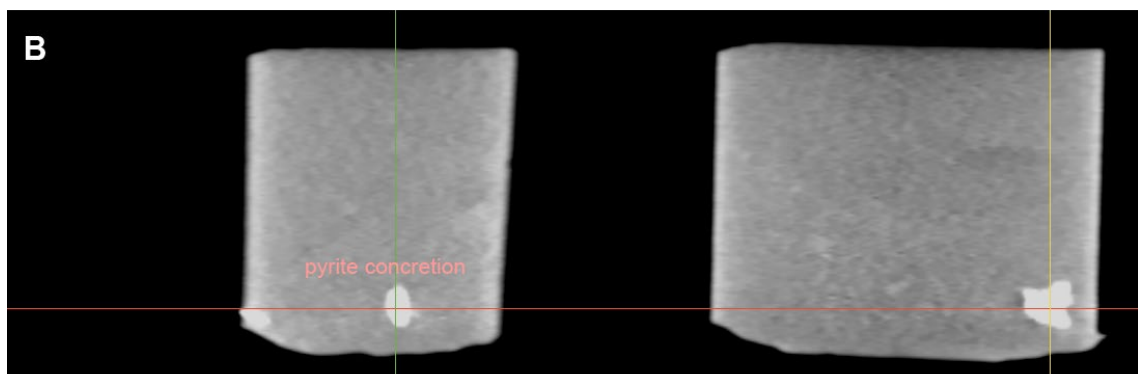
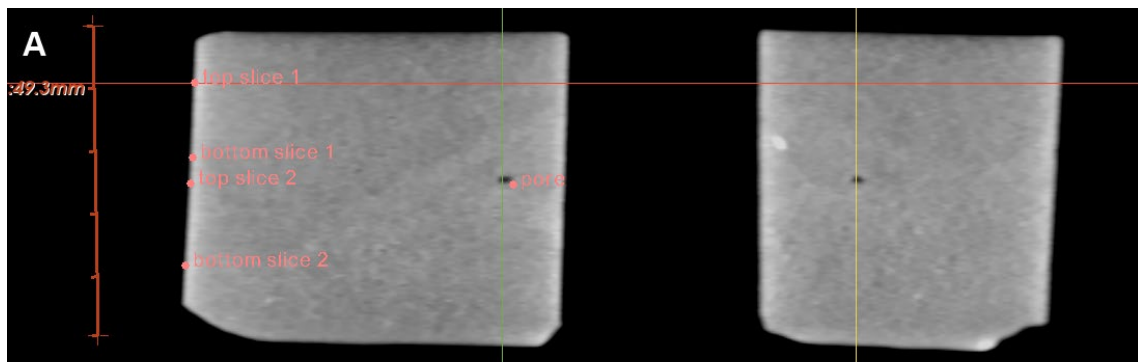
BUL1-1-1008.42-DI

Mean depth [m]: 1,008.42
Stratigraphic position: Staffelegg Fm.
Lithological description: Heterogeneous claystone-calc. marl
CT-based slice selection:

Sample contains a large pyrite concretion at the base and a macropore in the middle of the sample, which can all be avoided for sampling (Fig. 25). What cannot be avoided are planar, low-density, subvertical-vertical features that cut across the sample. Presumably, these are tectonic stylolites. Moreover, there are two slightly different density domains: a comparatively lower density domain (clay-rich) in the upper part of the sample and a comparatively higher density domain (calc.-marl) in the lower part of the sample. Two slices were sampled: Slice 1 in the upper part of the sample is almost entirely composed of the low density material, whereas Slice 2 in the lower part of the sample represents a mixture of both and is considered more representative of the entire sample.

To be analysed: Slice 2

General comments regarding
sample preparation: Cut surfaces of DI slices may not be entirely parallel.



BUL1-1-1014.23-DI

Mean depth [m]: 1,014.23

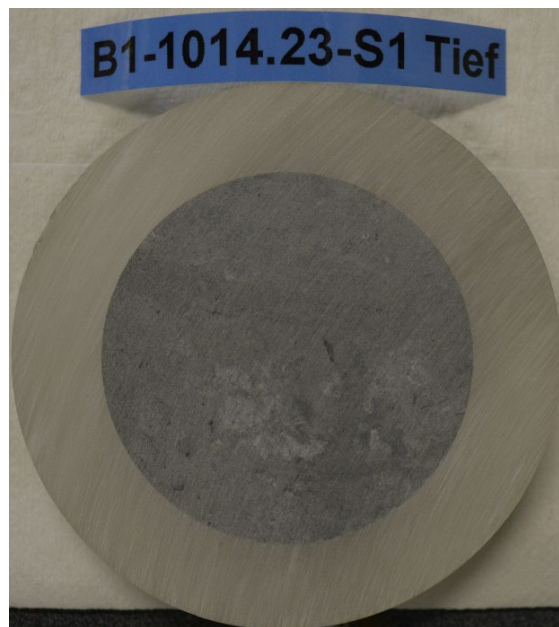
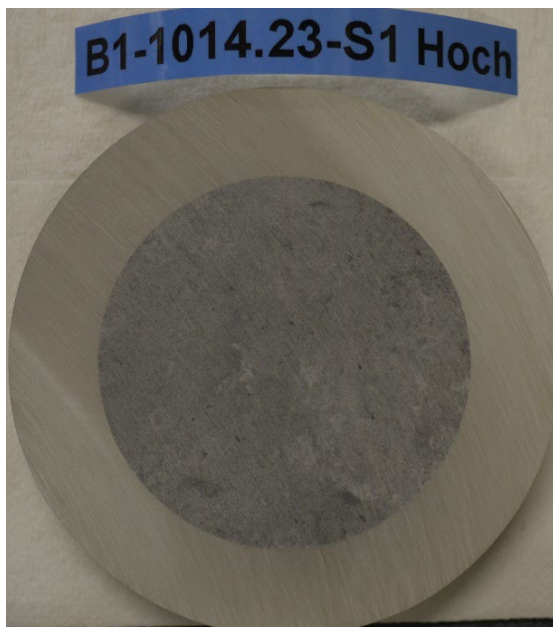
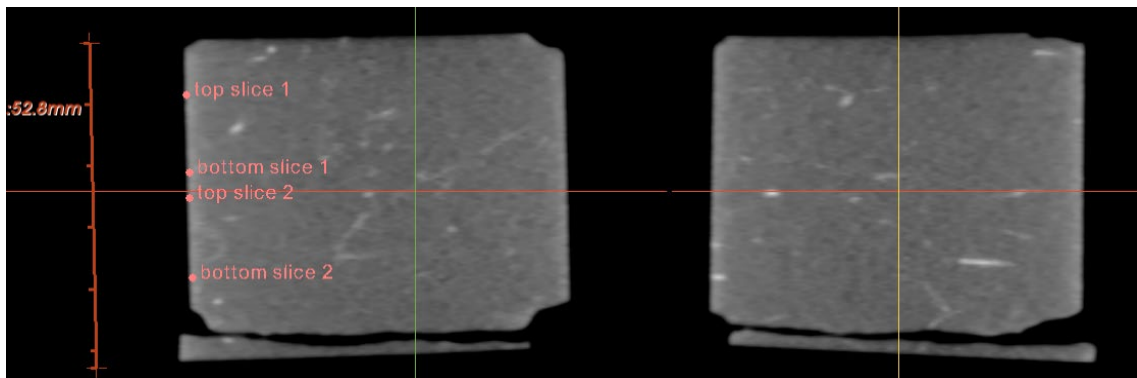
Stratigraphic position: Staffelegg Fm.

Lithological description: Claystone

CT-based slice selection: Sample is bioturbated and contains many small pyrite (or siderite?) concretions and burrows. Two slices were sampled. There is a relatively large, but bedding-parallel burrow located between the two slices. Overall, Slice 1 and Slice 2 appear similar.

To be analysed: Slice 1 (arbitrary selection)

General comments regarding sample preparation: Cut surfaces of DI slices may not be entirely parallel.



BUL1-1-1017.33-DI

Mean depth [m]: 1,017.33

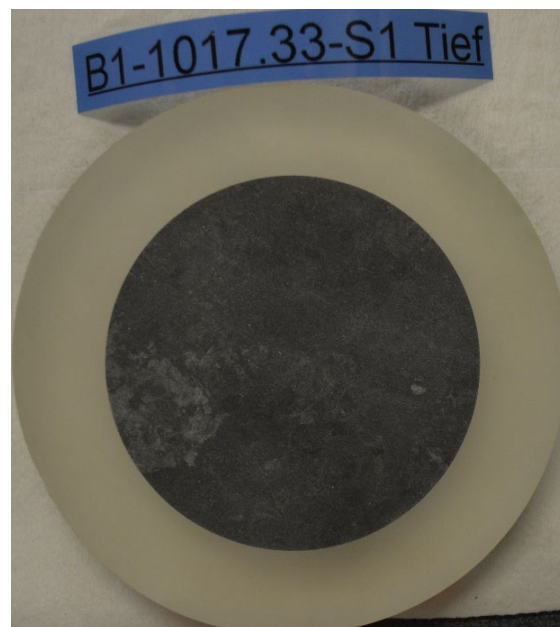
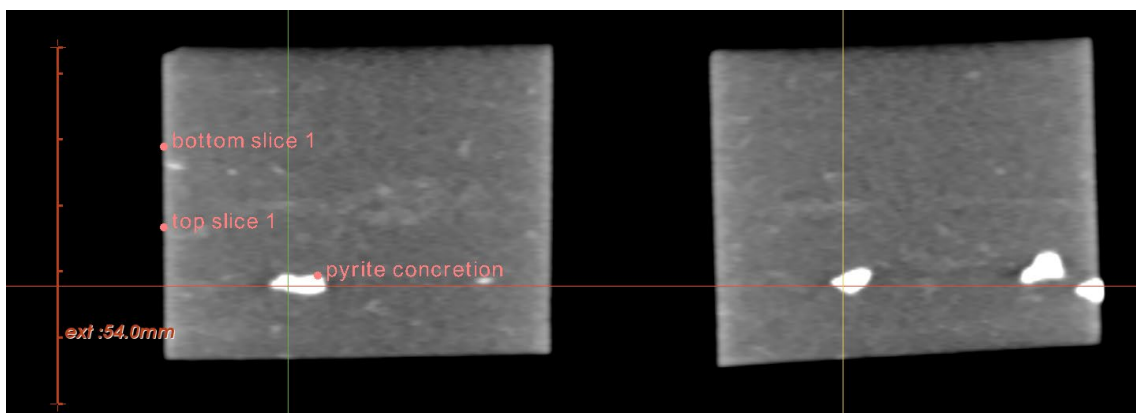
Stratigraphic position: Staffelegg Fm.

Lithological description: Claystone

CT-based slice selection: Sample contains some relatively large (0.8 cm) pyrite concretions in its lower part. Apart from that, this is a rather homogeneous sample. Only one slice was sampled.

To be analysed: Slice 1

General comments regarding sample preparation: Cut surfaces of DI slice may not be entirely parallel.



BUL1-1-1025.96-DI

Mean depth [m]: 1,025.96

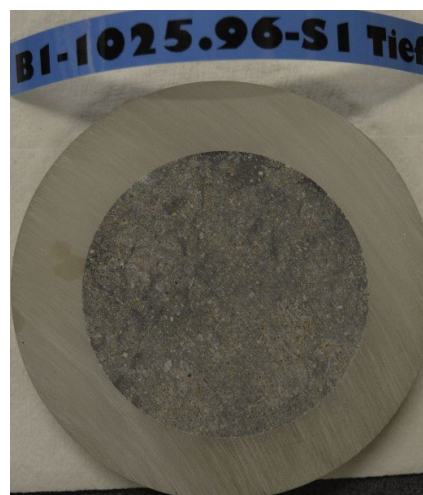
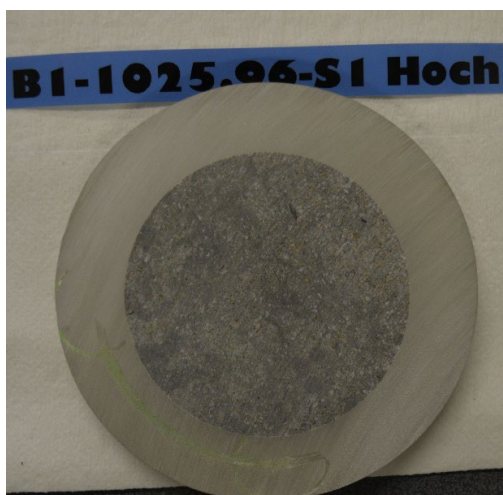
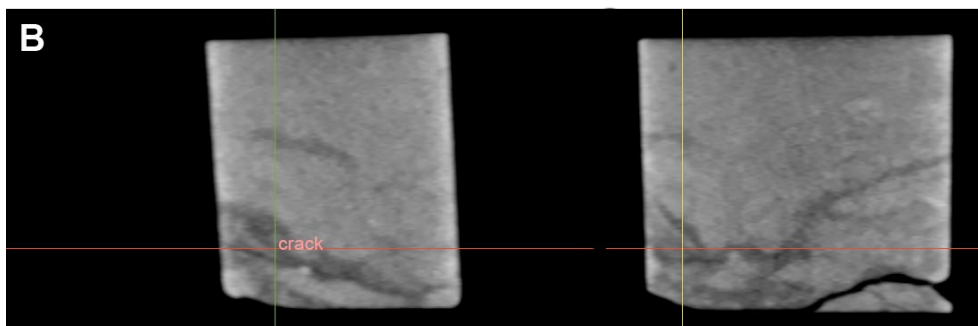
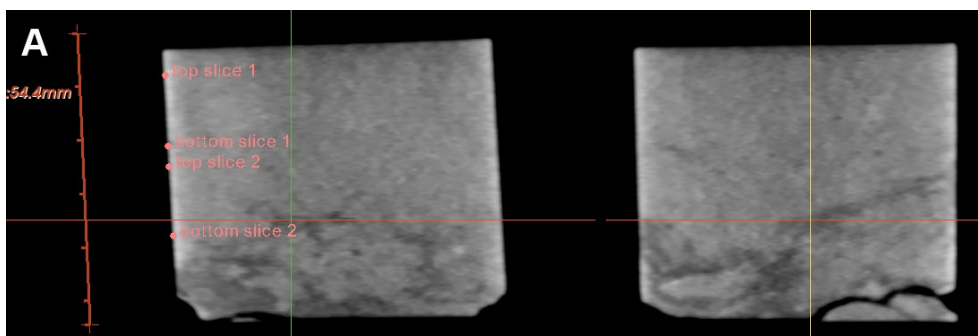
Stratigraphic position: Staffelegg Fm.

Lithological description: Heterogeneous arg. marl–calc. marl

CT-based slice selection: In its upper part, the sample appears rather homogeneous, whereas in its lower part it contains stylolites/clay seams. Two slices were sampled: Slice 1 is located in the relatively homogeneous upper part, whereas the underlying Slice 2 contains some stylolites/clay seams.

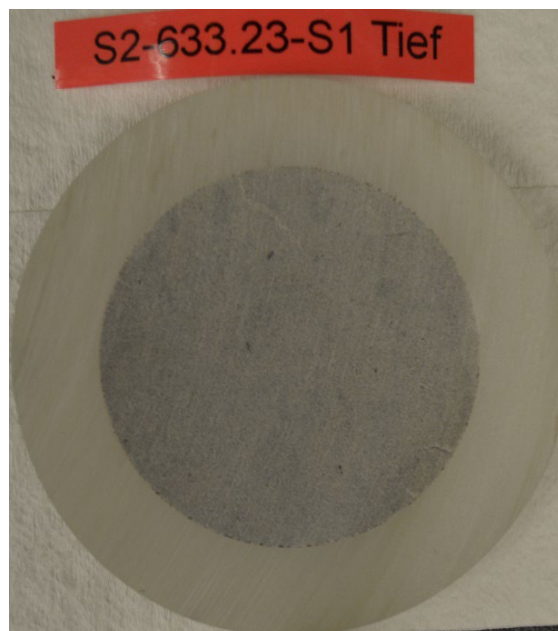
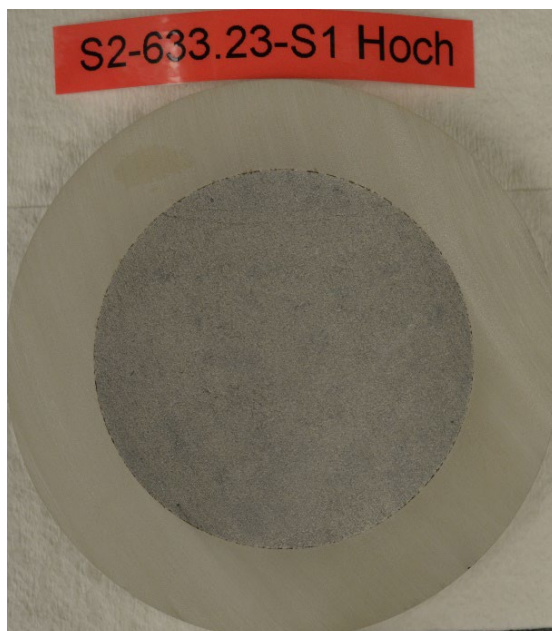
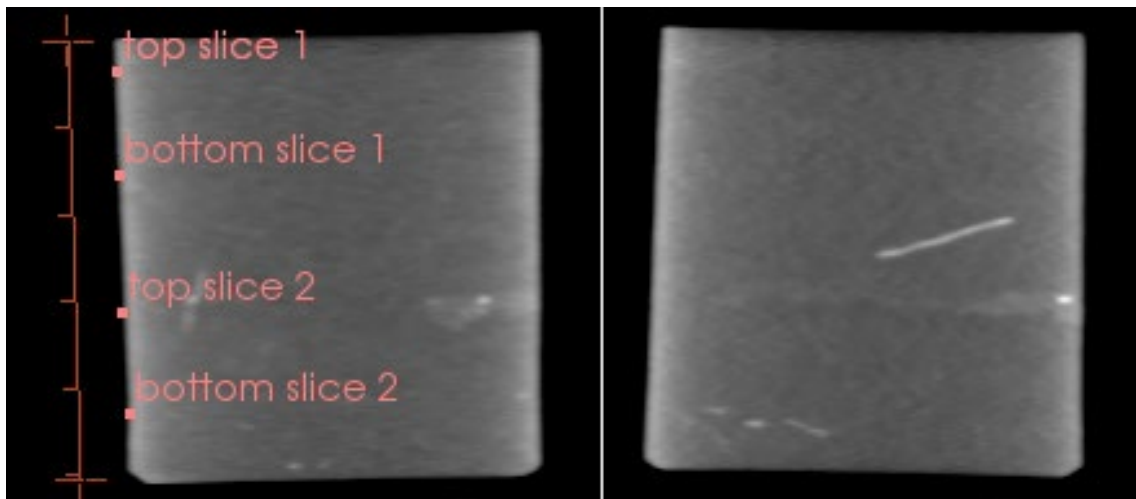
To be analysed: Slice 1

General comments regarding sample preparation: Cut surfaces of DI slices may not be entirely parallel.



H.2.3 Stadel 2-1**STA2-1-633.23-DI**

Mean depth [m]: 633.23
Stratigraphic position: Villigen Fm. (Malm)
CT-based slice selection: Slice 1 is more homogeneous.
To be analysed: Slice 1



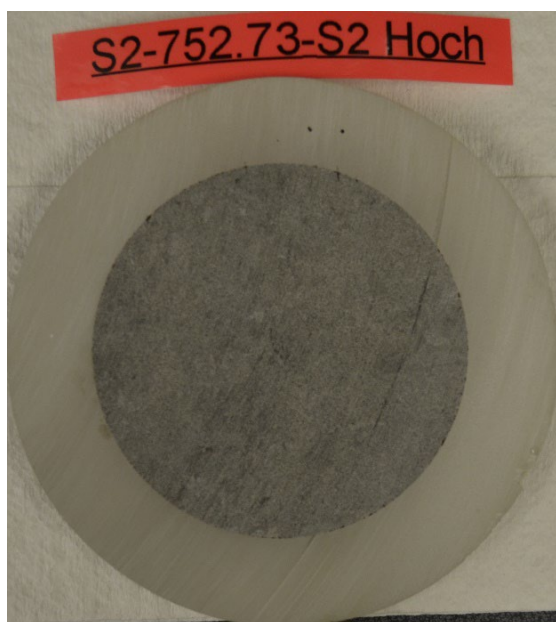
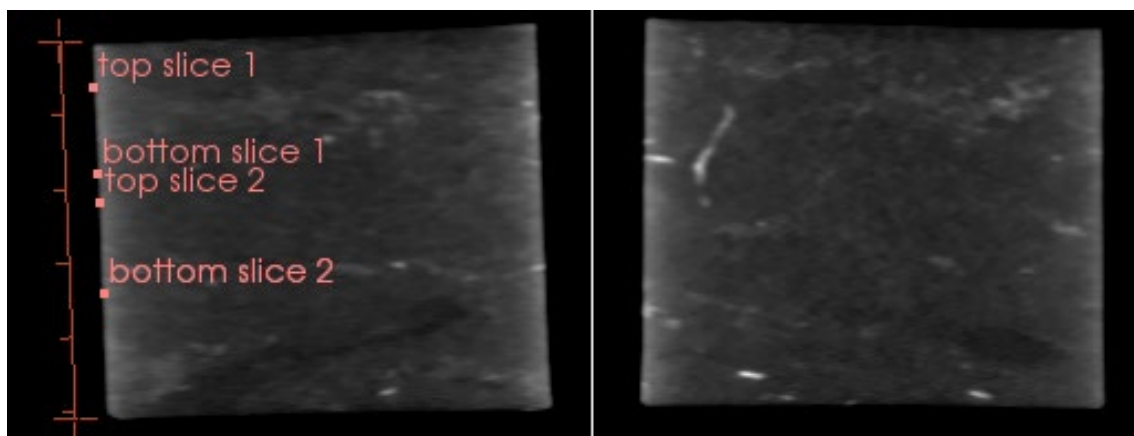
STA2-1-752.73-DI

Mean depth [m]: 752.73

Stratigraphic position: «Parkinsoni-Württembergica-Schichten» (Dogger)

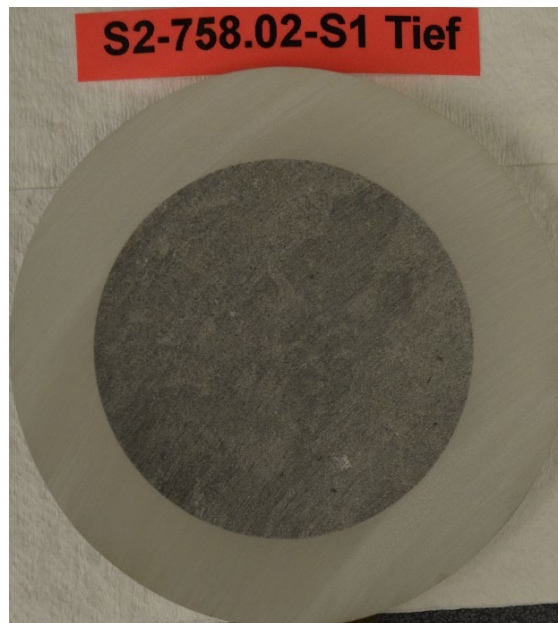
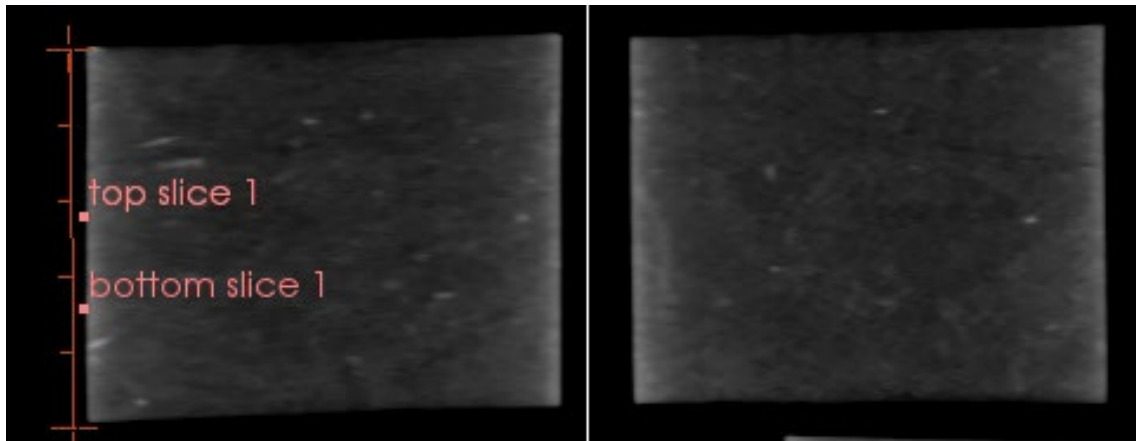
CT-based slice selection: Slice 2 is more homogeneous.

To be analysed: Slice 2



STA2-1-758.02-DI

Mean depth [m]: 758.02
Stratigraphic position: «Parkinsoni-Württembergica-Schichten» (Dogger)
CT-based slice selection: Only one slice was taken due to fractures.
To be analysed: Slice 1



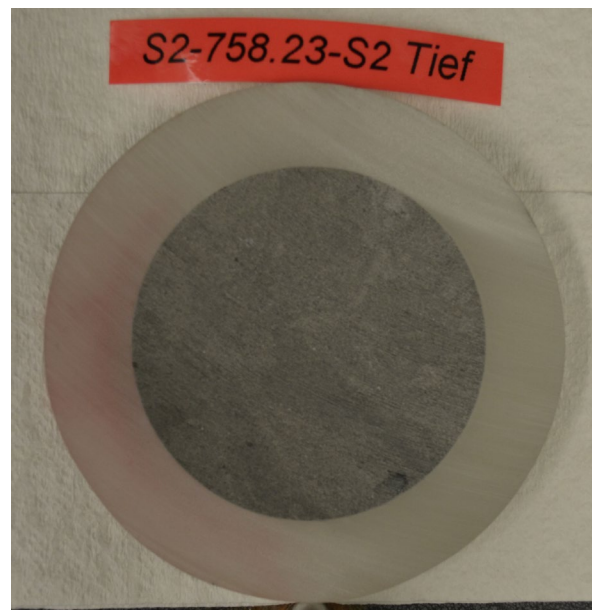
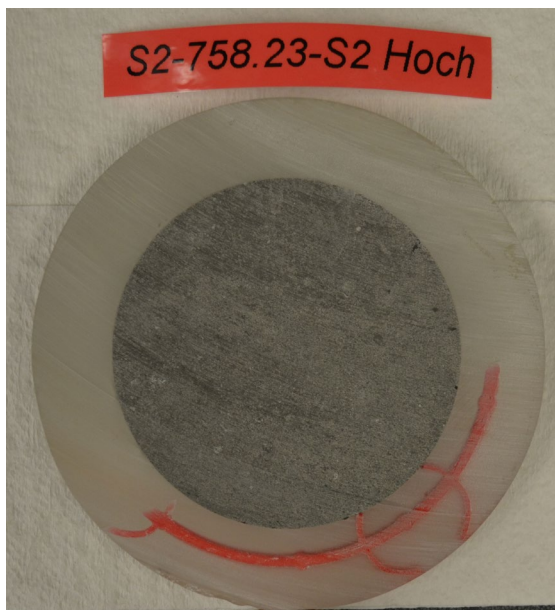
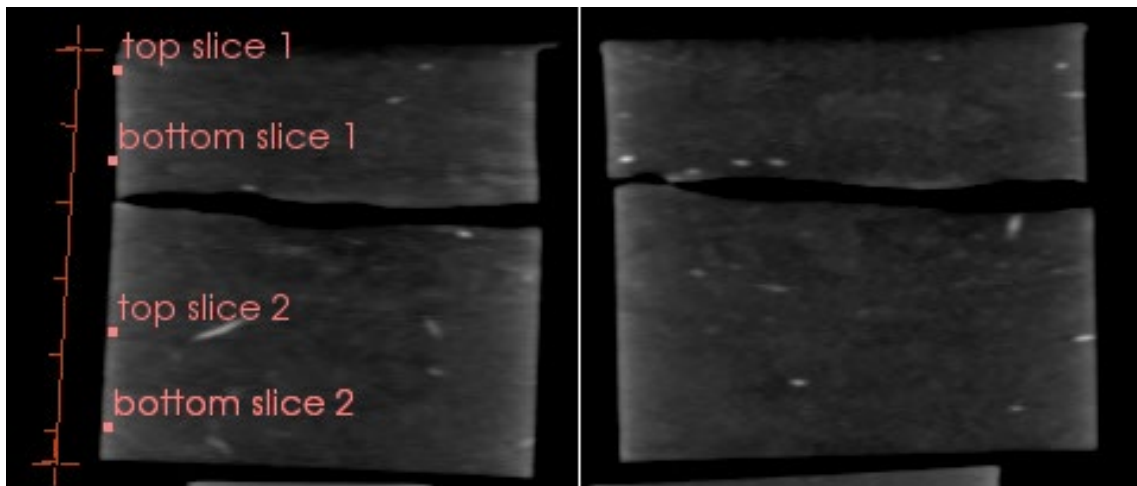
STA2-1-758.23-DI

Mean depth [m]: 758.23

Stratigraphic position: «Parkinsoni-Württembergica-Schichten» (Dogger)

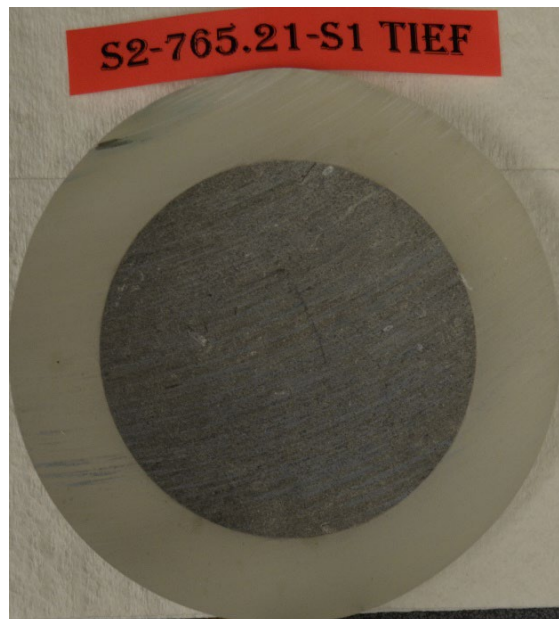
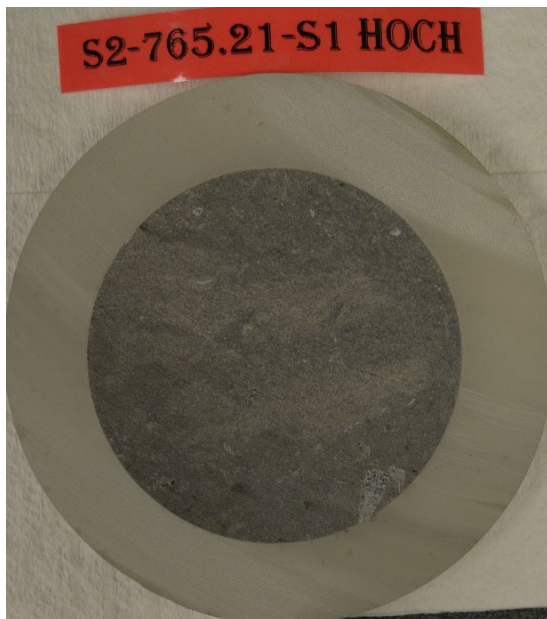
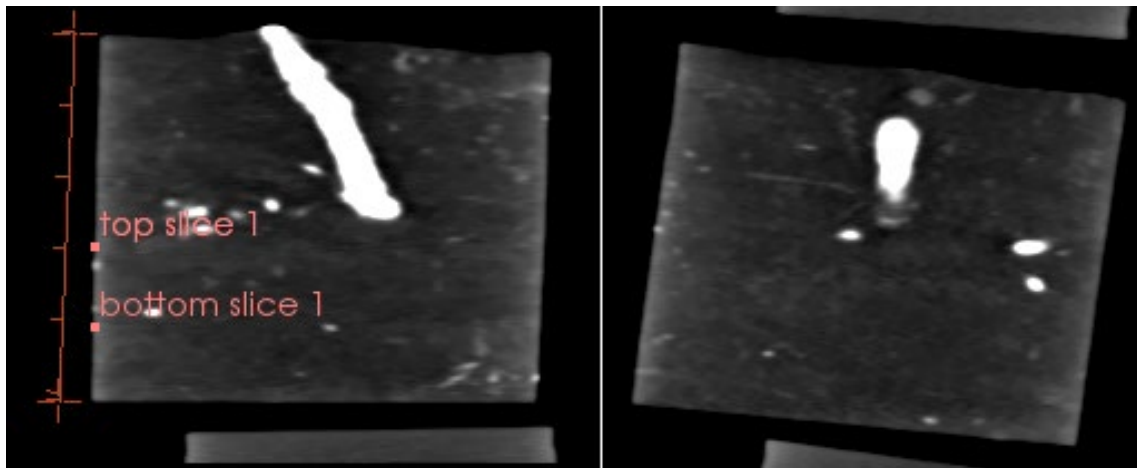
CT-based slice selection: Slice 2 was chosen due to fractures in the upper part of the sample

To be analysed: Slice 2



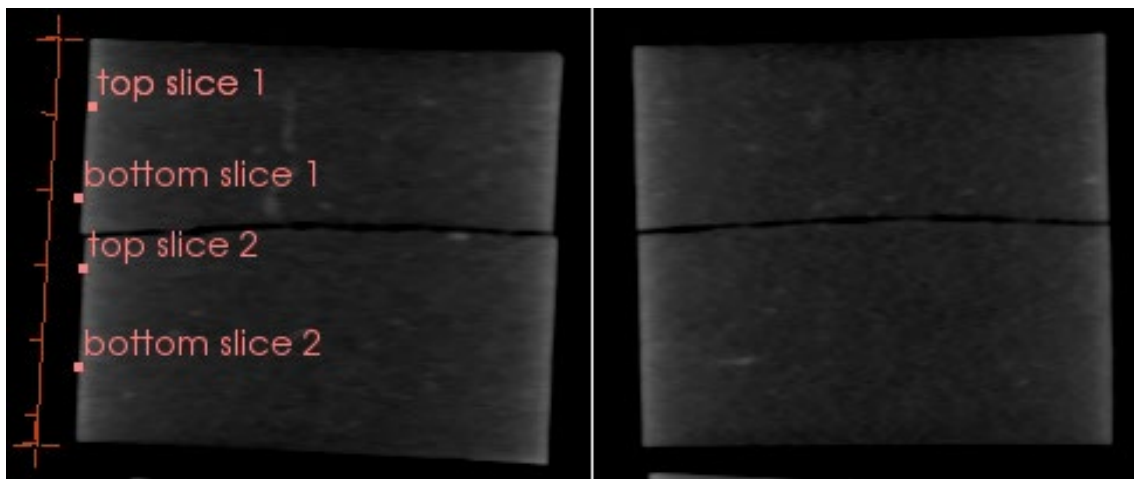
STA2-1-765.21-DI

Mean depth [m]: 765.21
Stratigraphic position: «Parkinsoni-Württembergica-Schichten» (Dogger)
CT-based slice selection: Only one slice was taken due to nodules.
To be analysed: Slice 1



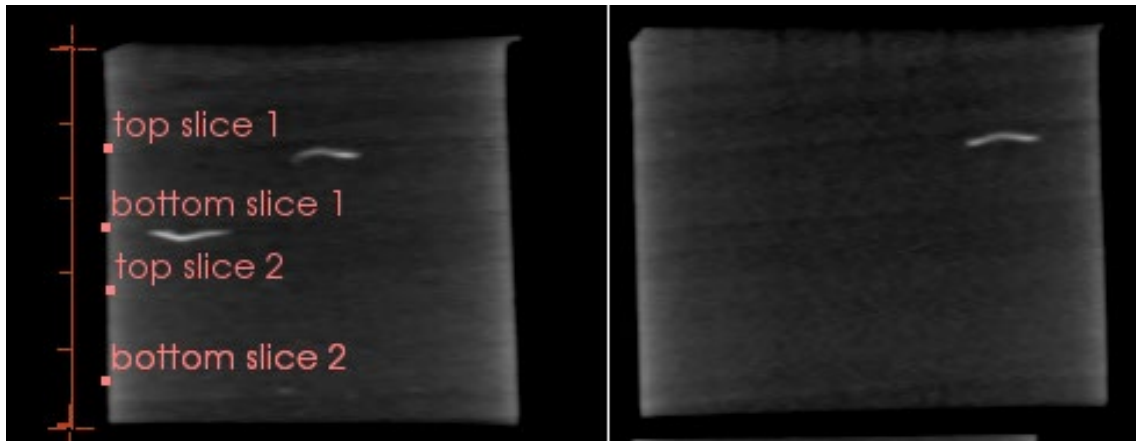
STA2-1-779.96-DI

Mean depth [m]: 779.96
Stratigraphic position: Wedelsandstein (Dogger)
CT-based slice selection: Slice 2 is more homogeneous.
To be analysed: Slice 2



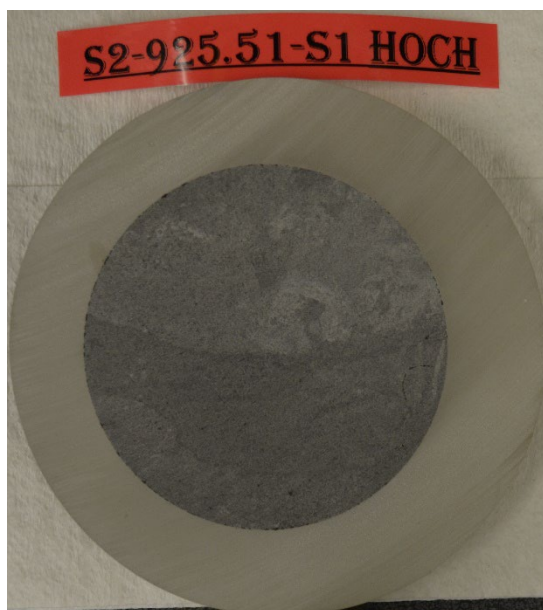
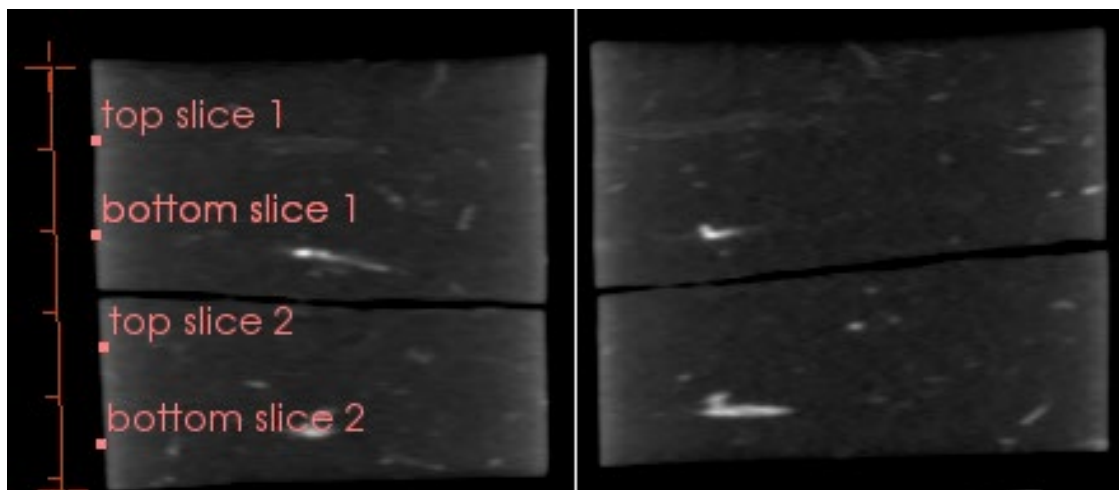
STA2-1-912.69-DI

Mean depth [m]: 912.69
Stratigraphic position: Staffelegg (Lias)
CT-based slice selection: Slice 2 is more homogeneous.
To be analysed: Slice 2



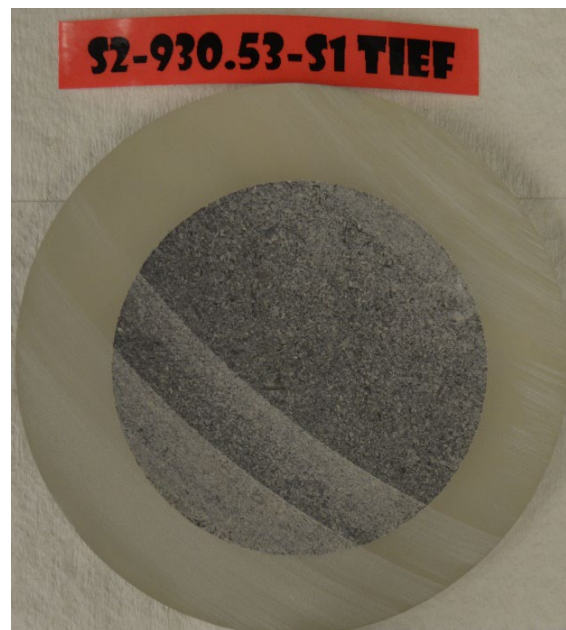
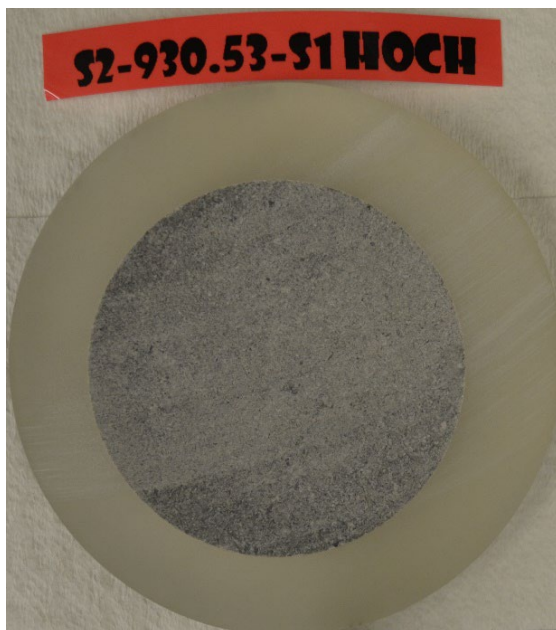
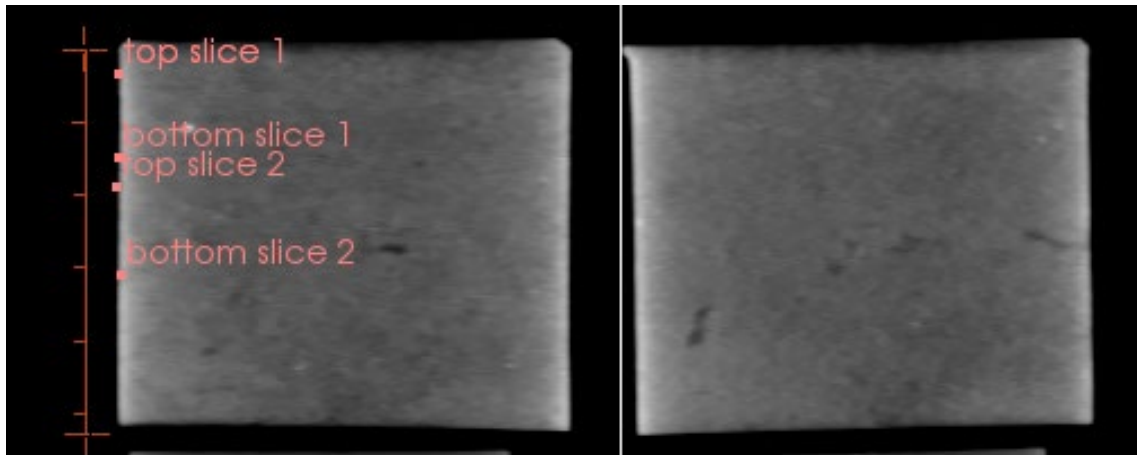
STA2-1-925.51-DI

Mean depth [m]: 925.51
Stratigraphic position: Staffelegg (Lias)
CT-based slice selection: Slice 1 is more homogeneous.
To be analysed: Slice 1



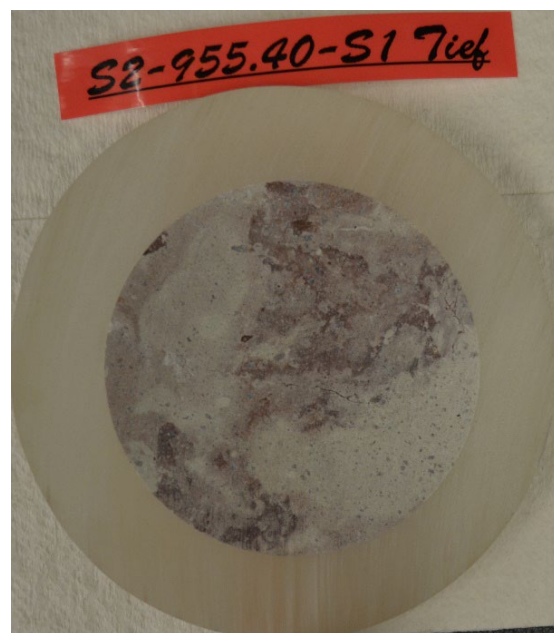
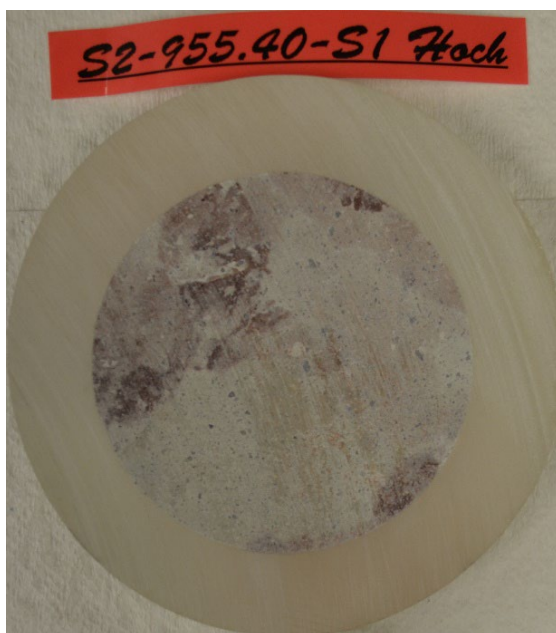
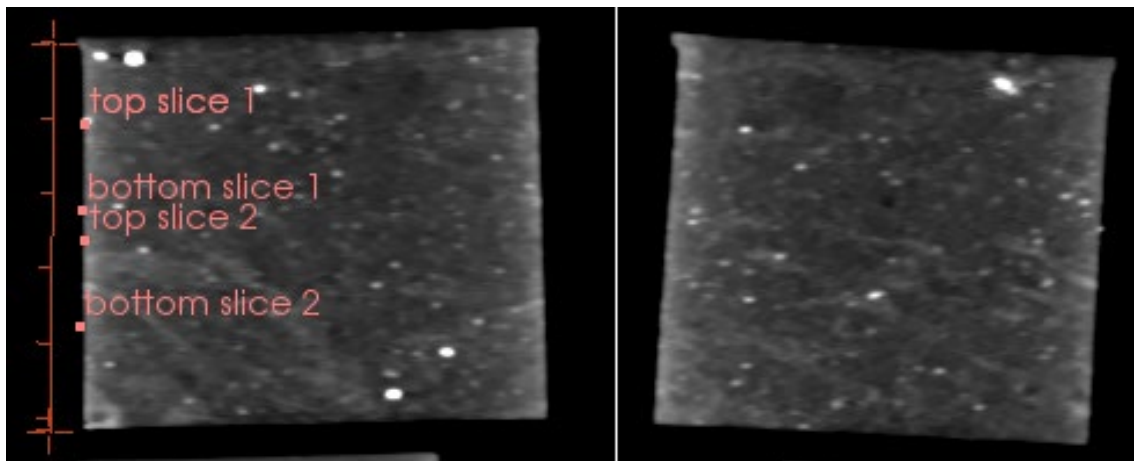
STA2-1-930.53-DI

Mean depth [m]: 930.53
Stratigraphic position: Staffelegg (Lias)
CT-based slice selection: Slice 1 is more homogeneous.
To be analysed: Slice 1



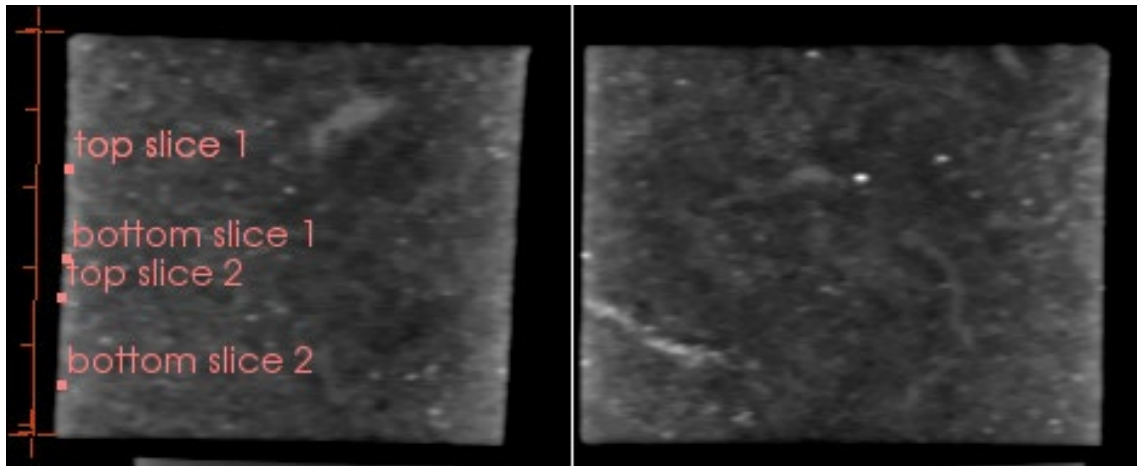
STA2-1-955.40-DI

Mean depth [m]: 955.40
Stratigraphic position: Klettgau (Keuper)
CT-based slice selection: Slice 1 is more homogeneous.
To be analysed: Slice 1



STA2-1-957.28-DI

Mean depth [m]: 957.28
Stratigraphic position: Klettgau (Keuper)
CT-based slice selection: Slice 1 is more homogeneous.
To be analysed: Slice 1



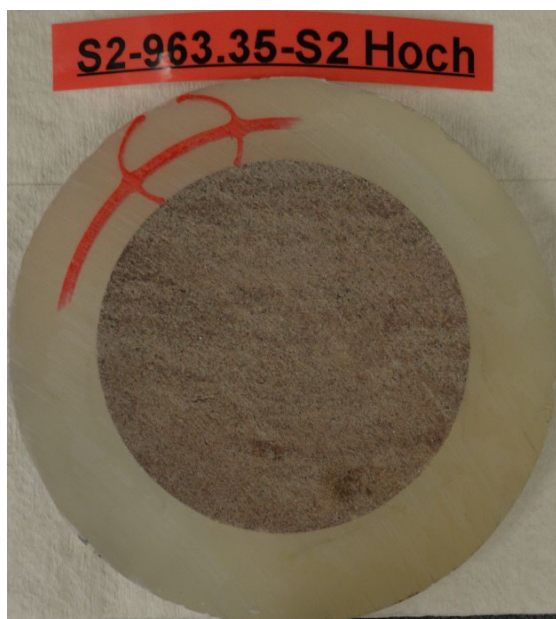
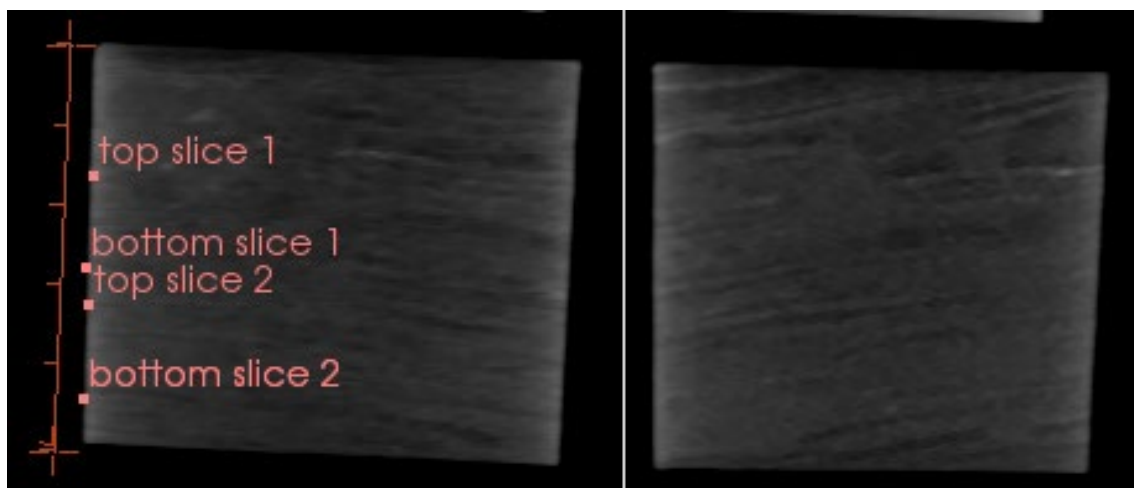
STA2-1-963.35-DI

Mean depth [m]: 963.35

Stratigraphic position: Klettgau (Keuper)

CT-based slice selection: Slice 2 is more homogeneous.

To be analysed: Slice 2



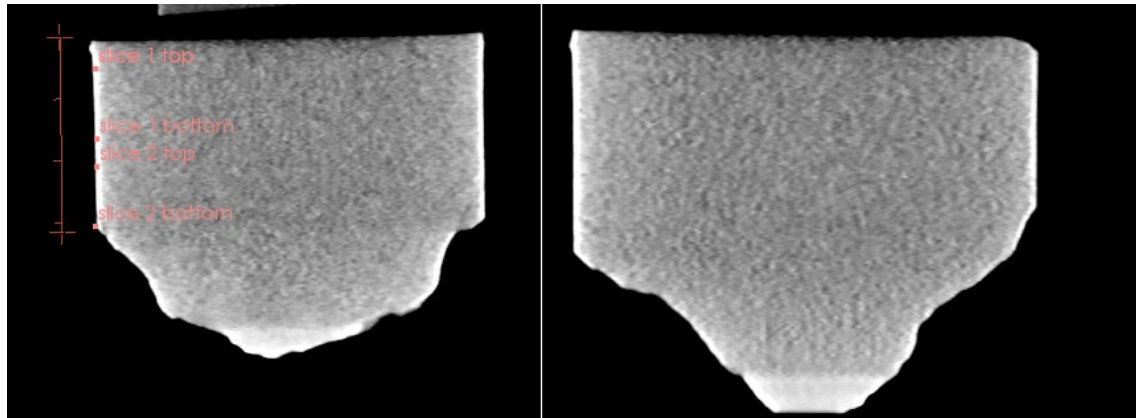
STA2-1-1126.66-DI

Mean depth [m]: 1126.66

Stratigraphic position: Zeglingen Fm. (Keuper)

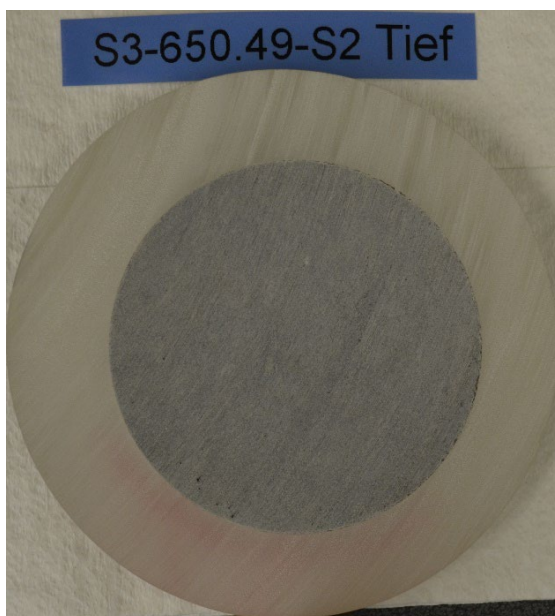
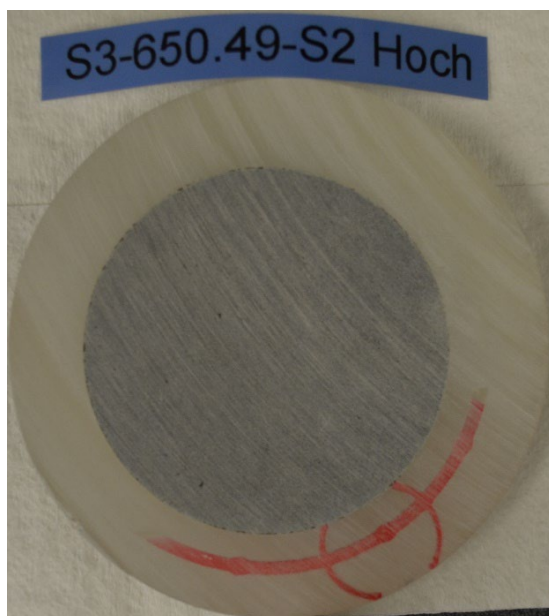
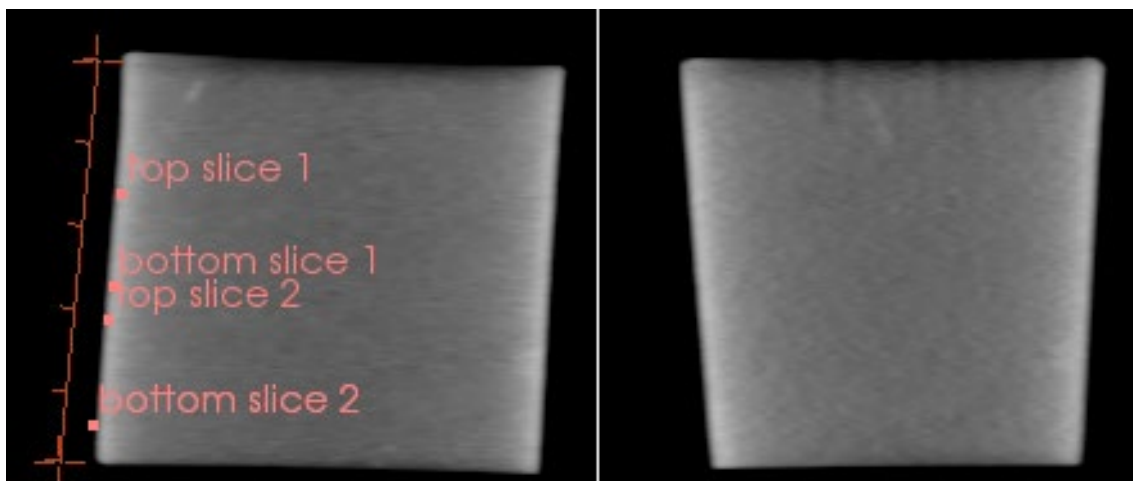
CT-based slice selection: Anhydrite with fine lines. Both slices are homogeonous but Slice 2 might be too thin.

To be analysed: Slice 1



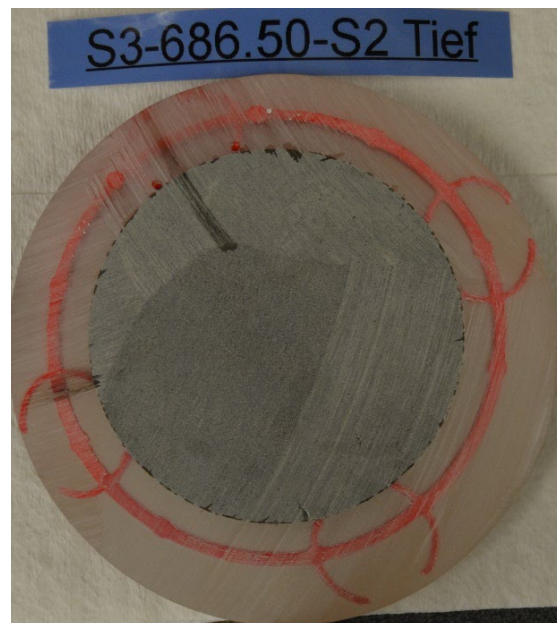
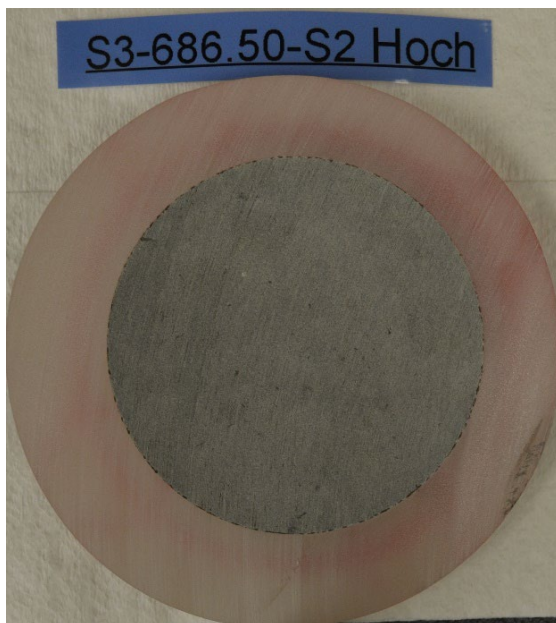
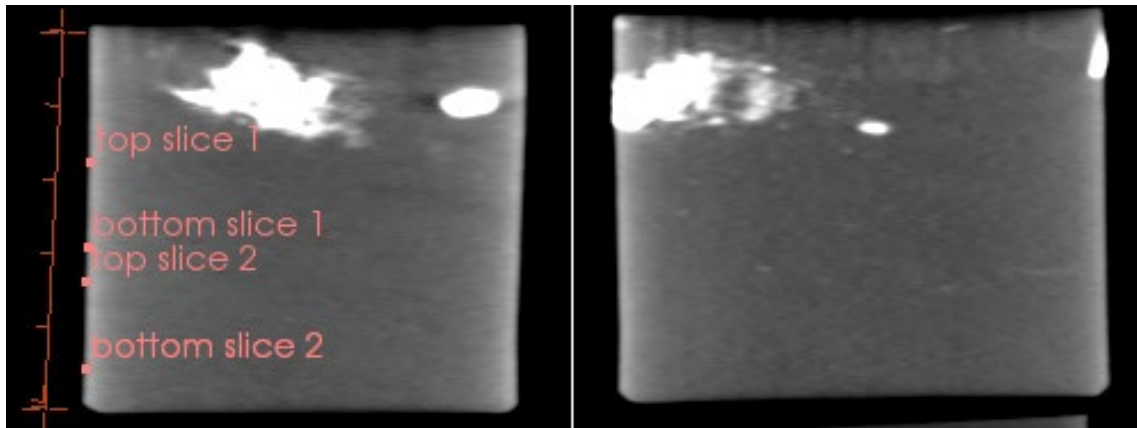
H.2.4 Stadel 3-1**STA3-1-650.49-DI**

Mean depth [m]: 650.49
Stratigraphic position: Wildegg Fm. (Malm)
CT-based slice selection: Slice 2 is more homogeneous.
To be analysed: Slice 2



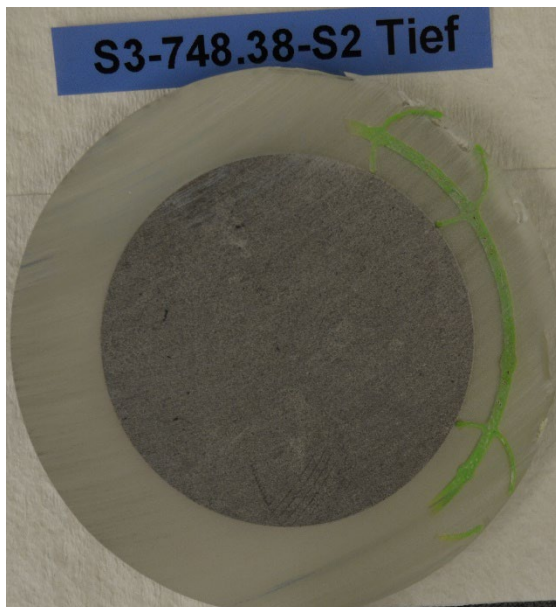
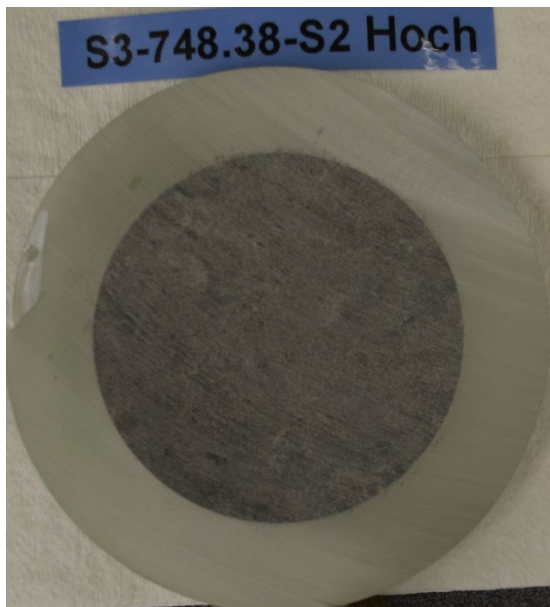
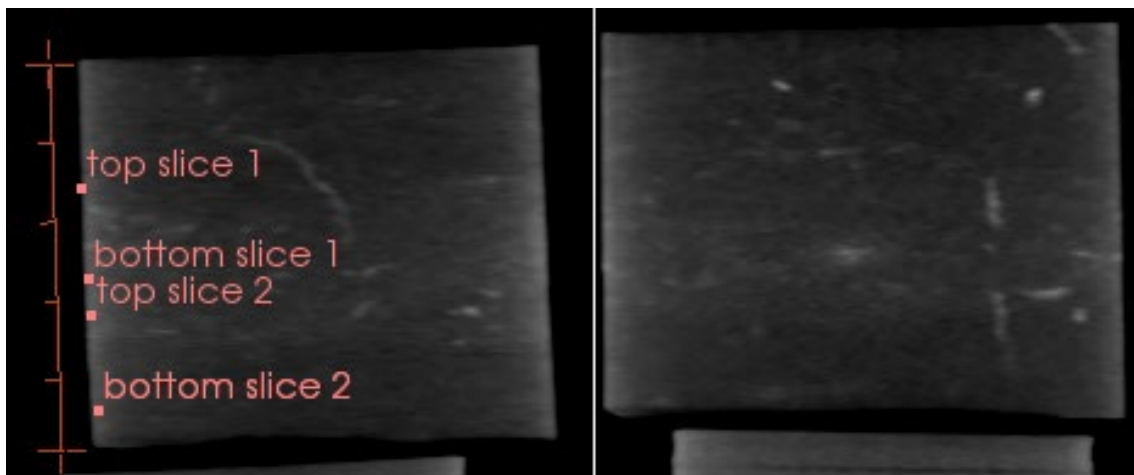
STA3-1-686.50-DI

Mean depth [m]: 686.50
Stratigraphic position: Wildegg Fm. (Malm)
CT-based slice selection: Slice 2 is more homogeneous.
To be analysed: Slice 2



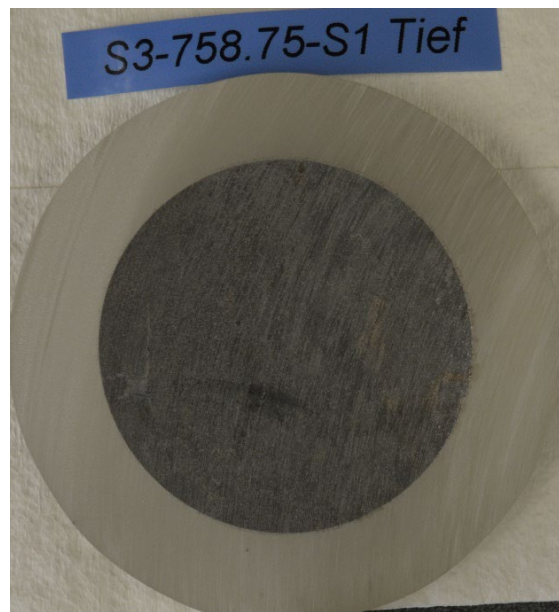
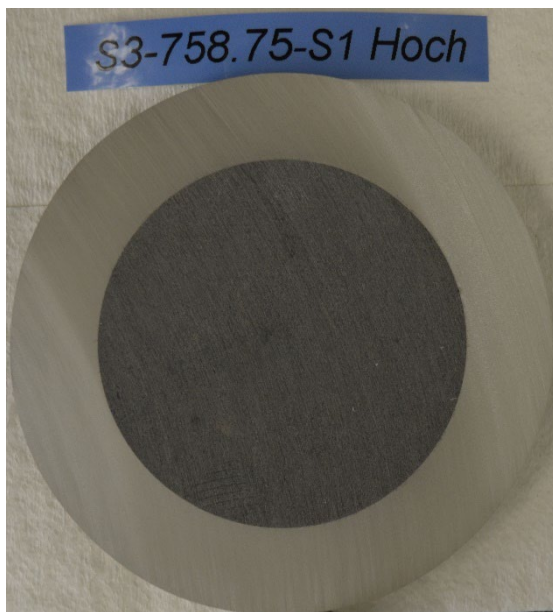
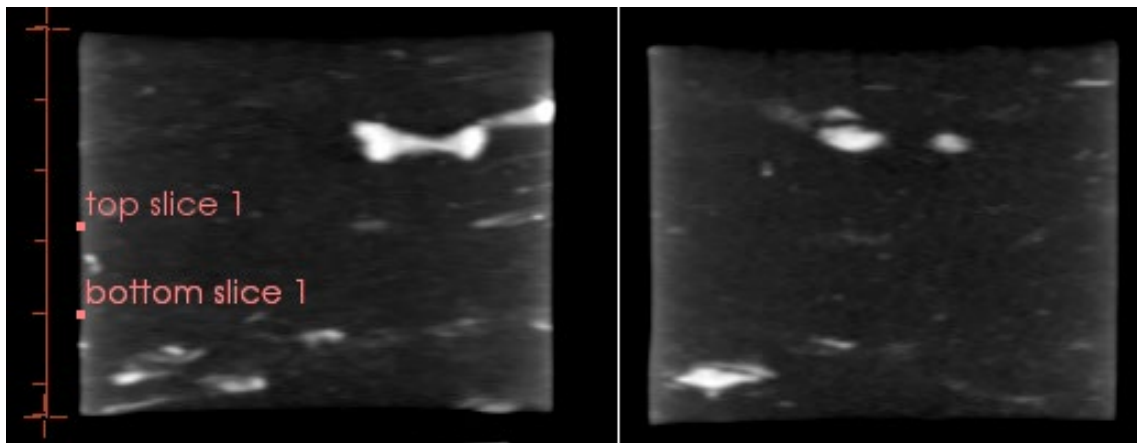
STA3-1-748.38-DI

Mean depth [m]: 748.38
Stratigraphic position: Wedelsandstein Fm. (Dogger)
CT-based slice selection: Slice 2 is more homogeneous.
To be analysed: Slice 2



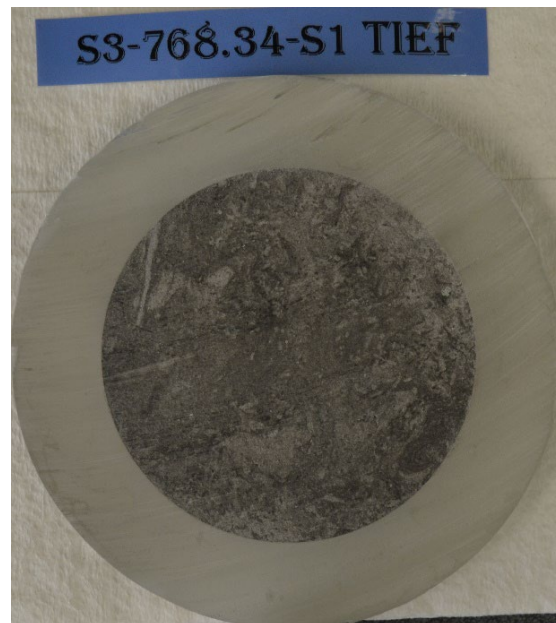
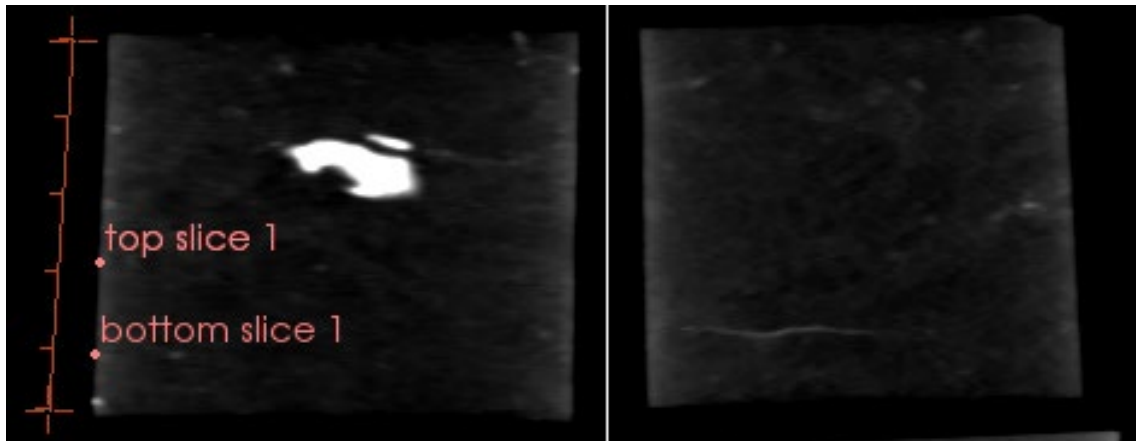
STA3-1-758.75-DI

Mean depth [m]: 758.75
Stratigraphic position: Wedelsandstein Fm. (Dogger)
CT-based slice selection: Only one slice was taken due to nodules and fractures.
To be analysed: Slice 1



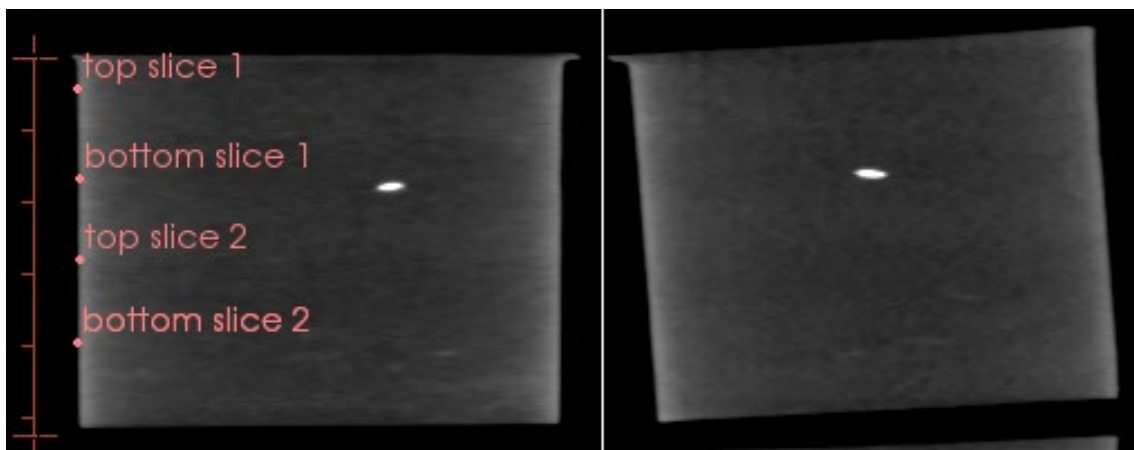
STA3-1-768.34-DI

Mean depth [m]: 768.34
Stratigraphic position: Murchisonae-Oolith Fm. (Dogger)
CT-based slice selection: Only one slice was taken due to nodules.
To be analysed: Slice 1



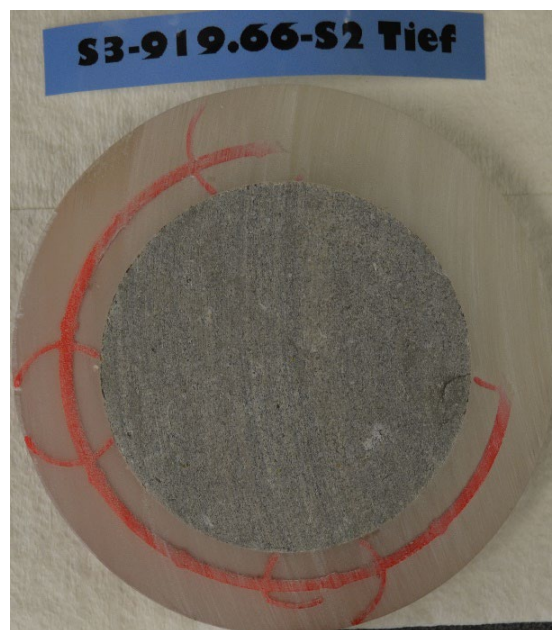
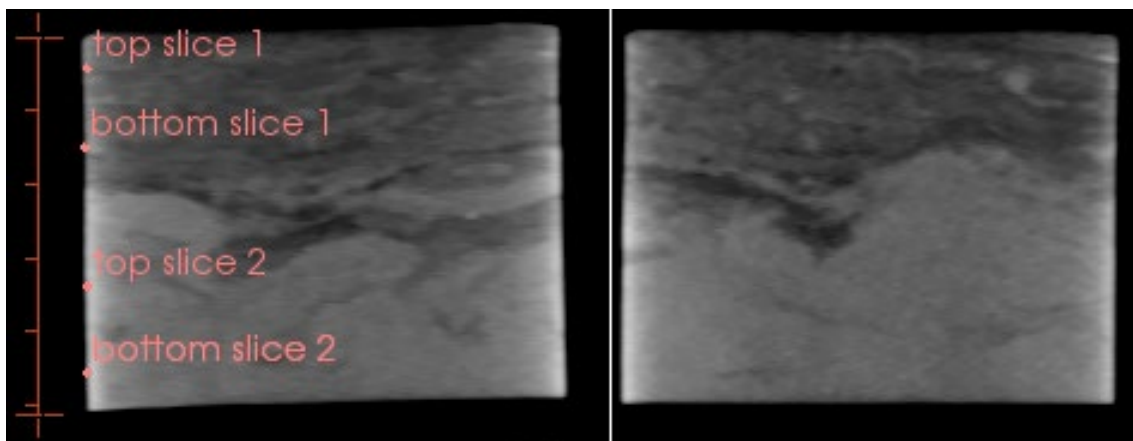
STA3-1-896.82-DI

Mean depth [m]: 896.82
Stratigraphic position: Staffelegg Fm. (Lias)
CT-based slice selection: Slice 2 is more homogeneous.
To be analysed: Slice 2



STA3-1-919.66-DI

Mean depth [m]: 919.66
Stratigraphic position: Staffelegg Fm. (Lias)
CT-based slice selection: Slice 2 is more homogeneous.
To be analysed: Slice 2



H.3 Samples from the Zürich Nordost siting region

This section documents the final step of the sampling campaign for diffusion experiments (DI) of the Marthalen and Trüllikon (MAR1-1 and TRU1-1, respectively) boreholes. In detail, it documents the positioning and selection of the final core slices, which were used for the experiments at PSI, from the subcored and epoxy-embedded samples. The axis of the subcores is parallel to that of the original full core, i.e. normal to bedding. The positioning, selection and description of the slices is mainly based on CT scans performed at the Institute of Forensic Medicine at the University of Bern. While scans of the whole DI samples were already available, new scans were acquired on the subcored samples and served as a basis for the positioning of the DI slices. On the one hand, these scans allow the assessment of sample homogeneity (or heterogeneity) and, thus, aid the selection of slices such that they are as representative as possible. Moreover, these scans make it possible to identify features such as cracks or fractures (either natural or artificially induced), large pyrite concretions, siderite lenses, macropores (i.e. in calcareous lithologies), stylolites or burrows, which could affect the experiments. Such features were avoided.

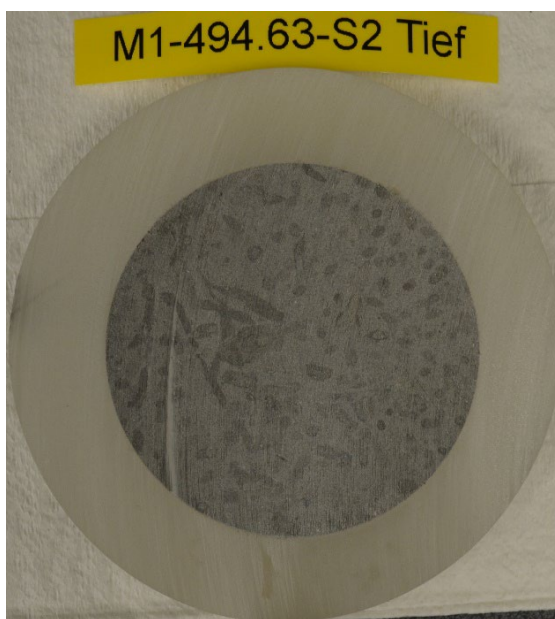
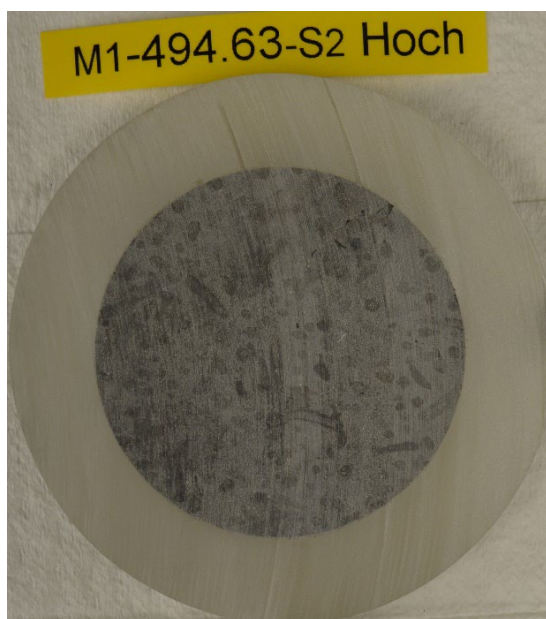
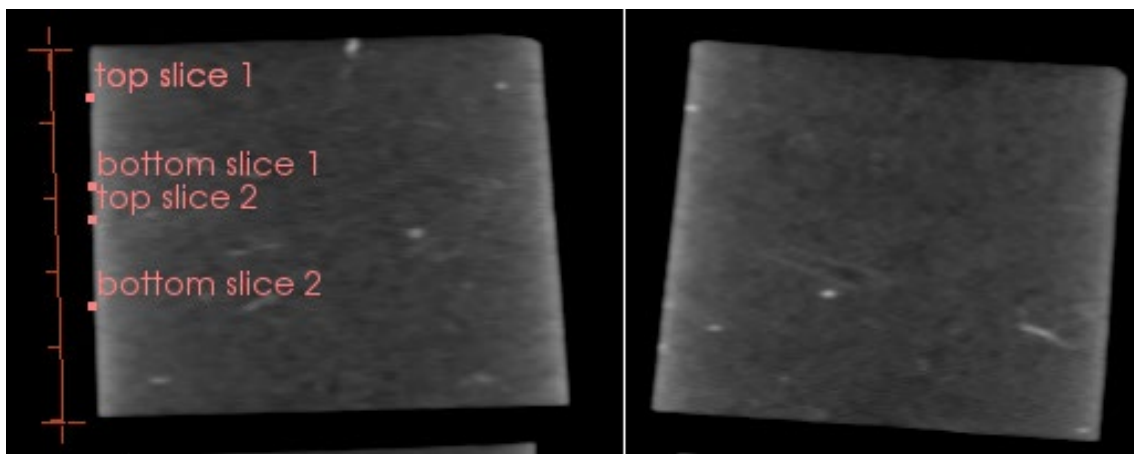
In general, 1 to 2 DI slices were collected per sample, of which one slice is selected for the experiments at PSI. The slice selection is documented in various CT images, generally arranged as image pairs; one on the left side and one on the right side. Such an image pair illustrates two sections of the subcores parallel to the core axis, which are oriented orthogonally with respect to each other. The position of the DI slices within the subcored samples (i.e. top and bottom of slices) is shown in the CT image on the left-hand side, whereas special features relevant to sampling may be illustrated in the second CT image on the right-hand side.

Tab. H-3: Depth information for all samples from Marthalen and Trüllikon used for diffusion experiments. The values in grey text are samples for which no diffusion data are available.

MAR1-1	TRU1-1
Sample mean depth [m]	
494.63	620.33
566.33	660.04
570.12	710.95
588.40	728.55
712.65	735.62
717.76	748.98
729.30	750.88
738.21	756.39
742.25	768.29
862.35	778.32
	788.76
	794.56
	802.48
	807.74
	814.82
	815.28
	831.37
	839.97
	870.23
	882.36
	890.39
	902.85
	913.07
	923.34
	930.99
	938.90
	943.21
	955.02
	961.72
	1,026.55

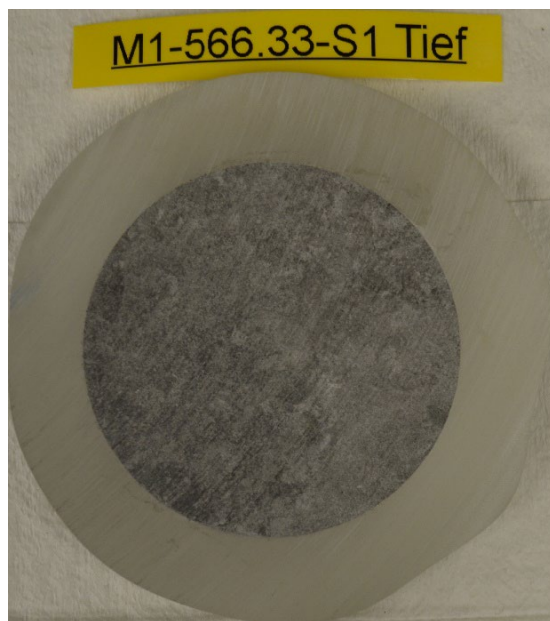
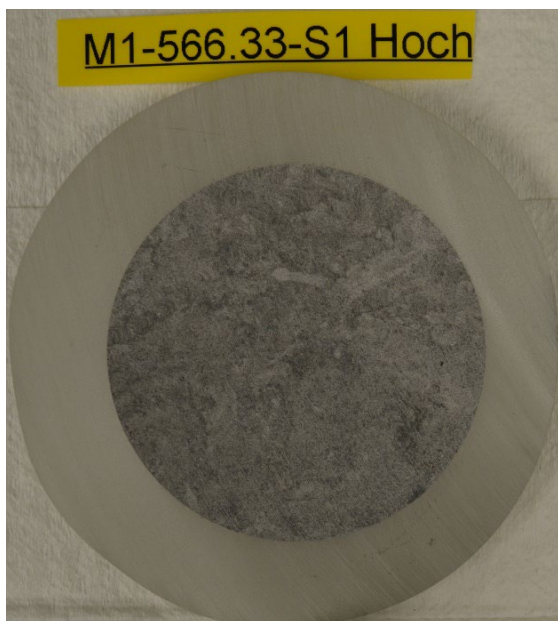
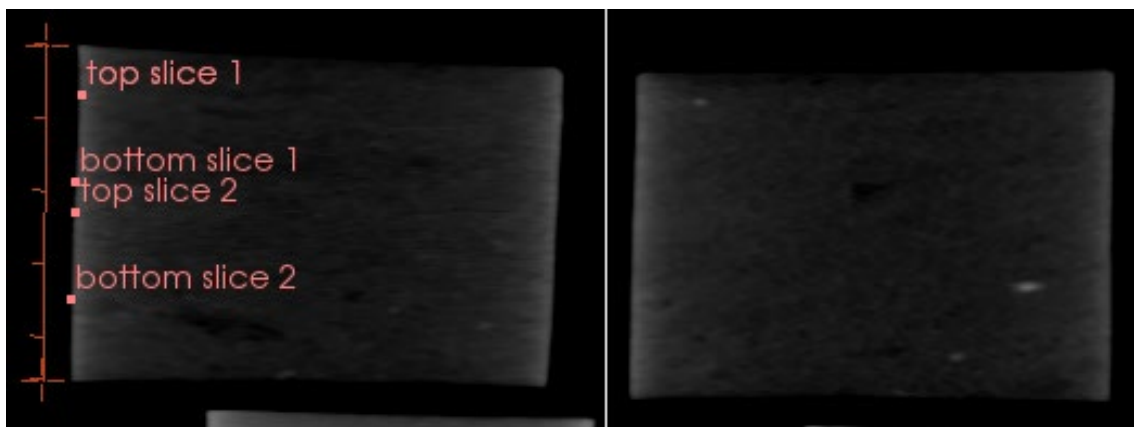
H.3.1 Marthalen 1-1**MAR1-1-494.63-DI**

Mean depth [m]: 494.63
Stratigraphic position: Wildegg Fm. (Malm)
CT-based slice selection: Slice 2 is more homogeneous.
To be analysed: Slice 2



MAR1-1-566.33-DI

Mean depth [m]: 566.33
Stratigraphic position: Wedelsandstein Fm. (Dogger)
CT-based slice selection: Slice 1 is slightly more homogeneous.
To be analysed: Slice 1



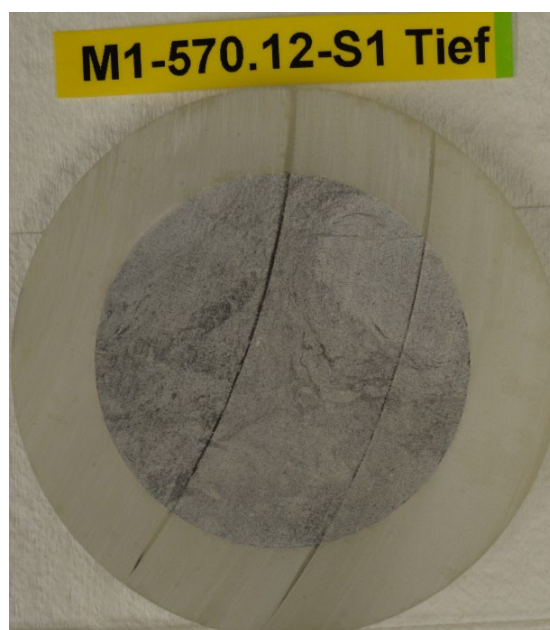
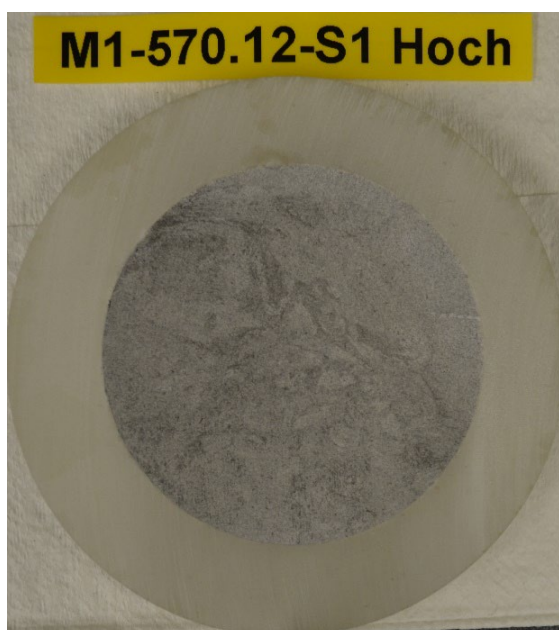
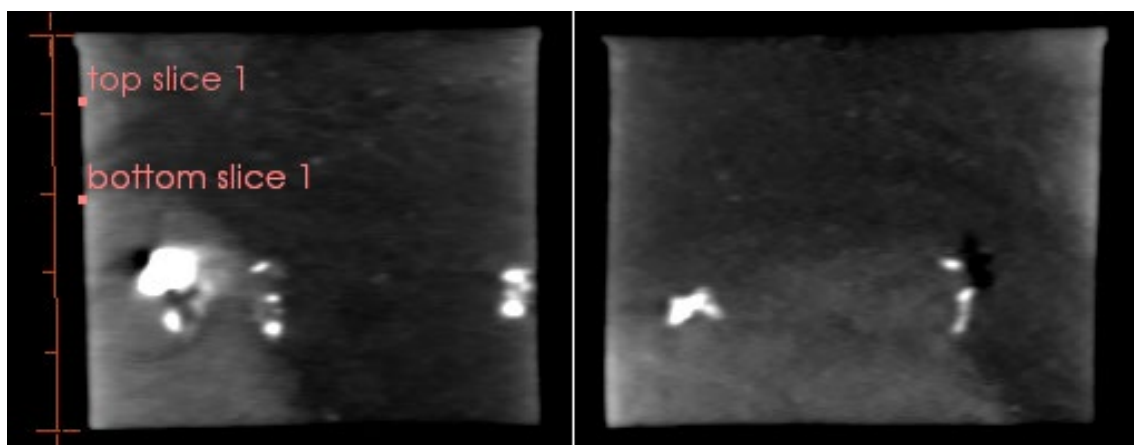
MAR1-1-570.12-DI

Mean depth [m]: 570.12

Stratigraphic position: Wedelsandstein Fm. (Dogger)

CT-based slice selection: Only one slice was chosen due to inclusions.

To be analysed: Slice 1



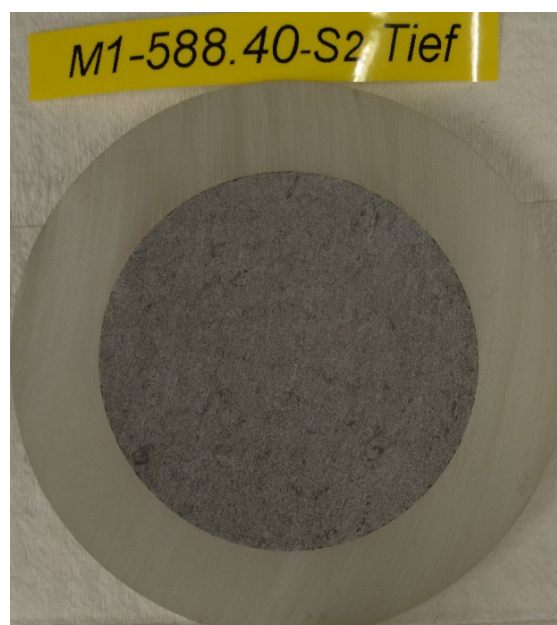
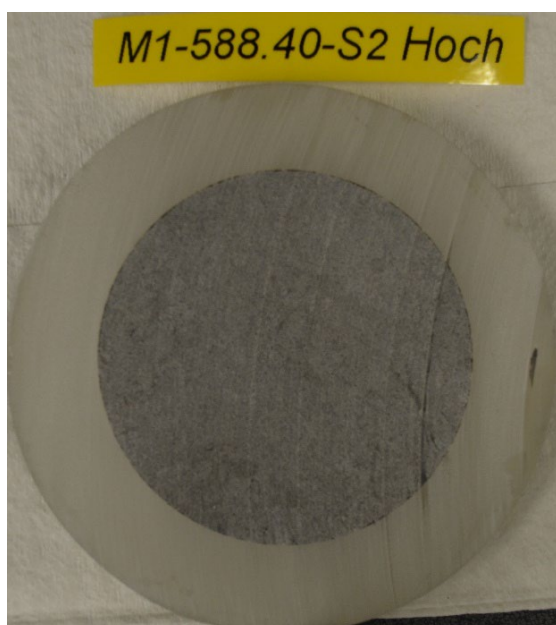
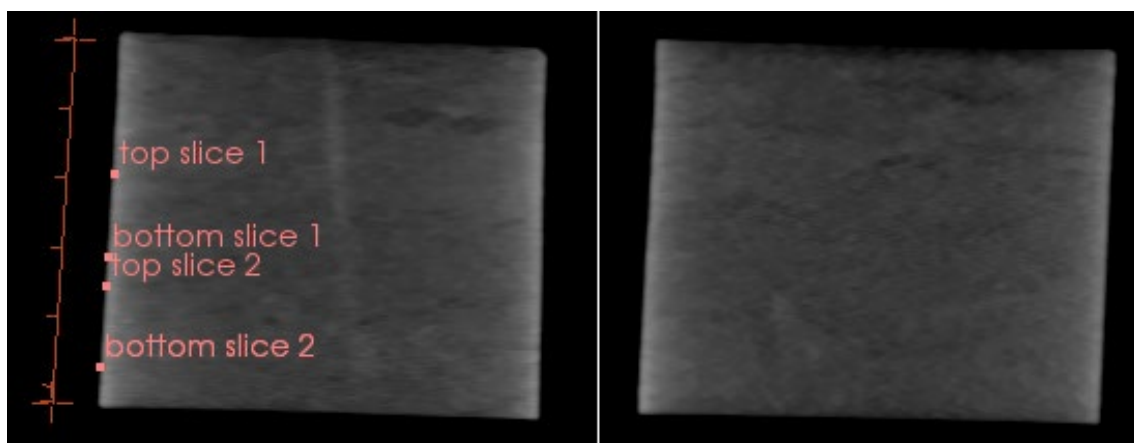
MAR1-1-588.40-DI

Mean depth [m]: 588.40

Stratigraphic position: Wedelsandstein Fm. (Dogger)

CT-based slice selection: Both slices are good but Slice 2 is slightly more homogeneous.

To be analysed: Slice 2



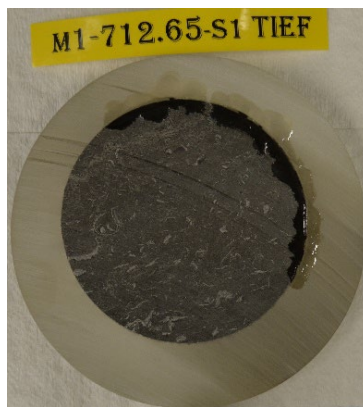
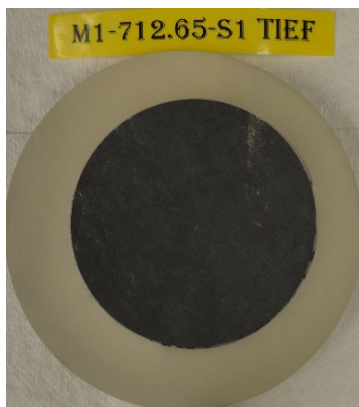
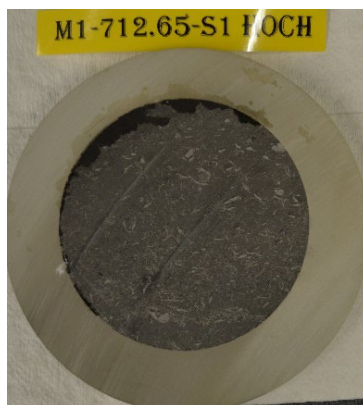
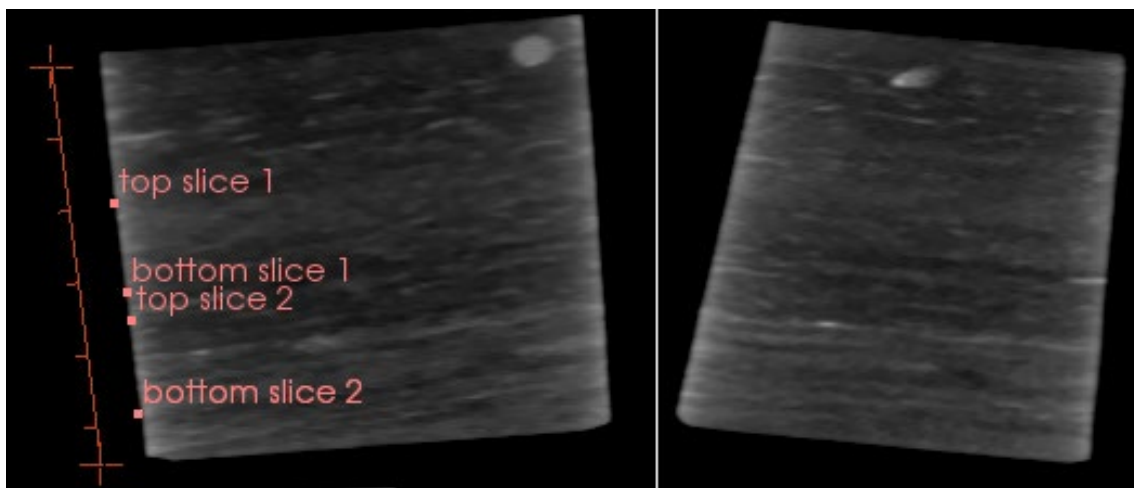
MAR1-1-712.65-DI

Mean depth [m]: 712.65

Stratigraphic position: Staffelegg Fm. (Lias)

CT-based slice selection: Both slices are good but Slice 1 is slightly more homogeneous.

To be analysed: Slice 1



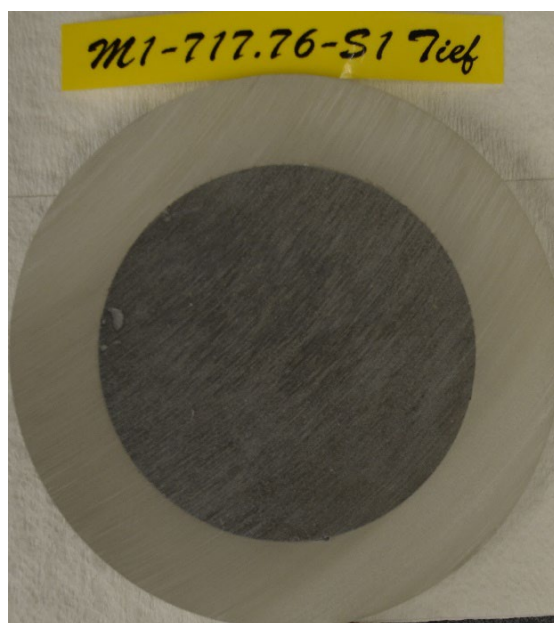
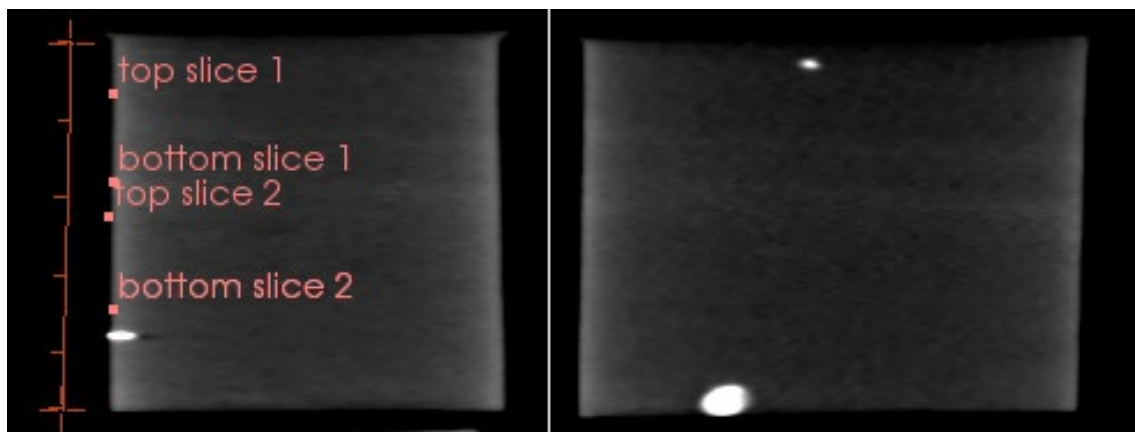
MAR1-1-717.76-DI

Mean depth [m]: 717.76

Stratigraphic position: Staffelegg Fm. (Lias)

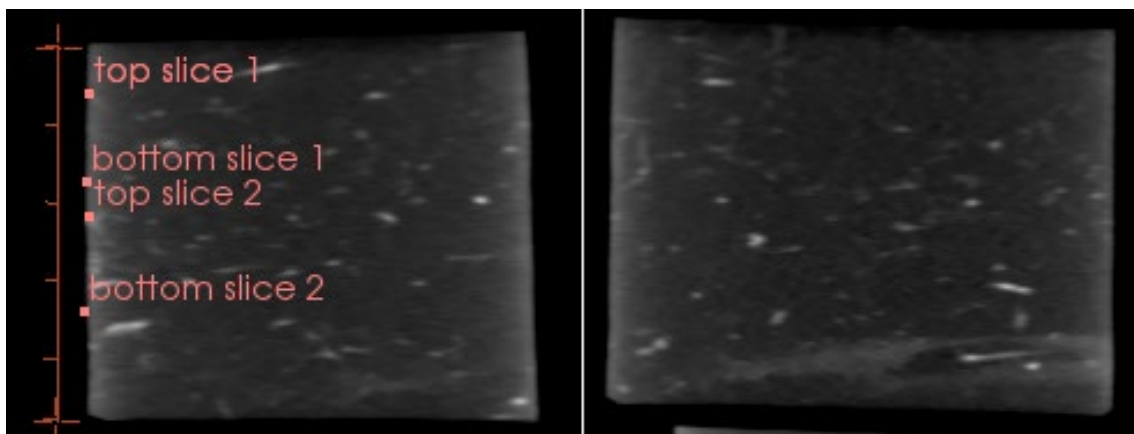
CT-based slice selection: Both slices are good but Slice 1 is slightly more homogeneous.

To be analysed: Slice 1



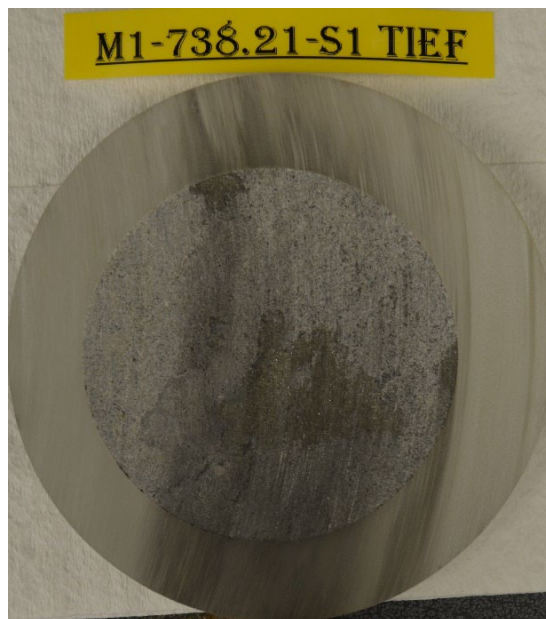
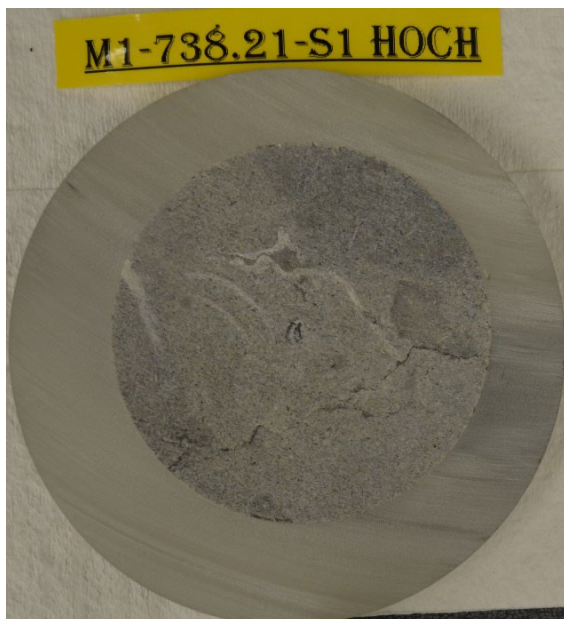
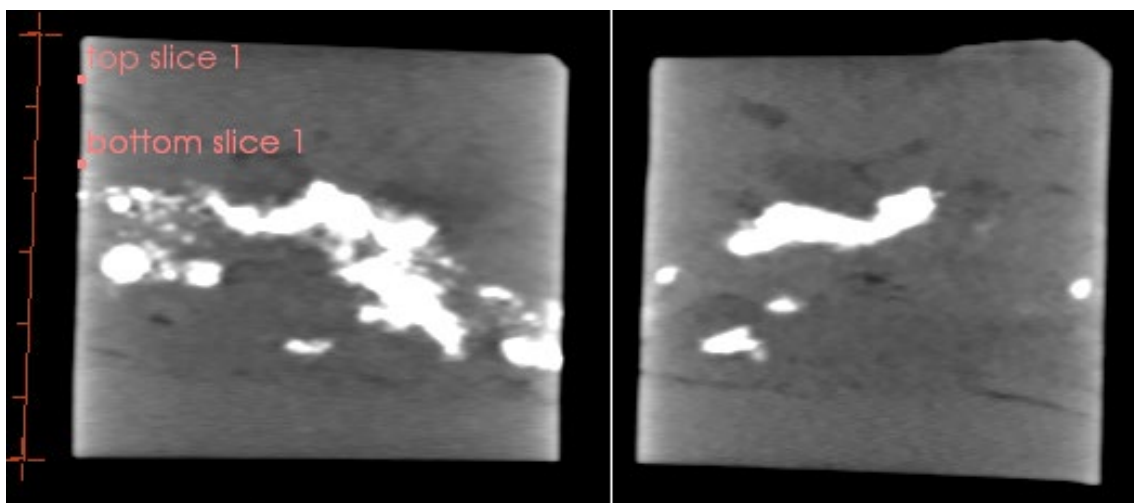
MAR1-1-729.30-DI

Mean depth [m]: 729.30
Stratigraphic position: Staffelegg Fm. (Lias)
CT-based slice selection: Slice 1 is more homogeneous.
To be analysed: Slice 1



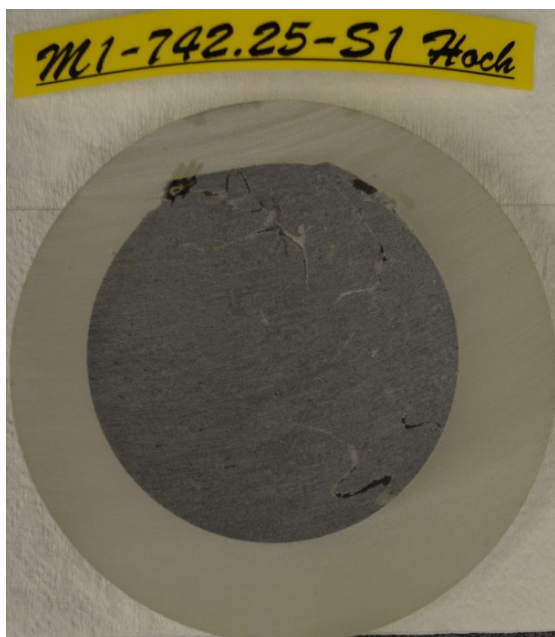
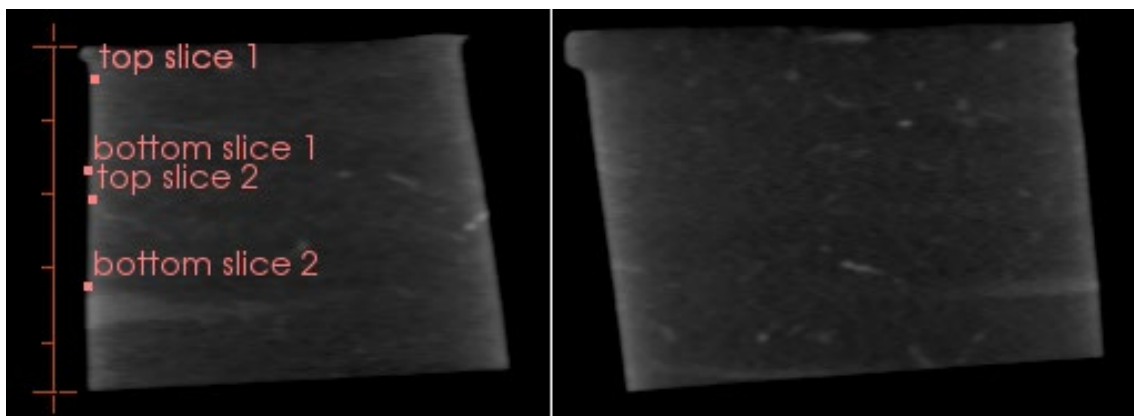
MAR1-1-738.21-DI

Mean depth [m]: 738.21
Stratigraphic position: Staffelegg Fm. (Lias)
CT-based slice selection: Only one slice was taken due to nodules.
To be analysed: Slice 1



MAR1-1-742.25-DI

Mean depth [m]: 742.25
Stratigraphic position: Staffelegg Fm. (Lias)
CT-based slice selection: Slice 1 is more homogeneous.
To be analysed: Slice 1



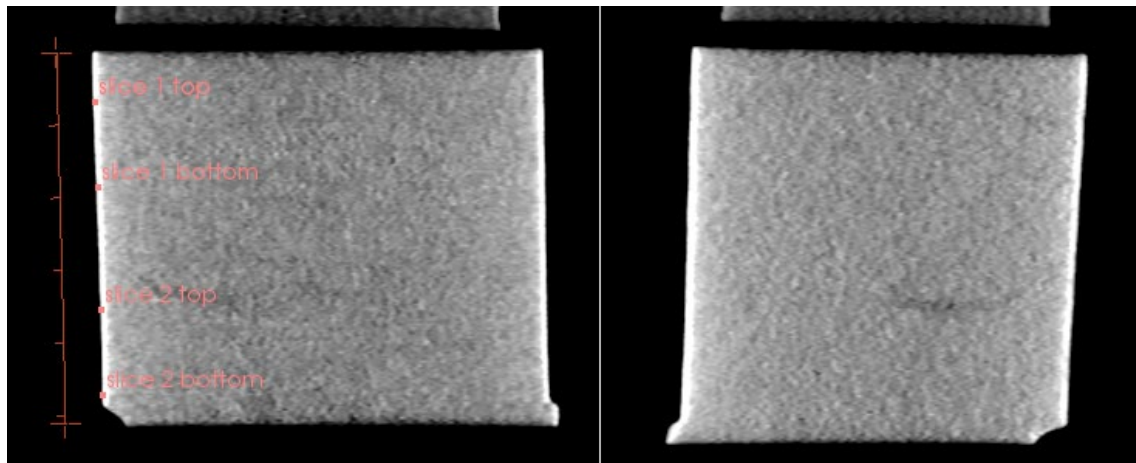
MAR1-1-862.35-DI

Mean depth [m]: 862.35

Stratigraphic position: Bänkerjoch Fm. (Keuper)

CT-based slice selection: Massive anhydrite with a fine structure or clay band at the top of Slice 2. Therefore Slice 1 is more homogeneous.

To be analysed: Slice 1



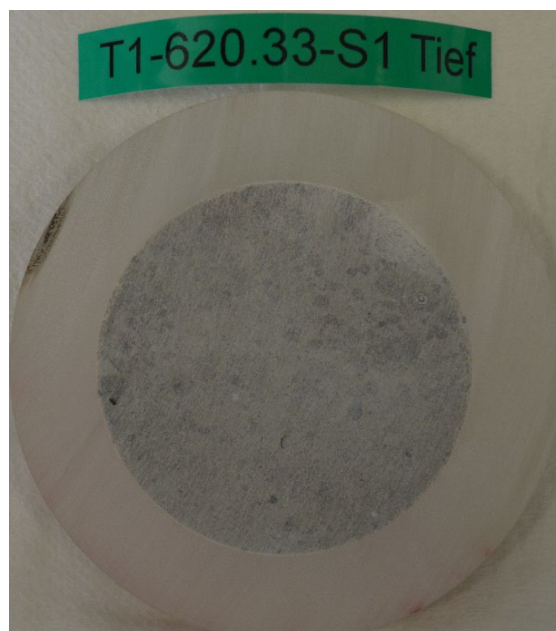
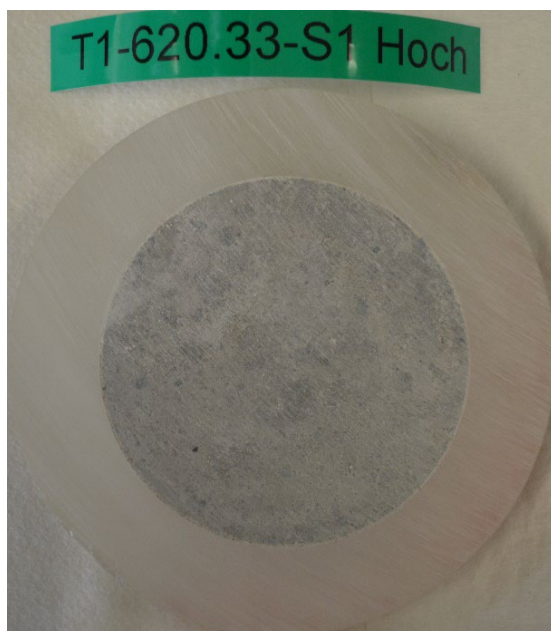
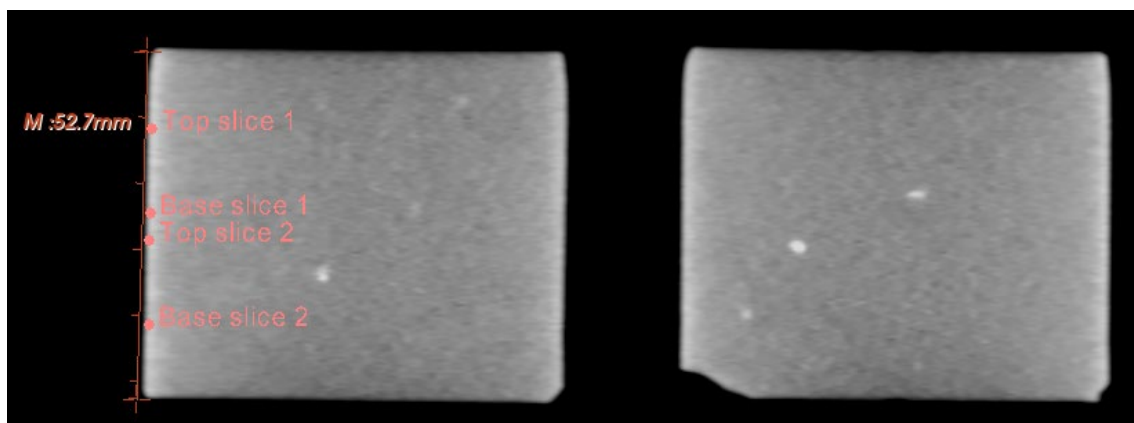
H.3.2 Trüllikon 1-1**TRU1-1-620.33-OD**

Mean depth [m]: 620.33

Stratigraphic position: Felsenkalke (Malm)

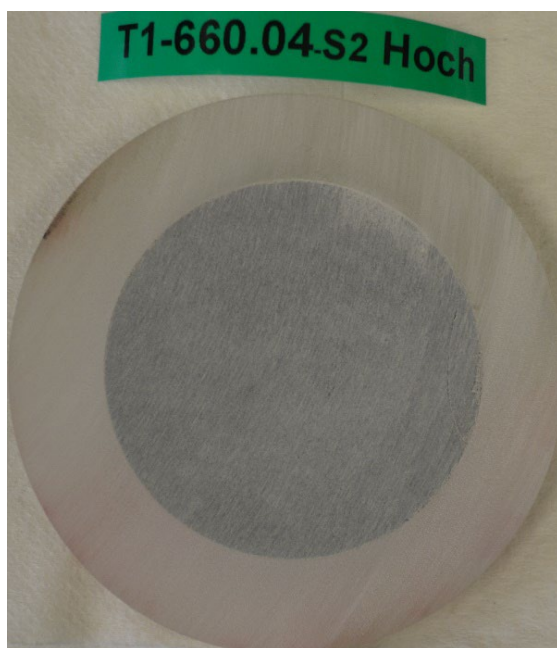
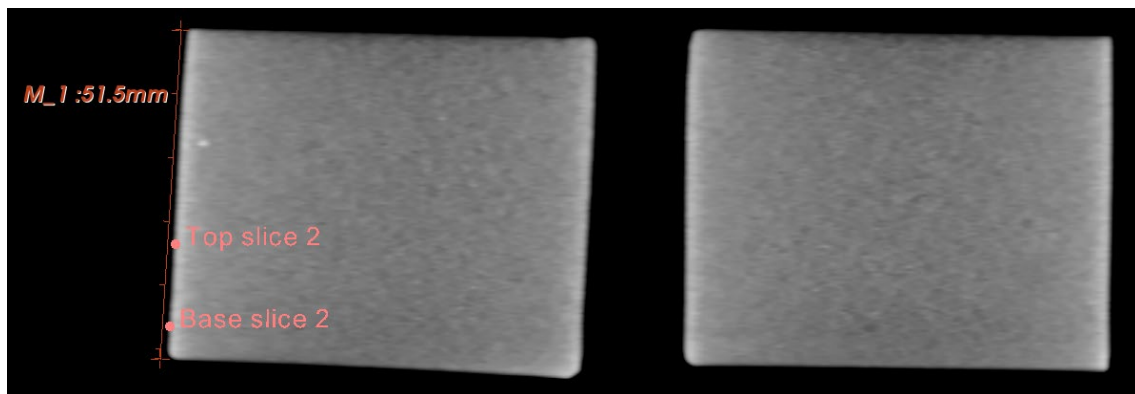
CT-based slice selection: Sample is bioclastic and slightly bioturbated. It contains few small pyrite concretions. Two slices were sampled. Slice 2 may contain more pyrite concretions than Slice 1.

To be analysed: Slice 1



TRU1-1-660.04-OD

Mean depth [m]: 660.04
Stratigraphic position: Villigen Fm. (Malm)
CT-based slice selection: Homogeneous sample
To be analysed: Slice 2



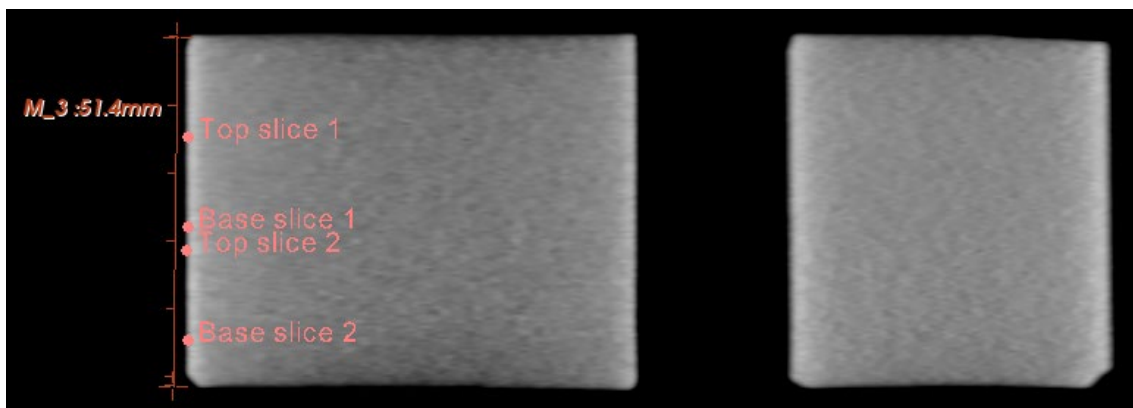
TRU1-1-710.95-OD

Mean depth [m]: 710.95

Stratigraphic position: Hornbuck Fm. (Malm)

CT-based slice selection: Minimally bioturbated sample. Two slices were collected. Slice 2 contains slightly less burrows and appears more representative for the overall rather homogeneous whole core sample.

To be analysed: Slice 2



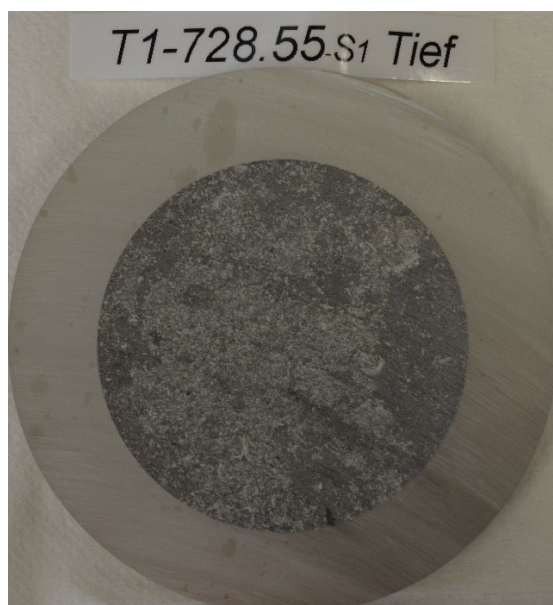
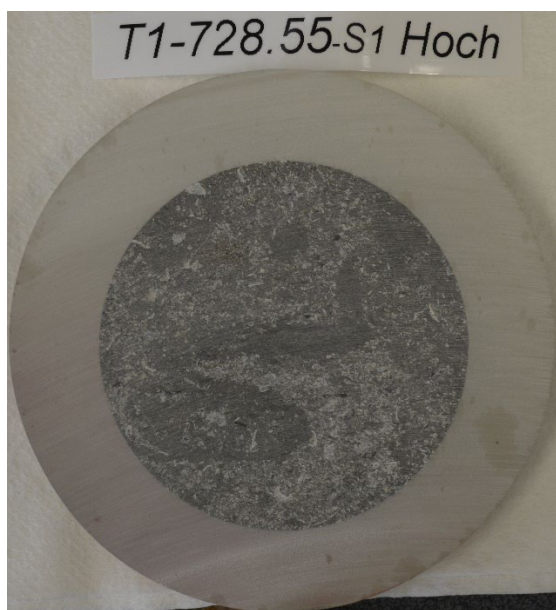
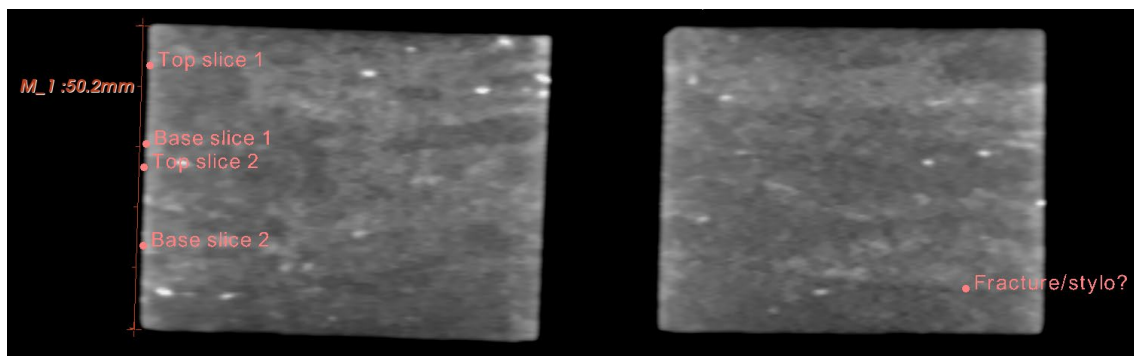
TRU1-1-728.55-DI

Mean depth [m]: 728.55

Stratigraphic position: Varians Fm ('Brauner Dogger')

CT-based slice selection: Heterogeneous, bioclastic sample with nodular texture. It contains numerous, small pyrite concretions and burrows. Two samples were collected.

To be analysed: Slice 1



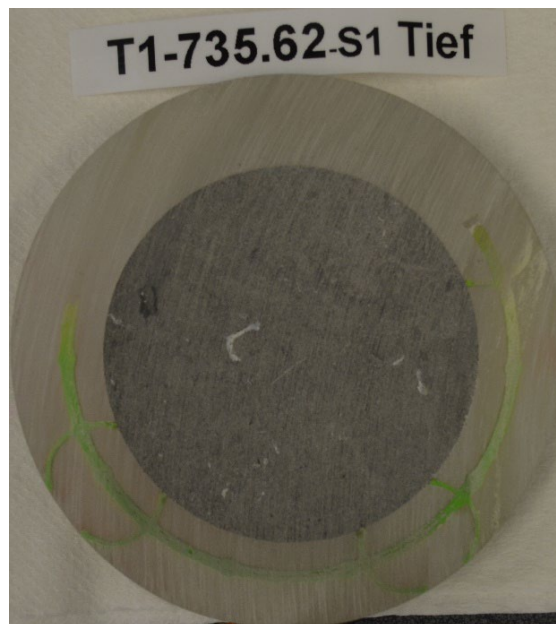
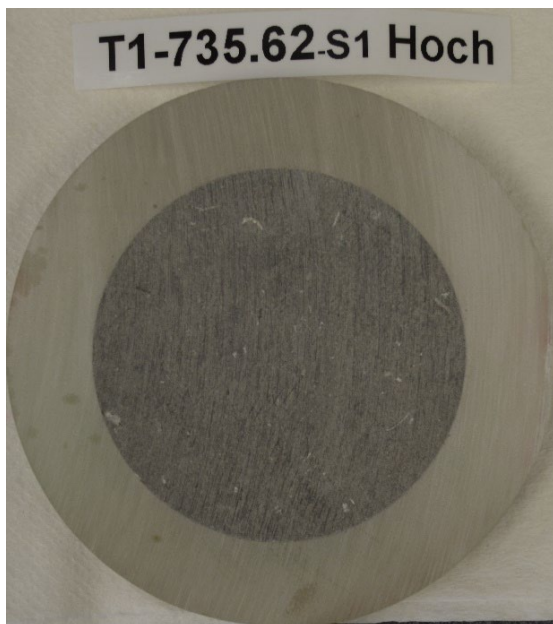
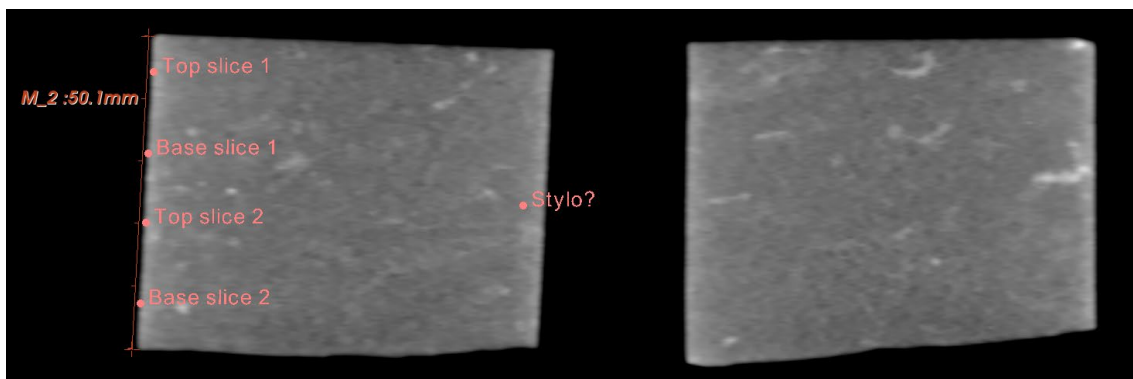
TRU1-1-735.62-DI

Mean depth [m]: 735.62

Stratigraphic position: «Parkinsoni-Württembergica-Schichten» ('Brauner Dogger')

CT-based slice selection: Heterogeneous, bioclastic sample, which contains some burrows and a diagenetic stylolite. Two slices were collected.

To be analysed: Slice 1



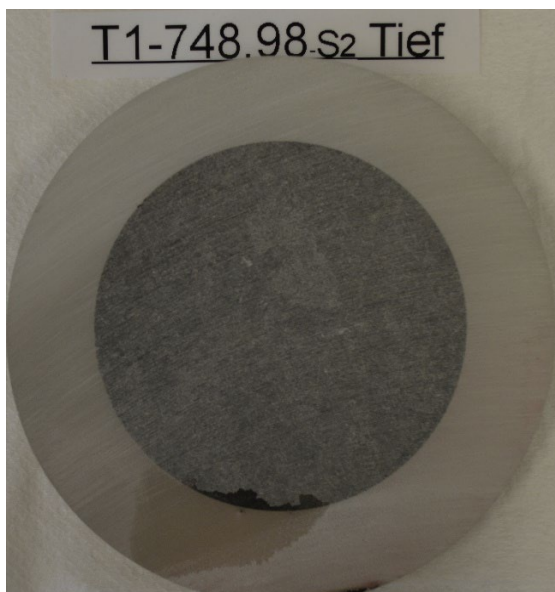
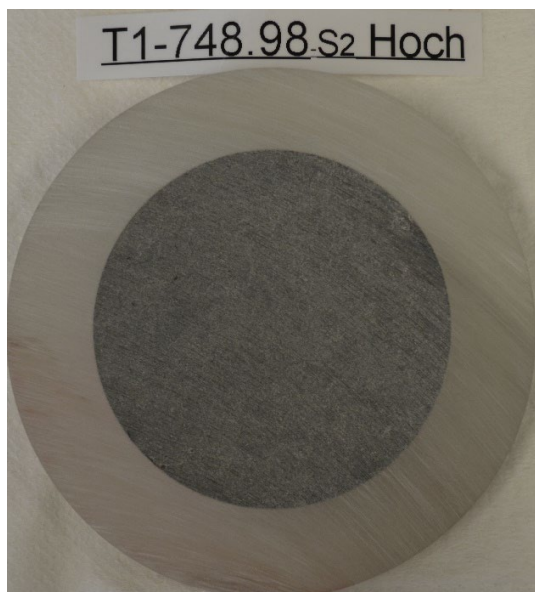
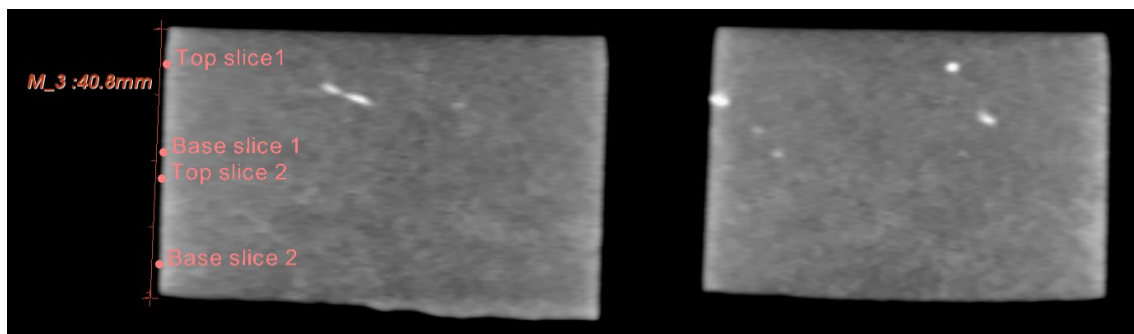
TRU1-1-748.98-DI

Mean depth [m]: 748.98

Stratigraphic position: «Parkinsoni-Württembergica-Schichten» ('Brauner Dogger')

CT-based slice selection: Overall rather homogeneous sample. Two slices were collected. Slice 1 contains some small pyrite concretions and a pyritised burrow.

To be analysed: Slice 2



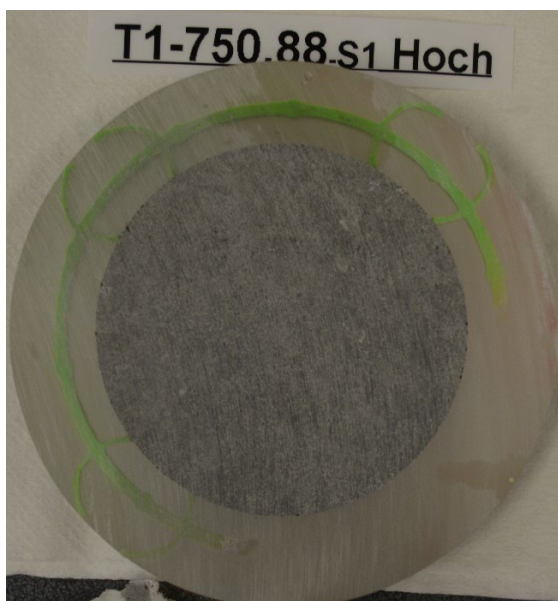
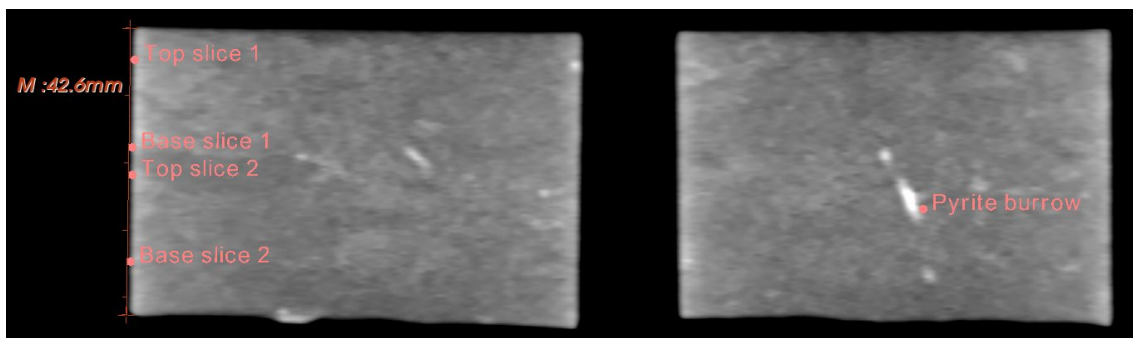
TRU1-1-750.88-DI

Mean depth [m]: 750.88

Stratigraphic position: «Parkinsoni-Württembergica-Schichten» ('Brauner Dogger')

CT-based slice selection: Heterogeneous, bioturbated sample. Two slices were collected. Slice 2 contains a pyritised burrow.

To be analysed: Slice 1



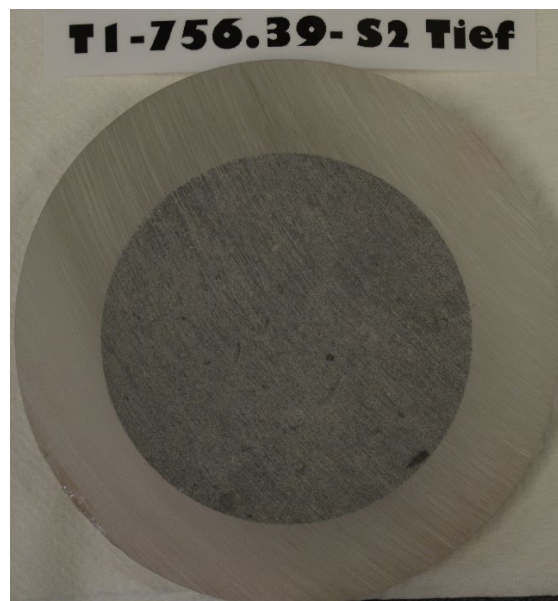
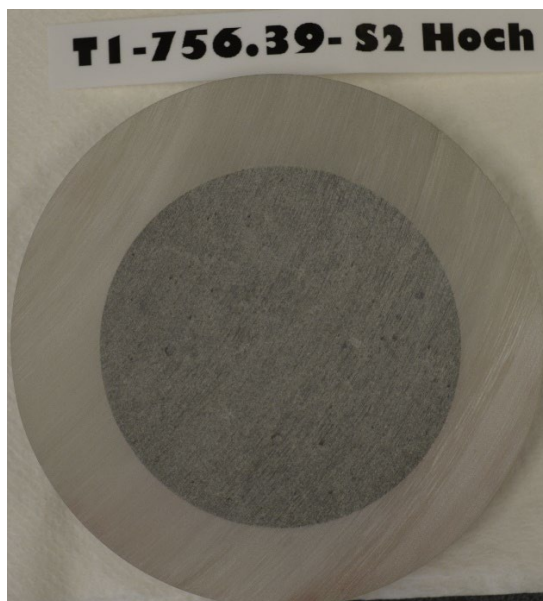
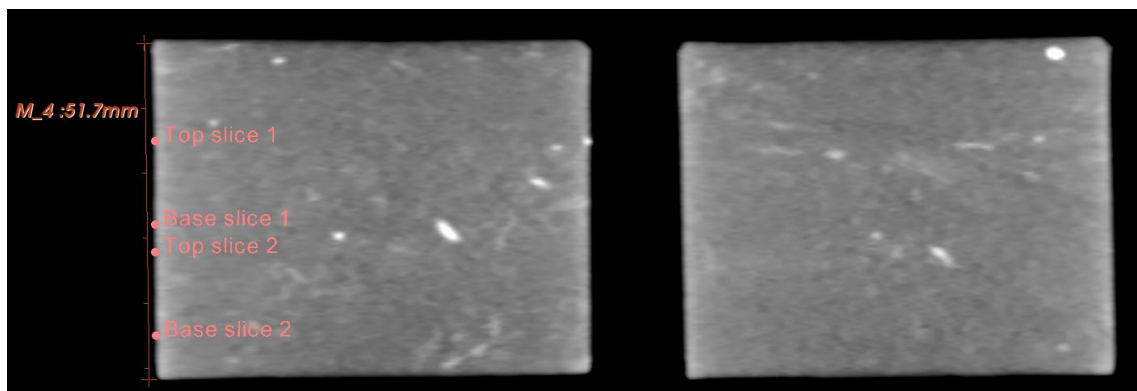
TRU1-1-756.39-DI

Mean depth [m]: 756.39

Stratigraphic position: «Parkinsoni-Württembergica-Schichten» ('Brauner Dogger')

CT-based slice selection: Heterogeneous, strongly bioturbated sample, which contains considerable amounts of pyritised burrows. Two slices were collected. Slice 2 appears slightly more representative for the entire whole core sample.

To be analysed: Slice 2



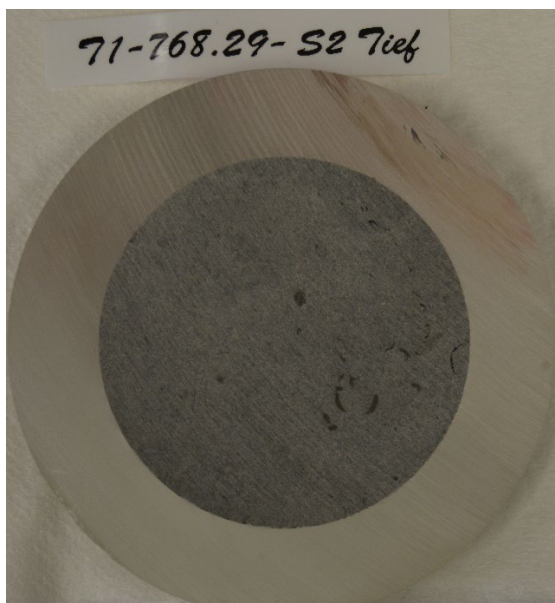
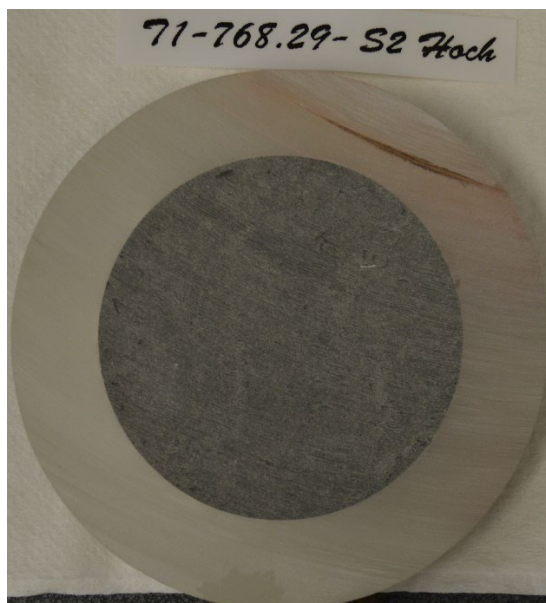
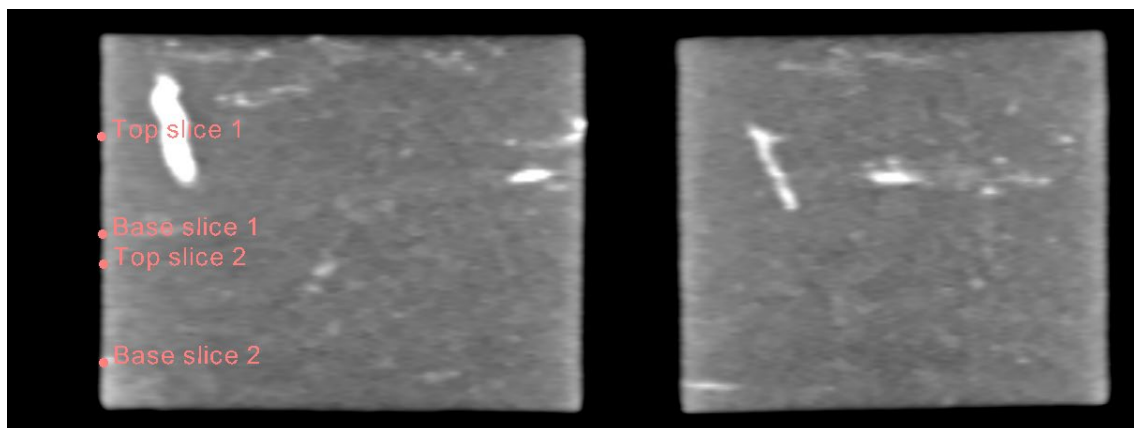
TRU1-1-768.29-DI

Mean depth [m]: 768.29

Stratigraphic position: «Parkinsoni-Württembergica-Schichten» ('Brauner Dogger')

CT-based slice selection: Heterogeneous, strongly bioturbated sample. Two slices were collected. Slice 1 contains a rather large (cm-scale) pyritised burrow.

To be analysed: Slice 2



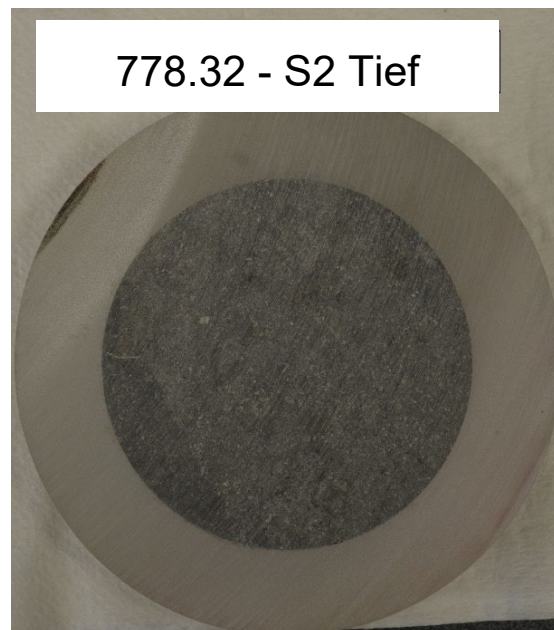
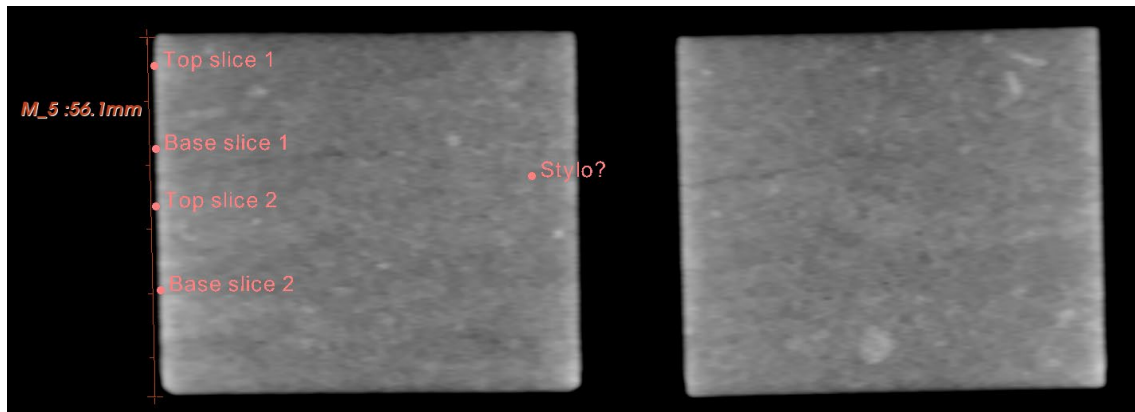
TRU1-1-778.32-DI

Mean depth [m]: 778.32

Stratigraphic position: Humphriesi-Oolith ('Brauner Dogger')

CT-based slice selection: Rather heterogeneous, bioclastic sample. Two slices were collected. Slice 1 may contain parts of a diagenetic stylolite.

To be analysed: Slice 2



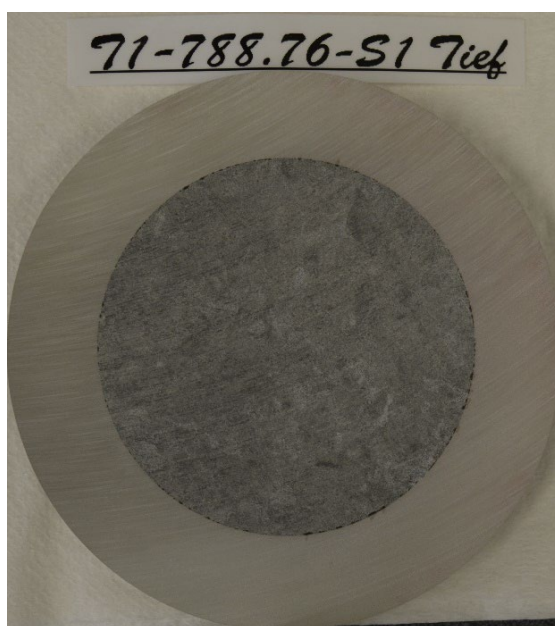
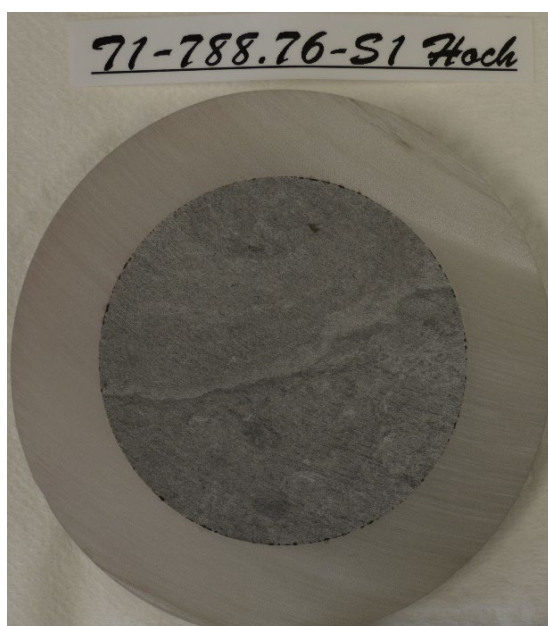
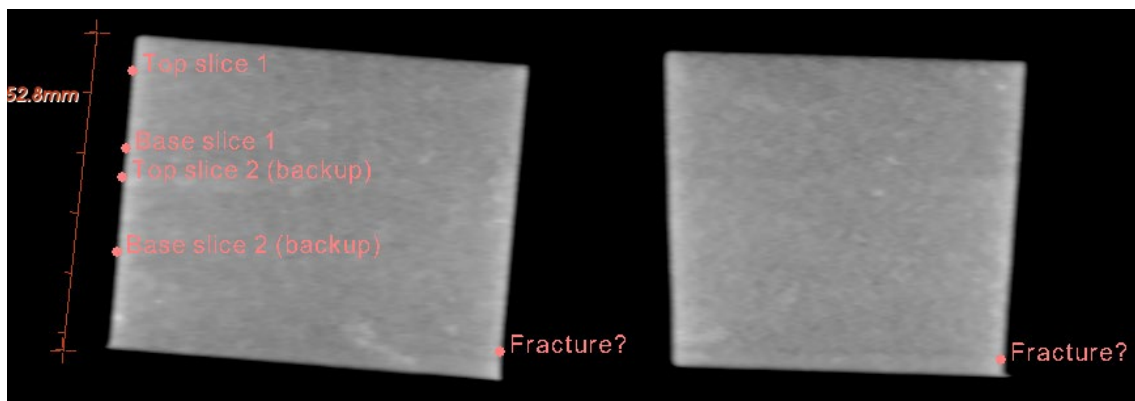
TRU1-1-788.76-DI

Mean depth [m]: 788.76

Stratigraphic position: Wedelsandstein ('Brauner Dogger')

CT-based slice selection: Rather homogeneous sample. Two slices were collected, which both could be used for the diffusion experiments.

To be analysed: Slice 1 (arbitrary selection)



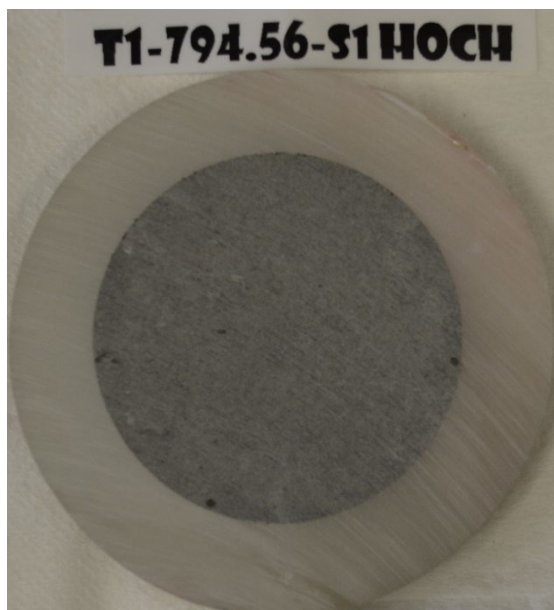
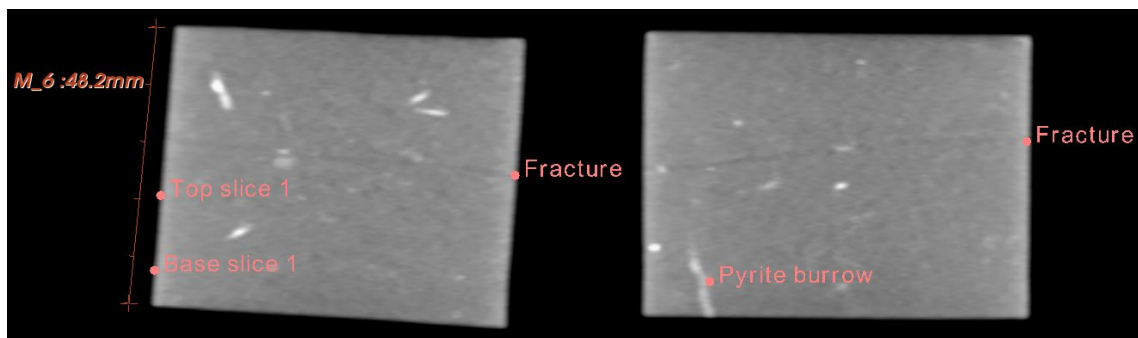
TRU1-1-794.56-DI

Mean depth [m]: 794.56

Stratigraphic position: Wedelsandstein ('Brauner Dogger')

CT-based slice selection: Bioturbated sample containing considerable amounts of small, randomly oriented, pyritised burrows. Owing to a drilling induced, bedding-parallel hair fracture, only one slice was collected.

To be analysed: Slice 1



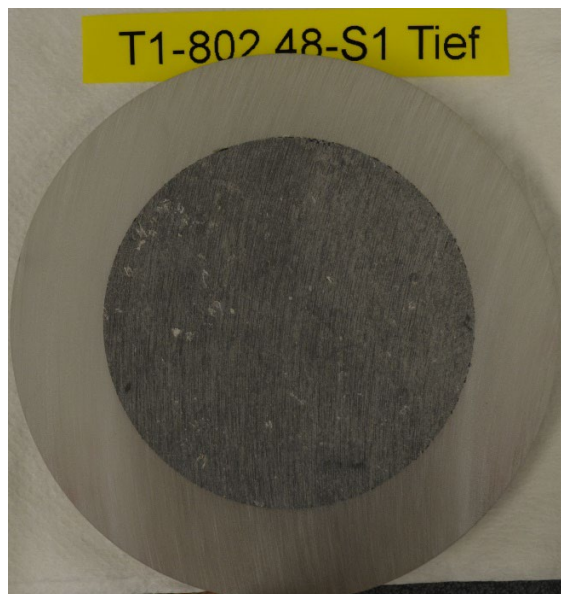
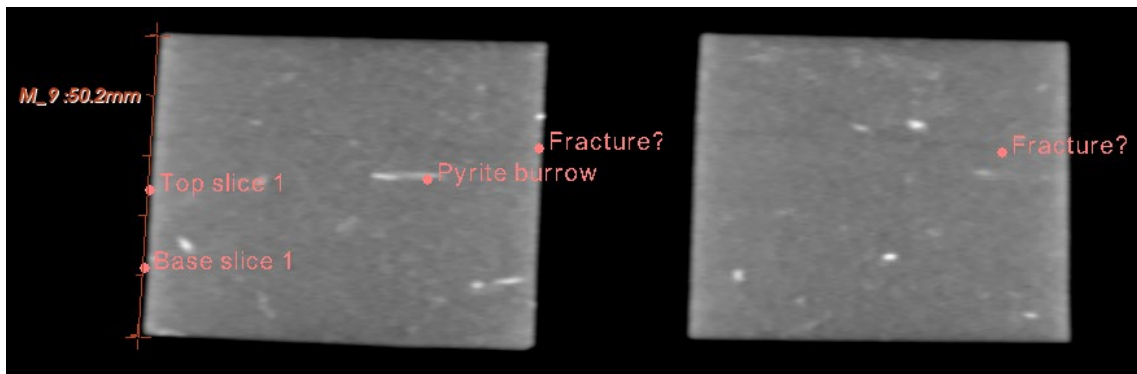
TRU1-1-802.48-DI

Mean depth [m]: 802.48

Stratigraphic position: Wedelsandstein ('Brauner Dogger')

CT-based slice selection: Bioturbated sample containing small, randomly oriented, pyritised burrows. Owing to a drilling induced, bedding-parallel hair fracture, only one slice was collected.

To be analysed: Slice 1



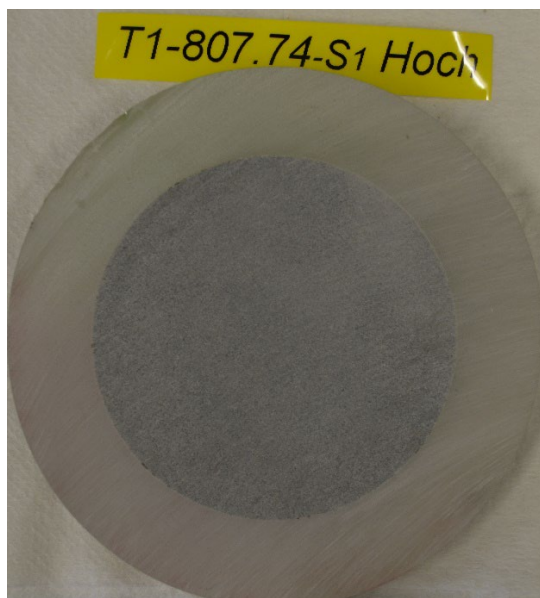
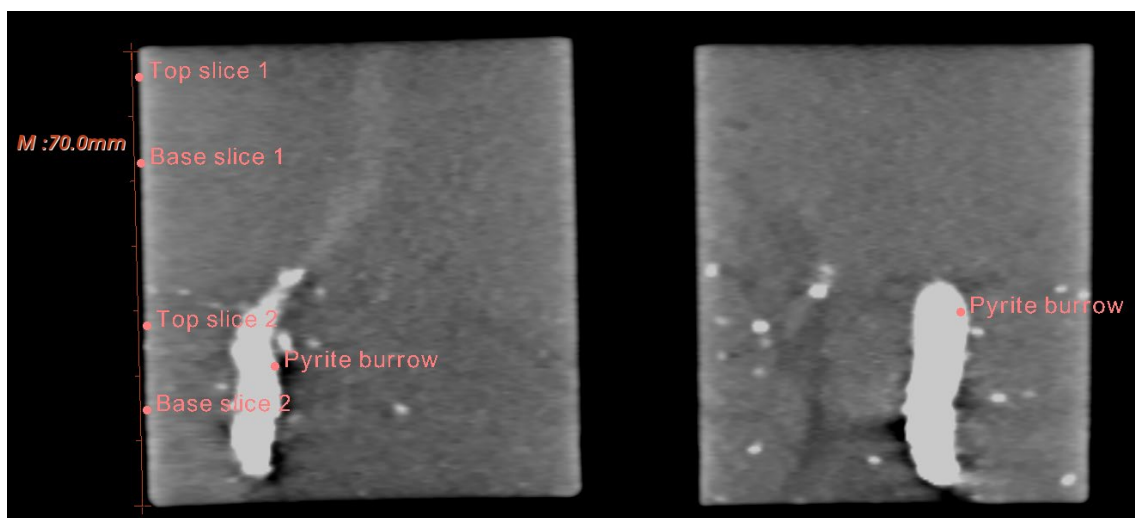
TRU1-1-807.74-DI

Mean depth [m]: 807.74

Stratigraphic position: Wedelsandstein ('Brauner Dogger')

CT-based slice selection: Heterogeneous sample: Upper part is rather homogeneous, whereas the lower part contains considerable amounts of small pyrite concretions. Moreover, there is a thick (cm-scale), vertical pyritised burrow. Two slices were selected: one in the homogeneous upper part and another one in the heterogeneous lower part.

To be analysed: Slice 1



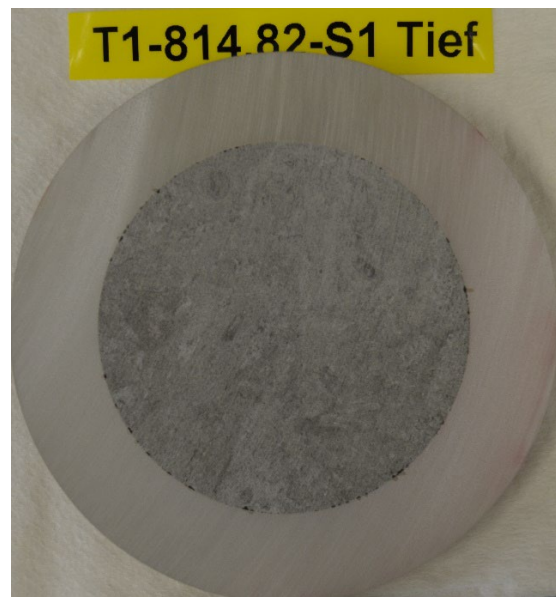
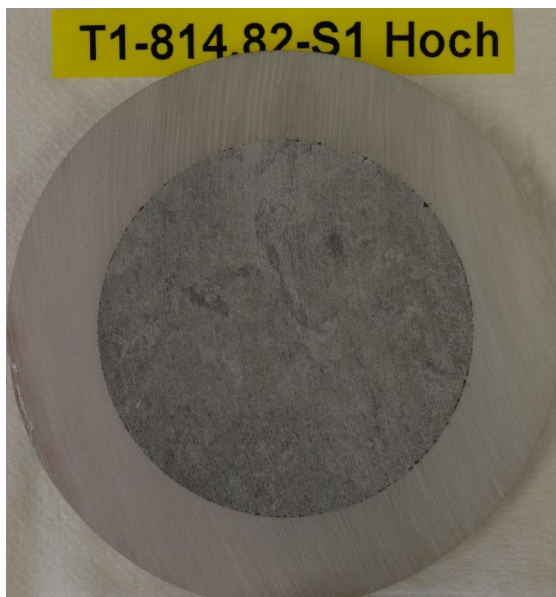
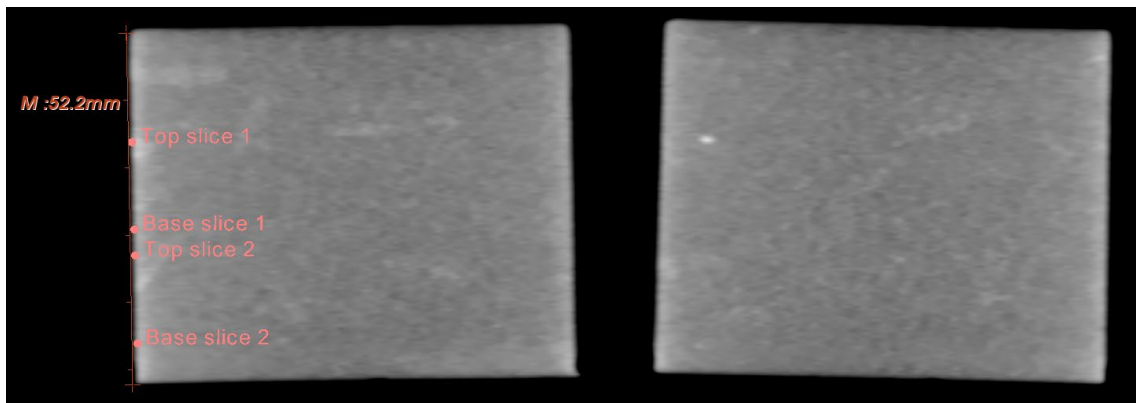
TRU1-1-814.82-OD

Mean depth [m]: 814.82

Stratigraphic position: Wedelsandstein ('Brauner Dogger')

CT-based slice selection: Rather homogeneous sample with lenses of slightly denser material (probably calcite cementing detrital quartz grains). Two slices were collected, however, Slice 2 seems to be slightly more representative for the entire drill core sample.

To be analysed: Slice 2



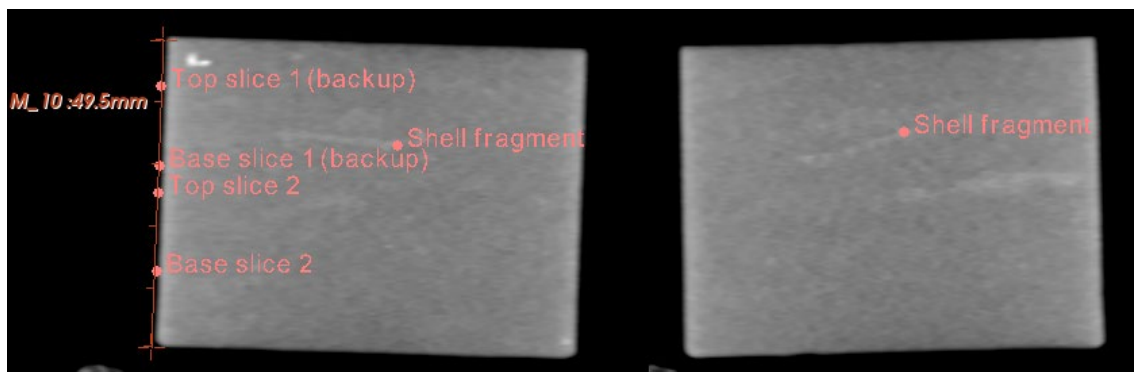
TRU1-1-815.28-DI

Mean depth [m]: 815.28

Stratigraphic position: Wedelsandstein ('Brauner Dogger')

CT-based slice selection: Bioclastic sample. Two slices were collected. Slice 1 contains a rather large shell fragment.

To be analysed: Slice 2



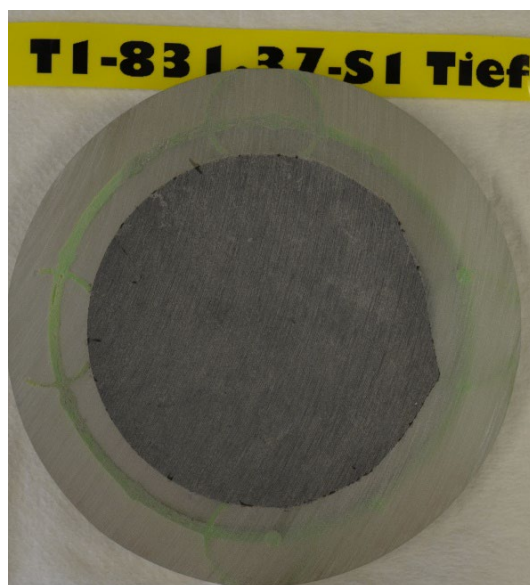
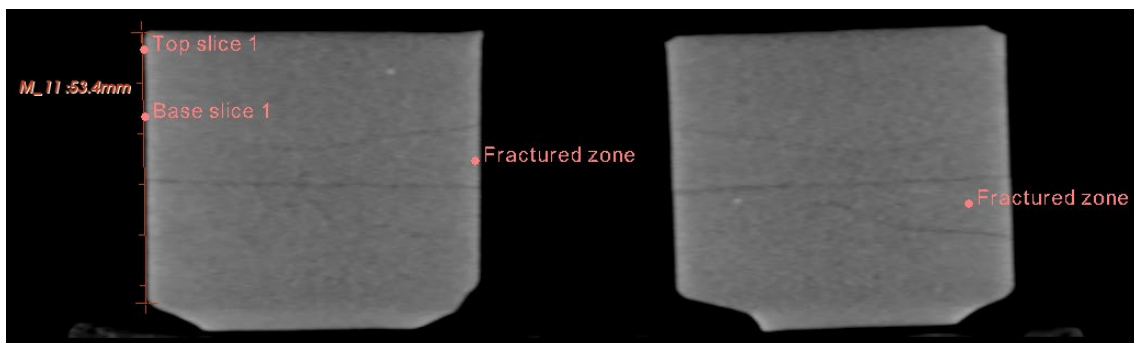
TRU1-1-831.37-DI

Mean depth [m]: 831.37

Stratigraphic position: OPA (Dogger)

CT-based slice selection: Rather homogeneous sample. However, owing to drilling induced fractures, only one slice was collected.

To be analysed: Slice 1



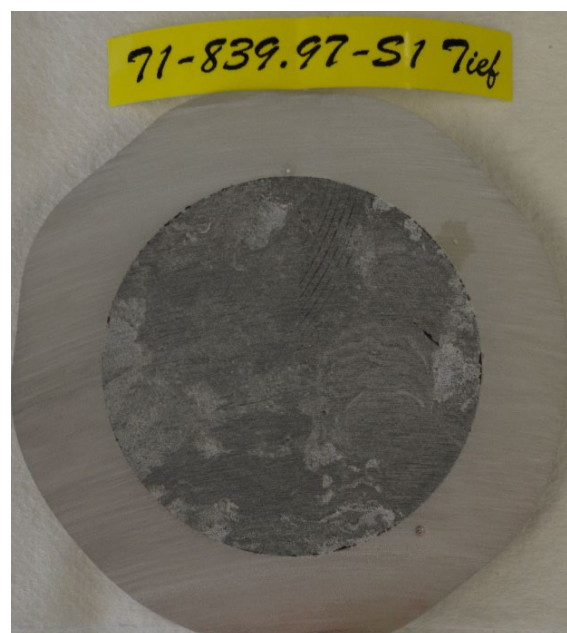
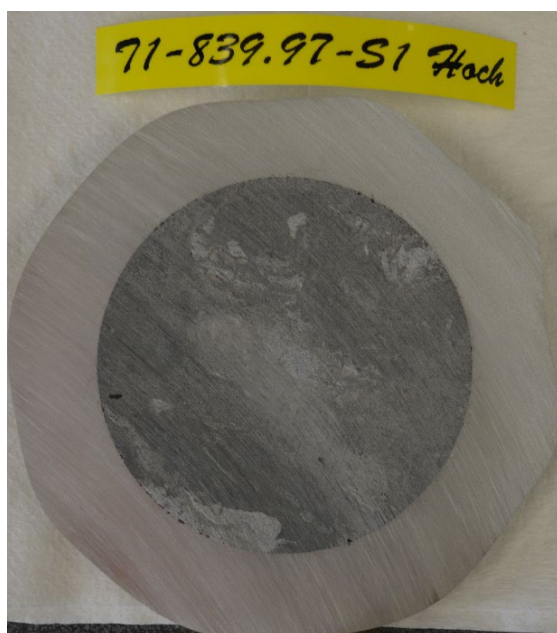
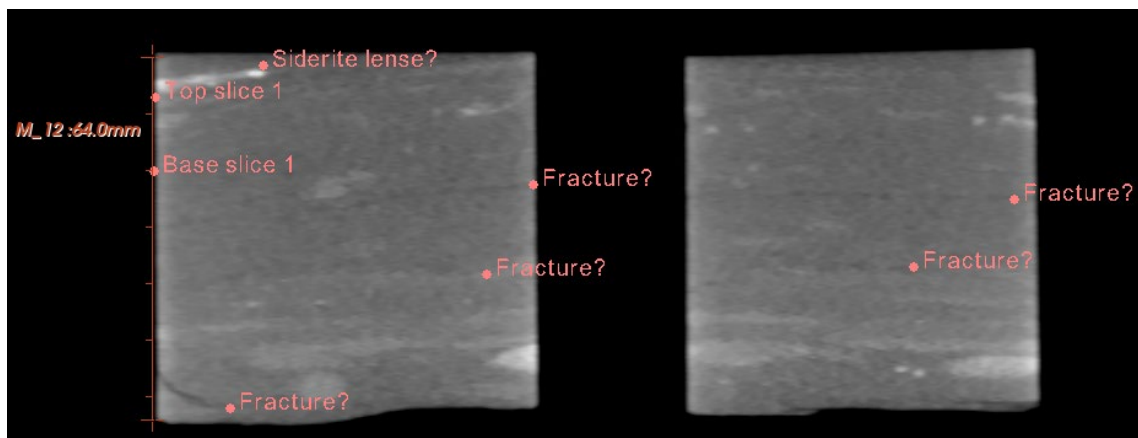
TRU1-1-839.97-DI

Mean depth [m]: 839.97

Stratigraphic position: OPA (Dogger)

CT-based slice selection: Sample contains some siderite lenses and silty beds. Owing to drilling induced fractures (actually, these could also be diagenetic stylolites) in the lower part of the sample, only one slice was collected.

To be analysed: Slice 1



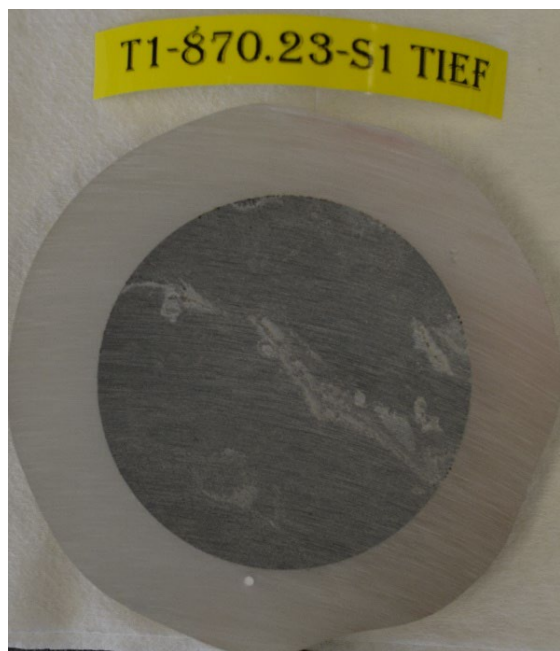
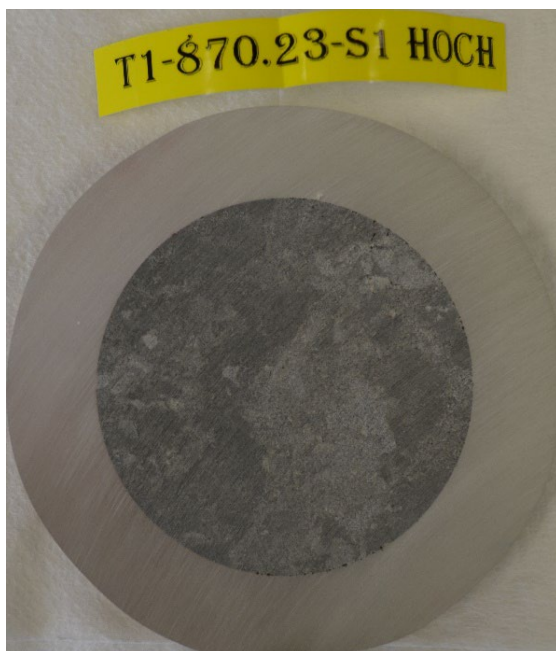
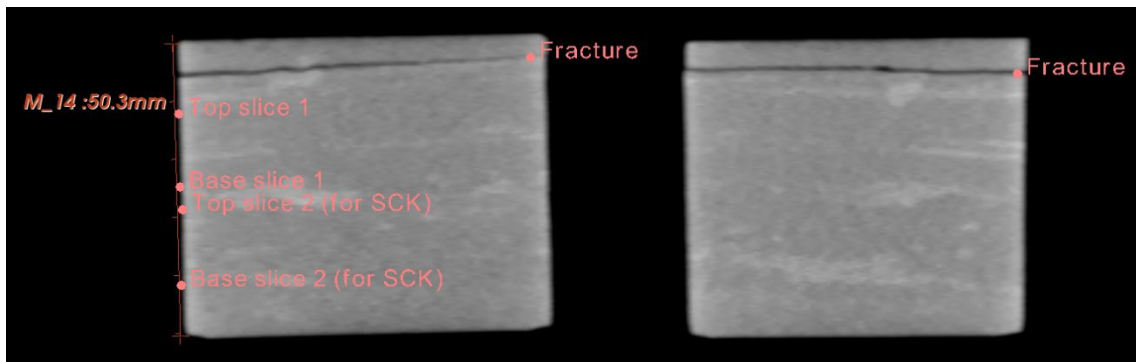
TRU1-1-870.23-DI

Mean depth [m]: 870.23

Stratigraphic position: OPA (Dogger)

CT-based slice selection: Overall, homogeneous sample, which contains some silty lenses. Two slices were collected, which are used for the benchmark study at SCK.

To be analysed: Slice 1 at PSI, Slice 2 at SCK



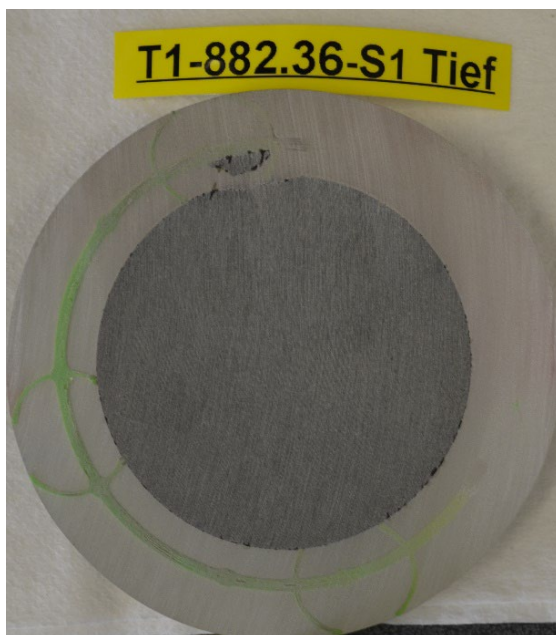
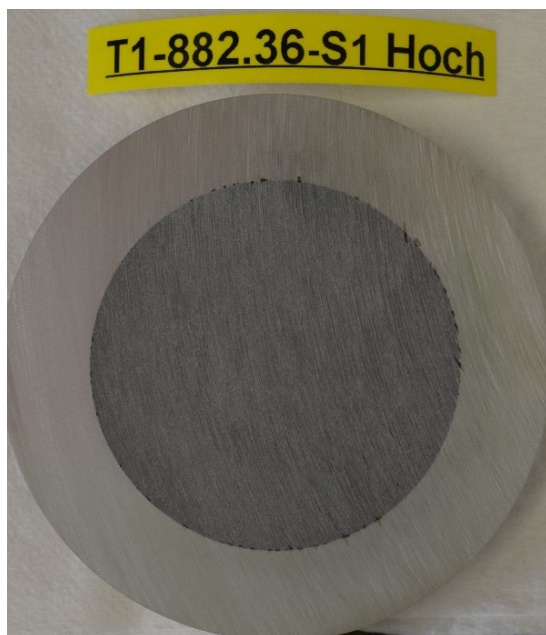
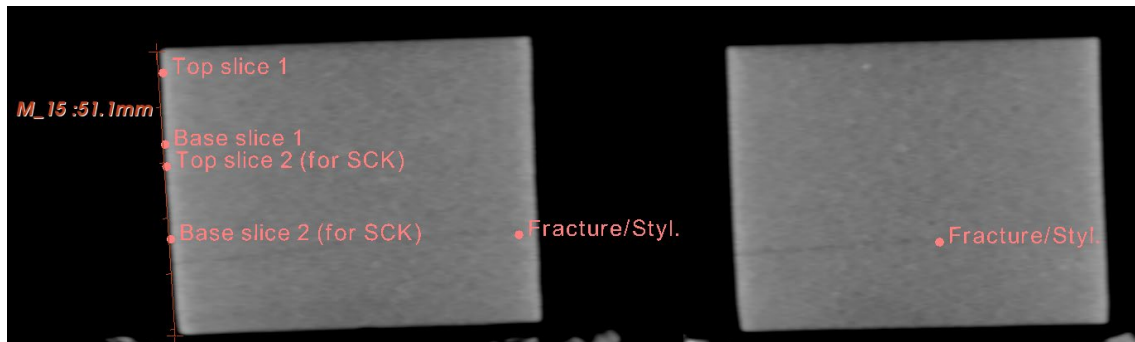
TRU1-1-882.36-DI

Mean depth [m]: 882.36

Stratigraphic position: OPA (Dogger)

CT-based slice selection: Rather homogeneous sample. Two slices were collected, which are used for the benchmark study at SCK.

To be analysed: Slice 1 at PSI, Slice 2 at SCK



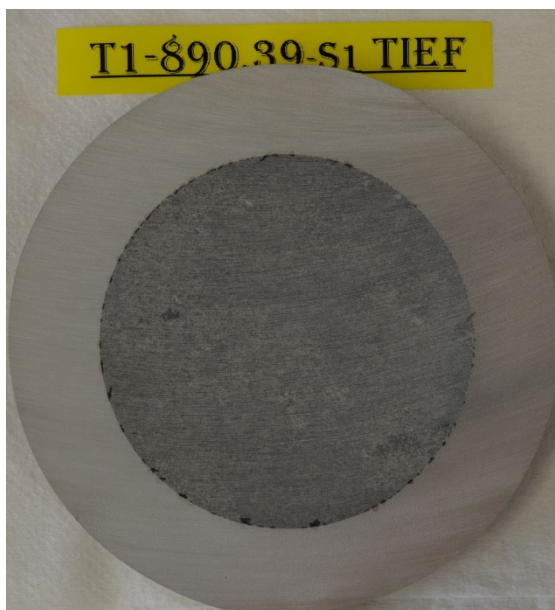
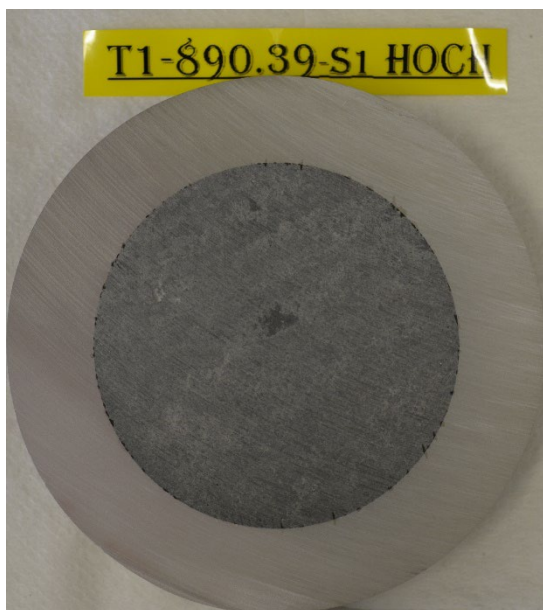
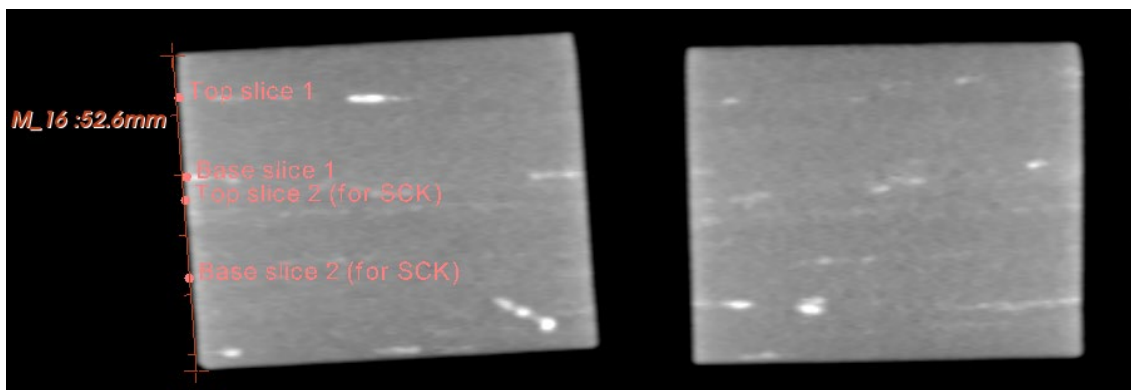
TRU1-1-890.39-DI

Mean depth [m]: 890.39

Stratigraphic position: OPA (Dogger)

CT-based slice selection: Overall, rather homogeneous sample, which contains some small siderite lenses. Two slices were collected, which are used for the benchmark study at SCK.

To be analysed: Slice 1 at PSI, Slice 2 at SCK



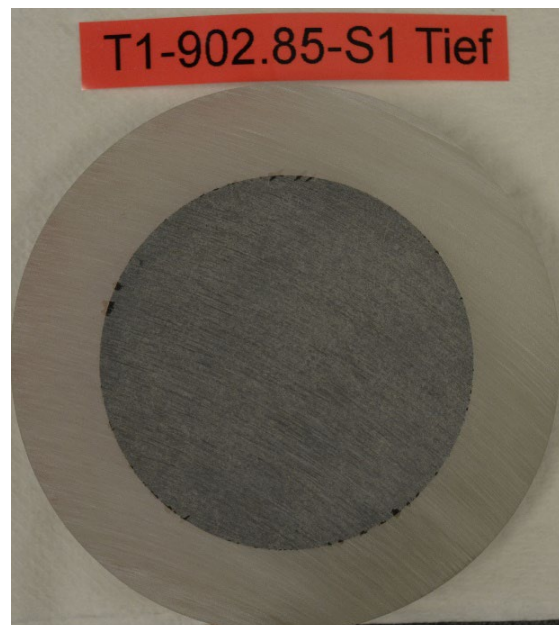
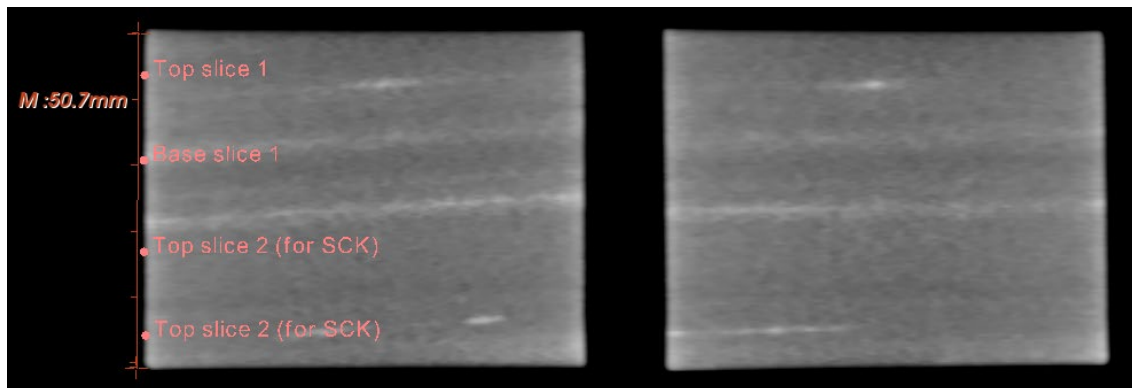
TRU1-1-902.85-DI

Mean depth [m]: 902.85

Stratigraphic position: OPA (Dogger)

CT-based slice selection: Overall, rather homogeneous sample, which contains some thin silty beds and lenses. Two slices were collected, which are used for the benchmark study at SCK.

To be analysed: Slice 1 at PSI, Slice 2 at SCK



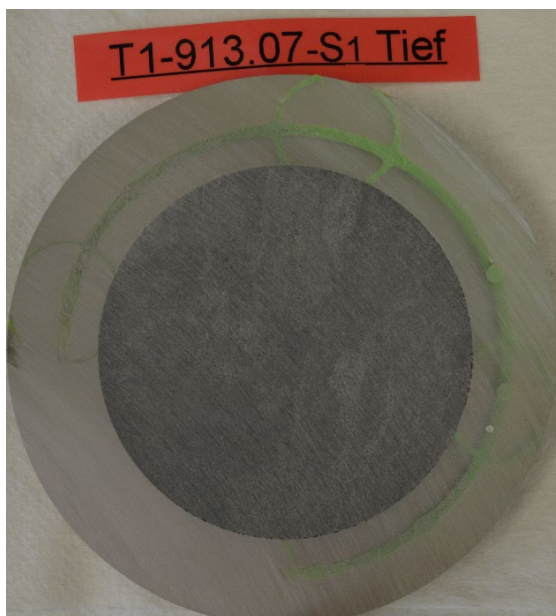
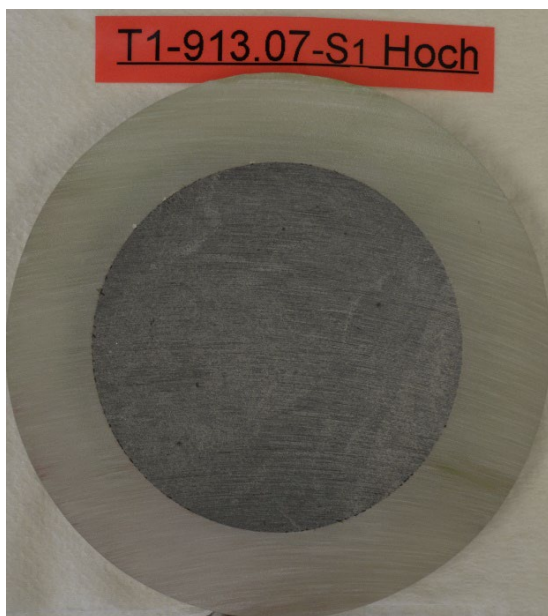
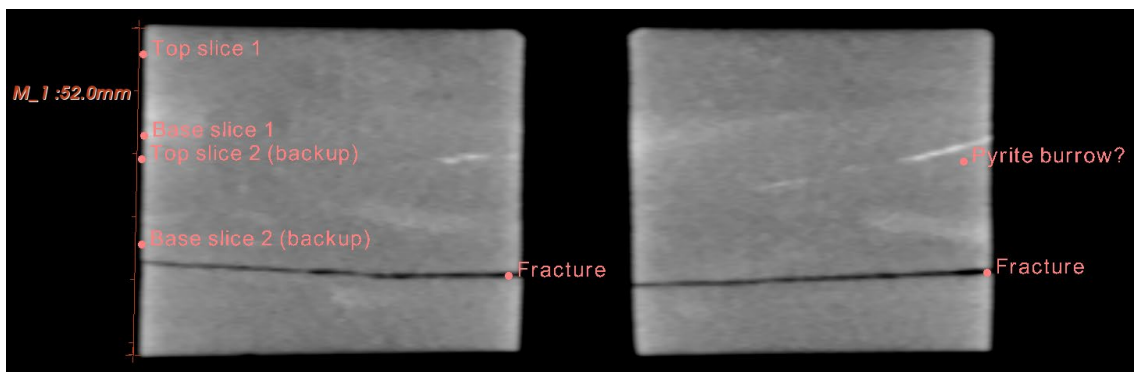
TRU1-1-913.07-DI

Mean depth [m]: 913.07

Stratigraphic position: OPA (Dogger)

CT-based slice selection: Overall, rather homogeneous, slightly bioturbated sample. Two slices were collected. Slice 2 contains a long, thin, sub-horizontal, pyritised burrow.

To be analysed: Slice 1



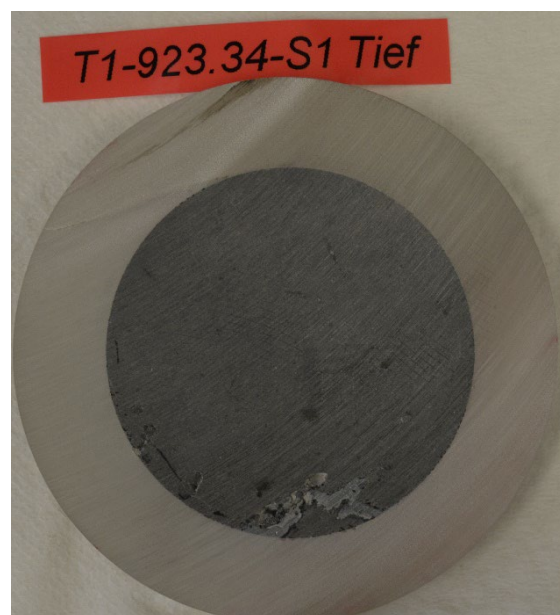
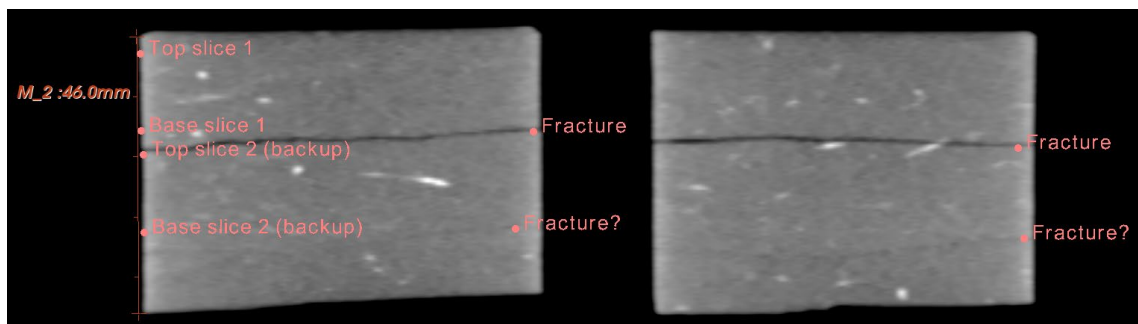
TRU1-1-923.34-DI

Mean depth [m]: 923.34

Stratigraphic position: OPA (Dogger)

CT-based slice selection: Bioturbated sample, which contains numerous small, thin, pyritised burrows. Moreover, it contains two drilling-induced fractures. Two slices were collected between the fractures. Slice 2 may be affected by such a fracture.

To be analysed: Slice 1



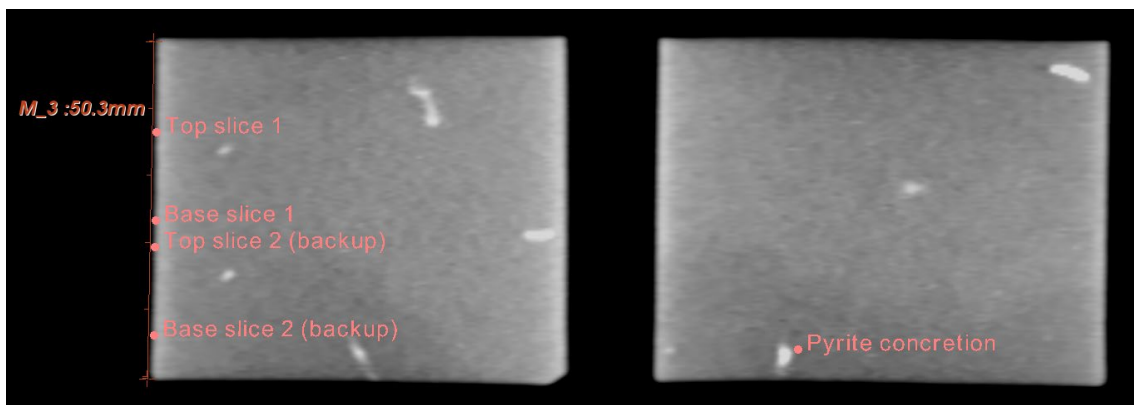
TRU1-1-930.99-DI

Mean depth [m]: 930.99

Stratigraphic position: Staffelegg Fm. (Lias)

CT-based slice selection: Overall, rather homogeneous sample, which contains some pyrite concretions. Two slices were sampled. Slice 2 contains distinctly more pyrite concretions than Slice 1.

To be analysed: Slice 1



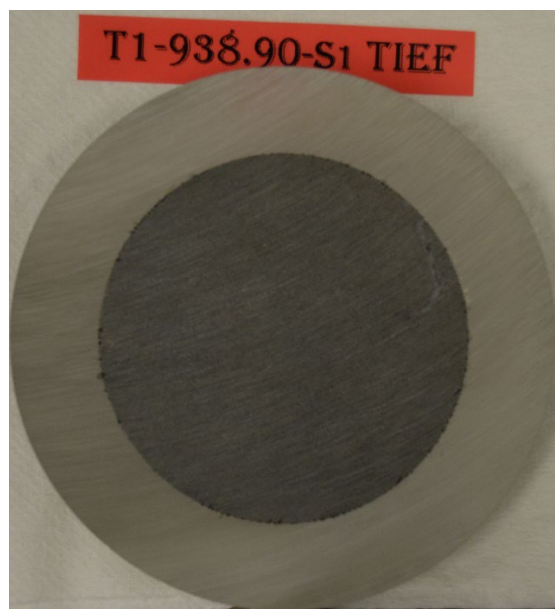
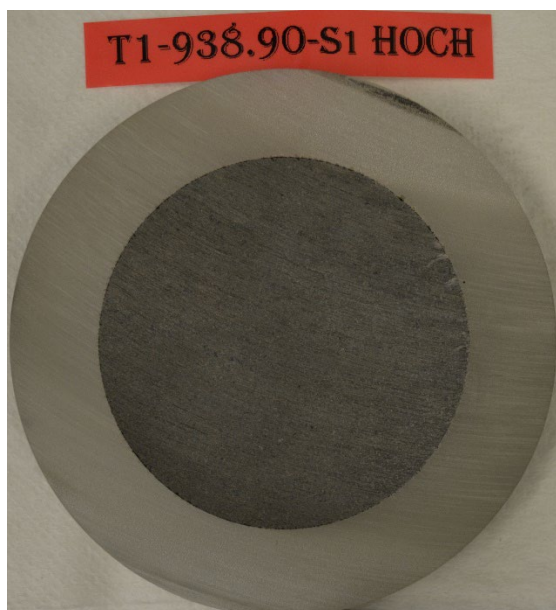
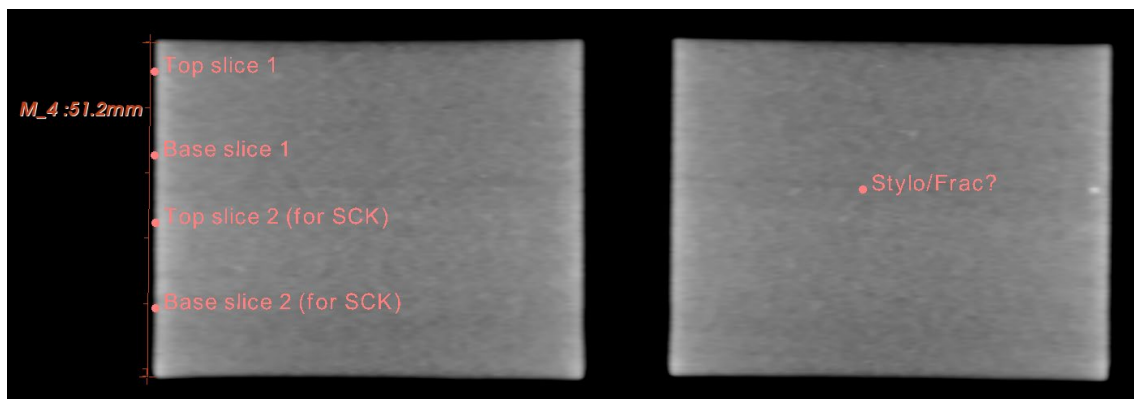
TRU1-1-938.90-DI

Mean depth [m]: 938.90

Stratigraphic position: Staffelegg Fm. (Lias)

CT-based slice selection: Homogeneous sample. Two slices were collected, which are used for the benchmark study at SCK.

To be analysed: Slice 1 at PSI, Slice 2 at SCK



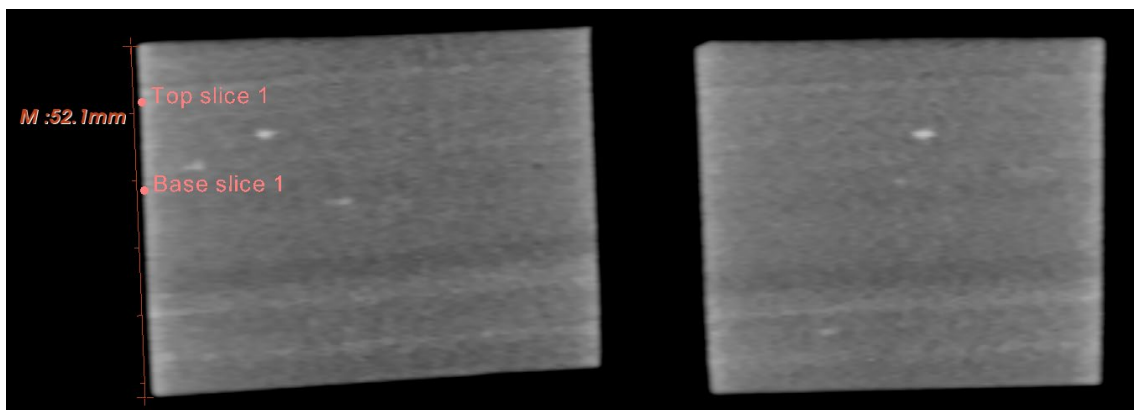
TRU1-1-943.21-DI

Mean depth [m]: 943.21

Stratigraphic position: Staffelegg Fm. (Lias)

CT-based slice selection: Rather homogeneous sample, which contains a few, small pyrite concretions.

To be analysed: Slice 1



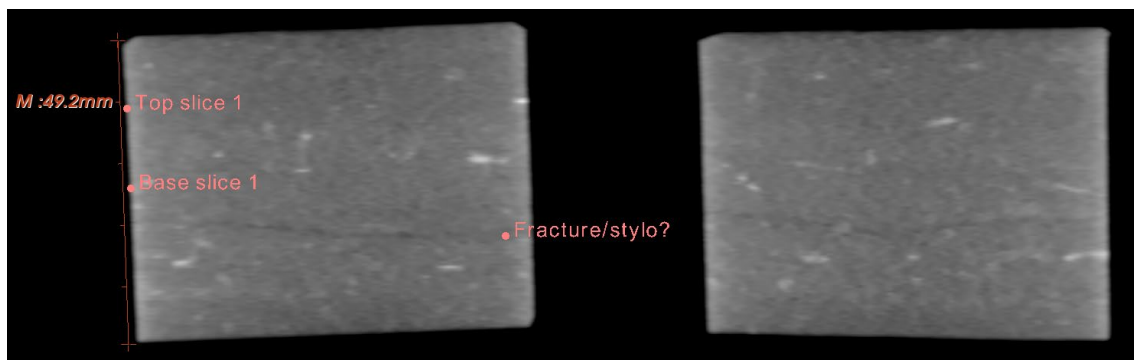
TRU1-1-955.02-DI

Mean depth [m]: 955.02

Stratigraphic position: Staffelegg Fm. (Lias)

CT-based slice selection: Bioturbated sample, which contains small, thin, pyritized burrows. Owing to a drilling induced fracture, only one slice was collected.

To be analysed: Slice 1



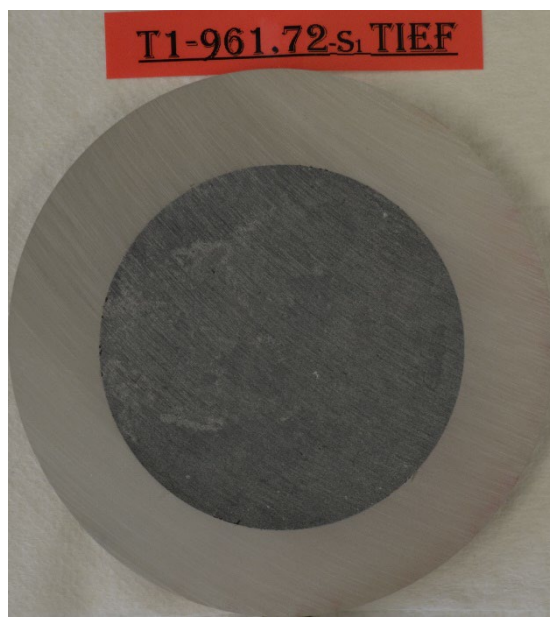
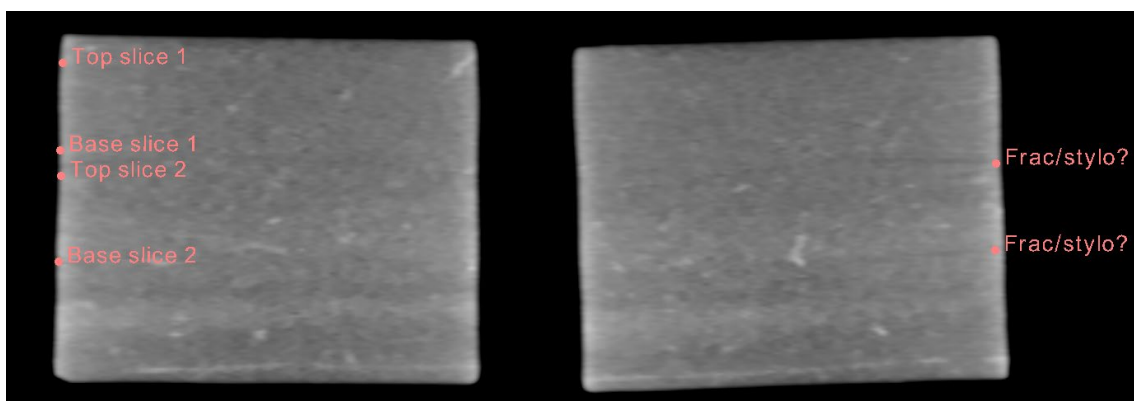
TRU1-1-961.72-DI

Mean depth [m]: 961.72

Stratigraphic position: Staffelegg Fm. (Lias)

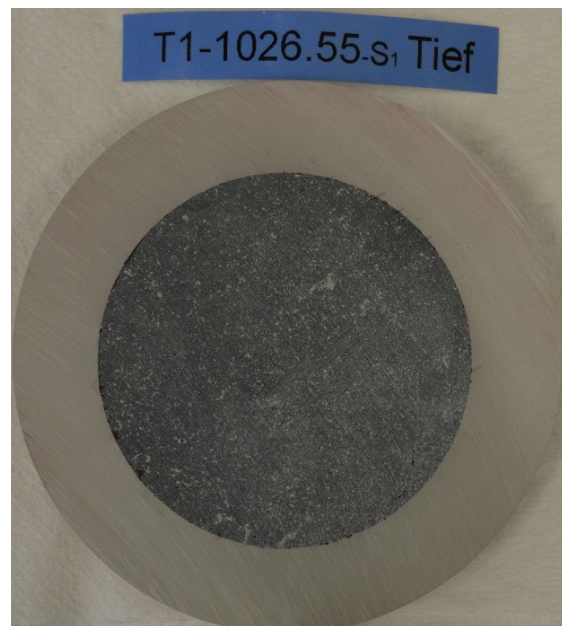
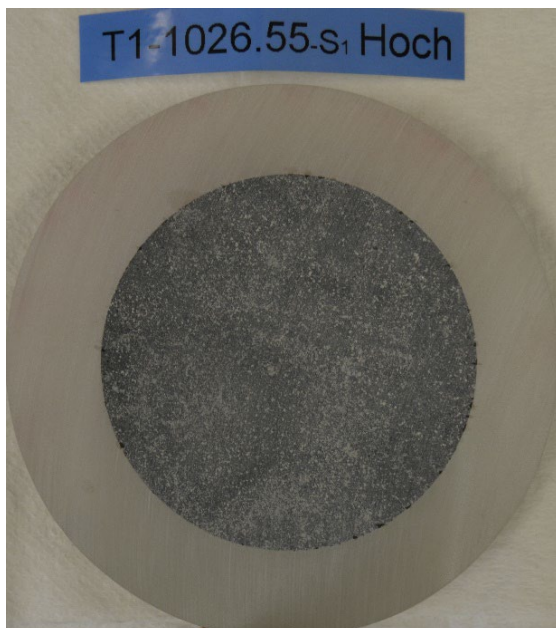
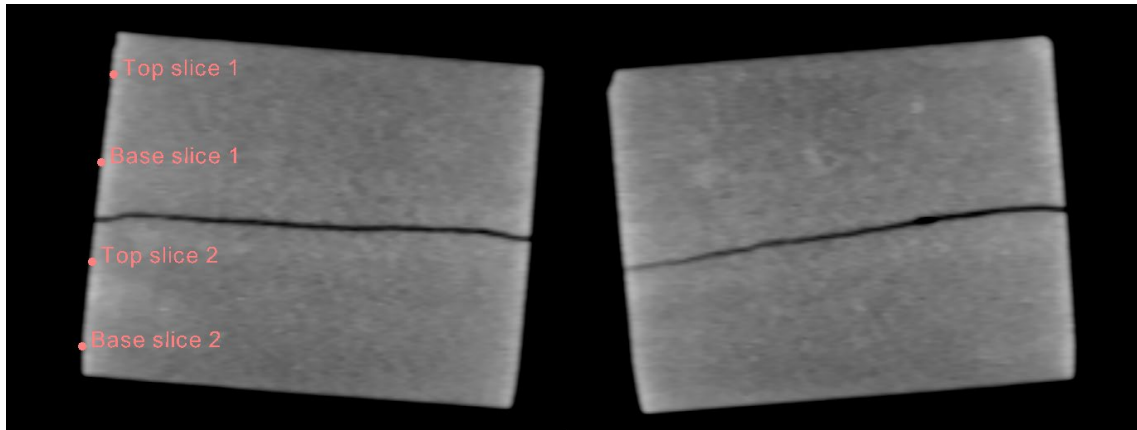
CT-based slice selection: Bioturbated sample, which contains a few small, thin, pyritised burrows. Moreover, there are two drilling-induced fractures (these could also be stylolites). Two slices were collected between these fractures. However, Slice 2 may contain some fractured rock material.

To be analysed: Slice 1



TRU1-1-1026.55-DI

Mean depth [m]: 1,026.55
Stratigraphic position: Klettgau Fm. (Keuper)
CT-based slice selection: Rather homogeneous sample. Two slices were collected, which can both be used for the experiments.
To be analysed: Slice 1 (arbitrary selection)



App. I Experimental data and model curves of the diffusion tests with HTO, $^{36}\text{Cl}^-$ and $^{22}\text{Na}^+$

This appendix shows the results of all diffusion tests performed with tritiated water (HTO) and the $^{36}\text{Cl}^-$ and $^{22}\text{Na}^+$ tracers. The data for each of the rock samples are represented on a single page including information on the origin (location of the borehole, mean depth of sample), geometry (diameter, length in direction of diffusion) and the type of synthetic porewater applied for the measurements. As mentioned in Section 2.4, the measurements of the HTO and $^{36}\text{Cl}^-$ tracers were carried out in parallel, followed by the measurements of the $^{22}\text{Na}^+$ tracer.

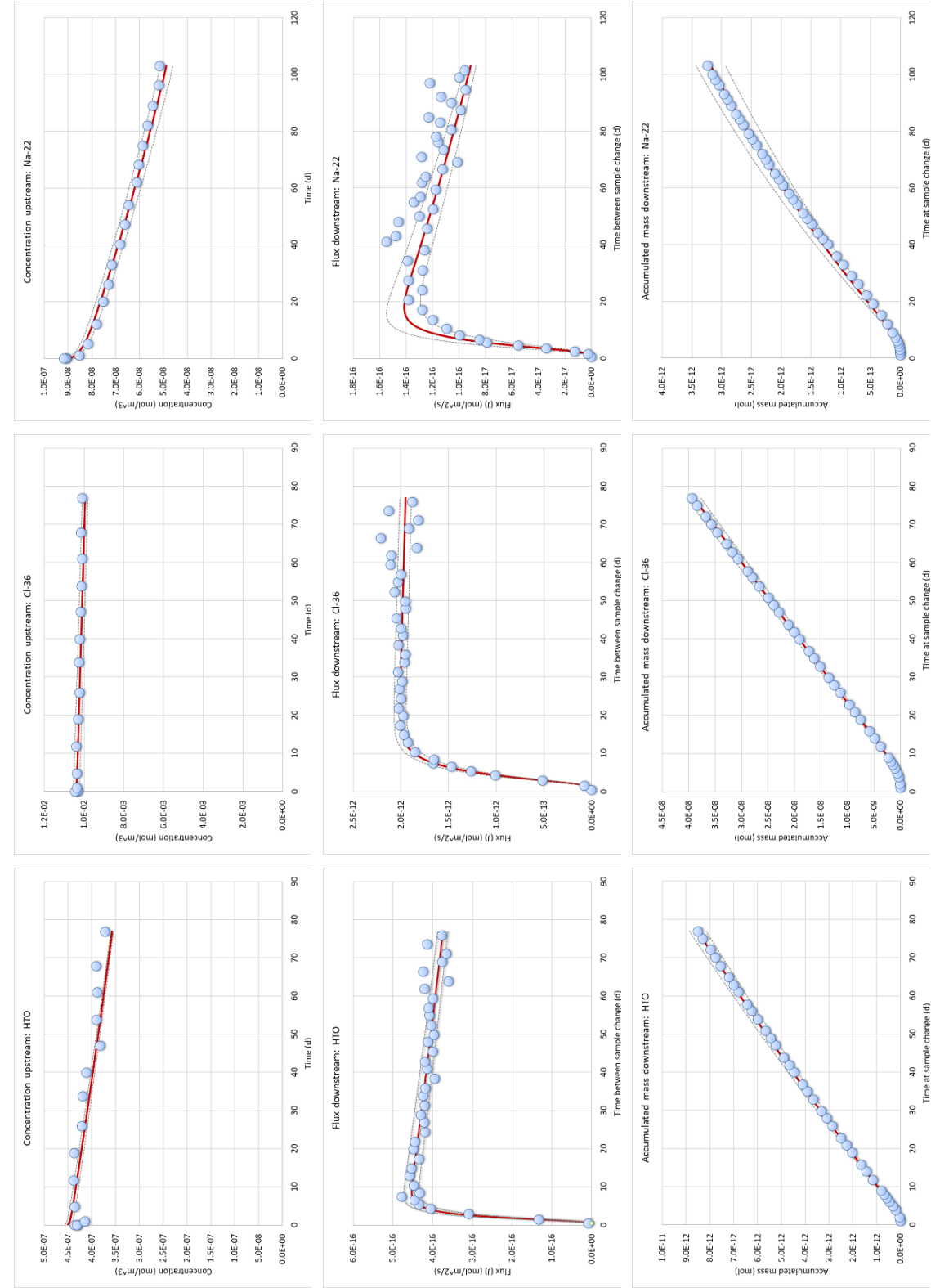
The plots are grouped in columns for each tracer (HTO, $^{36}\text{Cl}^-$ and $^{22}\text{Na}^+$ from left to right) and rows showing the depletion of tracer concentration in the upstream (source) reservoir and the average flux and accumulated mass in the downstream (target) reservoir, from top to bottom. The time scale of the plots for the concentration depletion and the accumulated mass refer to the times of sampling, while for the average flux plots, the average time of the interval between start and end of each of the downstream samples was used to be representative of the average flux. The measured data are shown by circles, and the fit curves are shown as solid lines for the best-fit parameter values and dashed lines for the upper and lower bounding combination of parameter values. The procedures involved have been described in Sections 2.7 and 2.8.

I.1 Samples from Jura Ost

- BOZ1-1
- BOZ2-1

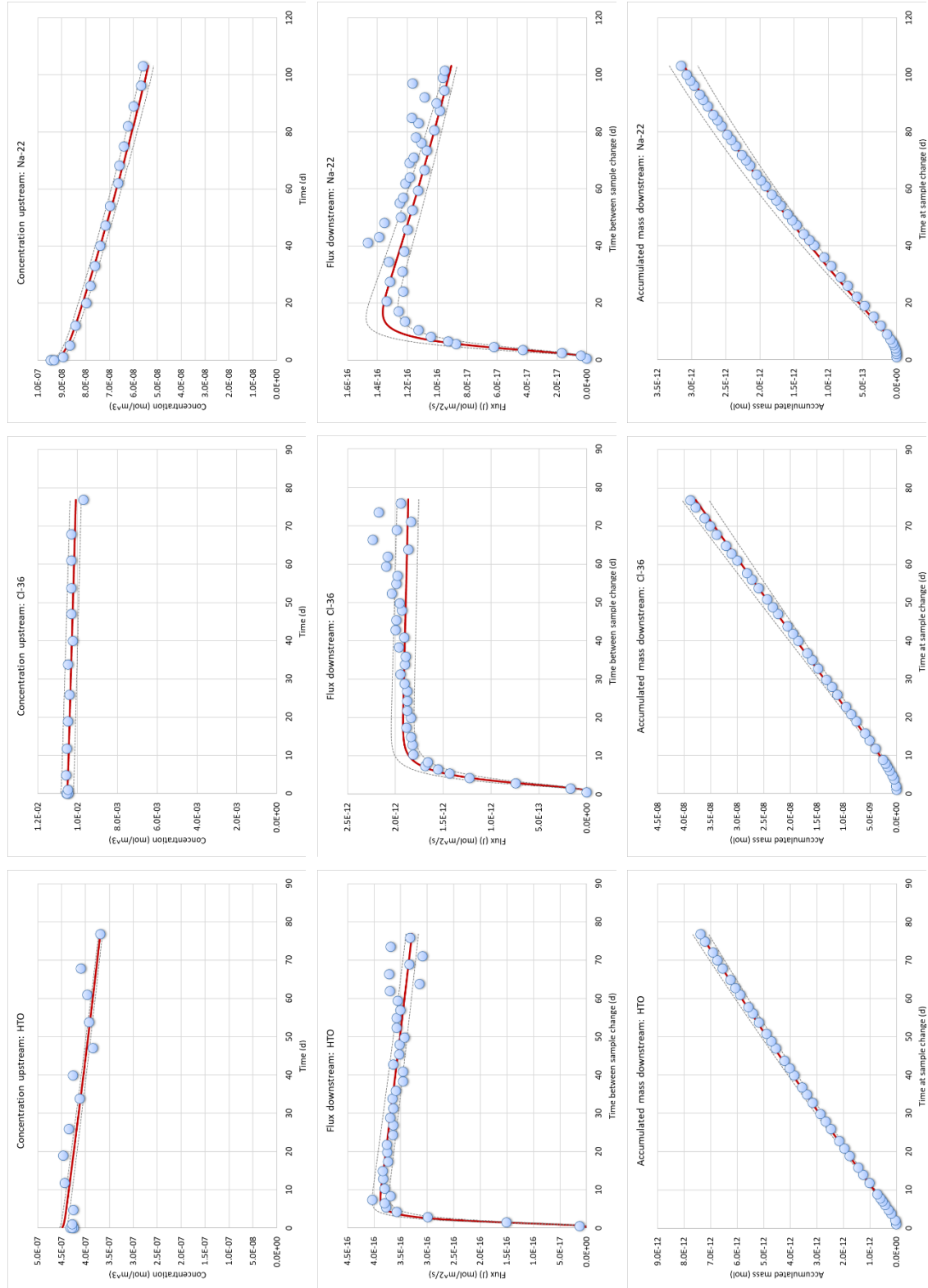
BOZ1-1-266.75-DI (S1) – Wildegg Formation – SPW-BOZ

Sample diameter 63.49 mm – sample thickness 13.13 mm



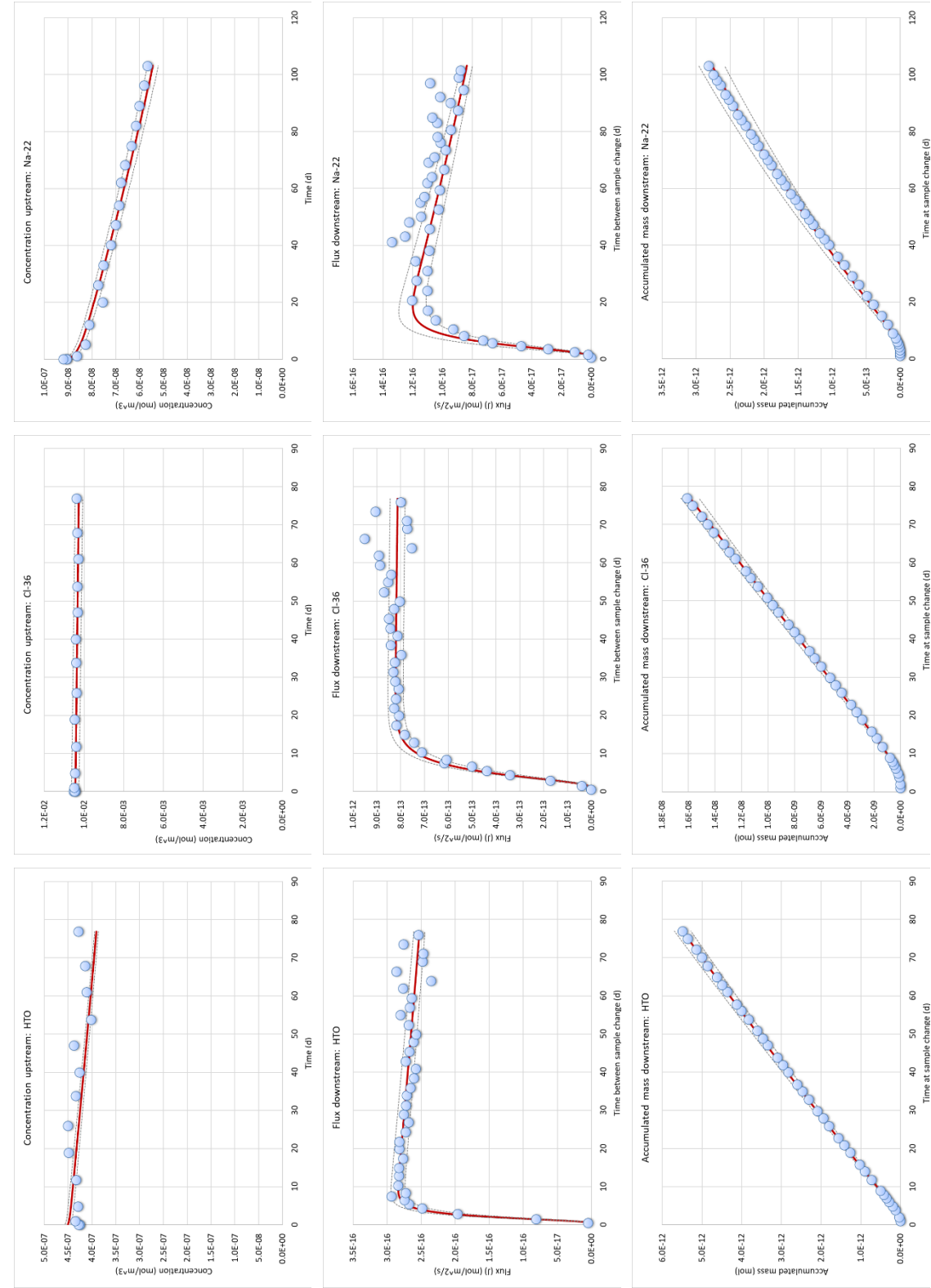
BOZ1-1-290.63-DI (S1) – Wildegg Formation – SPW-BOZ

Sample diameter 63.50 mm – sample thickness 12.47 mm



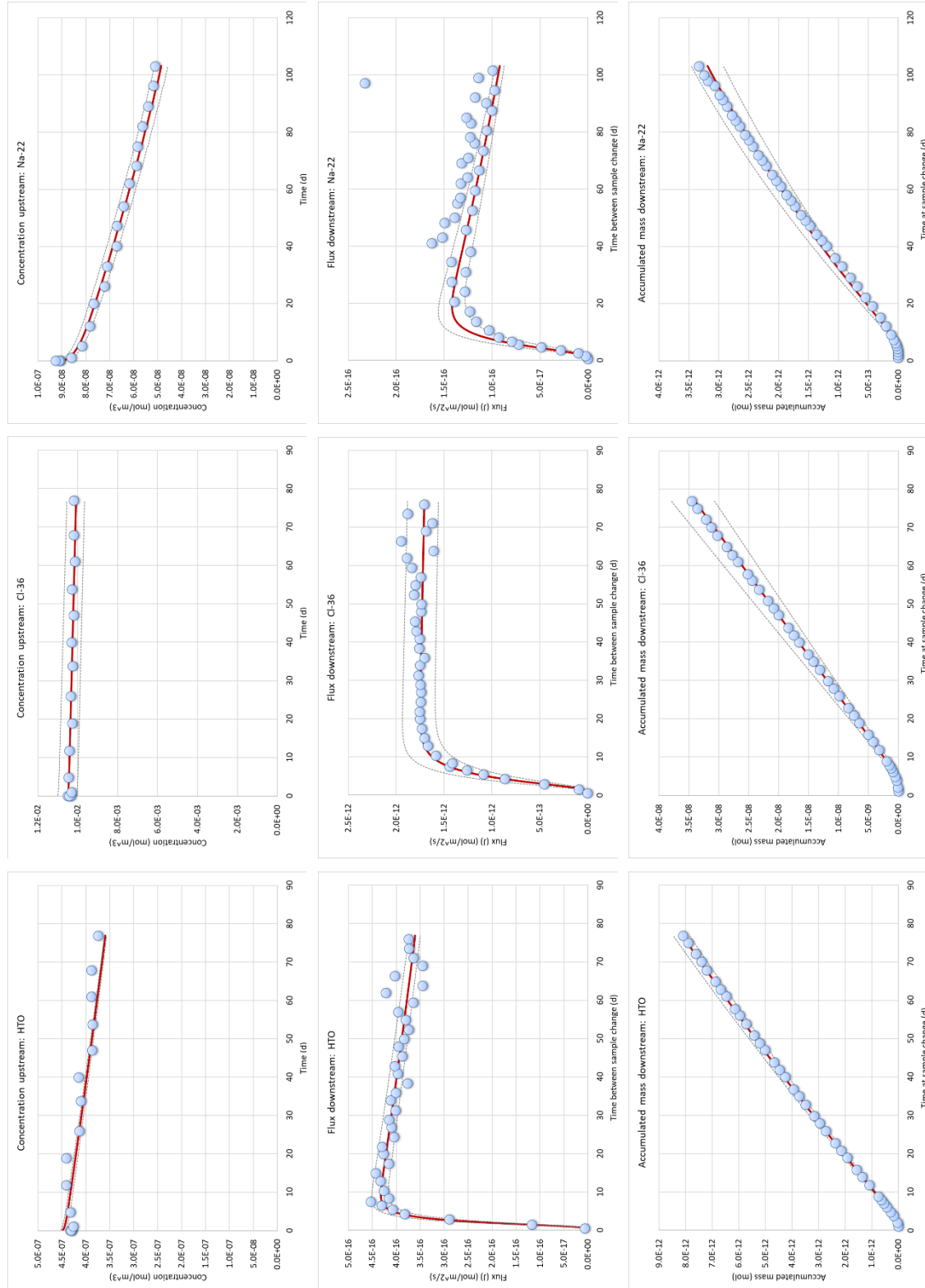
BOZ1-1-323.16-DI (S1) – Wildegg Formation – SPW-BOZ

Sample diameter 63.40 mm – sample thickness 12.33 mm



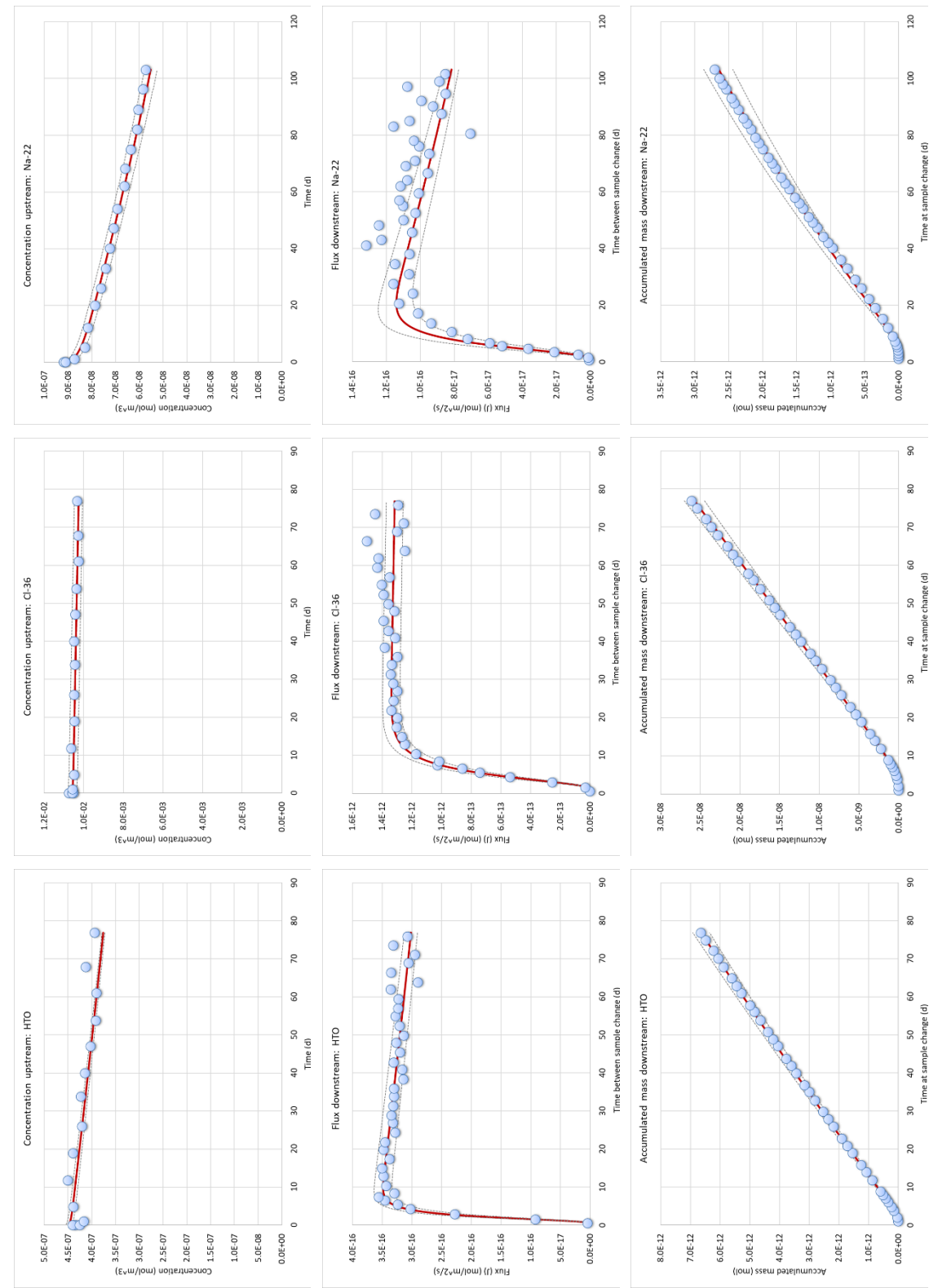
BOZ1-1-353.36-DI (S1) – Wildegg Formation – SPW-BOZ

Sample diameter 63.43 mm – sample thickness 12.44 mm



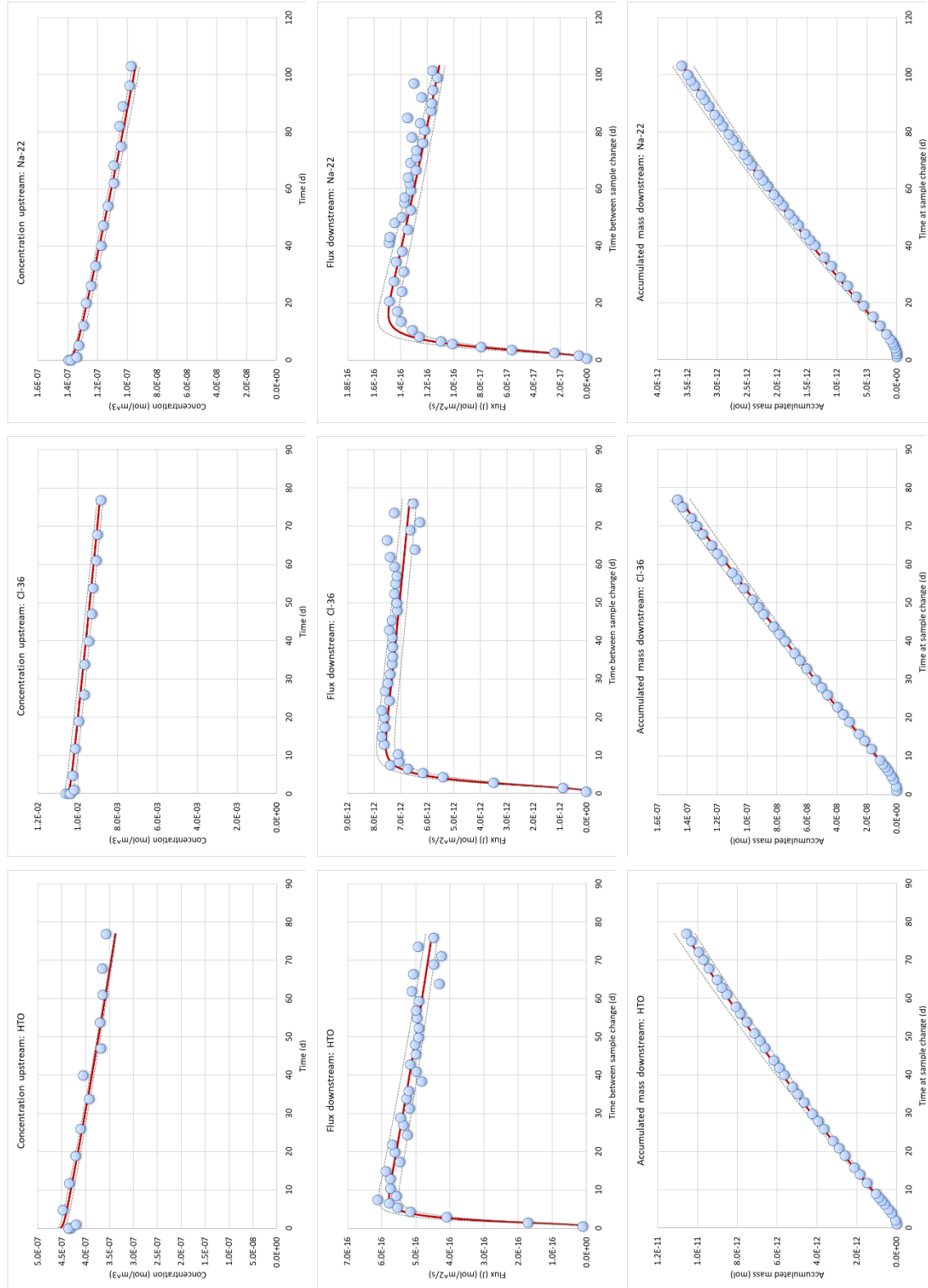
BOZ1-1-391.30-DI (S1) – Wildegg Formation – SPW-BOZ

Sample diameter 63.34 mm – sample thickness 12.76 mm



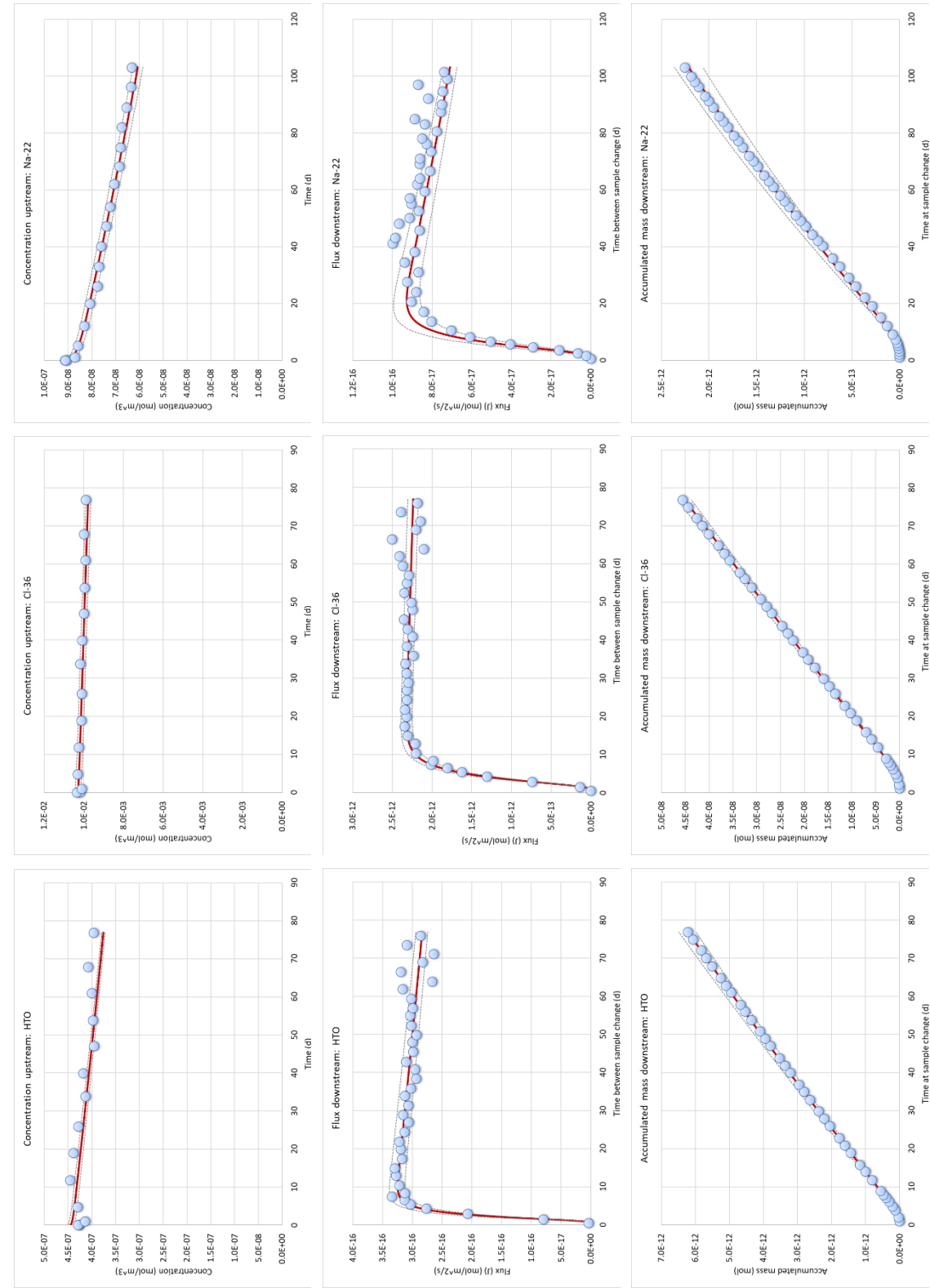
BOZ1-1-416.35-DI (S1) – Hauptrogenstein – SPW-BOZ

Sample diameter 63.55 mm – sample thickness 12.99 mm



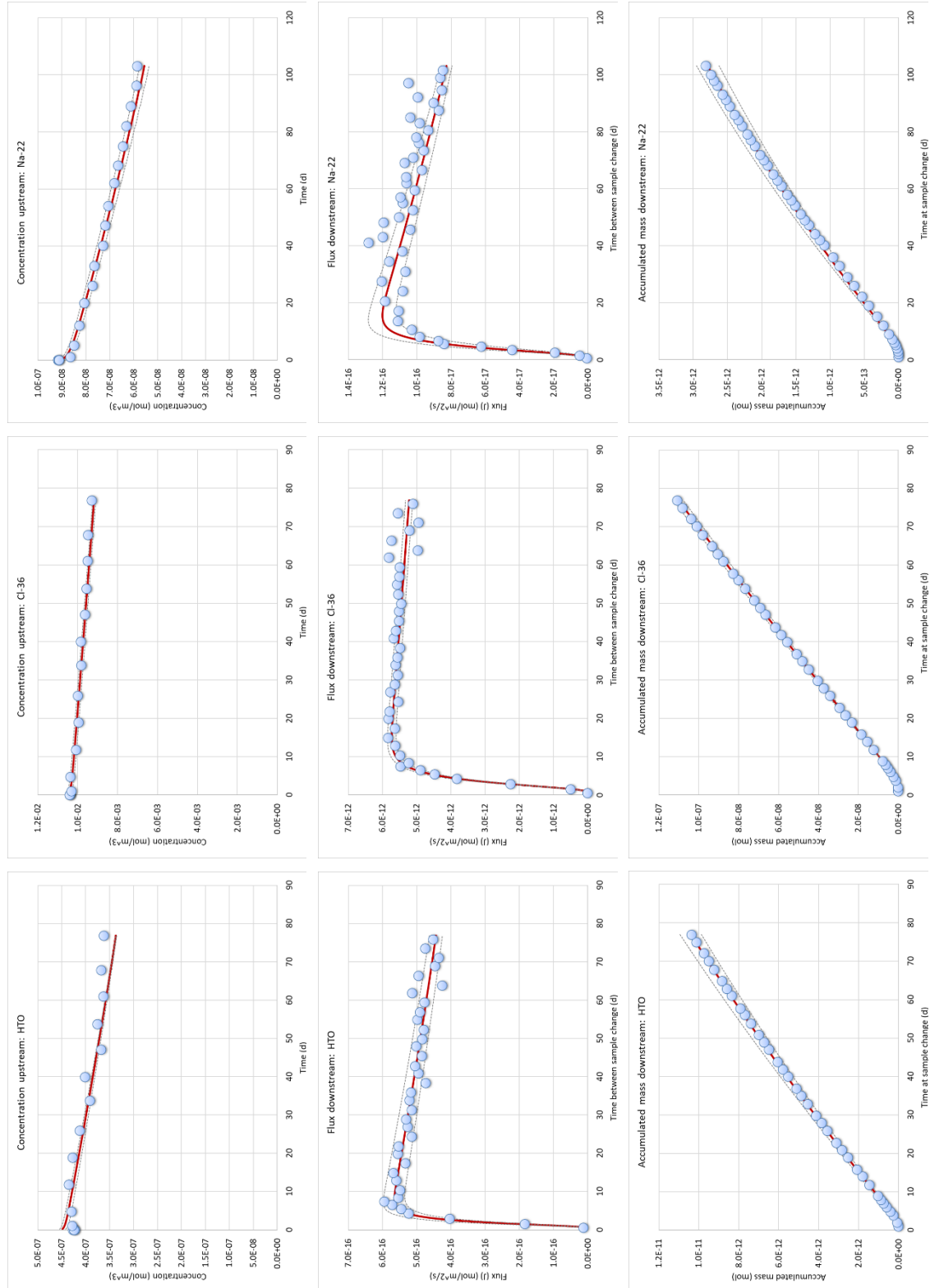
BOZ1-1-434.29-DI (S2) – Klingnau Formation – SPW-BOZ

Sample diameter 63.45 mm – sample thickness 13.23 mm



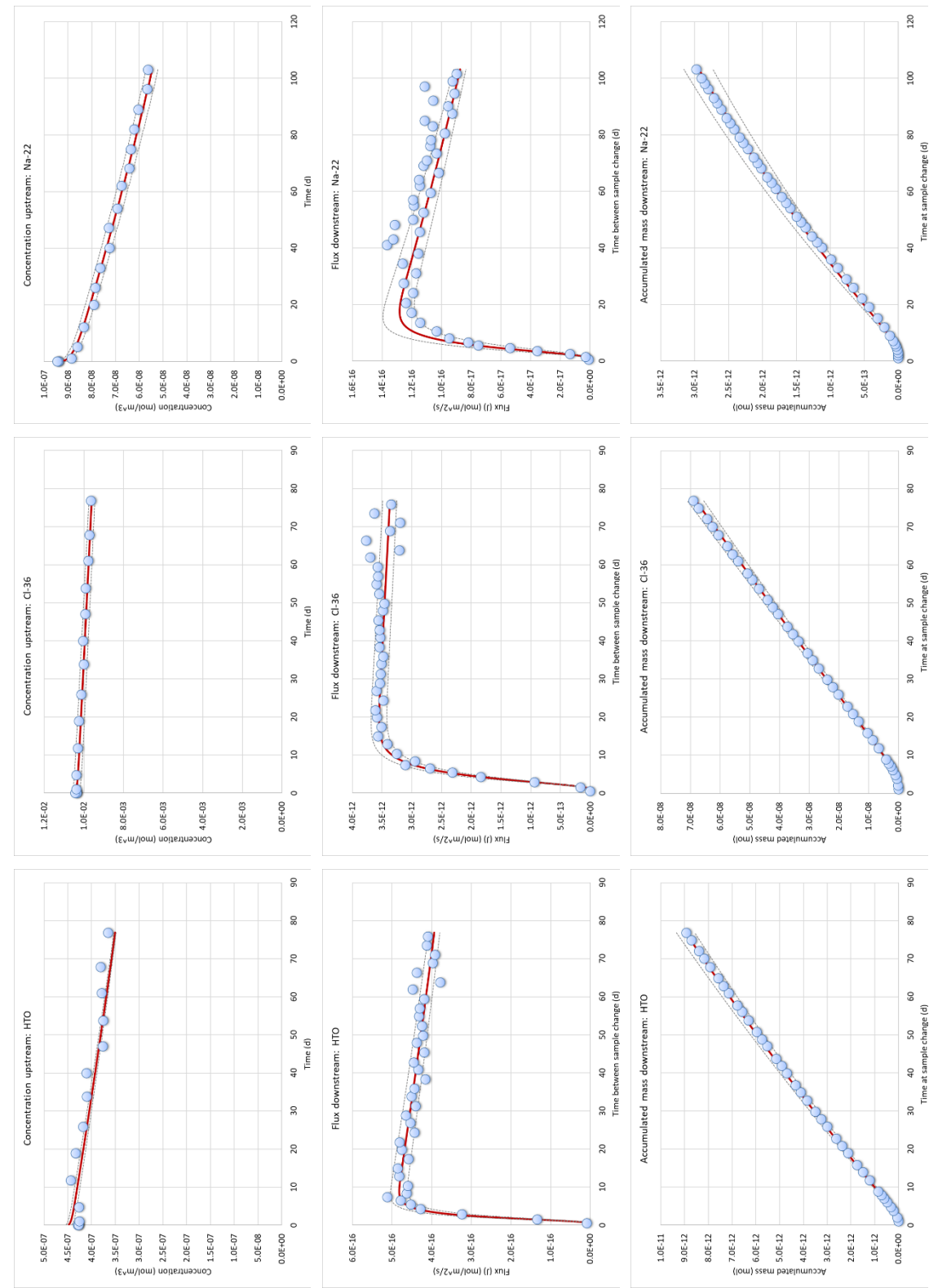
BOZ1-1-438.20-DI (S2) – Klingnau Formation – SPW-BOZ

Sample diameter 63.50 mm – sample thickness 12.09 mm



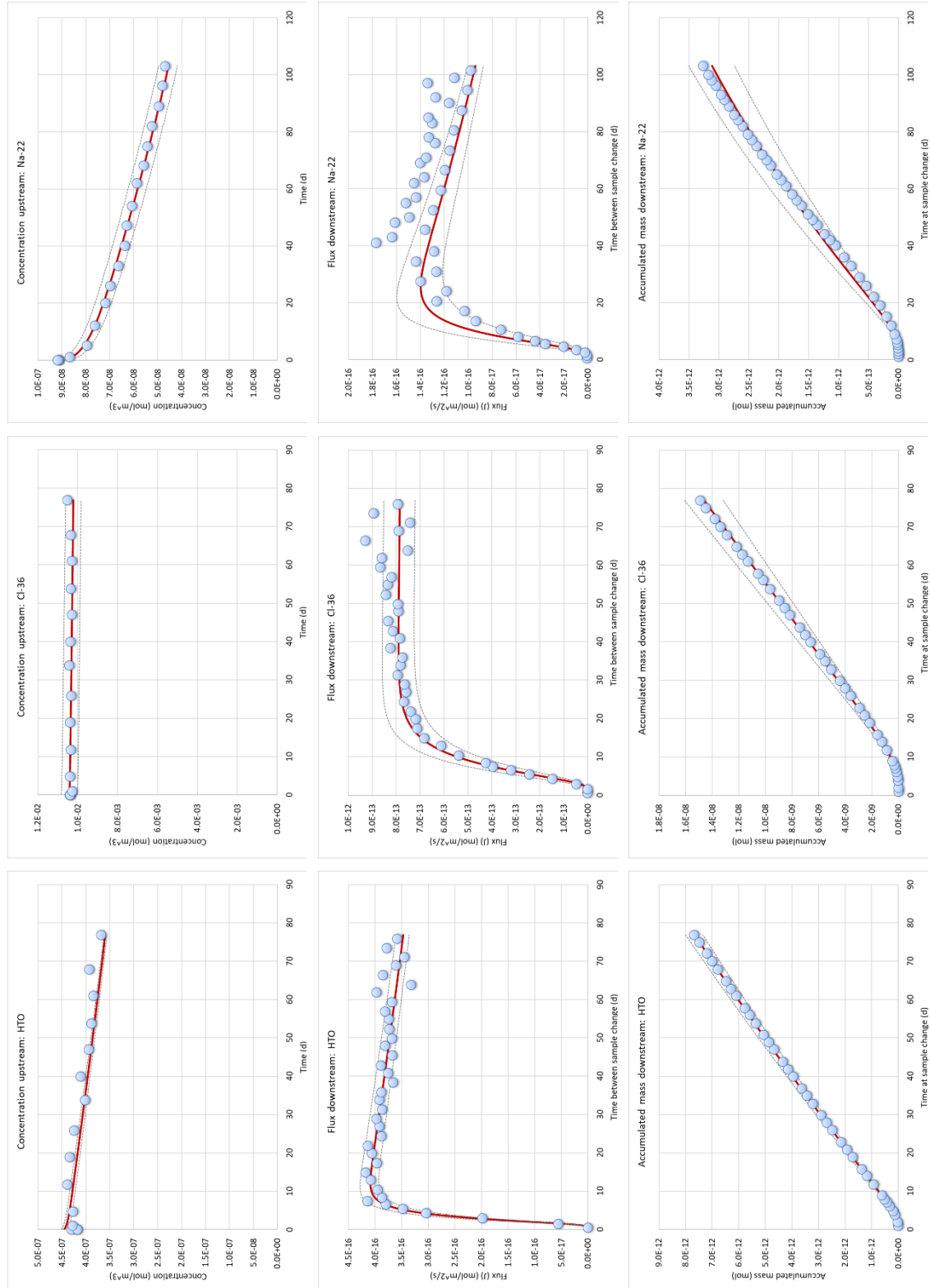
BOZ1-1-440.55-DI (S2) – Klingnau Formation – SPW-BOZ

Sample diameter 63.34 mm – sample thickness 11.88 mm



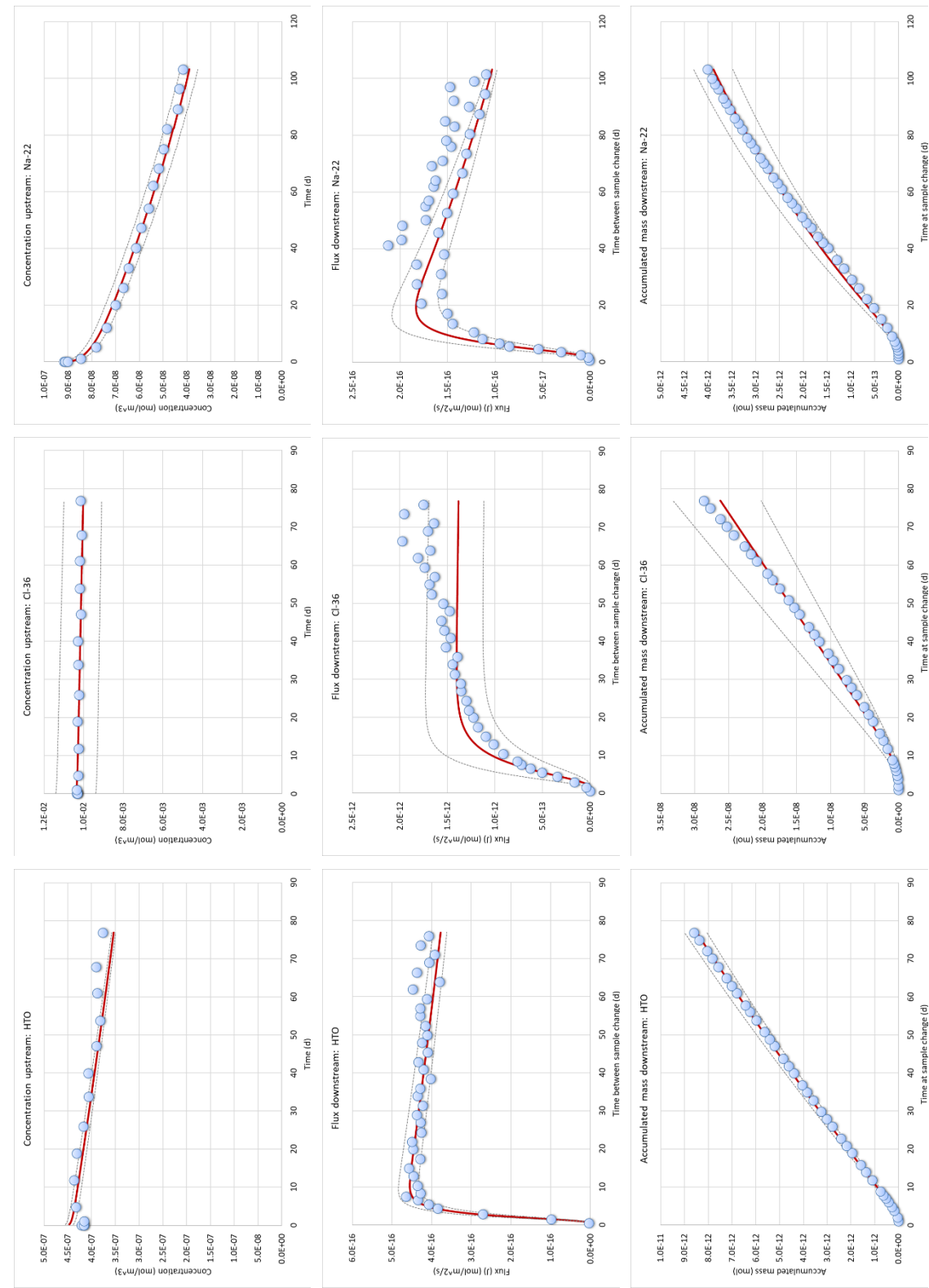
BOZ1-1-487.01-DI (S2) – Passwang Formation – SPW-BOZ

Sample diameter 63.29 mm – sample thickness 12.65 mm



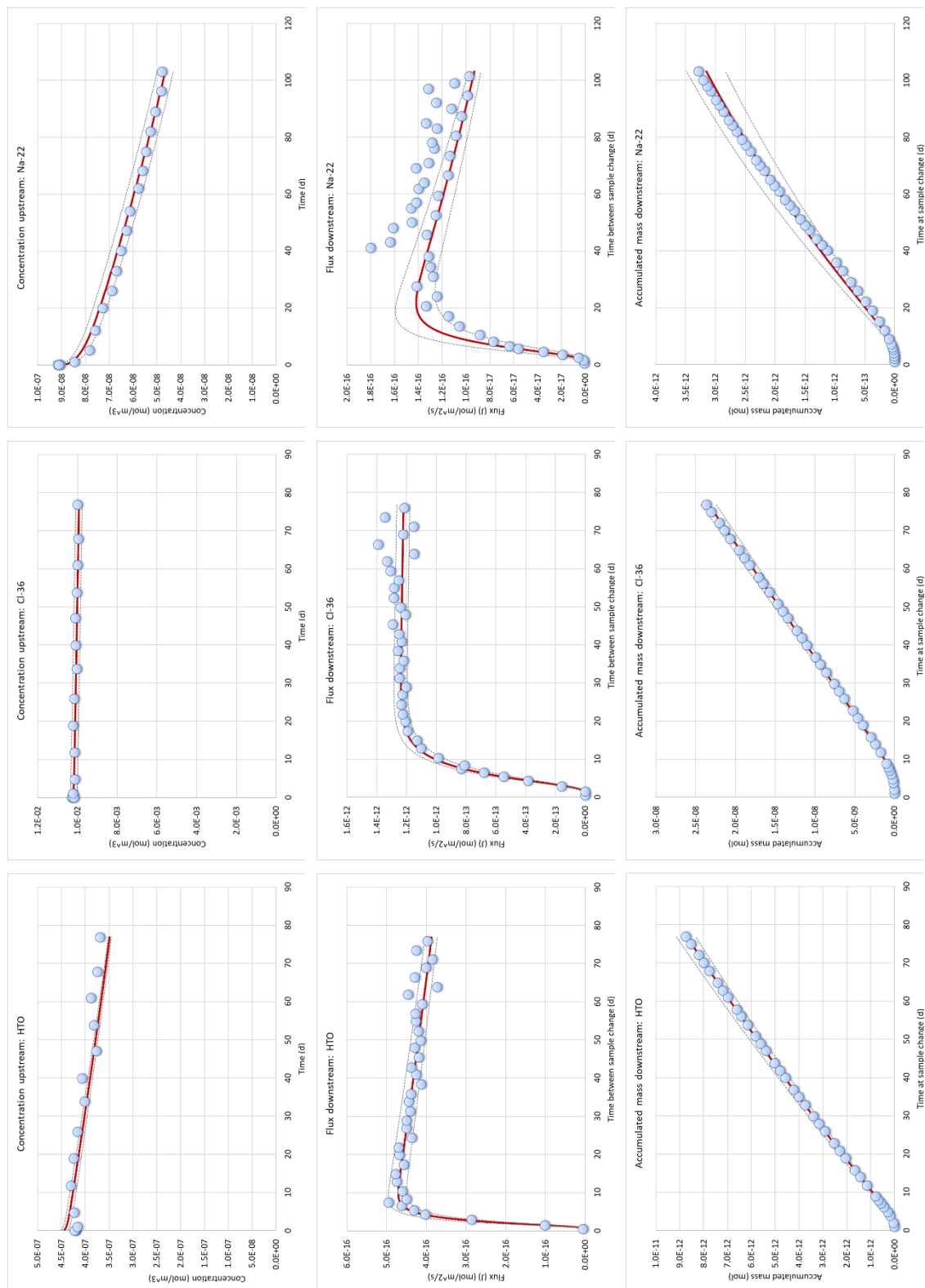
BOZ1-1-493.15-DI (S2) – Passwang Formation – SPW-BOZ

Sample diameter 63.33 mm – sample thickness 11.50 mm



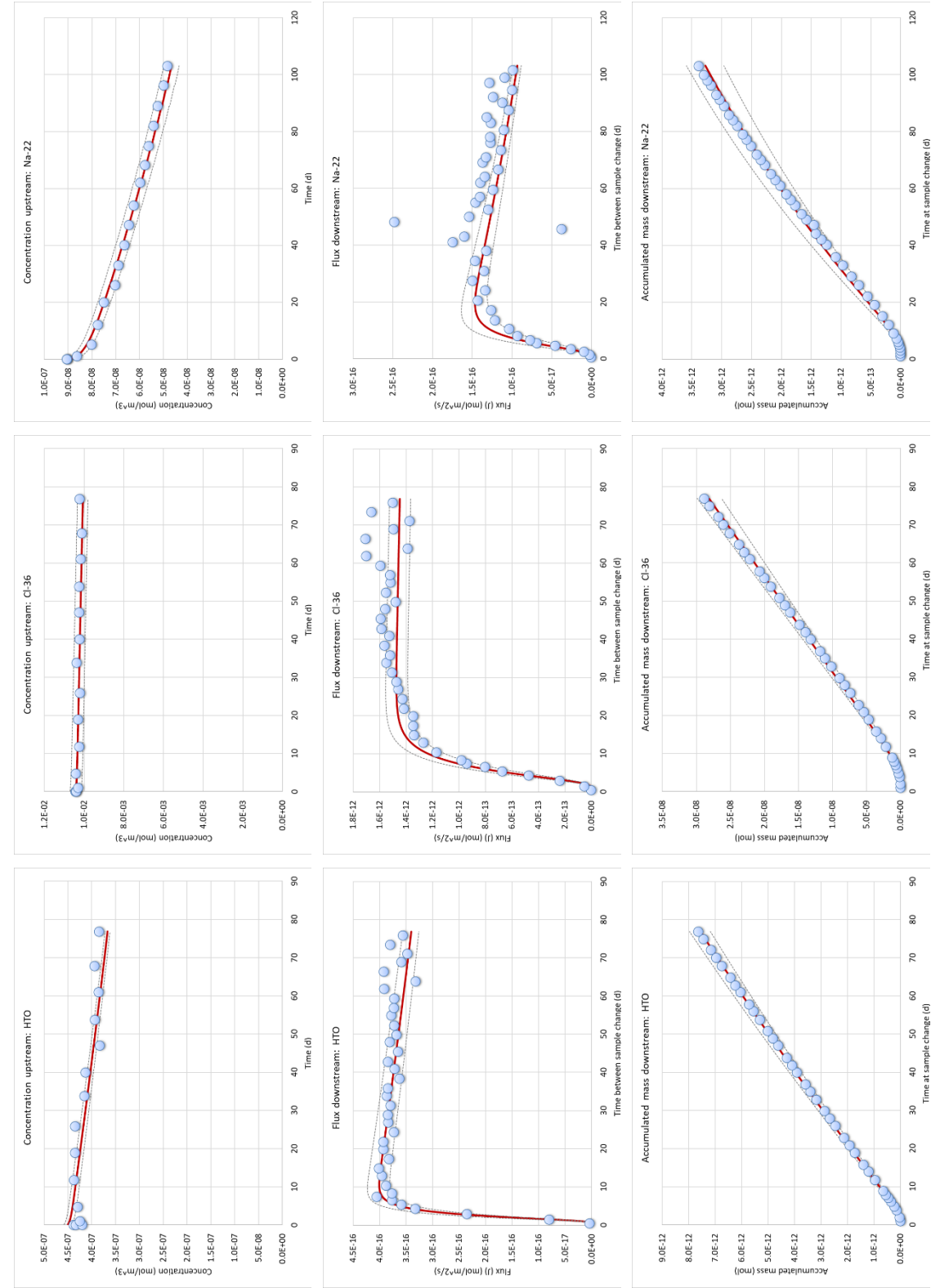
BOZ1-1-498.32-DI (S2) – Passwang Formation – SPW-BOZ

Sample diameter 63.20 mm – sample thickness 12.63 mm



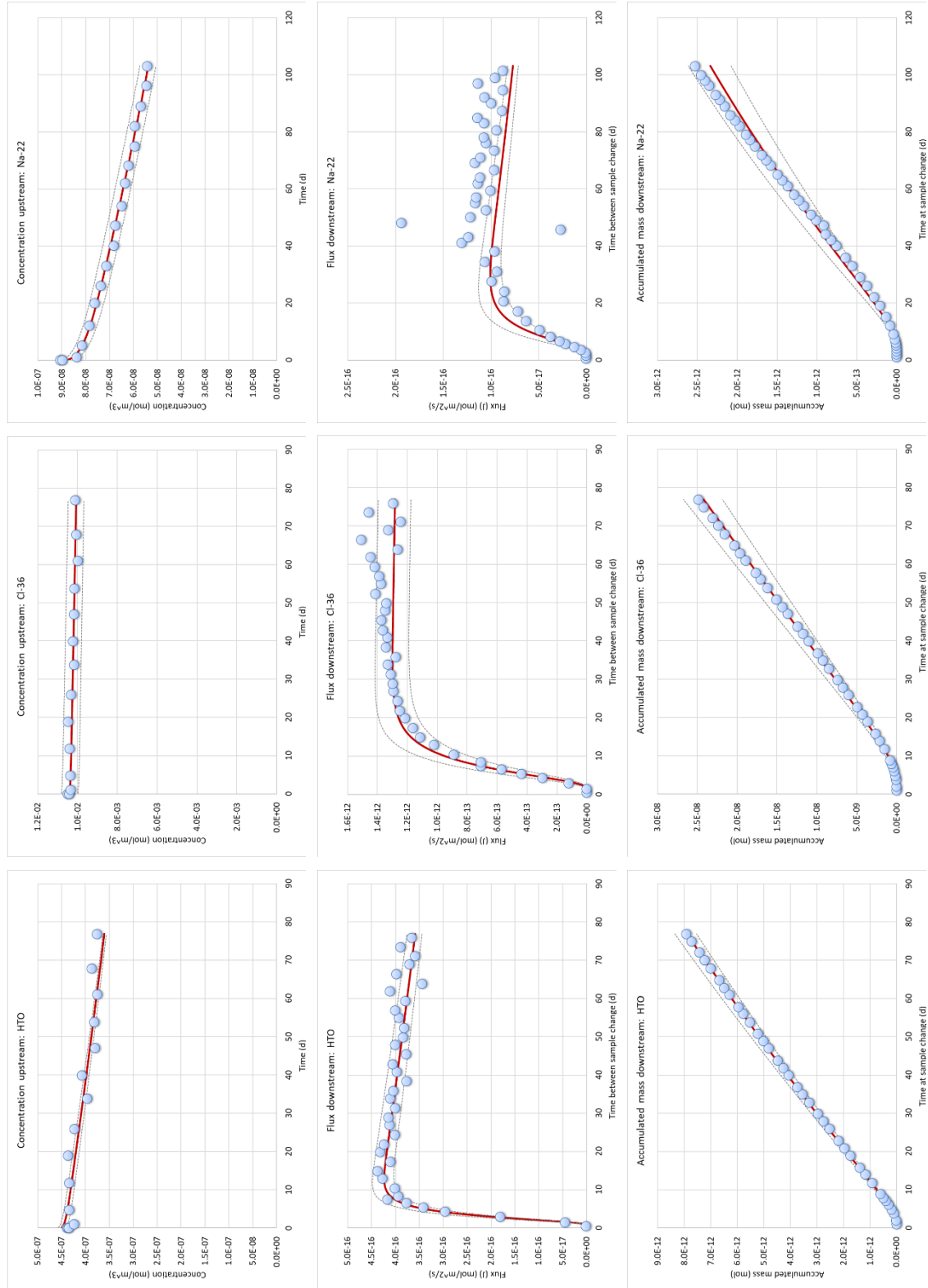
BOZ1-1-503.87-DI (S2) – Passwang Formation – SPW-BOZ

Sample diameter 63.42 mm – sample thickness 13.05 mm



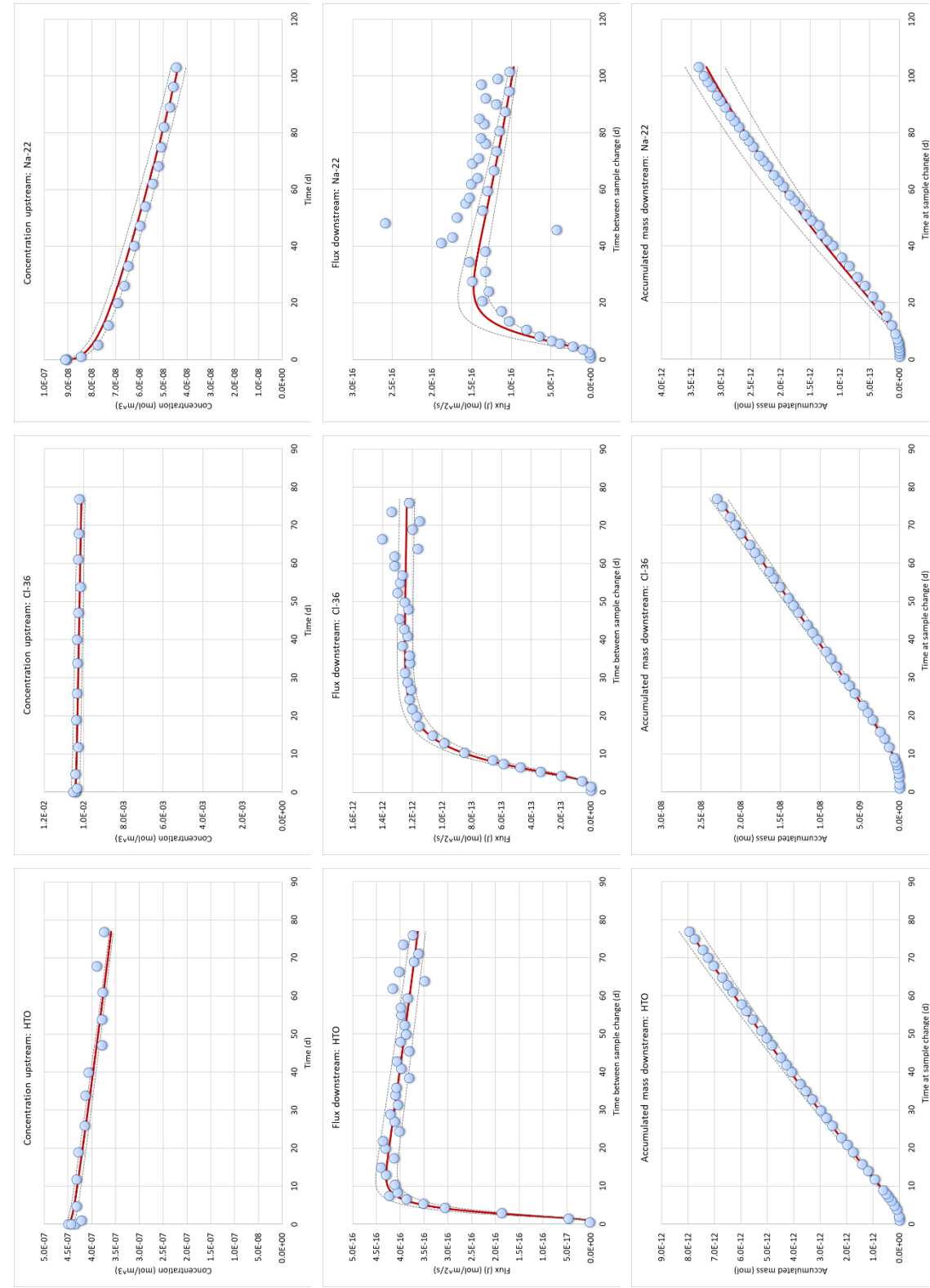
BOZ1-1-512.03-DI (S1) – Passwang Formation – SPW-BOZ

Sample diameter 63.32 mm – sample thickness 14.27 mm



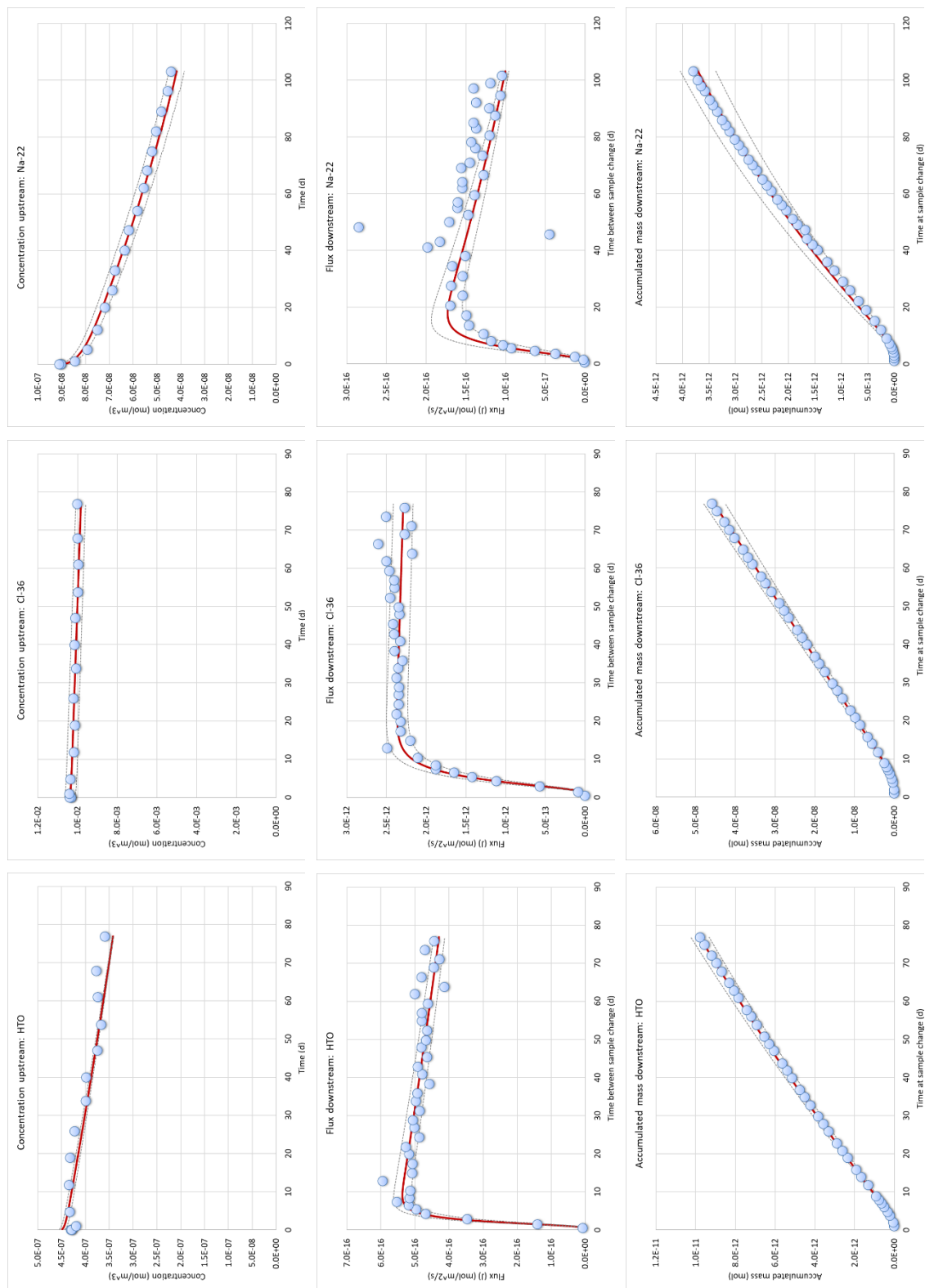
BOZ1-1-521.50-DI (S2) – Passwang Formation – SPW-BOZ

Sample diameter 63.05 mm – sample thickness 14.05 mm



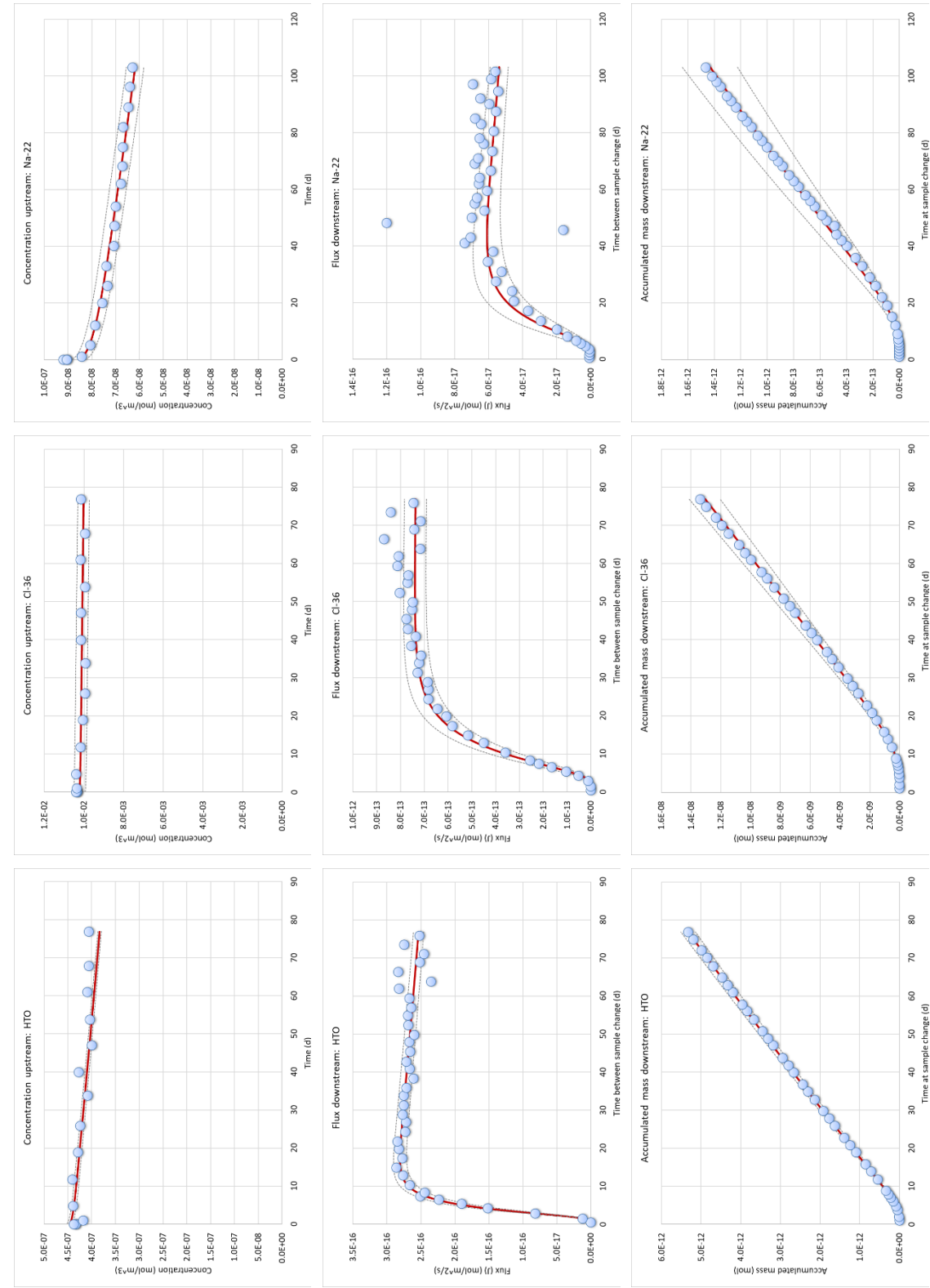
BOZ1-1-529.56-DI (S1) – Passwang Formation – SPW-BOZ

Sample diameter 63.10 mm – sample thickness 12.38 mm



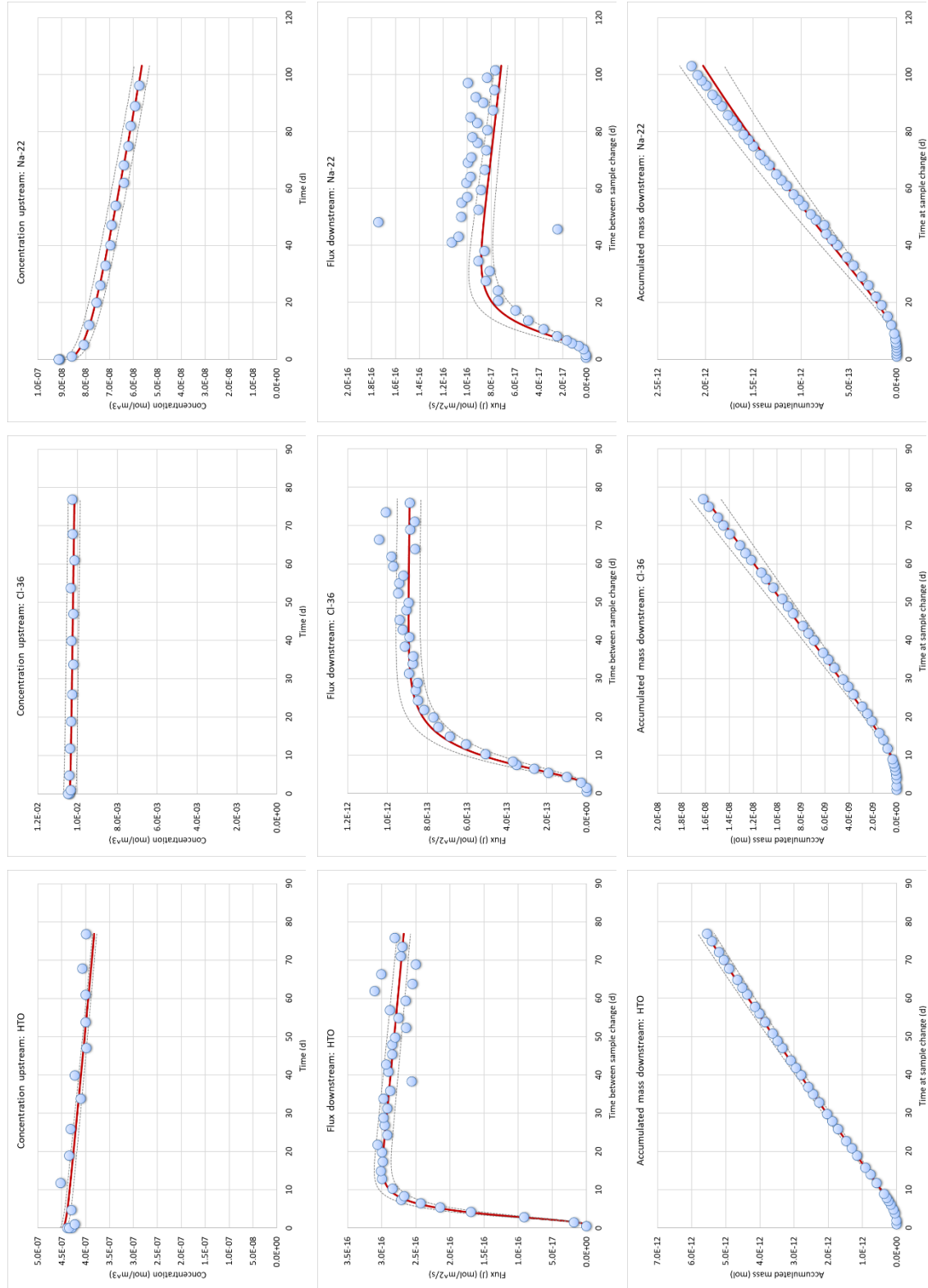
BOZ1-1-550.63-DI (S1) – Opalinus Clay – SPW-BOZ

Sample diameter 63.43 mm – sample thickness 13.08 mm



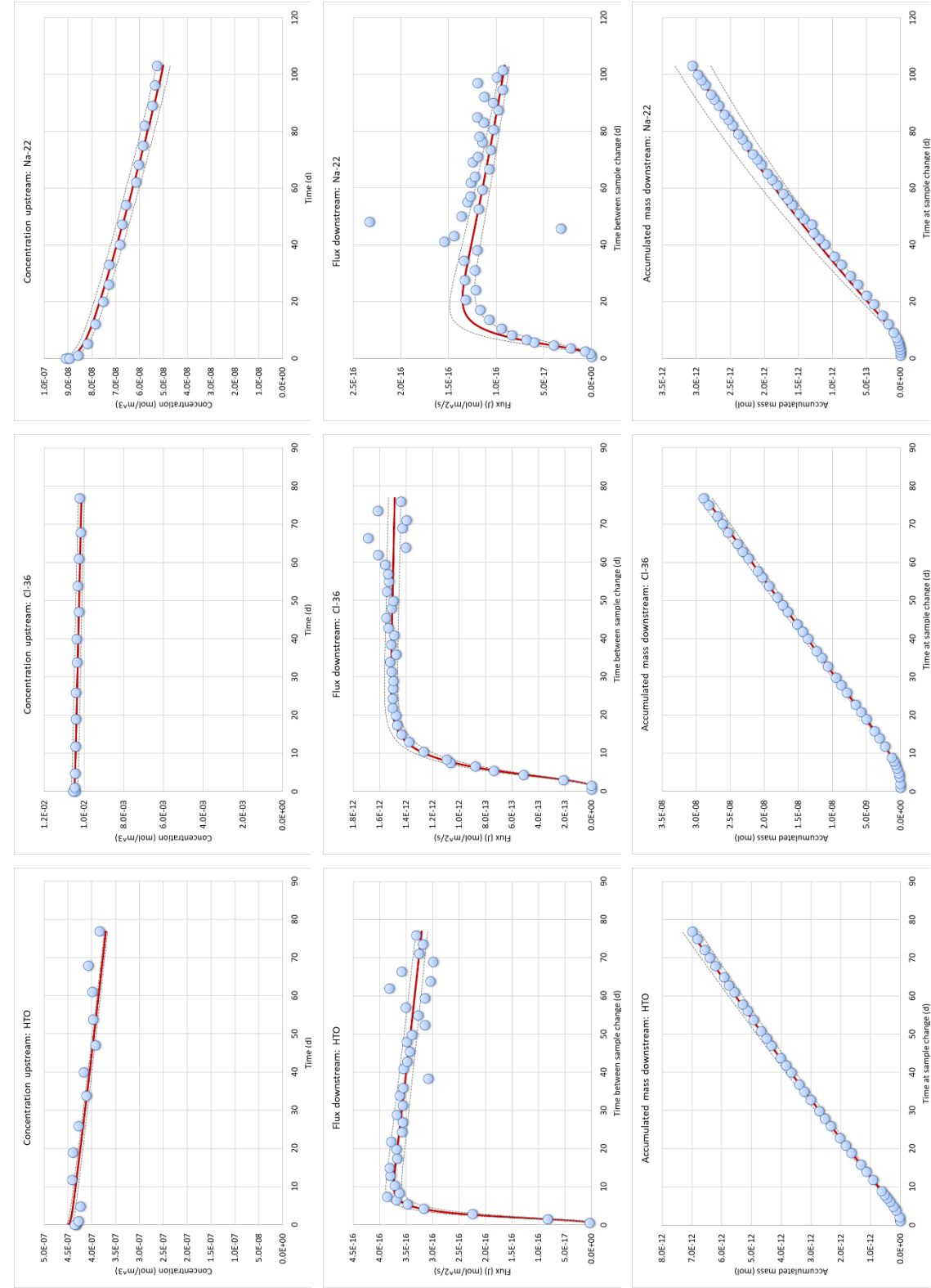
BOZ1-1-562.07-DI (S1) – Opalinus Clay – SPW-BOZ

Sample diameter 62.90 mm – sample thickness 13.20 mm



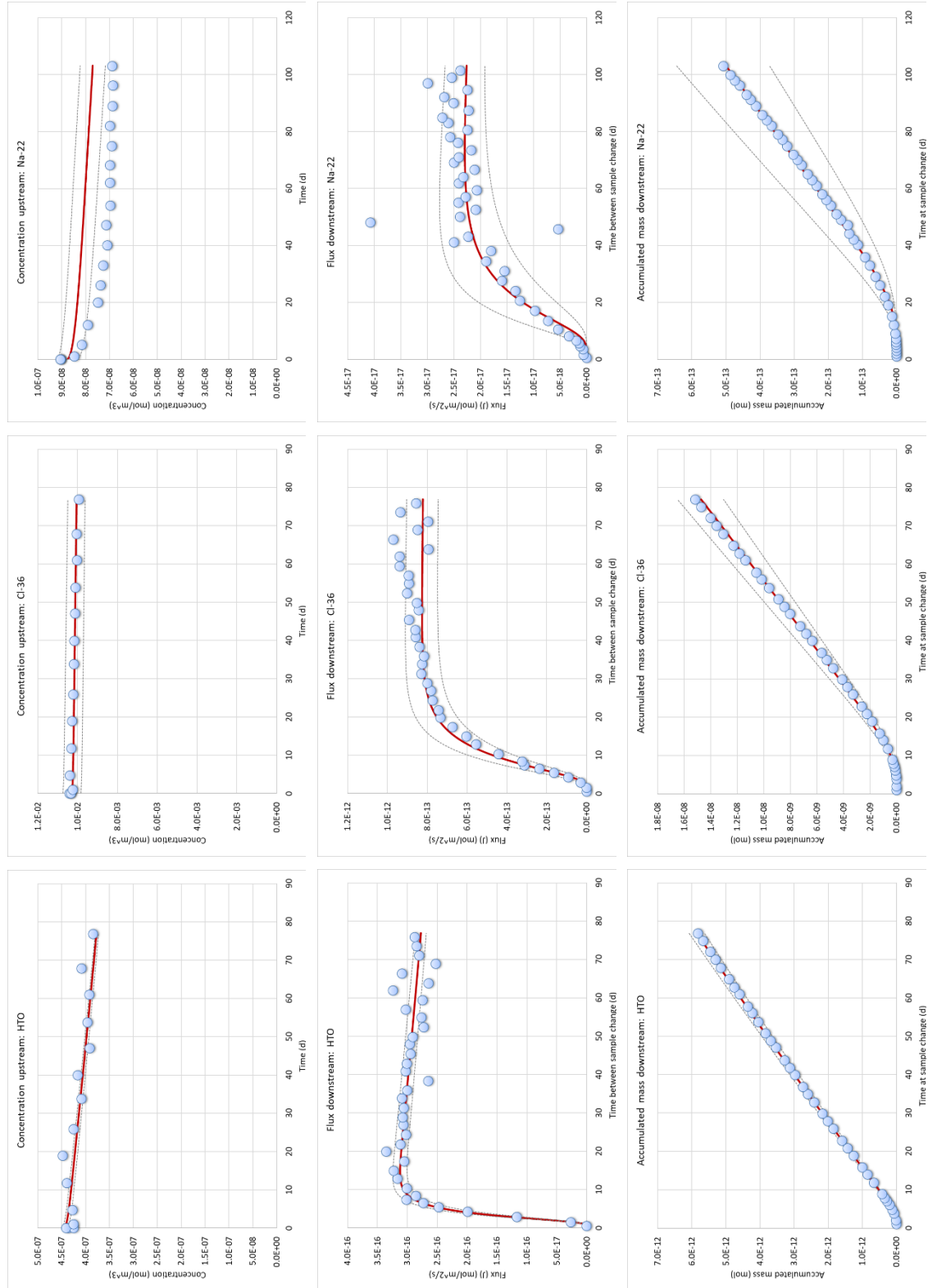
BOZ1-1-572.43-DI (S2) – Opalinus Clay – SPW-BOZ

Sample diameter 63.20 mm – sample thickness 12.53 mm



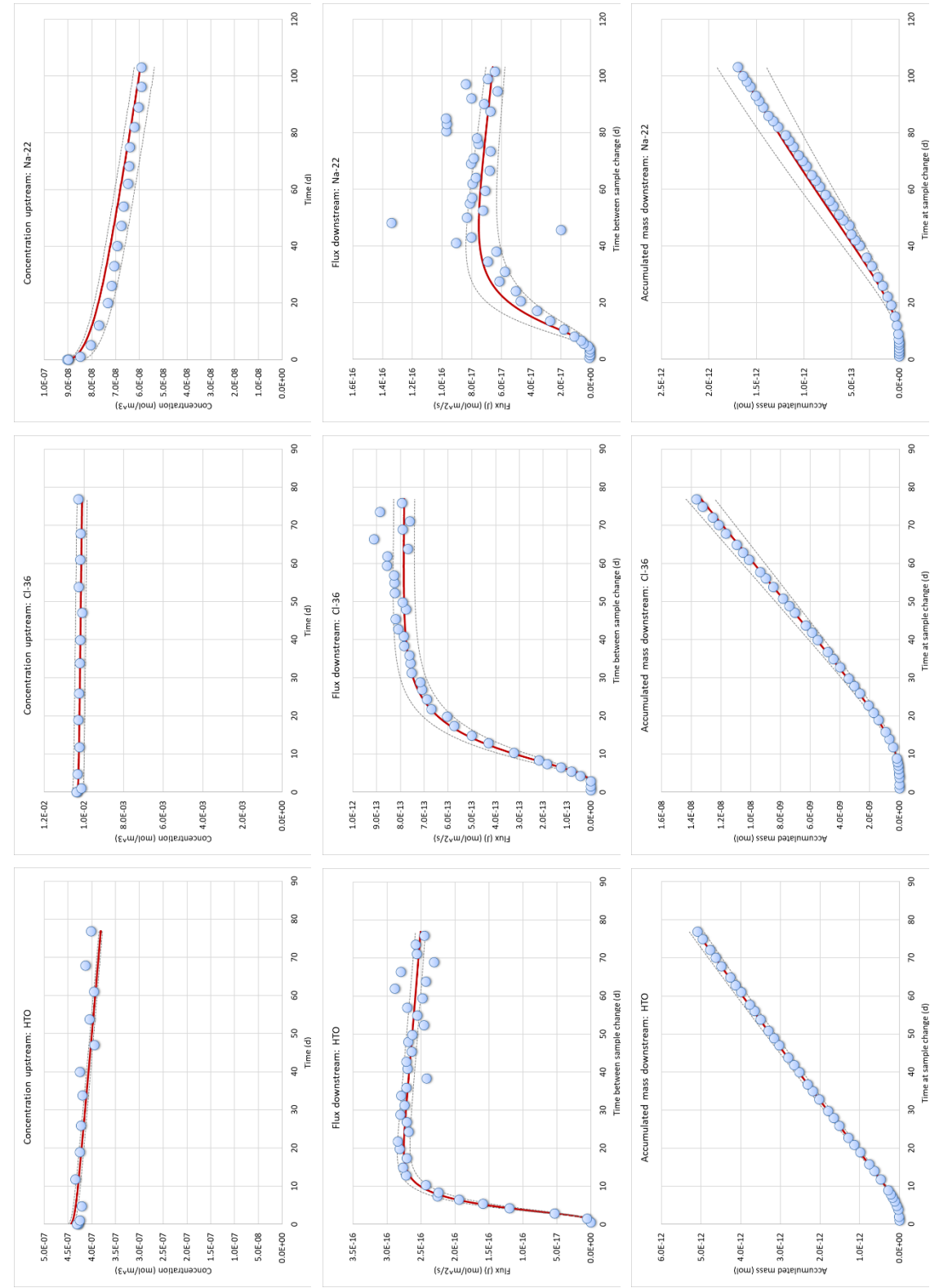
BOZ1-1-580.34-DI (S1) – Opalinus Clay – SPW-BOZ

Sample diameter 62.98 mm – sample thickness 12.12 mm



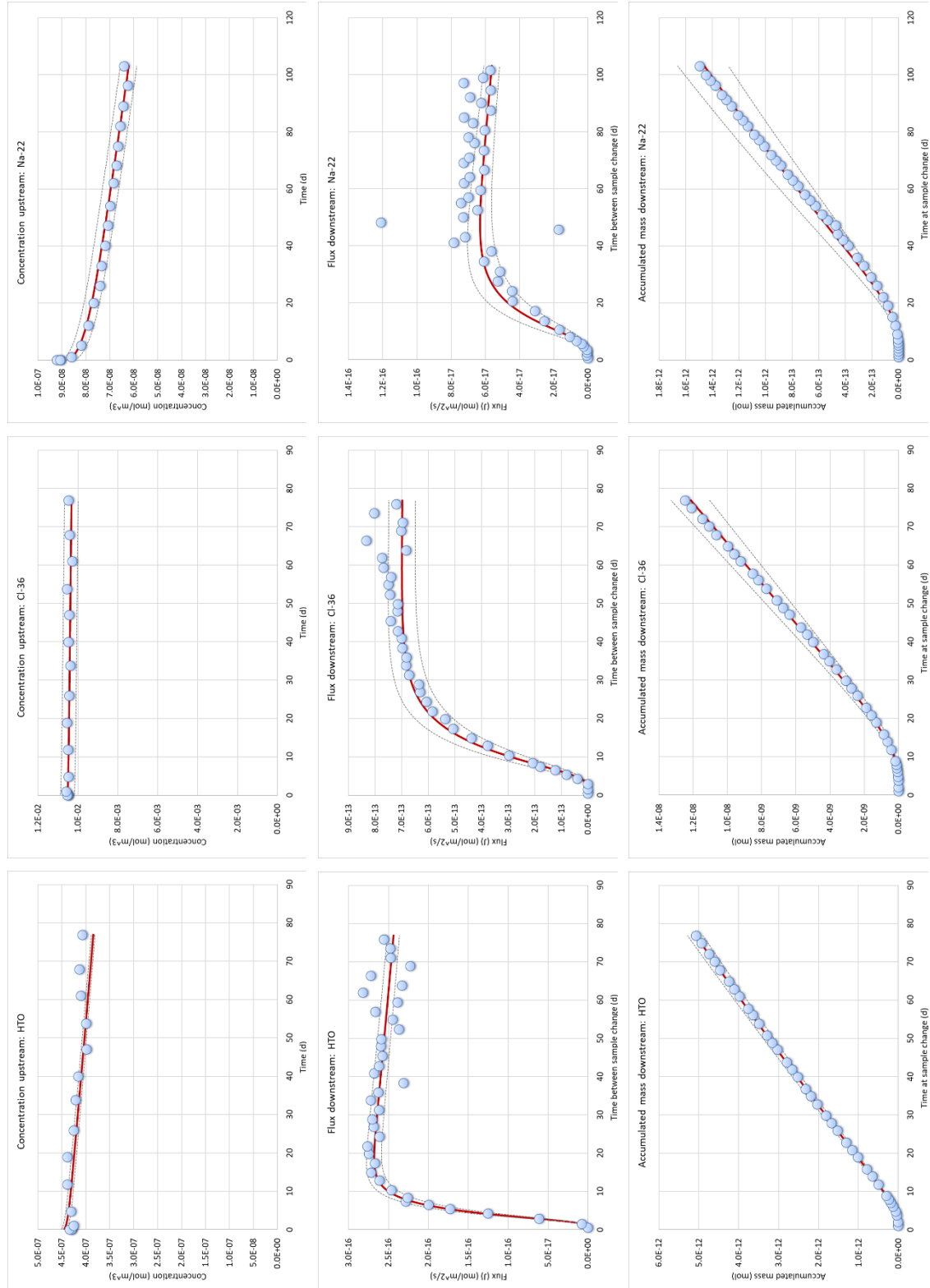
BOZ1-1-597.28-DI (S2) – Opalinus Clay – SPW-BOZ

Sample diameter 62.96 mm – sample thickness 13.75 mm



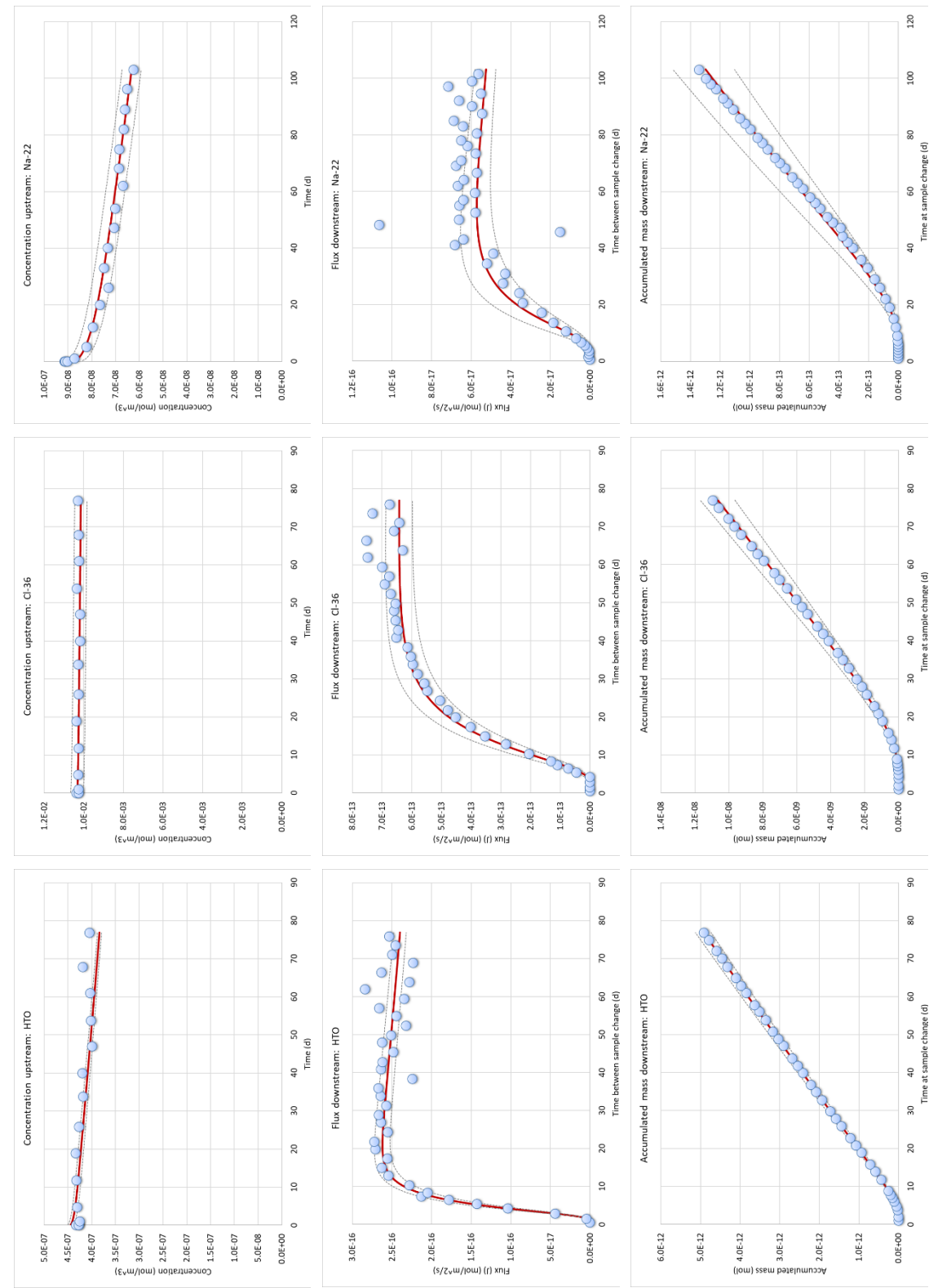
BOZ1-1-610.79-DI (S1) – Opalinus Clay – SPW-BOZ

Sample diameter 63.50 mm – sample thickness 12.47 mm



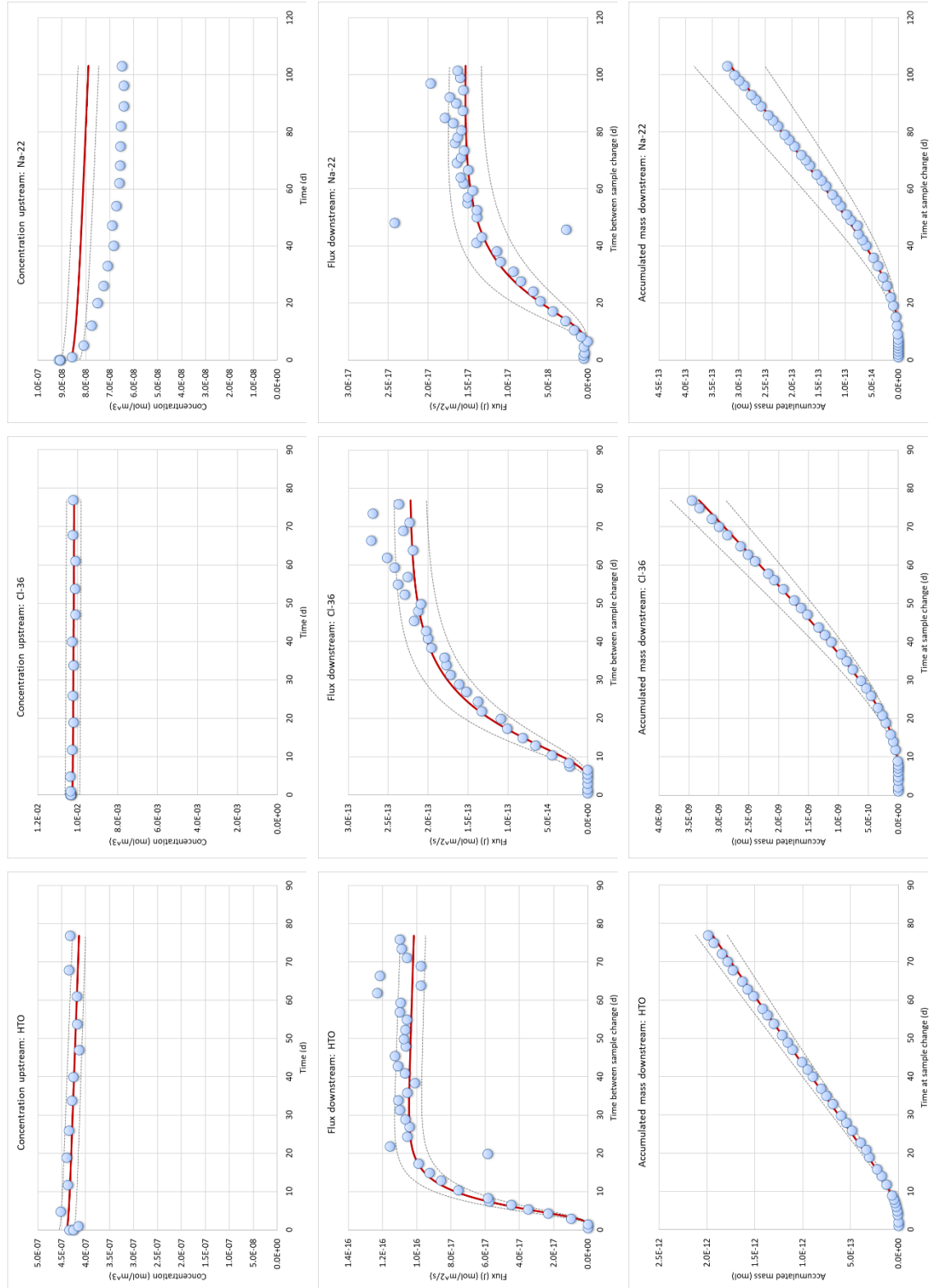
BOZ1-1-619.10-DI (S2) – Opalinus Clay – SPW-BOZ

Sample diameter 63.35 mm – sample thickness 13.15 mm



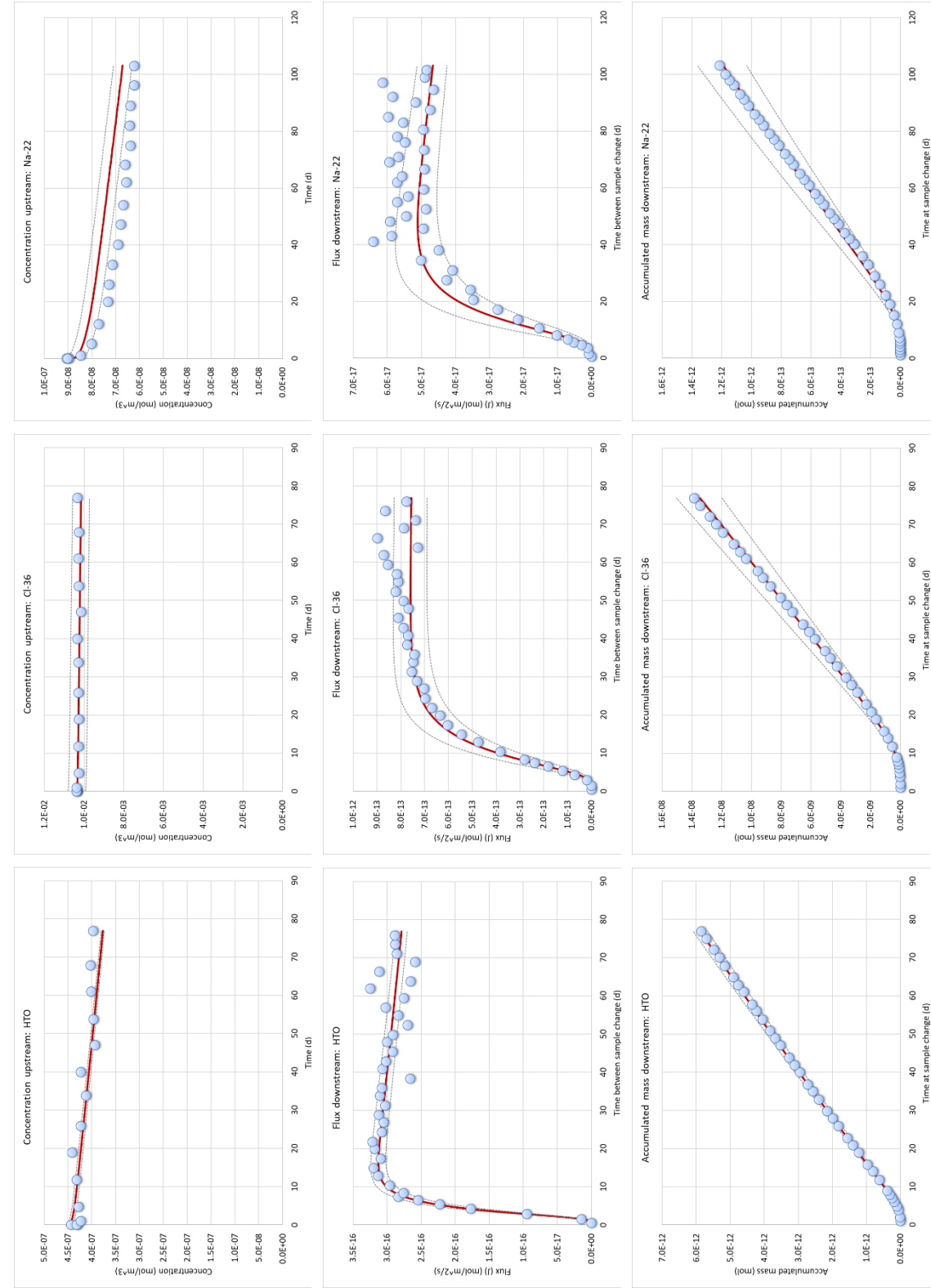
BOZ1-1-623.60-DI (S1) – Opalinus Clay – SPW-BOZ

Sample diameter 63.28 mm – sample thickness 12.80 mm



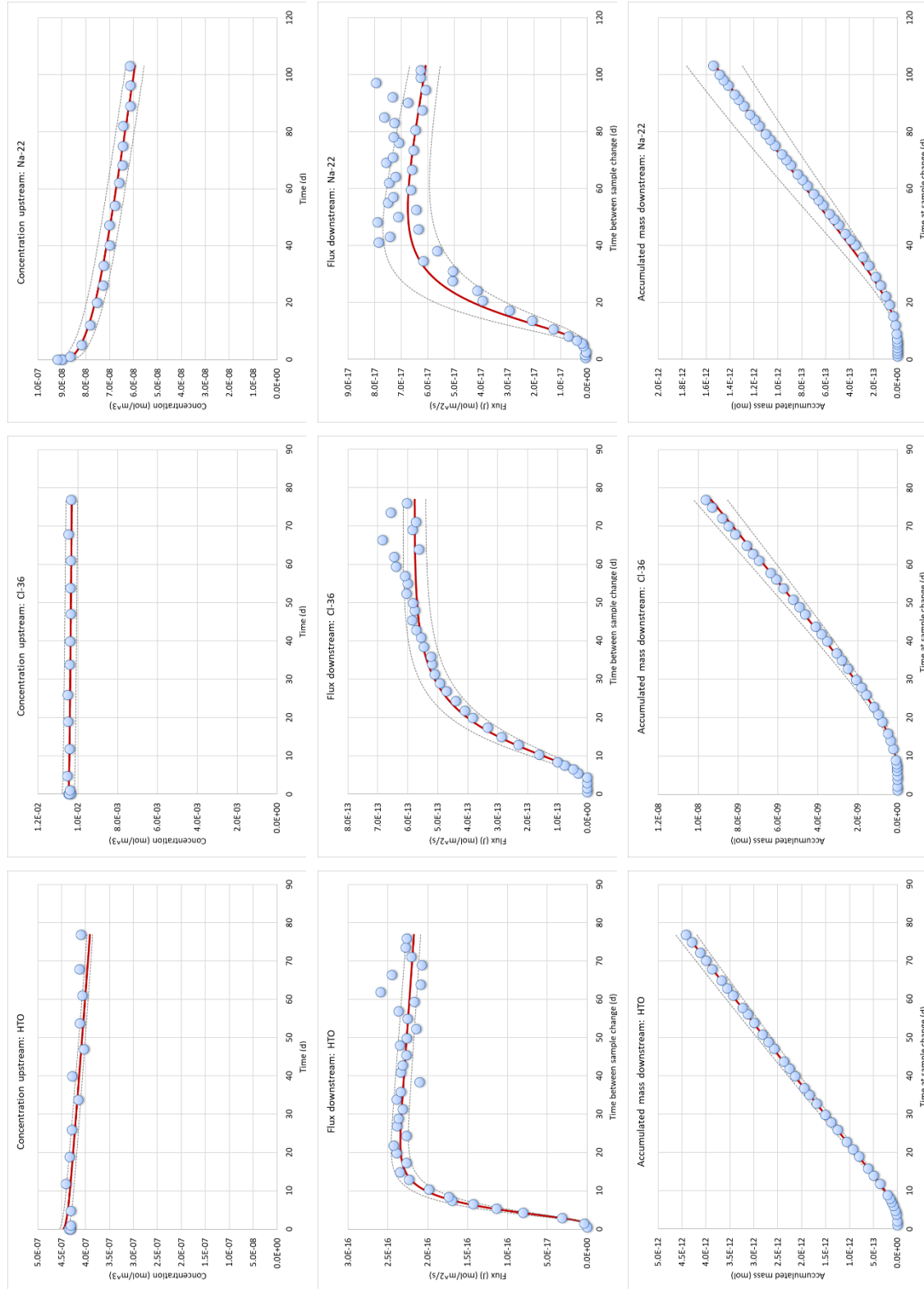
BOZ1-1-634.90-DI (S2) – Opalinus Clay – SPW-BOZ

Sample diameter 63.22 mm – sample thickness 11.46 mm



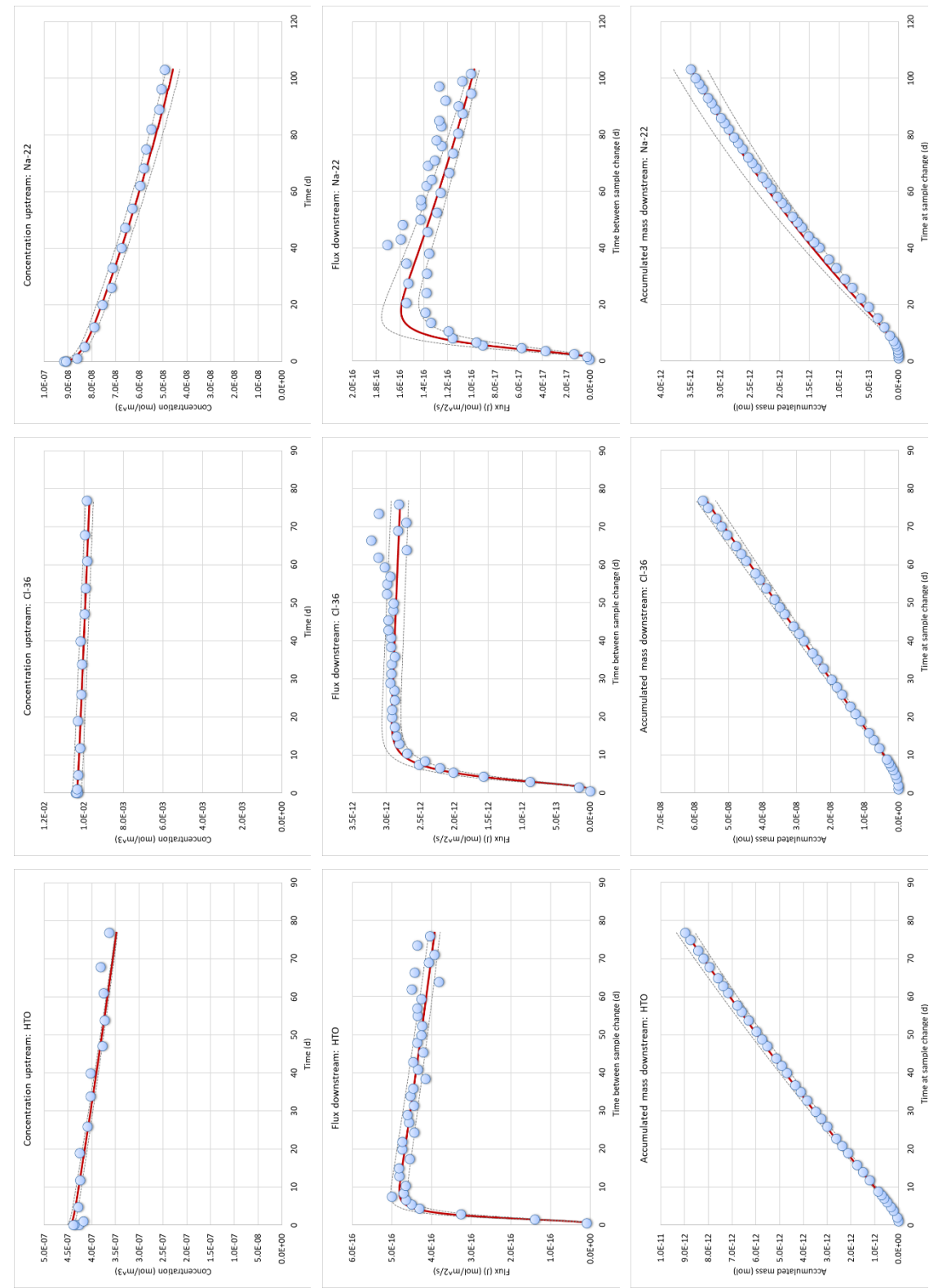
BOZ1-1-646.44-DI (S1) – Opalinus Clay – SPW-BOZ

Sample diameter 63.40 mm – sample thickness 13.50 mm



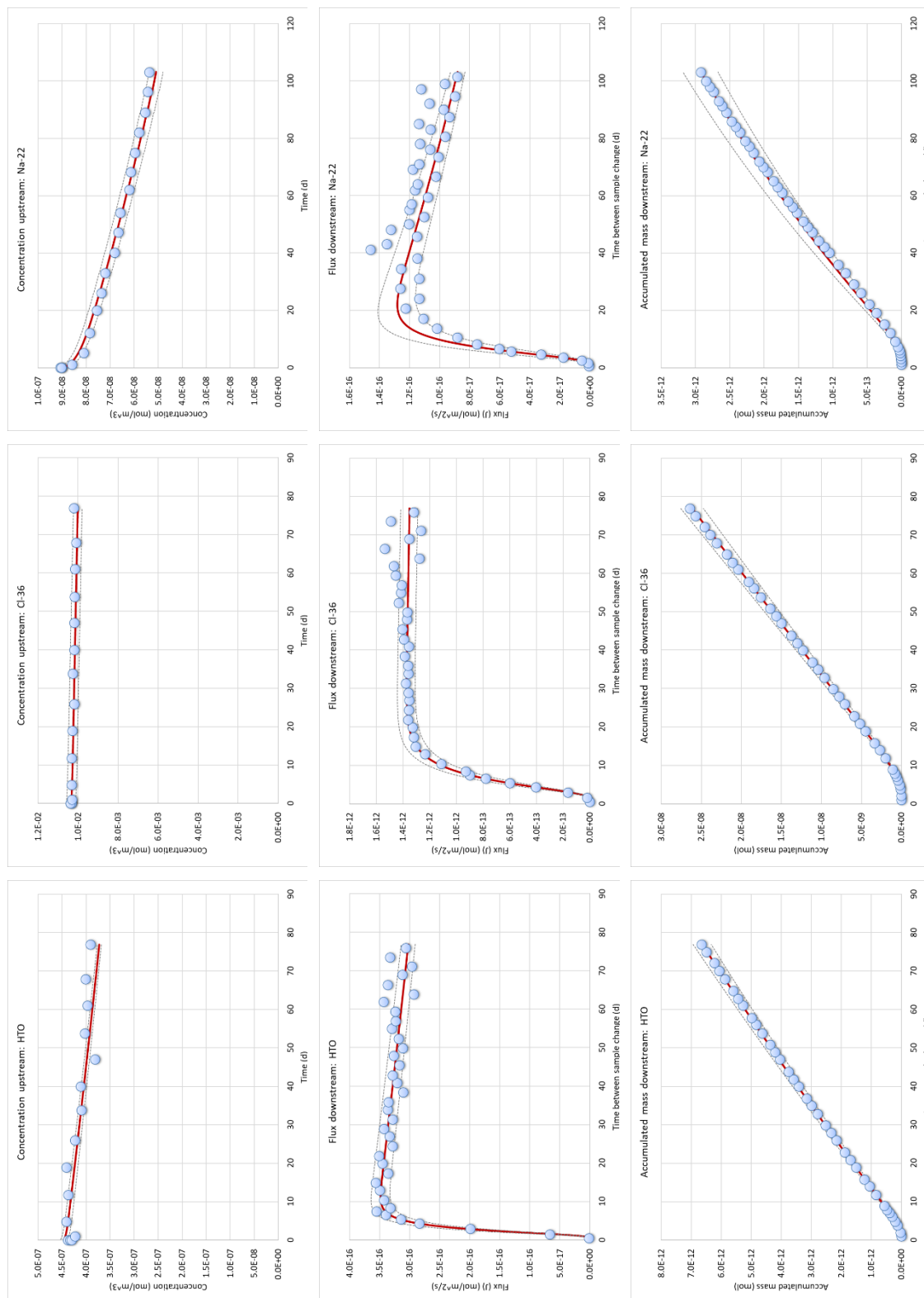
BOZ1-1-665.00-DI (S2) – Staffelegg Formation – SPW-BOZ

Sample diameter 63.37 mm – sample thickness 14.16 mm



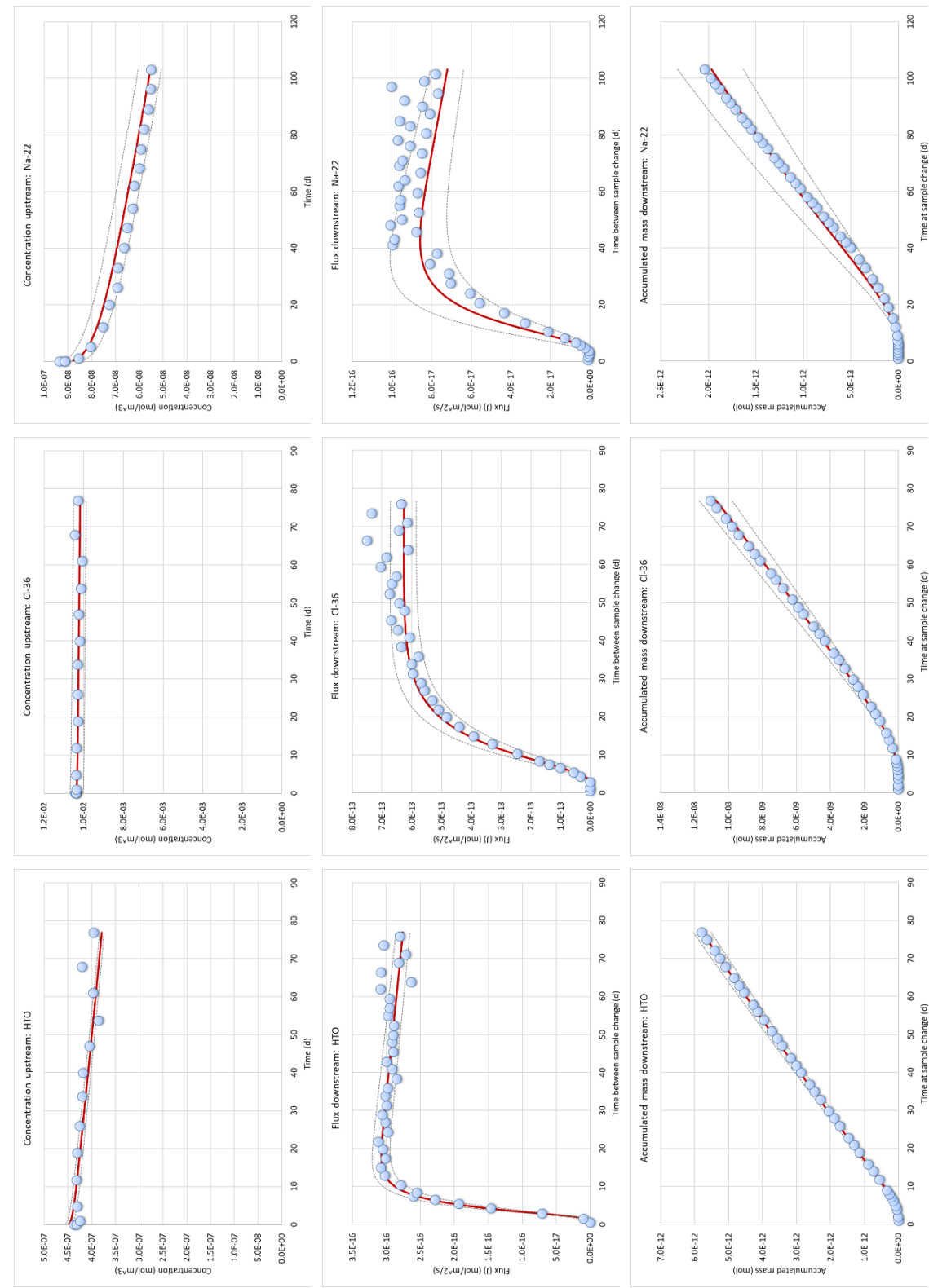
BOZ1-1-670.95-DI (S1) – Staffelegg Formation – SPW-BOZ

Sample diameter 63.42 mm – sample thickness 12.76 mm



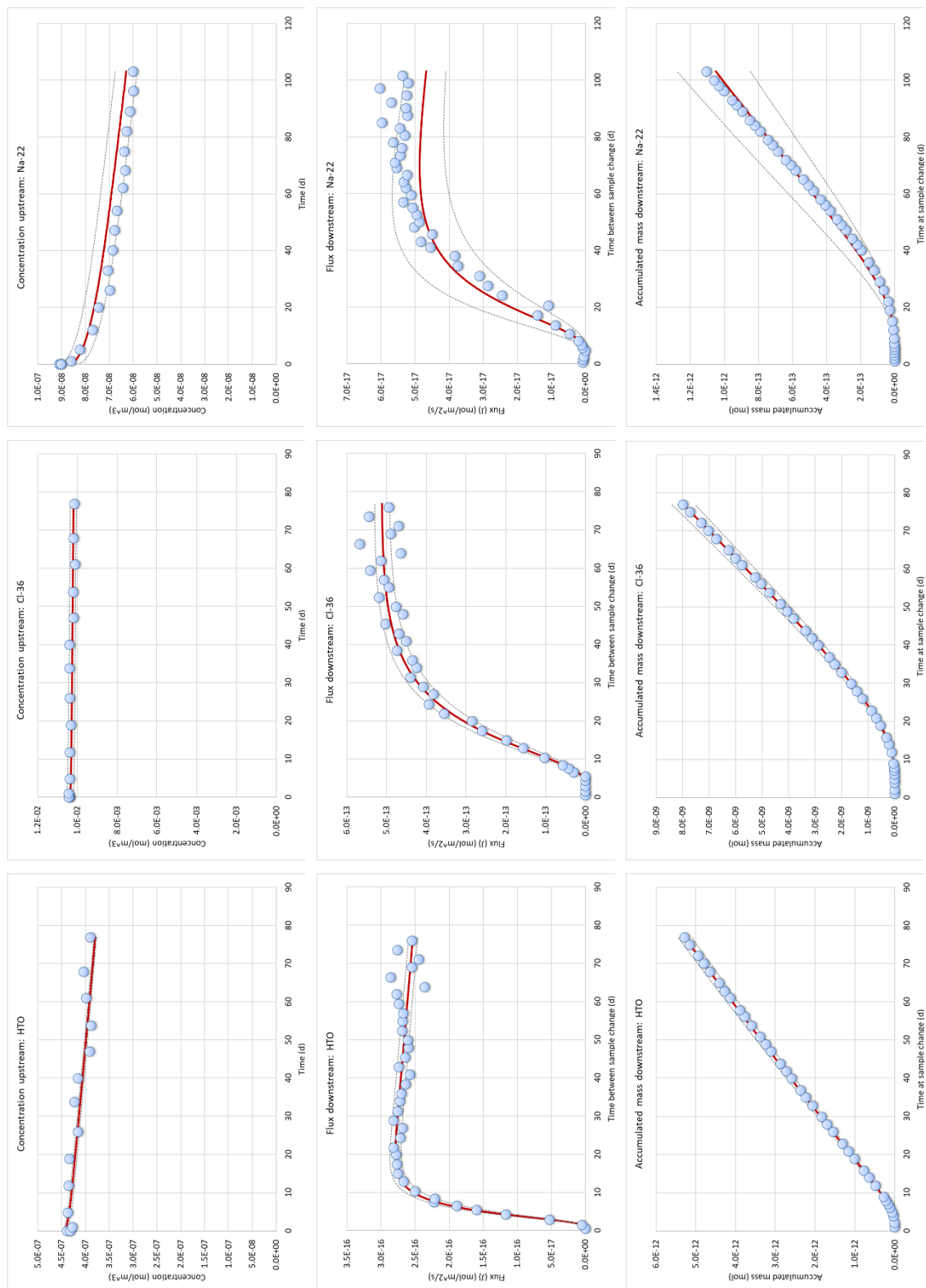
BOZ1-1-679.67-DI (S1) – Staffelegg Formation – SPW-BOZ

Sample diameter 63.36 mm – sample thickness 12.59 mm



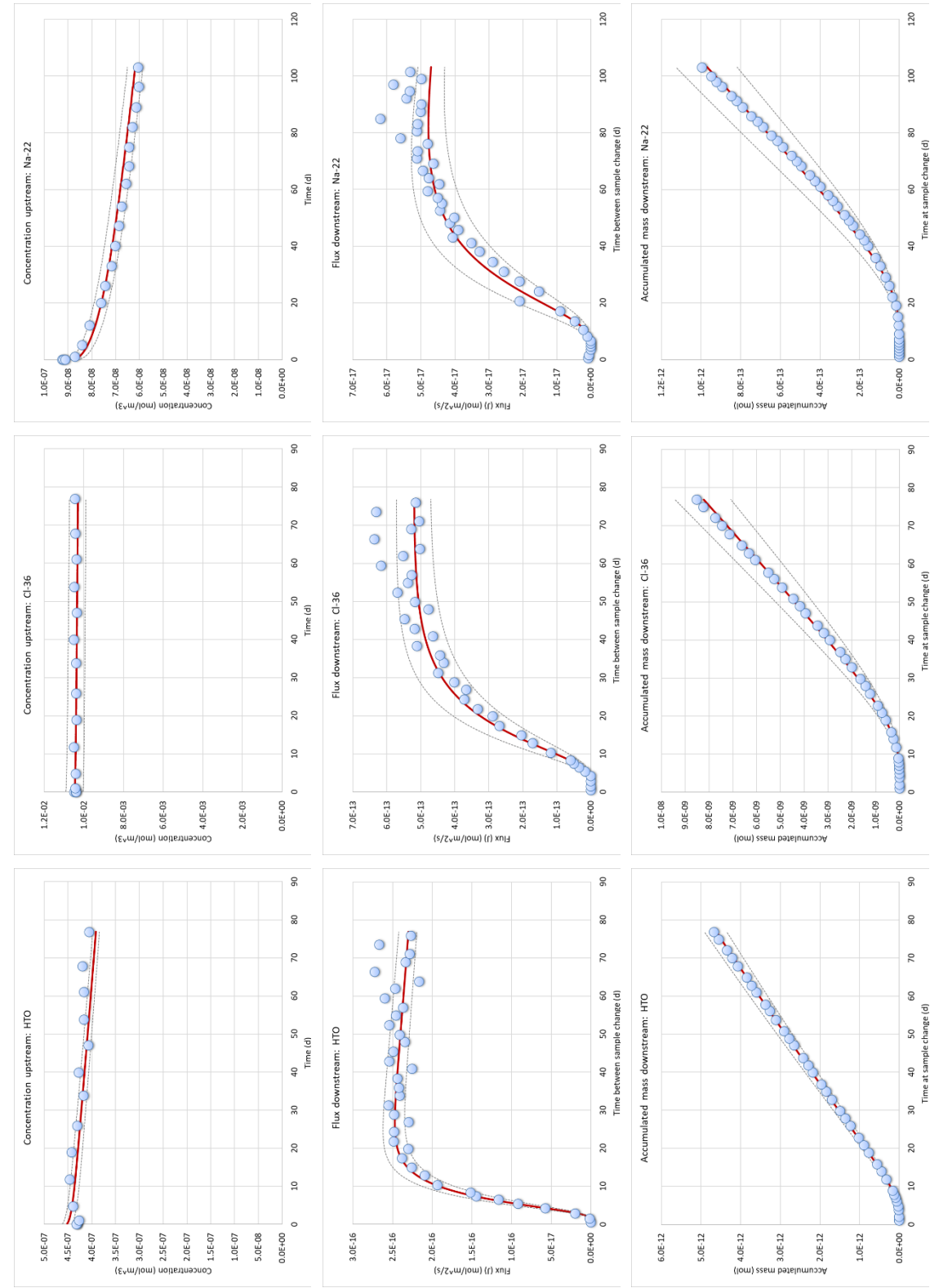
BOZ1-1-681.57-DI (S2) – Staffelegg Formation – SPW-BOZ

Sample diameter 63.65 mm – sample thickness 11.68 mm



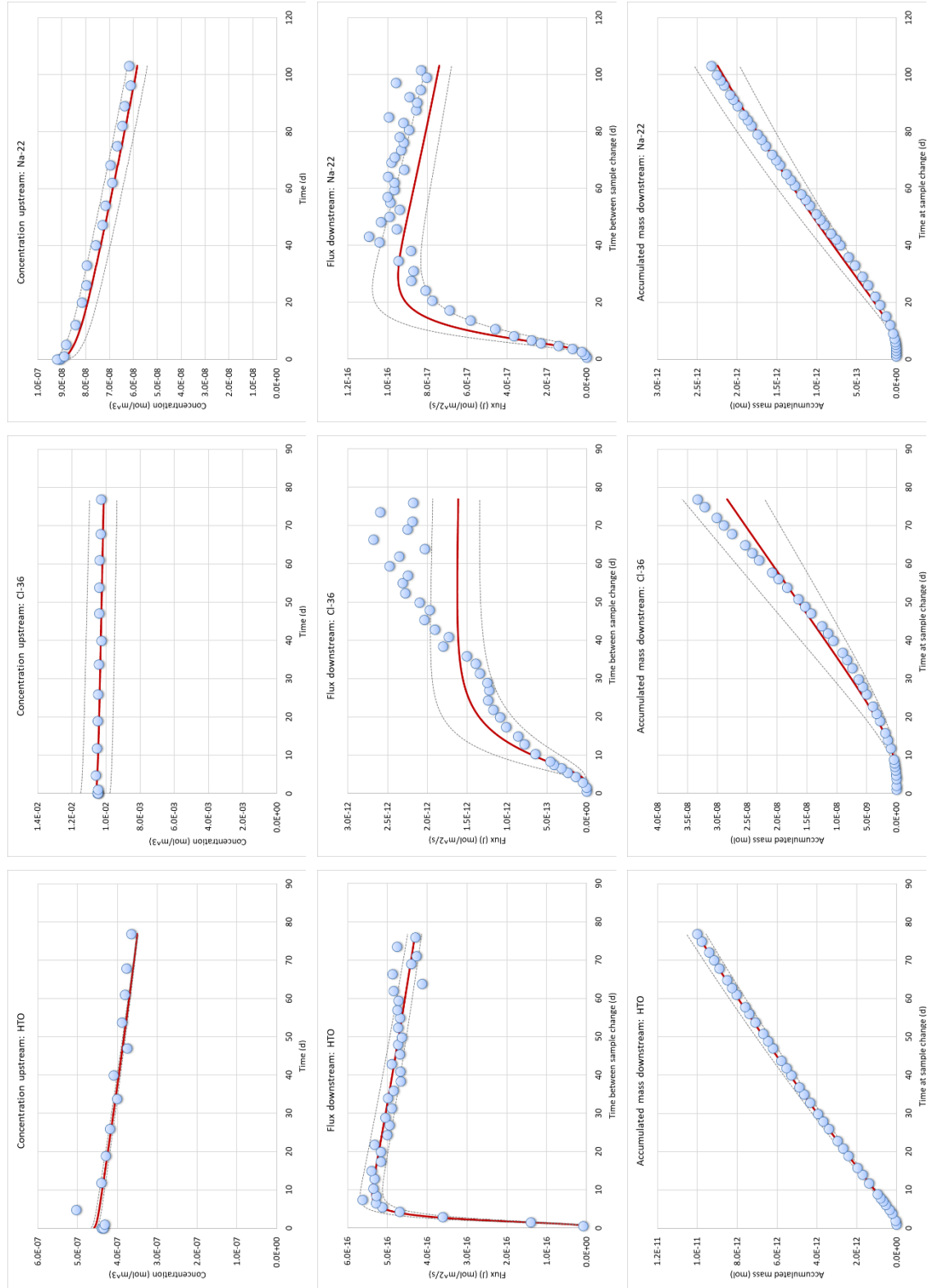
BOZ1-1-686.81-DI (S1) – Staffelegg Formation – SPW-BOZ

Sample diameter 63.69 mm – sample thickness 13.36 mm



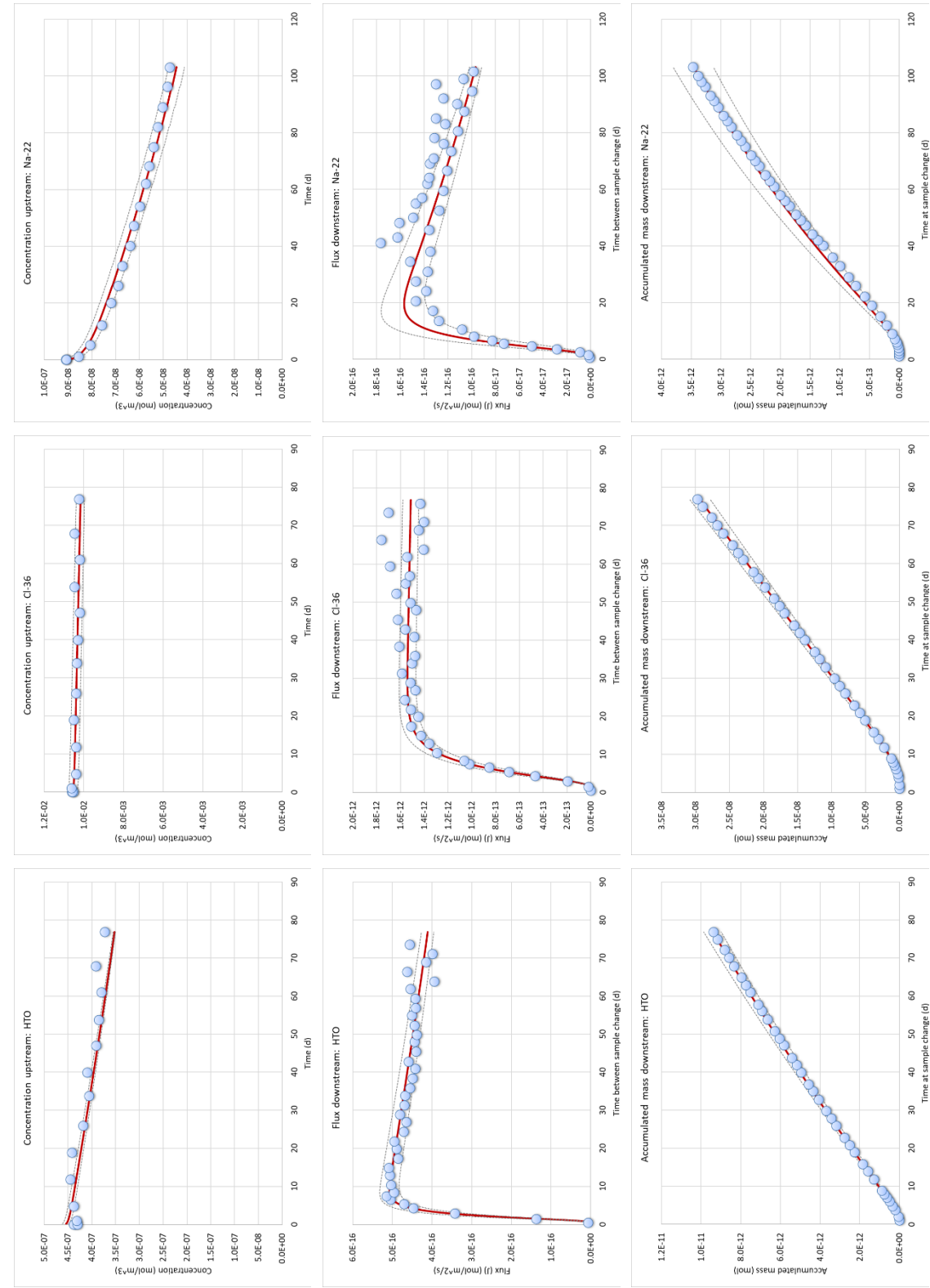
BOZ1-1-717.96-DI (S1) – Klettgau Formation – SPW-BOZ

Sample diameter 63.71 mm – sample thickness 12.10 mm



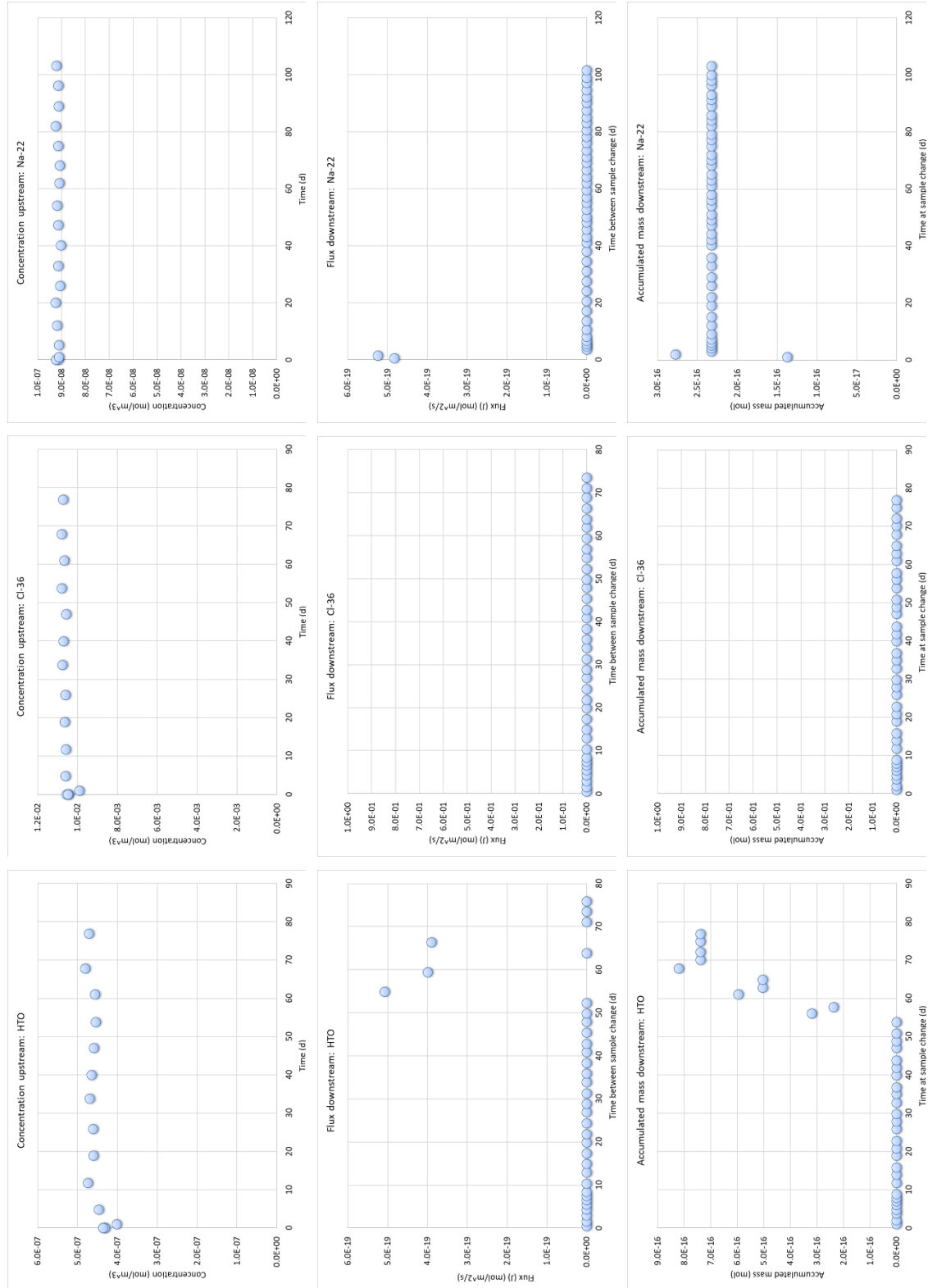
BOZ1-1-723.97-DI (S1) – Bänkerjoch Formation – SPW-BOZ

Sample diameter 63.49 mm – sample thickness 12.69 mm



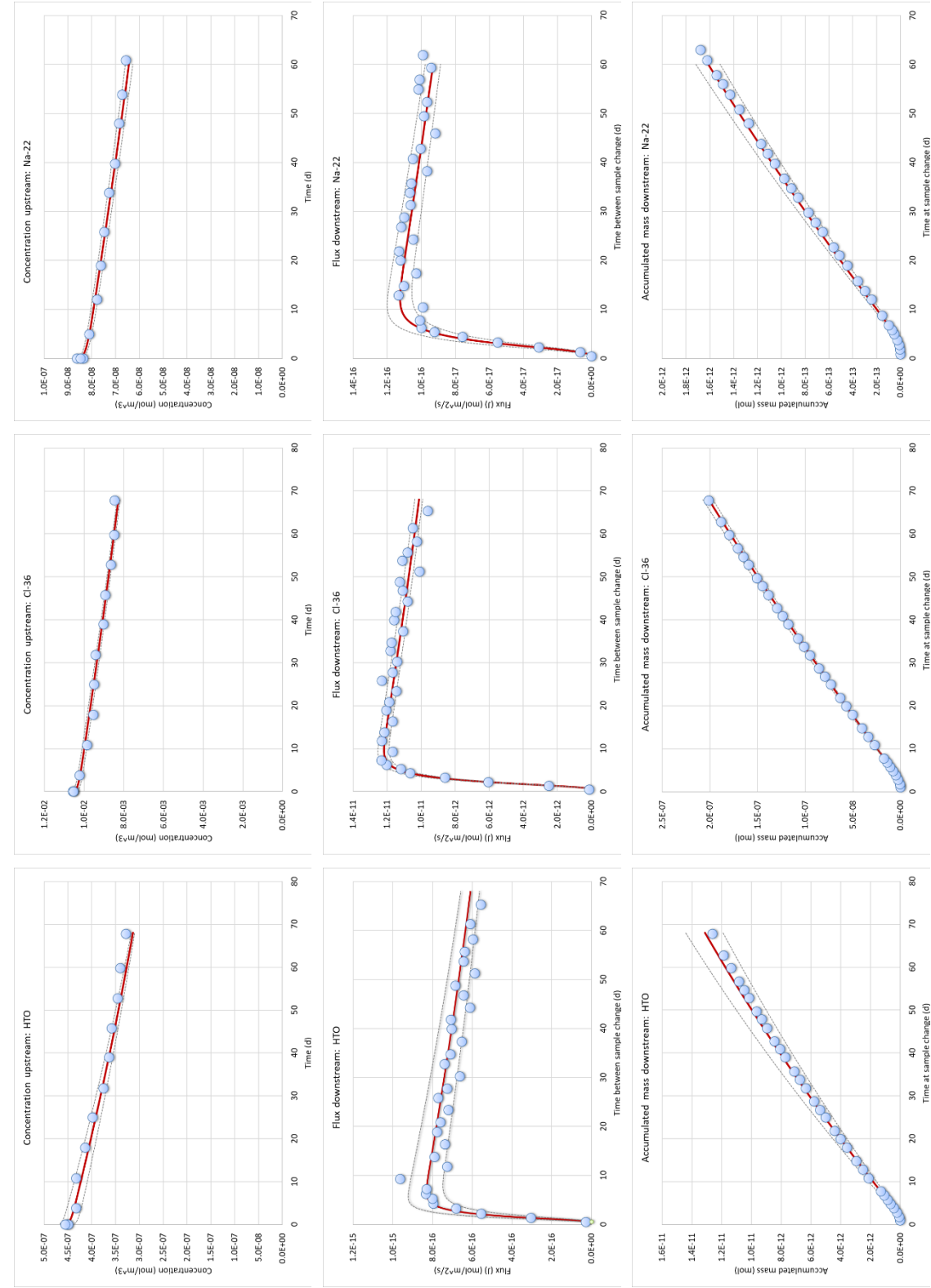
BOZ1-1-808.61-DI (S1) – Bänkerjoch Formation – SPW-BOZ

Sample diameter 63.75 mm – sample thickness 11.47 mm



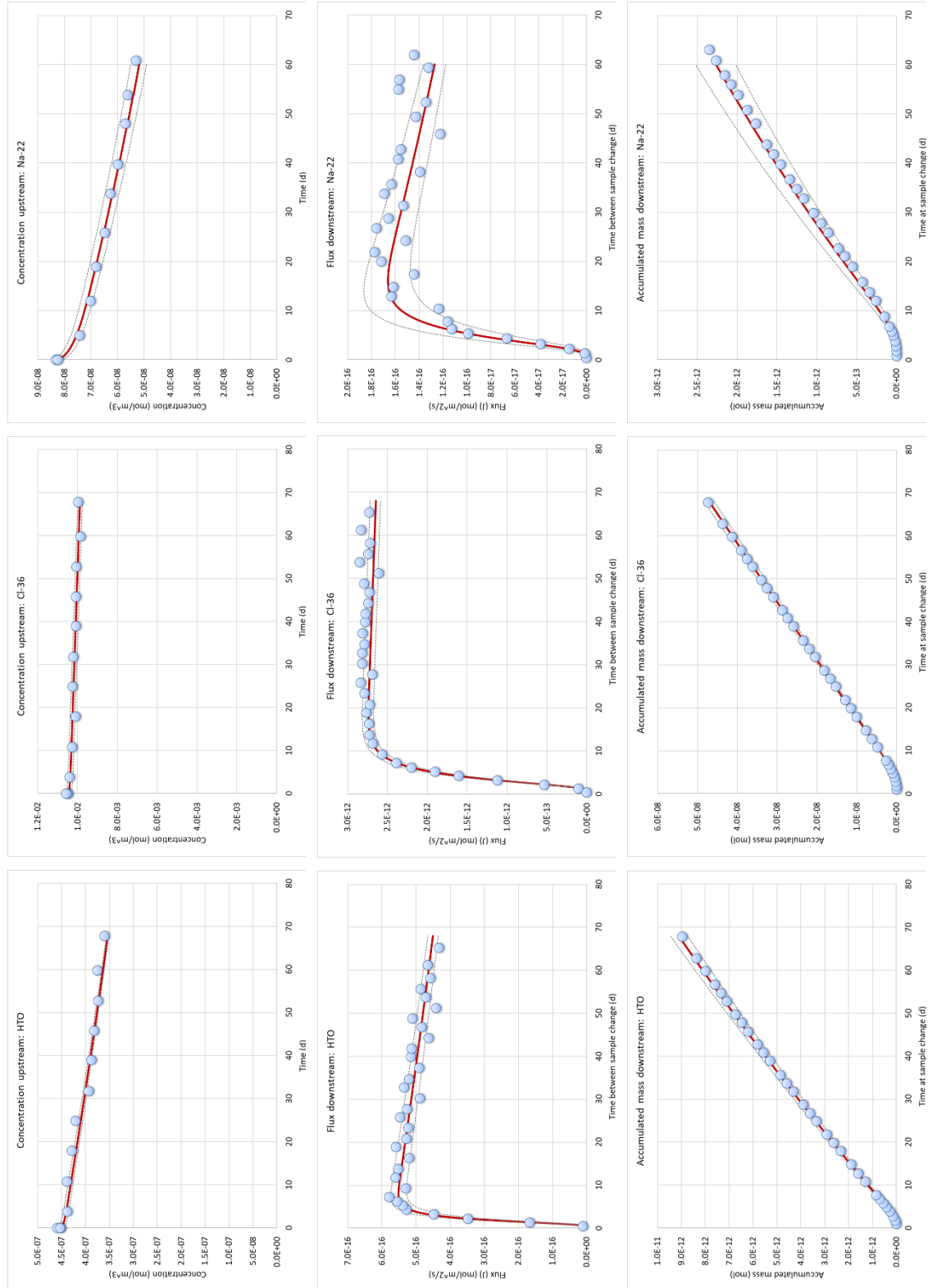
BOZ2-1-313.13-DI (S1) – Hauptrogenstein – SPW-BOZ

Sample diameter 63.59 mm – sample thickness 12.23 mm



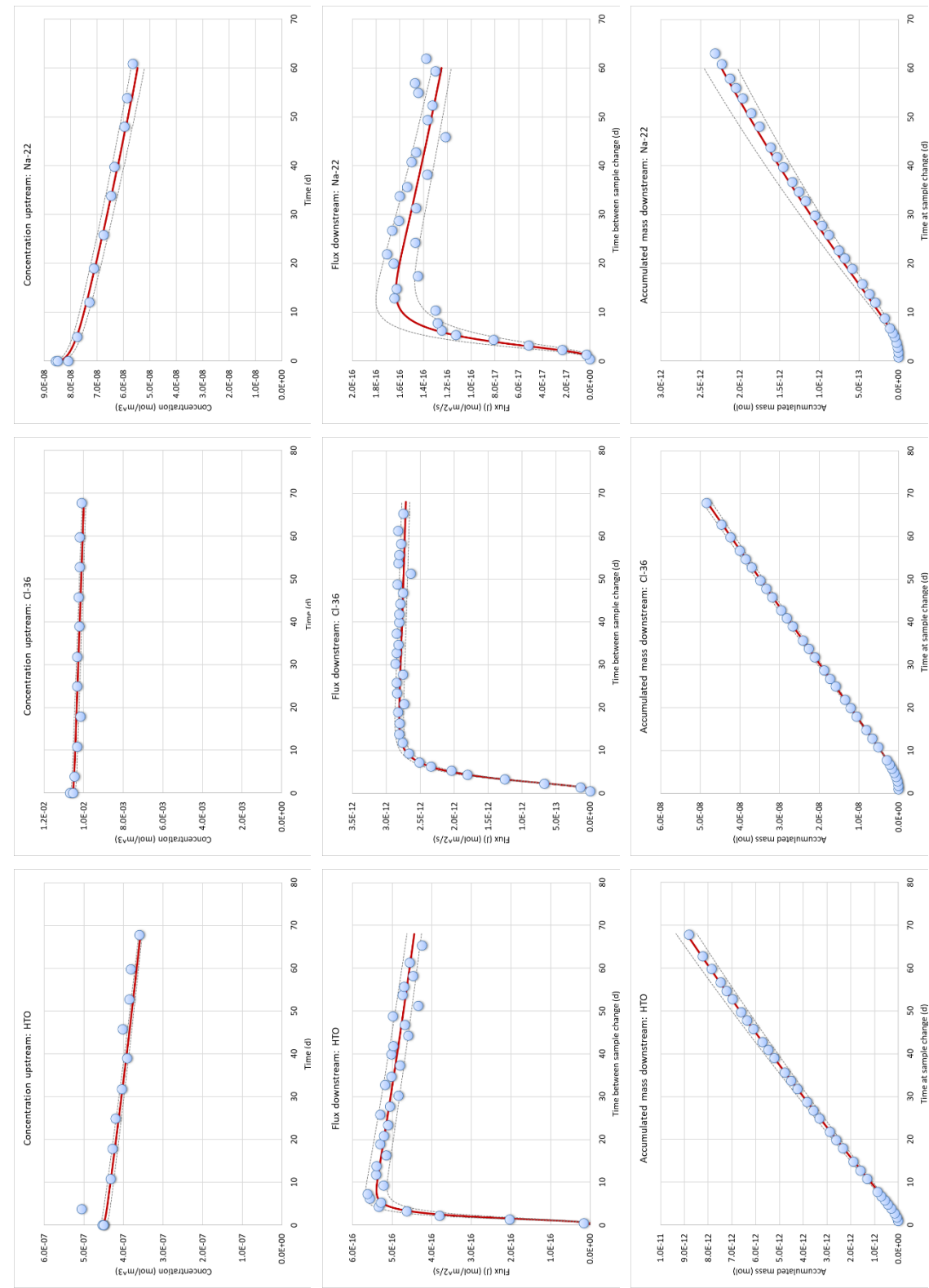
BOZ2-1-443.93-DI (S2) – Passwang Formation – SPW-BOZ

Sample diameter 63.20 mm – sample thickness 12.59 mm



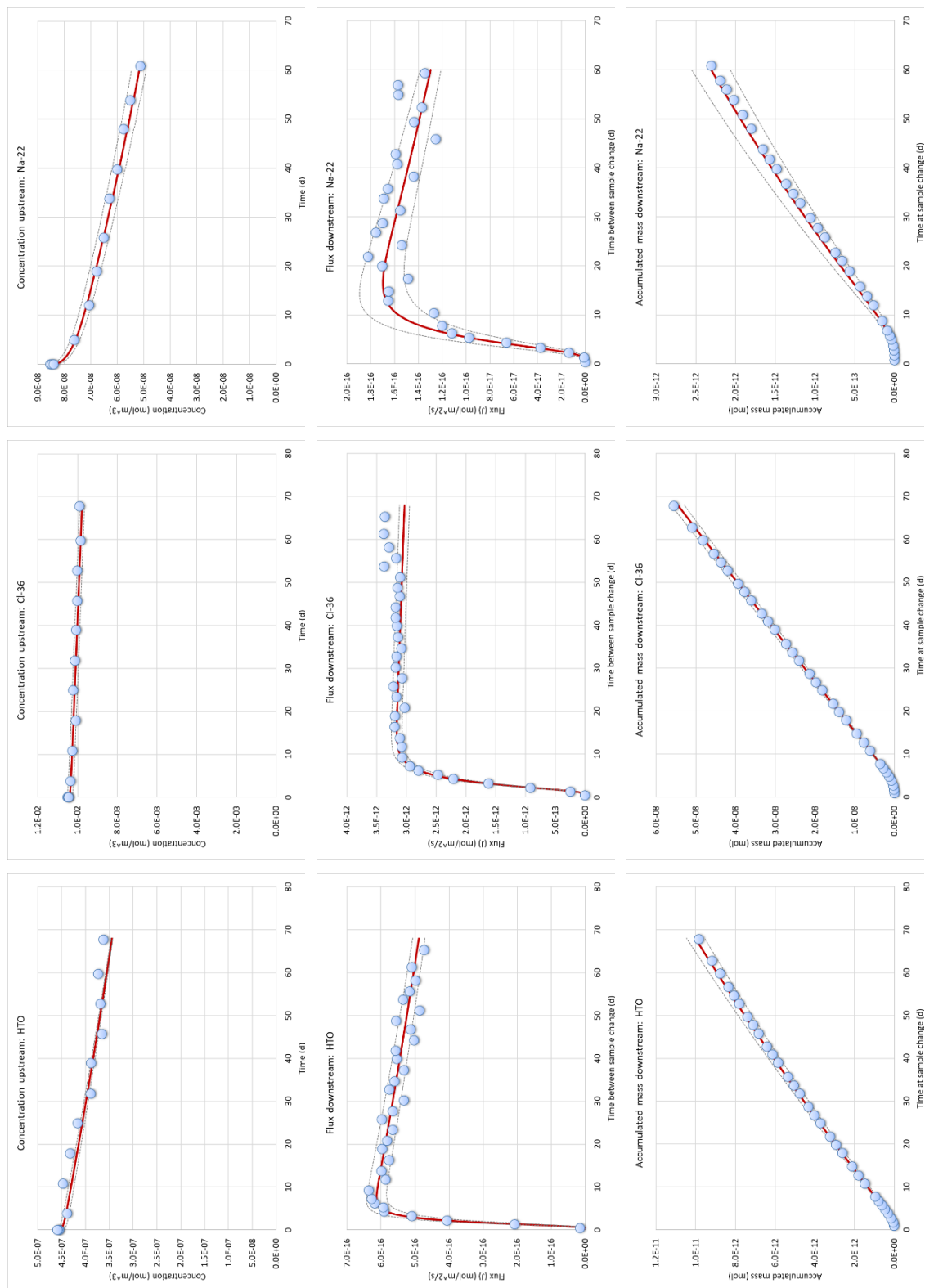
BOZ2-1-588.64-DI (S2) – Staffelegg Formation – SPW-BOZ

Sample diameter 63.26 mm – sample thickness 12.09 mm



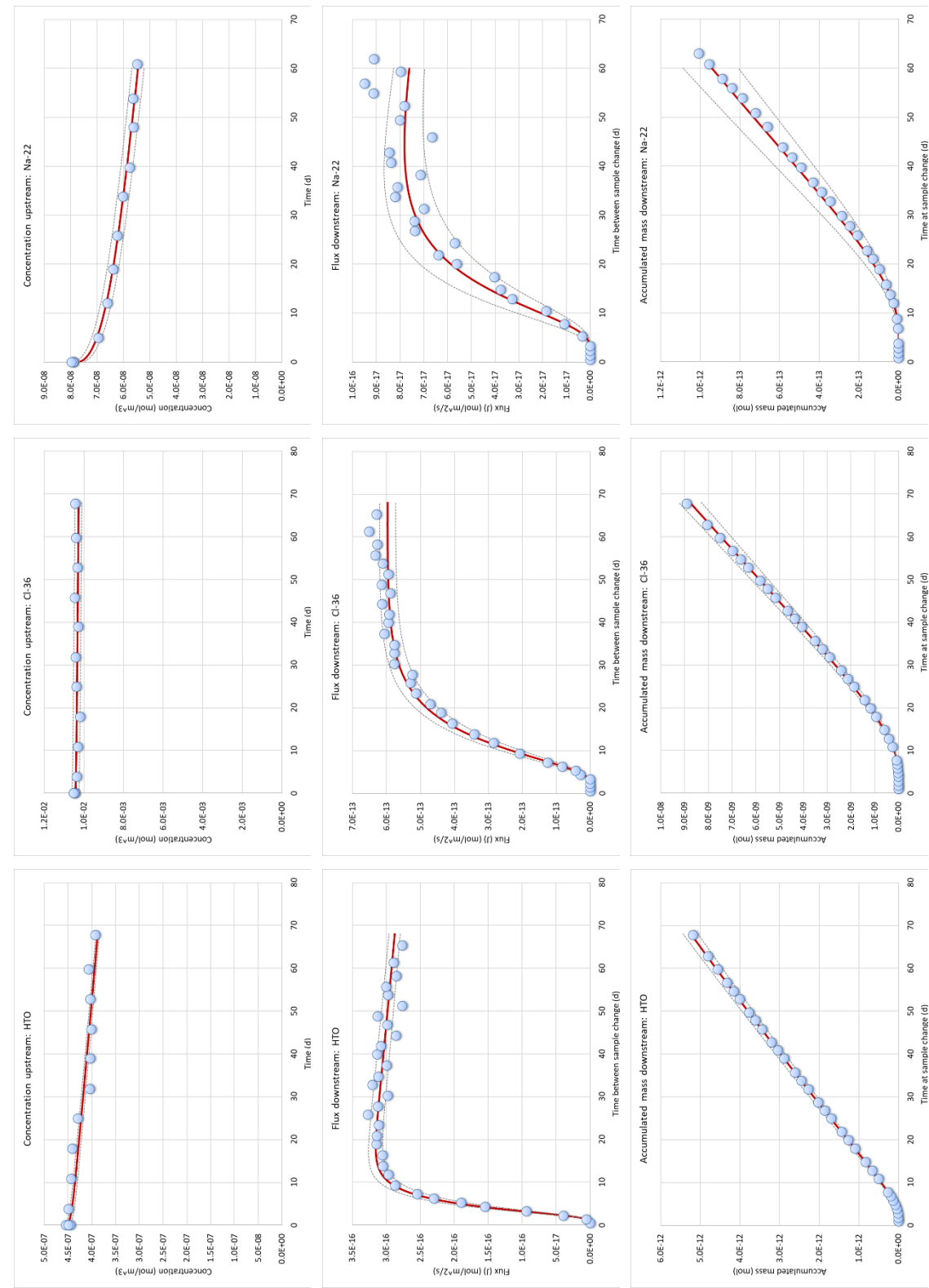
BOZ2-1-595.15-DI (S2) – Staffelegg Formation – SPW-BOZ

Sample diameter 63.28 mm – sample thickness 11.85 mm



BOZ2-1-606.95-DI (S2) – Staffelegg Formation – SPW-BOZ

Sample diameter 63.50 mm – sample thickness 12.13 mm

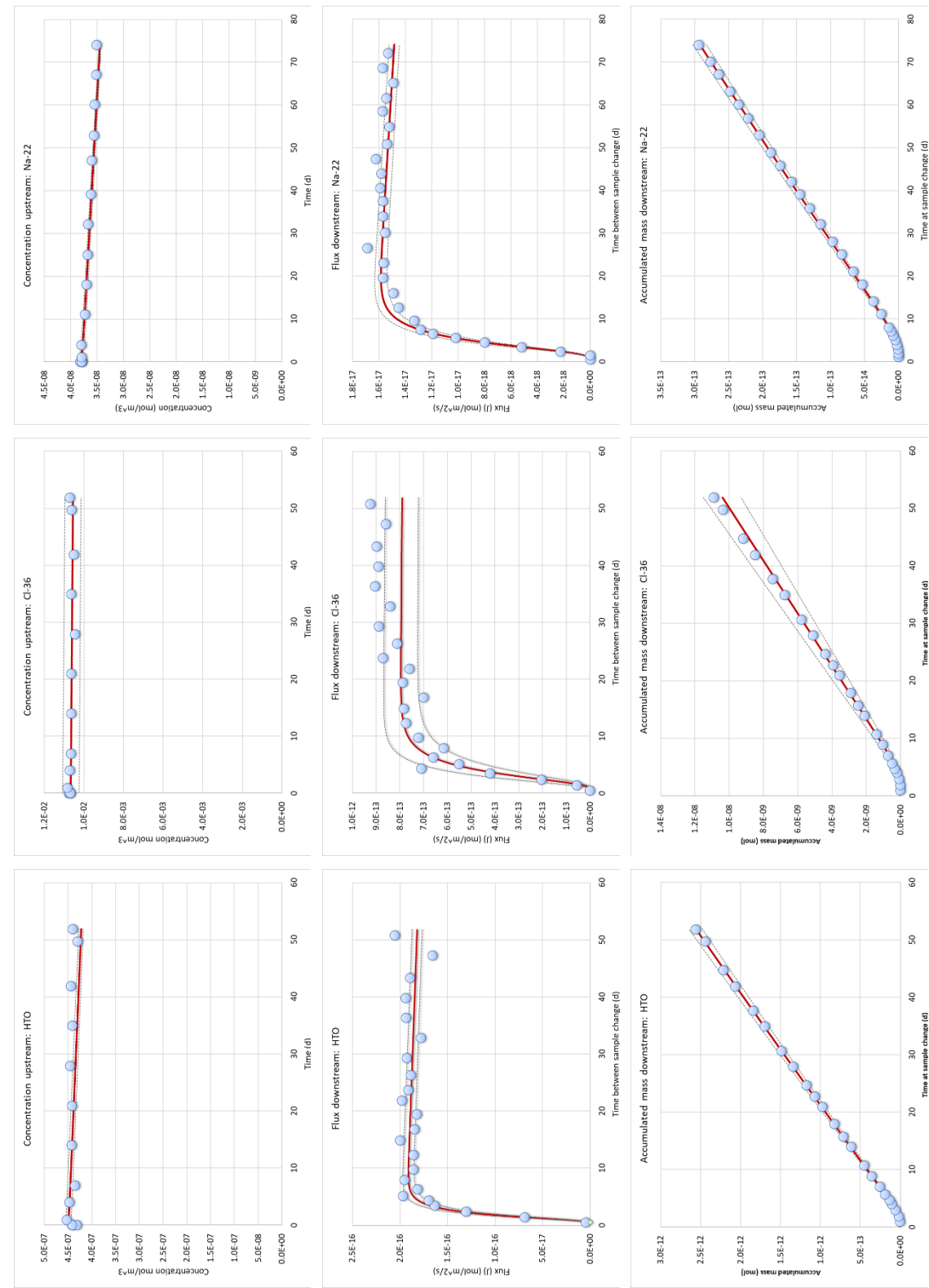


I.2 Samples from Nördlich Lägern

- BUL1-1
- STA2-1
- STA3-1

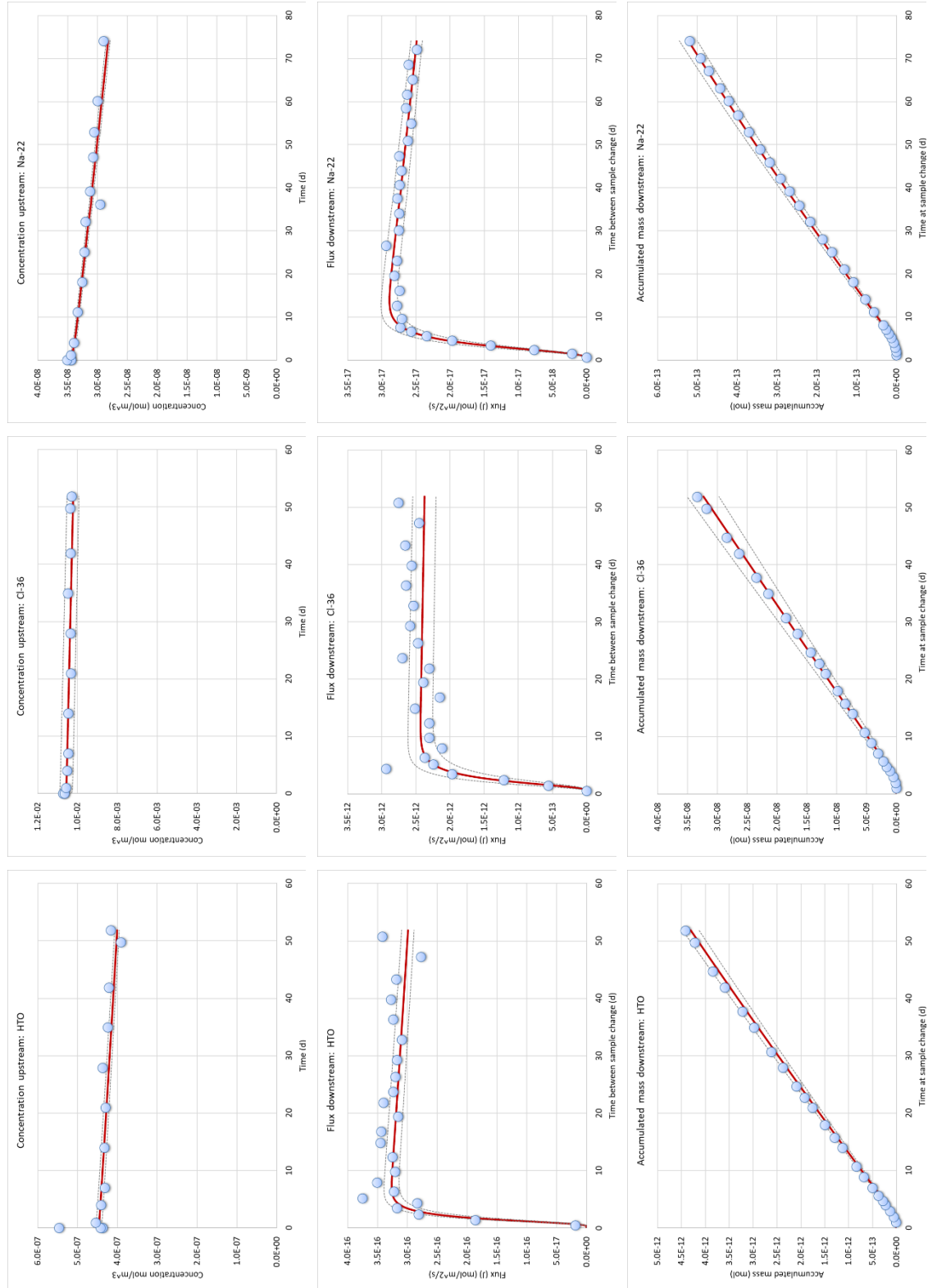
BUL1-1-734.45-DI (S1) – Villigen Formation – SPW-BUL

Sample diameter 63.60 mm – sample thickness 12.47 mm



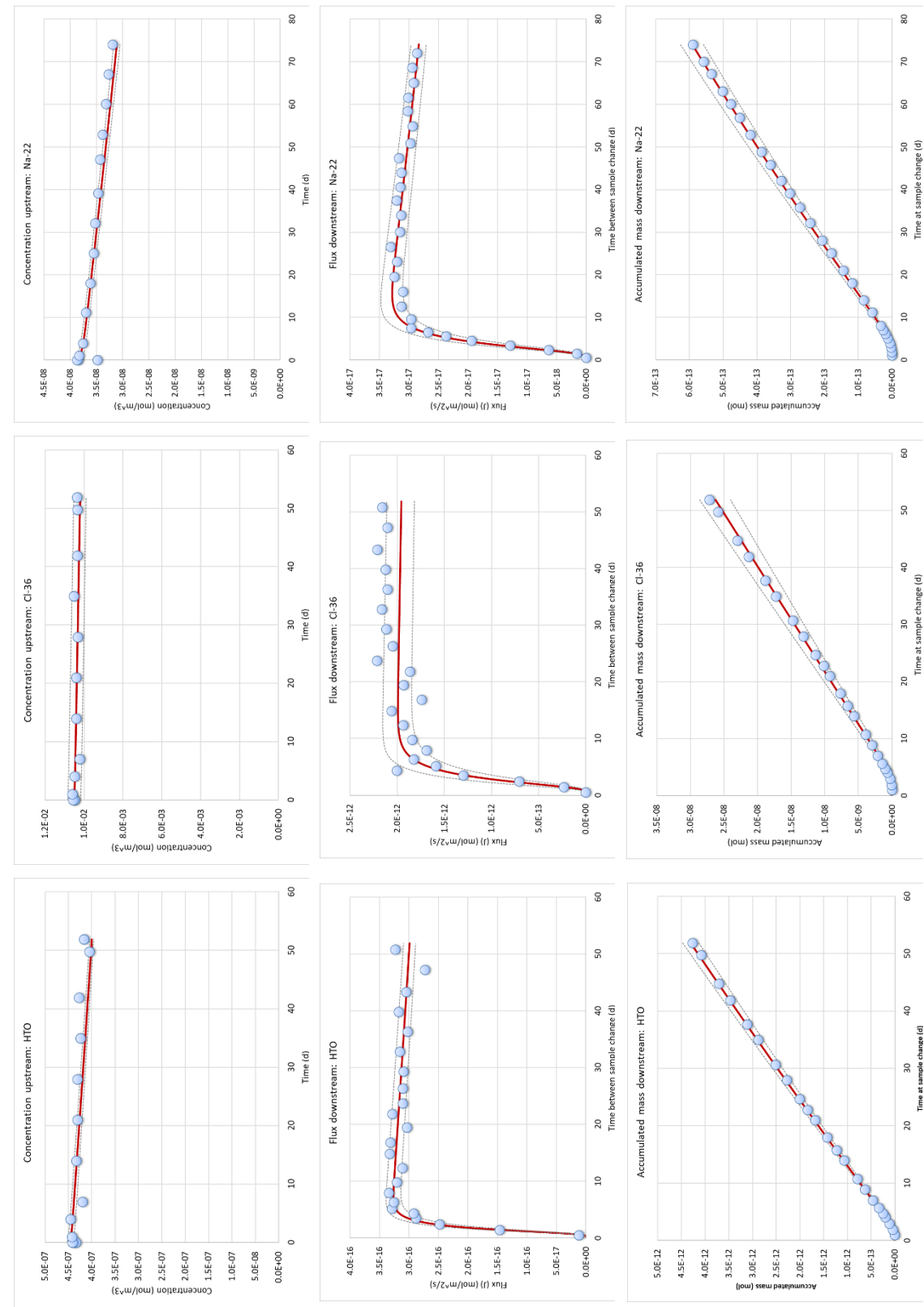
BUL1-1-752.48-DI (S2) – Wildegg Formation – SPW-BUL

Sample diameter 63.40 mm – sample thickness 12.47 mm



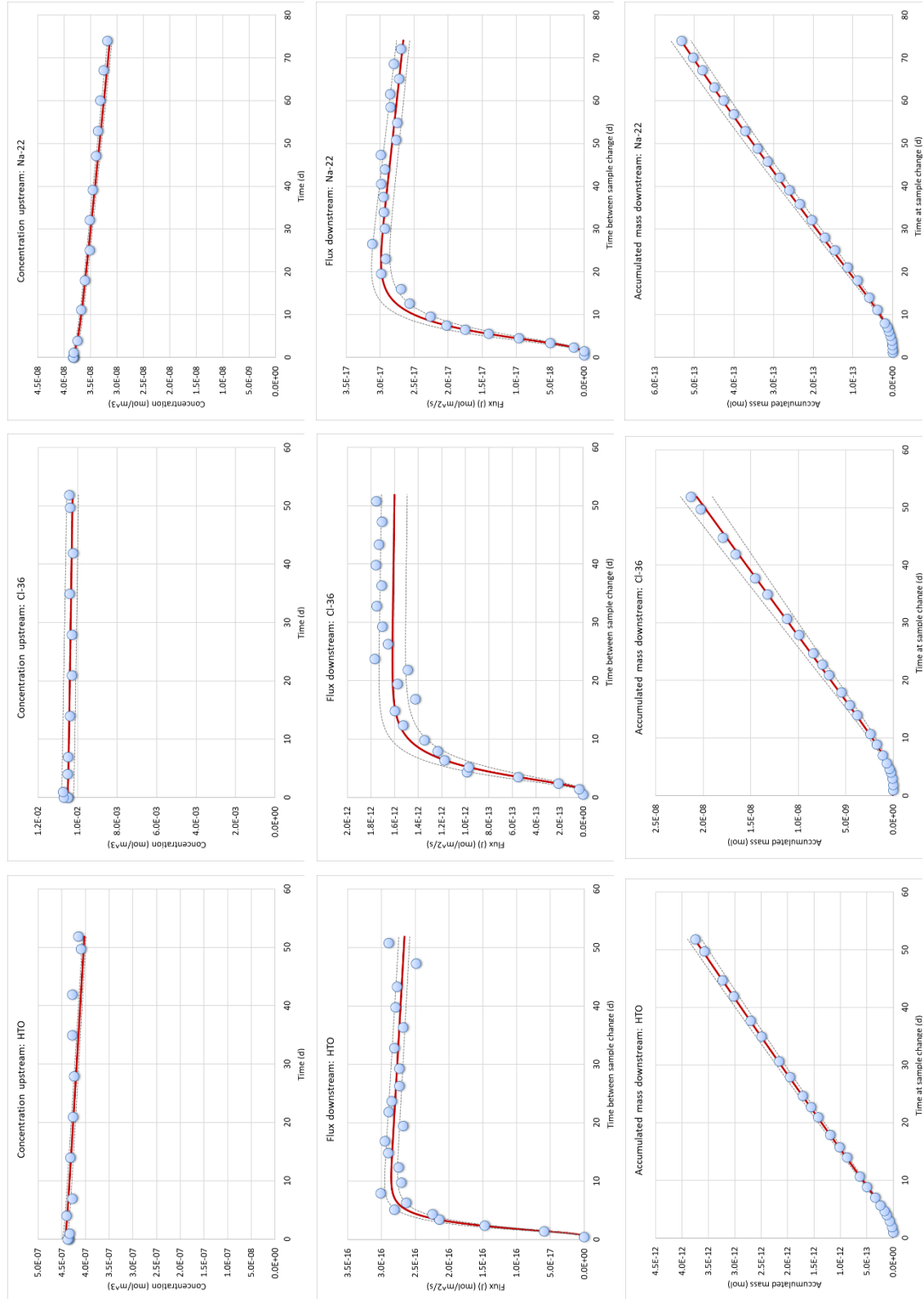
BUL1-1-766.65-DI (S1) – Wildegg-Formation – SPW-BUL

Sample diameter 63.46 mm – sample thickness 12.24 mm



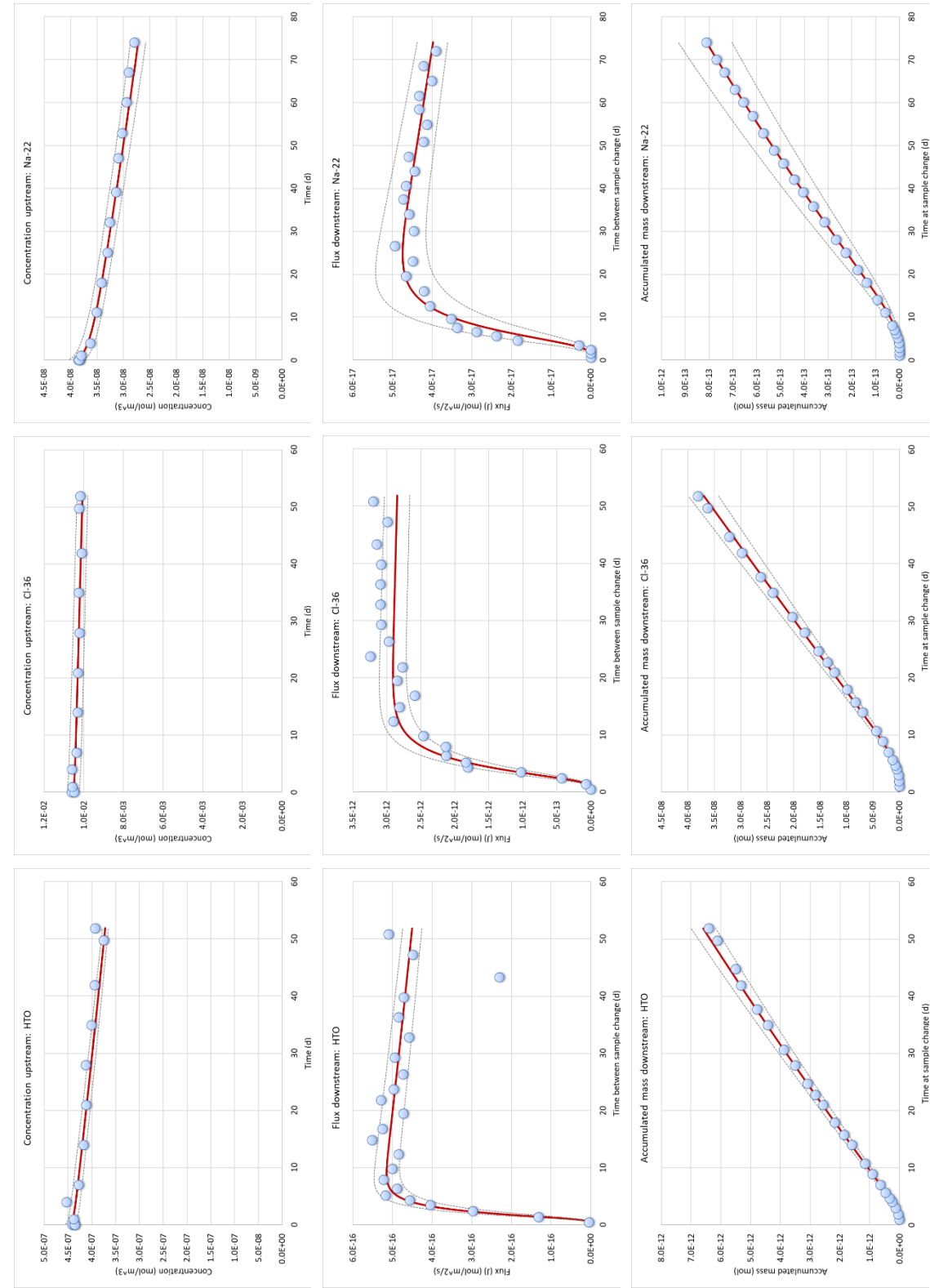
BUL1-1-794.35-DI (S1) – Wildegg-Formation – SPW-BUL

Sample diameter 63.70 mm – sample thickness 13.98 mm



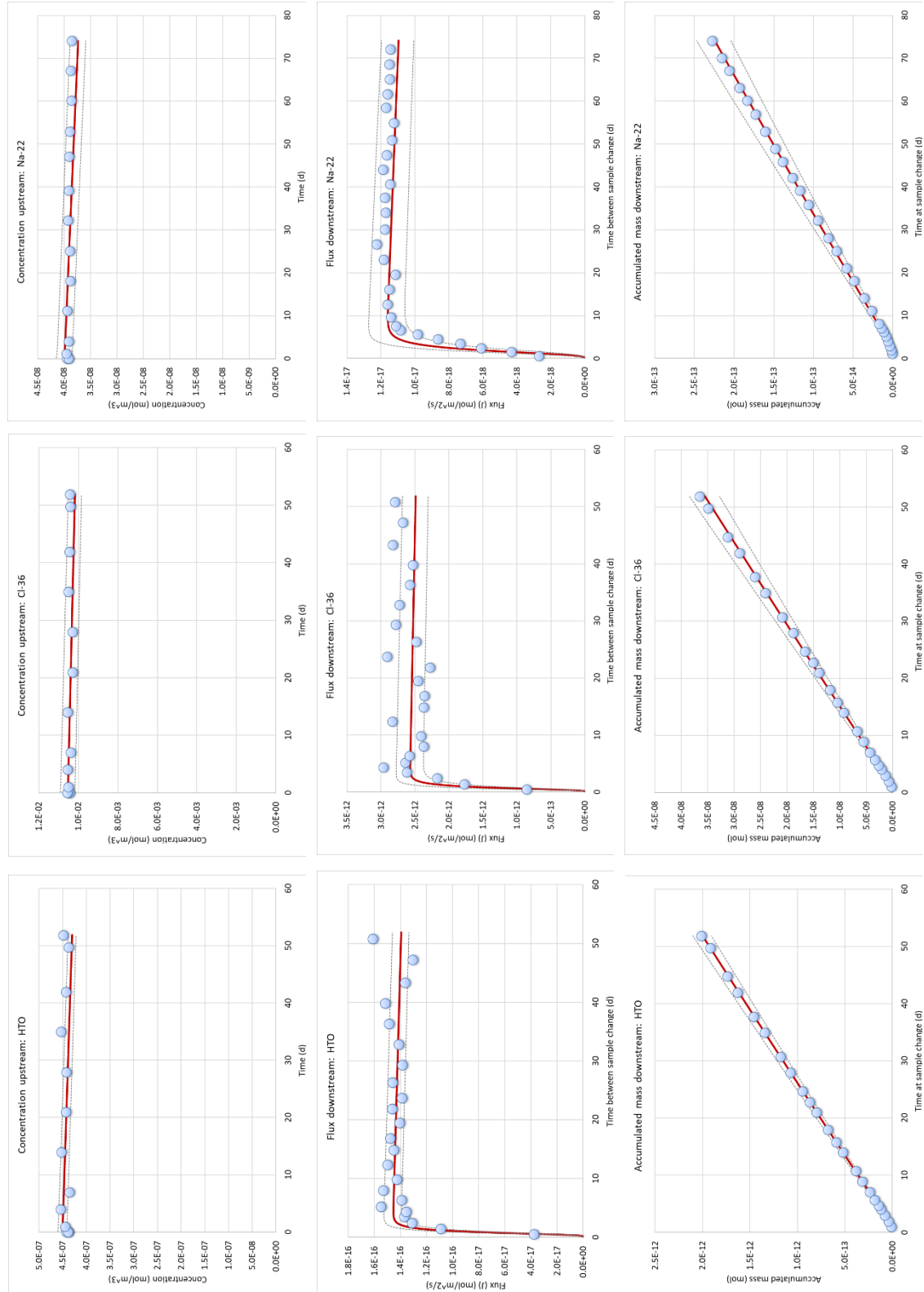
BUL1-1-808.92-DI (S1) – Parkinsoni-Wüttembergica-Schichten – SPW-BUL

Sample diameter 63.50 mm – sample thickness 11.40 mm



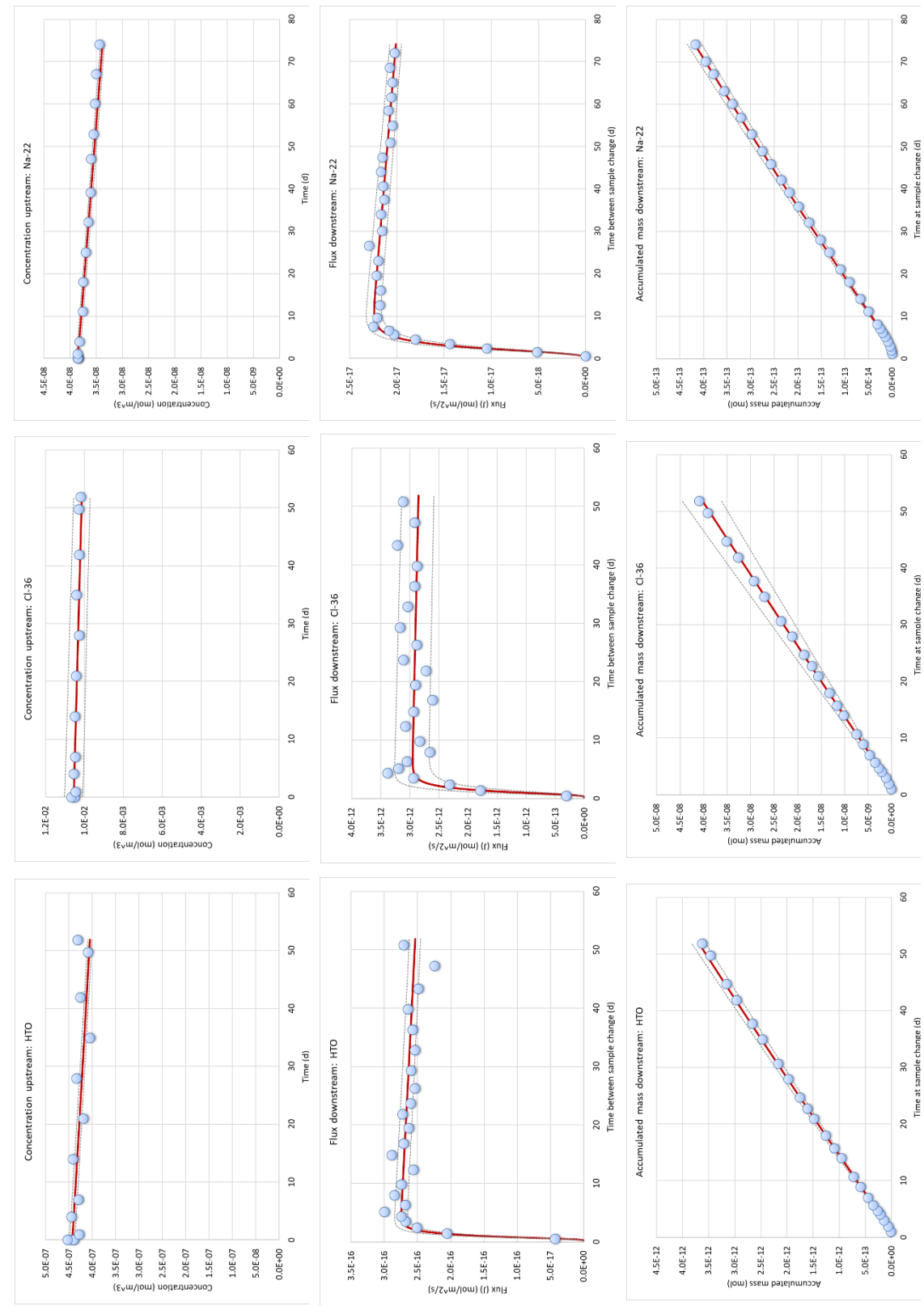
BUL1-1-840.45-DI (S1) – Herrenwis Unit – SPW-BUL

Sample diameter 63.70 mm – sample thickness 12.28 mm



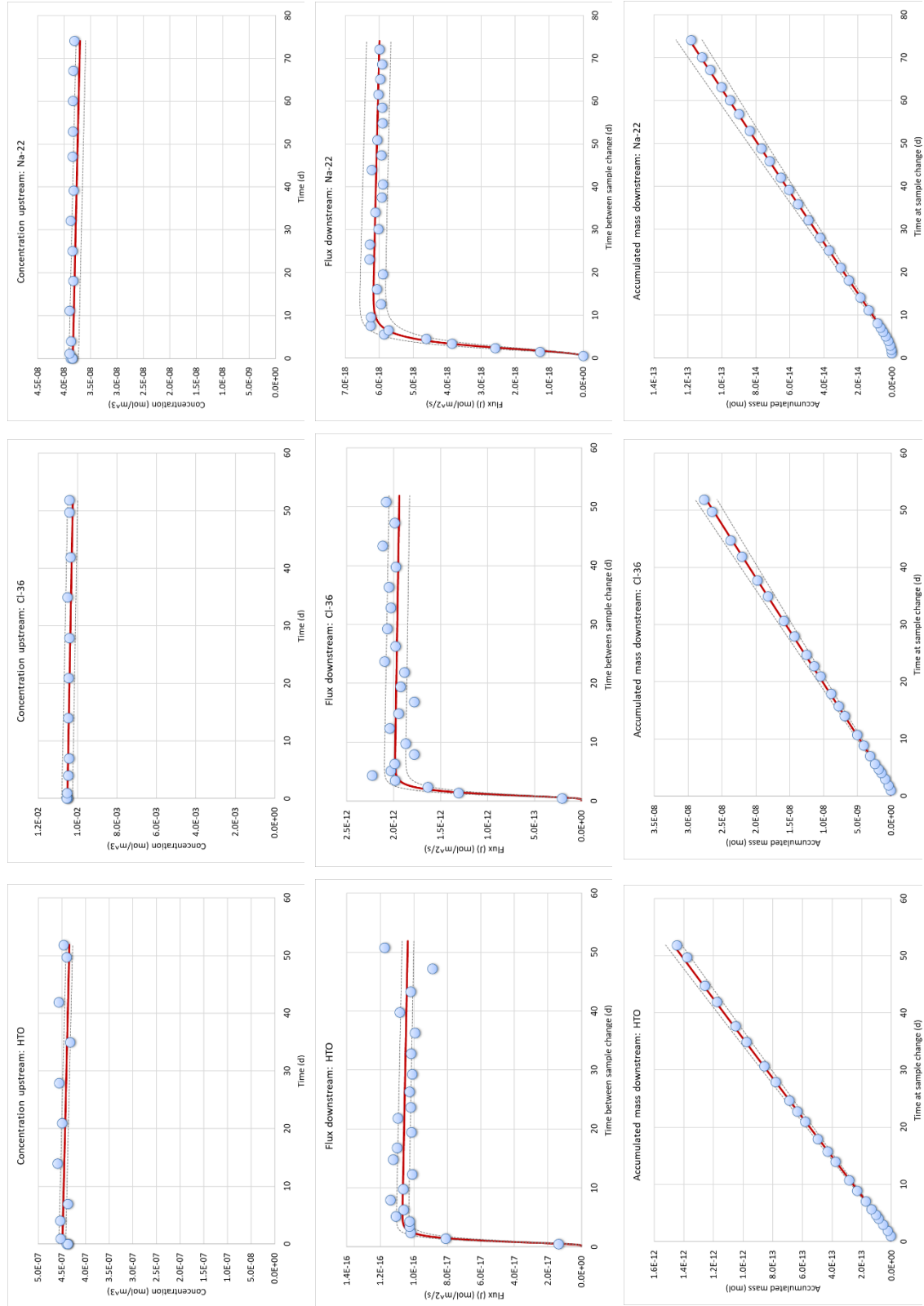
BUL1-1-840.45-DI (S2) – Herrenwis Unit – SPW-BUL

Sample diameter 63.56 mm – sample thickness 10.05 mm



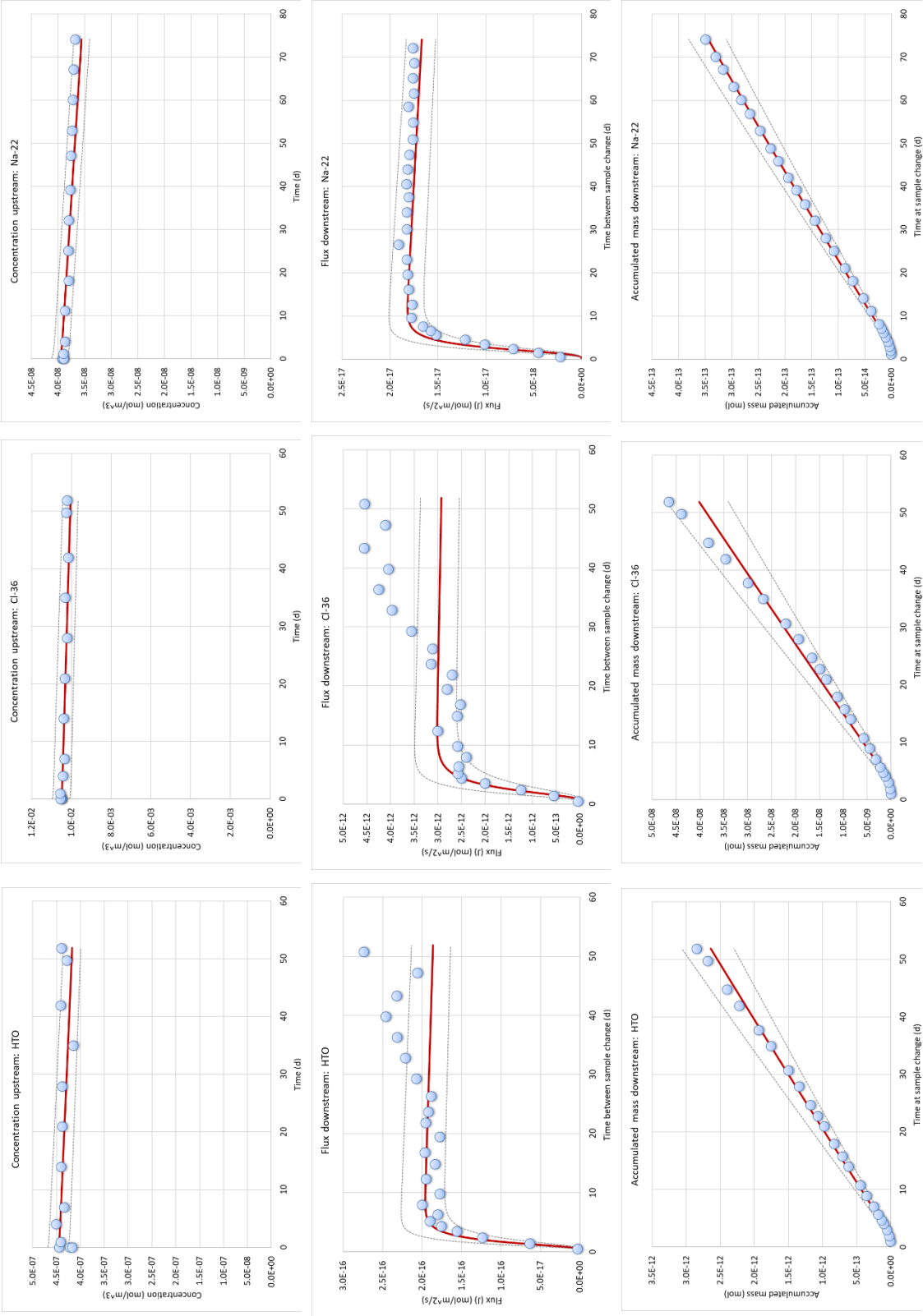
BUL1-1-845.05-DI S1 – Herrenwis Unit – SPW-BUL

Sample diameter 63.69 mm – sample thickness 11.80 mm



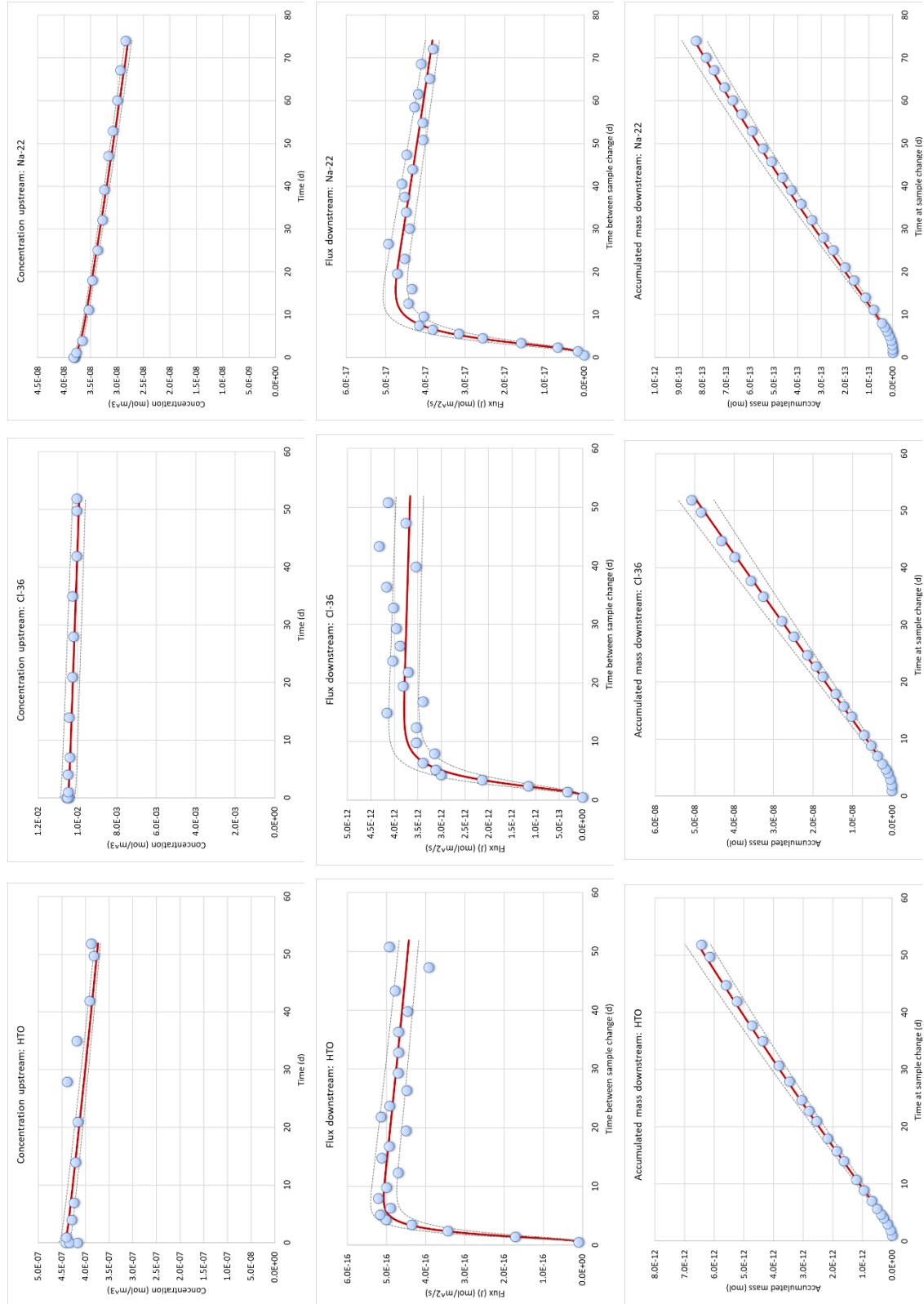
BUL1-1-851.50-DI (S1) – Herrenwis Unit – SPW-BUL

Sample diameter 63.74 mm – sample thickness 13.47 mm



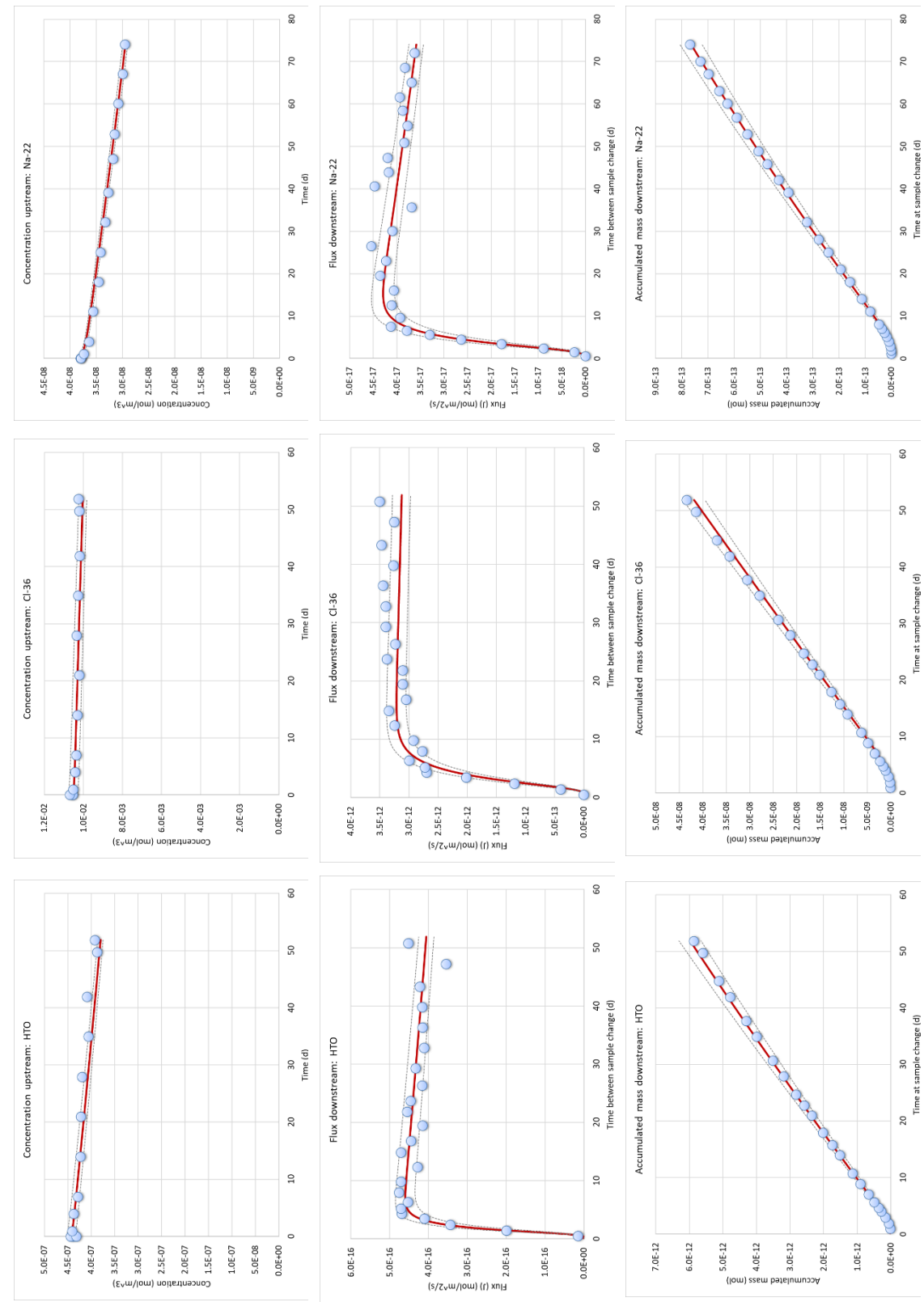
BUL1-1-872.35-DI (S1) – Wedelsandstein Formation – SPW-BUL

Sample diameter 63.60 mm – sample thickness 13.27 mm



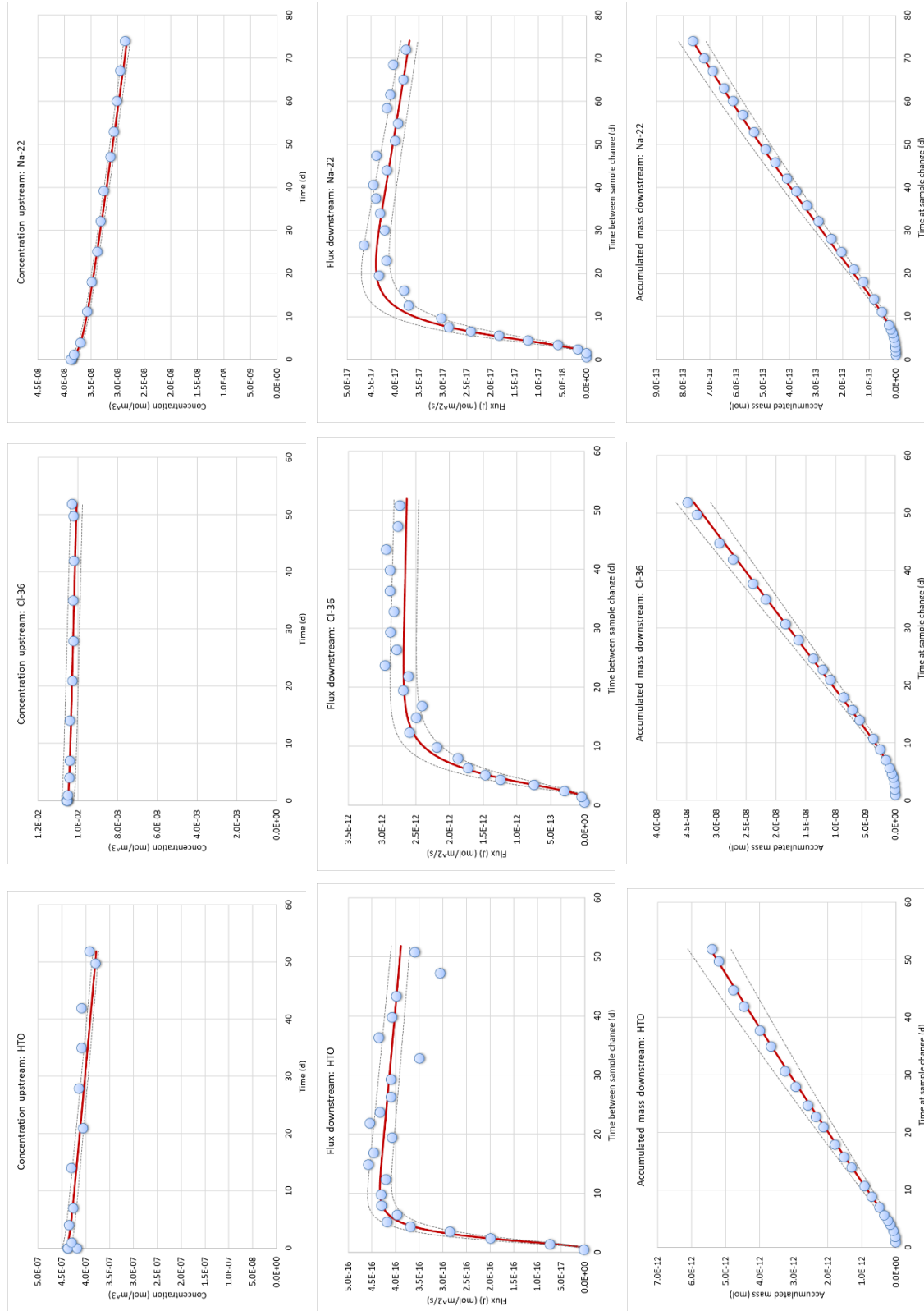
BUL1-1-879.20-DI (S1) – Wedelsandstein Formation – SPW-BUL

Sample diameter 63.55 mm – sample thickness 11.97 mm



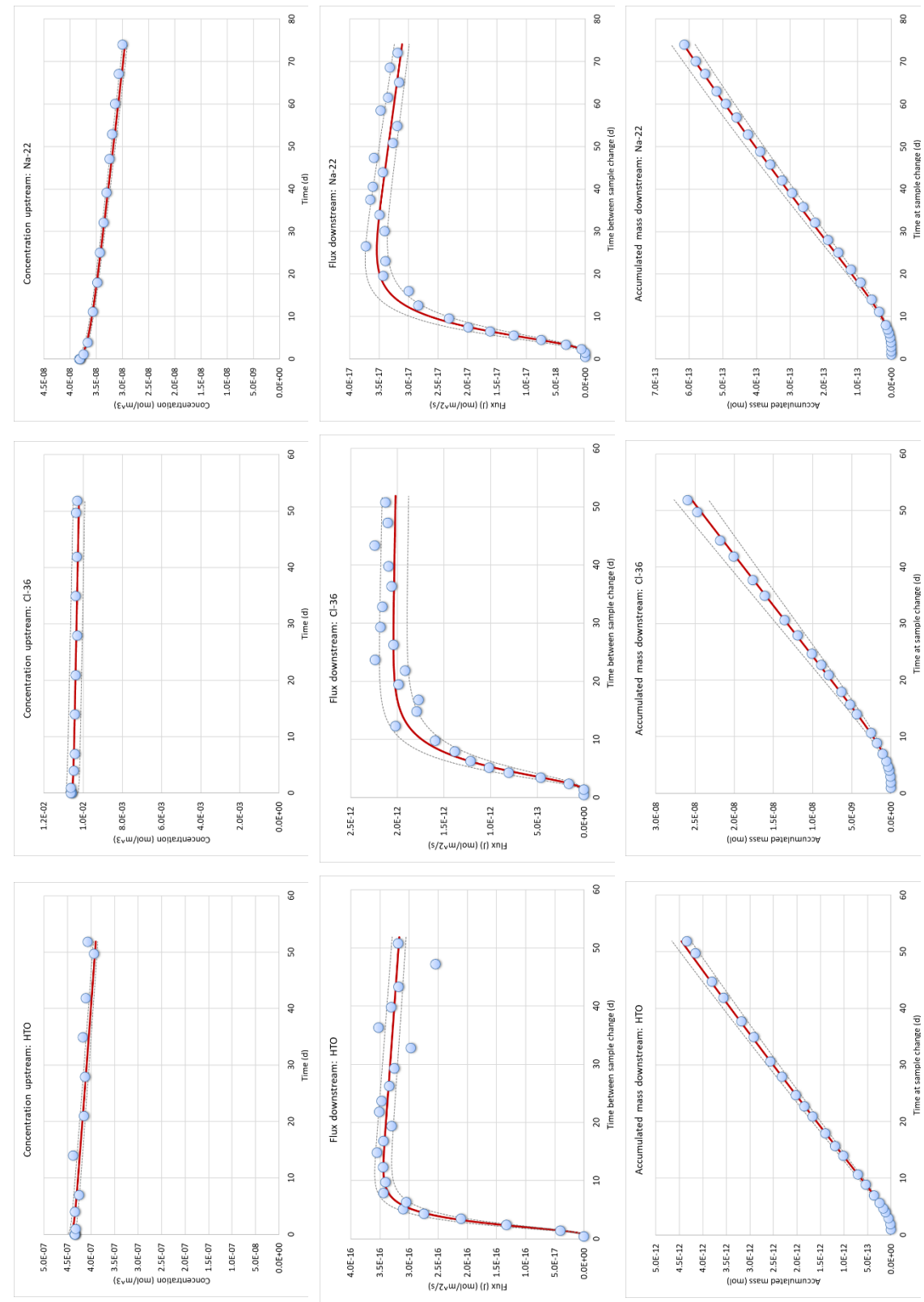
BUL1-1-908.93-DI (S1) – Opalinus Clay – SPW-BUL

Sample diameter 63.60 mm – sample thickness 13.42 mm



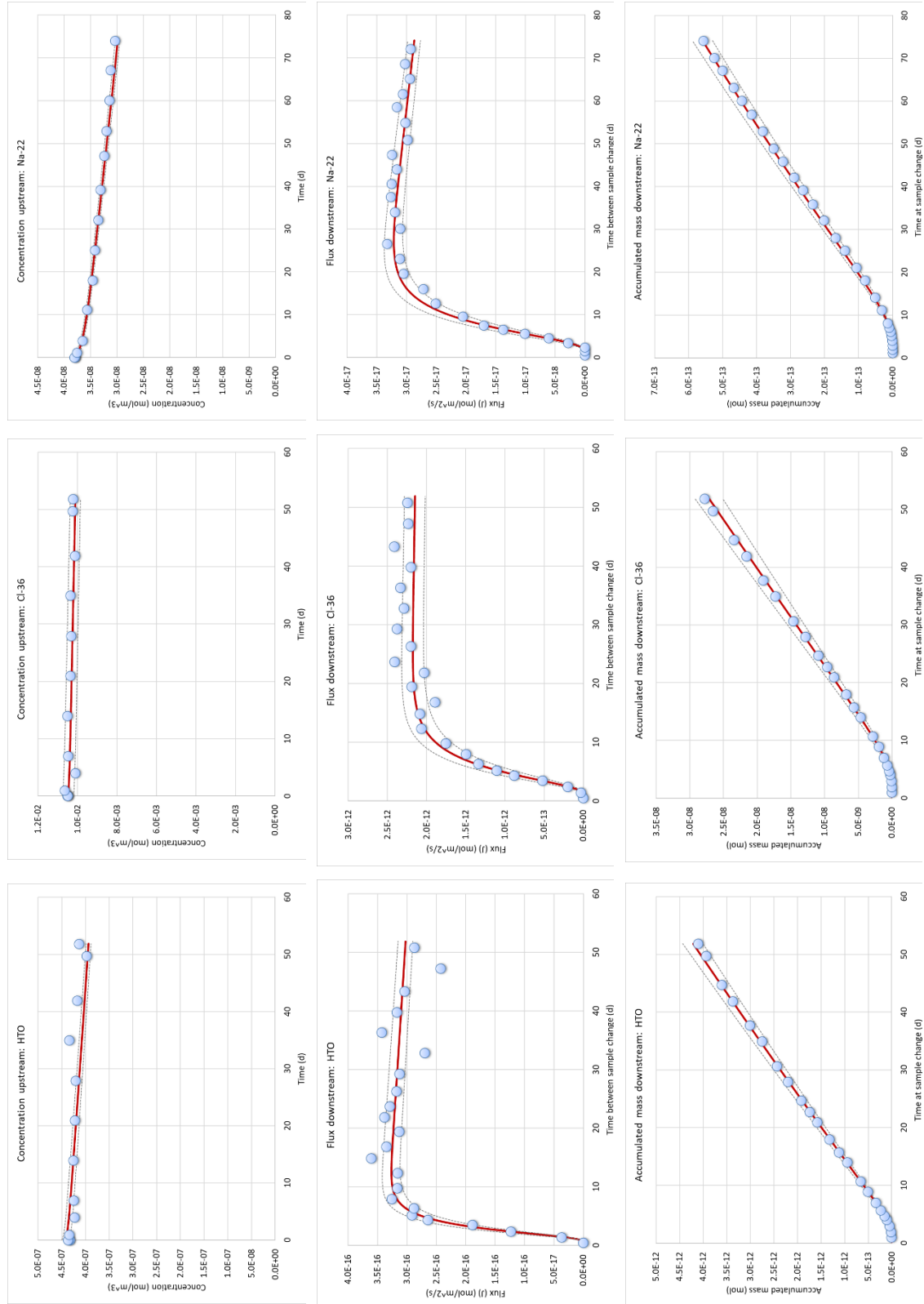
BUL1-1-914.61-DI (S1) – Opalinus Clay – SPW-BUL

Sample diameter 63.60 mm – sample thickness 11.70 mm



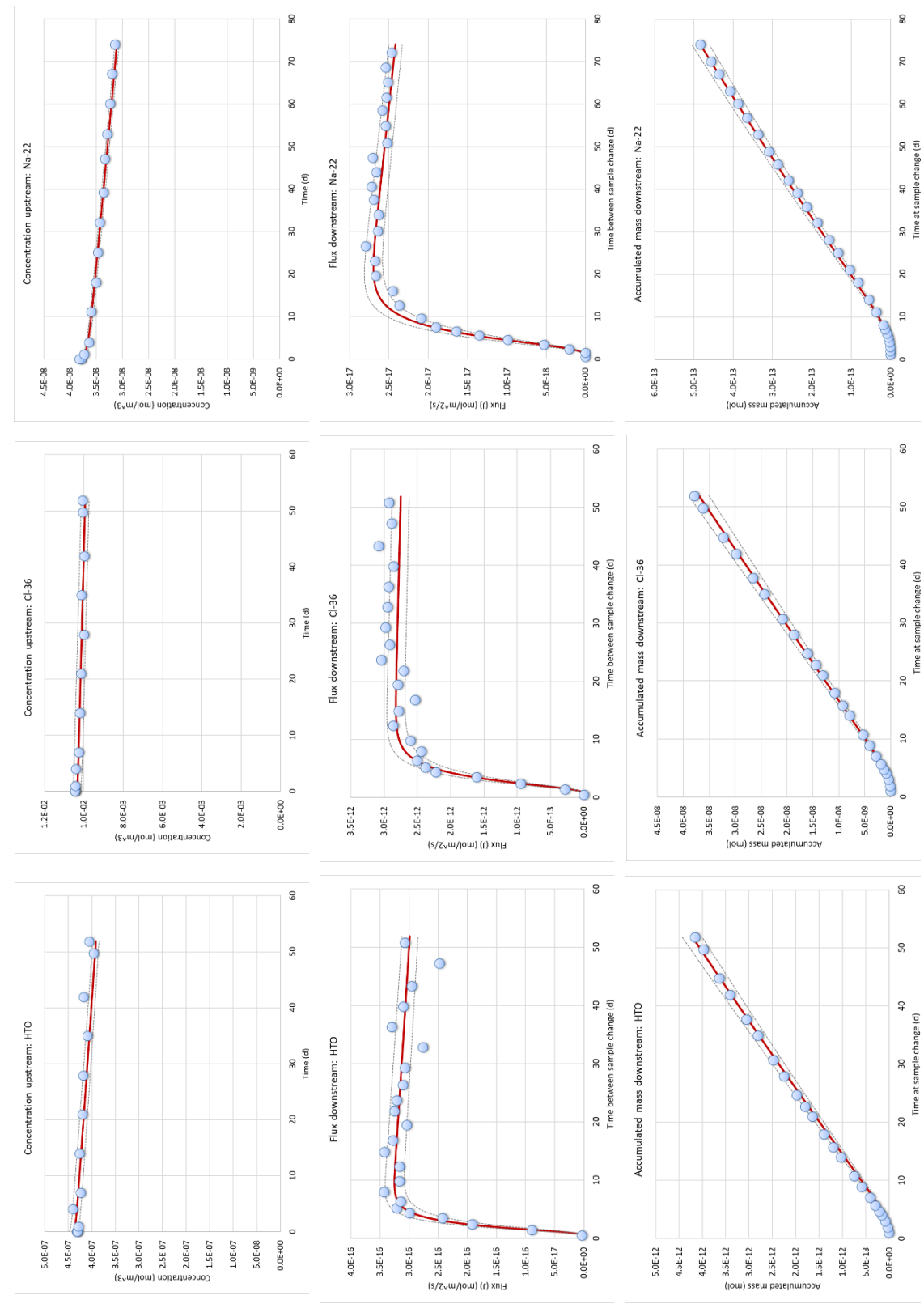
BUL1-1-921.43-DI (S1) – Opalinus Clay – SPW-BUL

Sample diameter 63.50 mm – sample thickness 11.63 mm



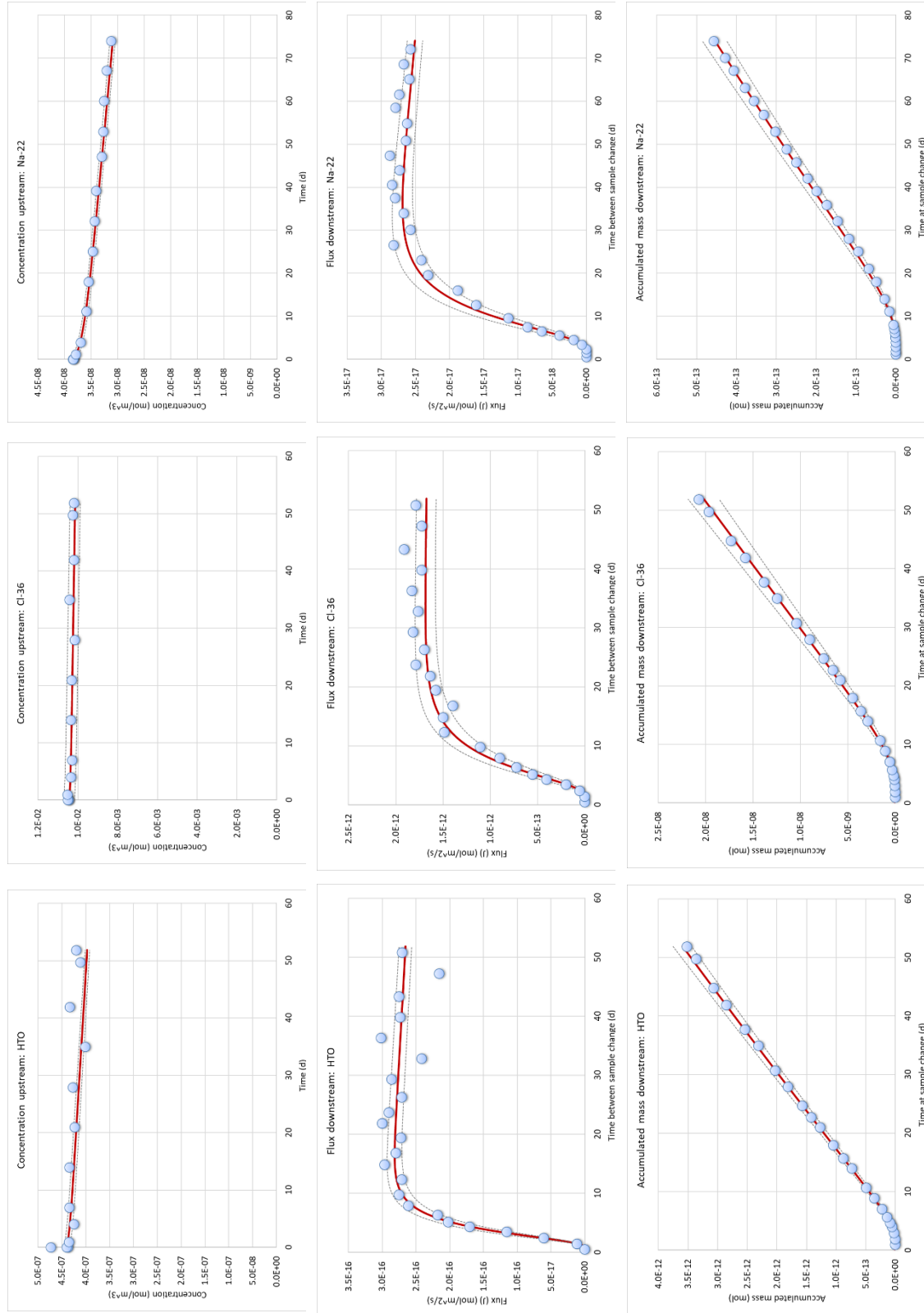
BUL1-1-921.43-DI (S2) – Opalinus Clay – SPW-BUL

Sample diameter 63.57 mm – sample thickness 10.61 mm



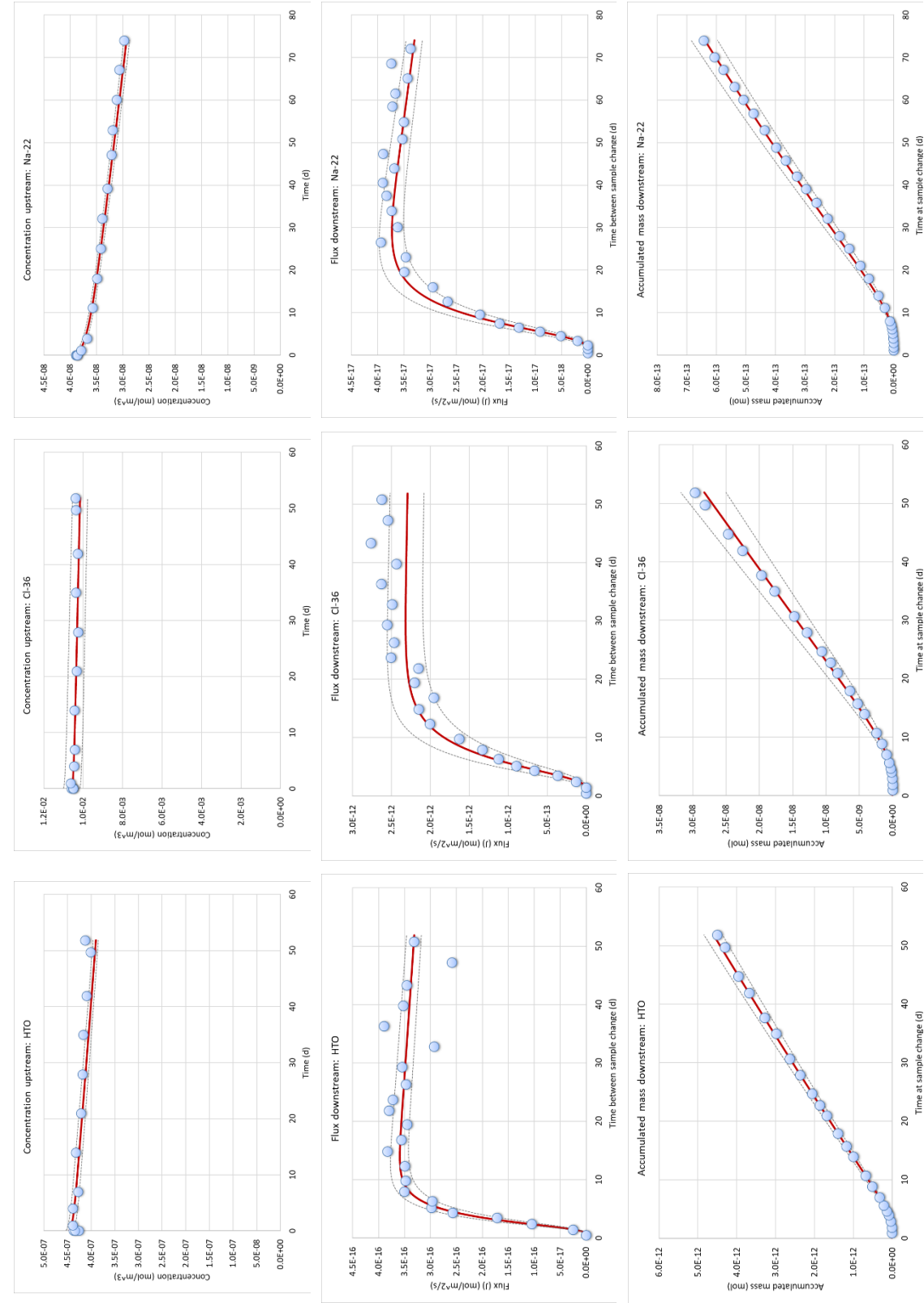
BUL1-1-932.82-DI (S1) – Opalinus Clay – SPW-BUL

Sample diameter 63.50 mm – sample thickness 12.68 mm



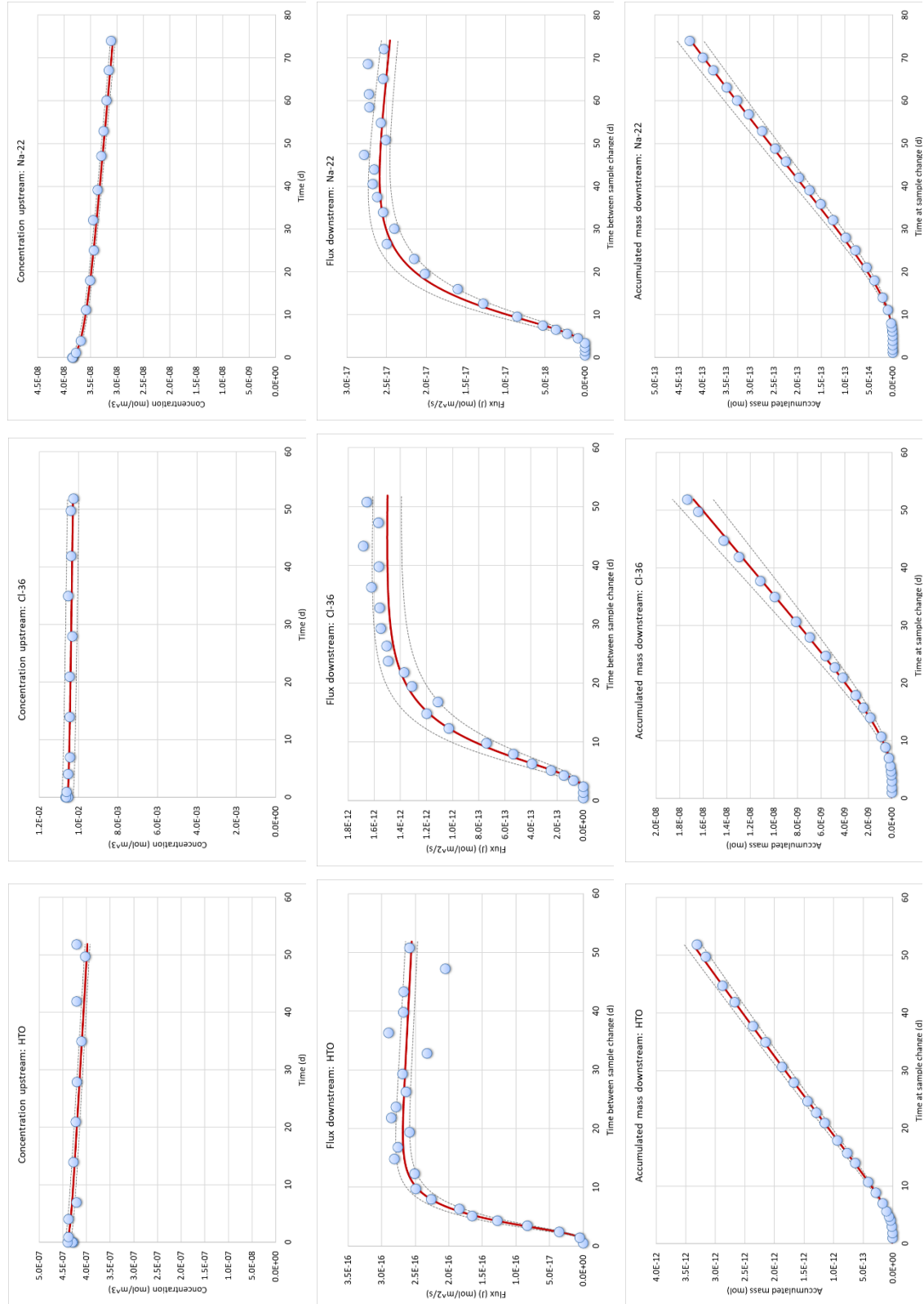
BUL1-1-945.46-DI (S1) – Opalinus Clay – SPW-BUL

Sample diameter 63.57 mm – sample thickness 12.68 mm



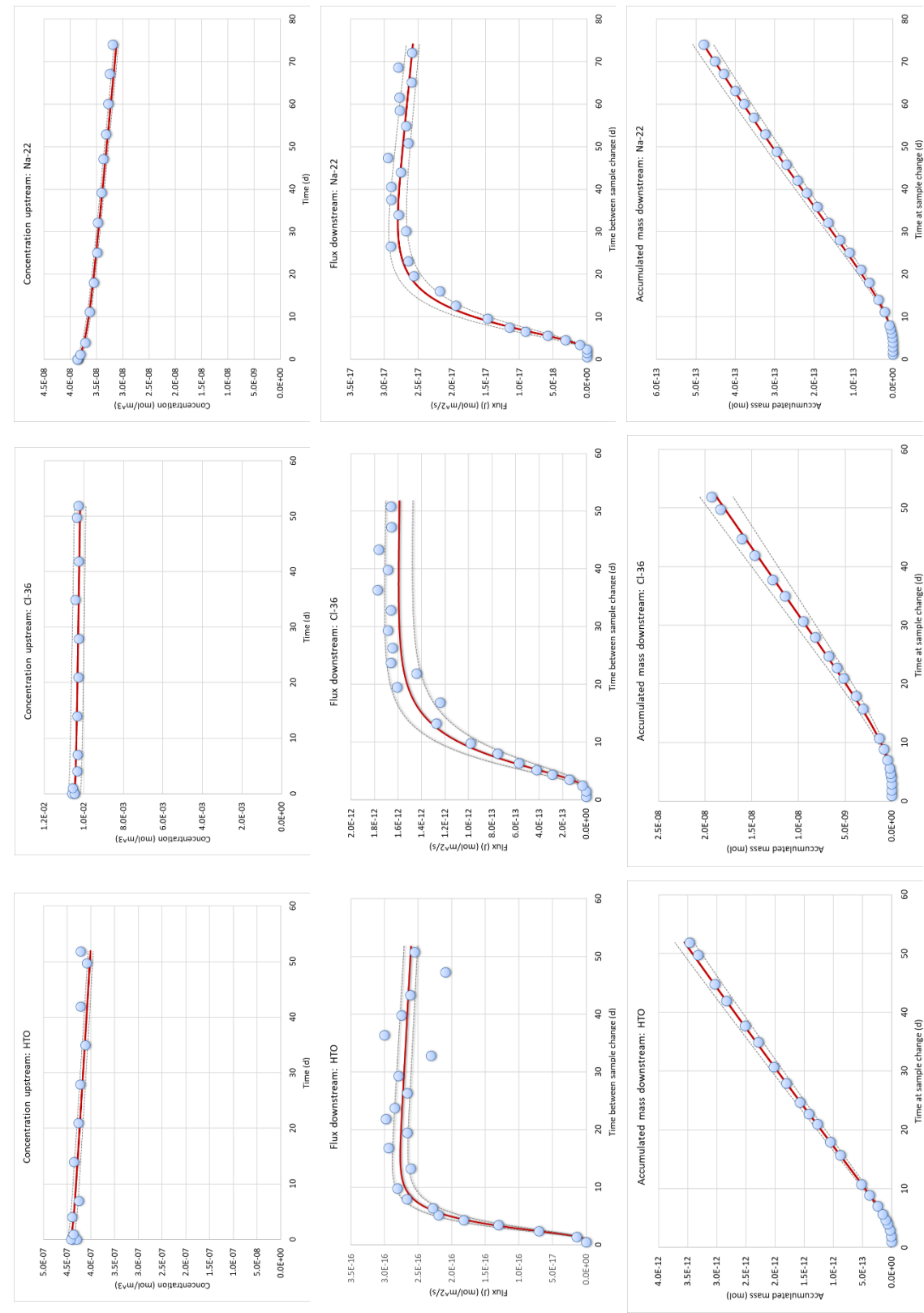
BUL1-1-955.62-DI (S1) – Opalinus Clay – SPW-BUL

Sample diameter 63.50 mm – sample thickness 13.08 mm



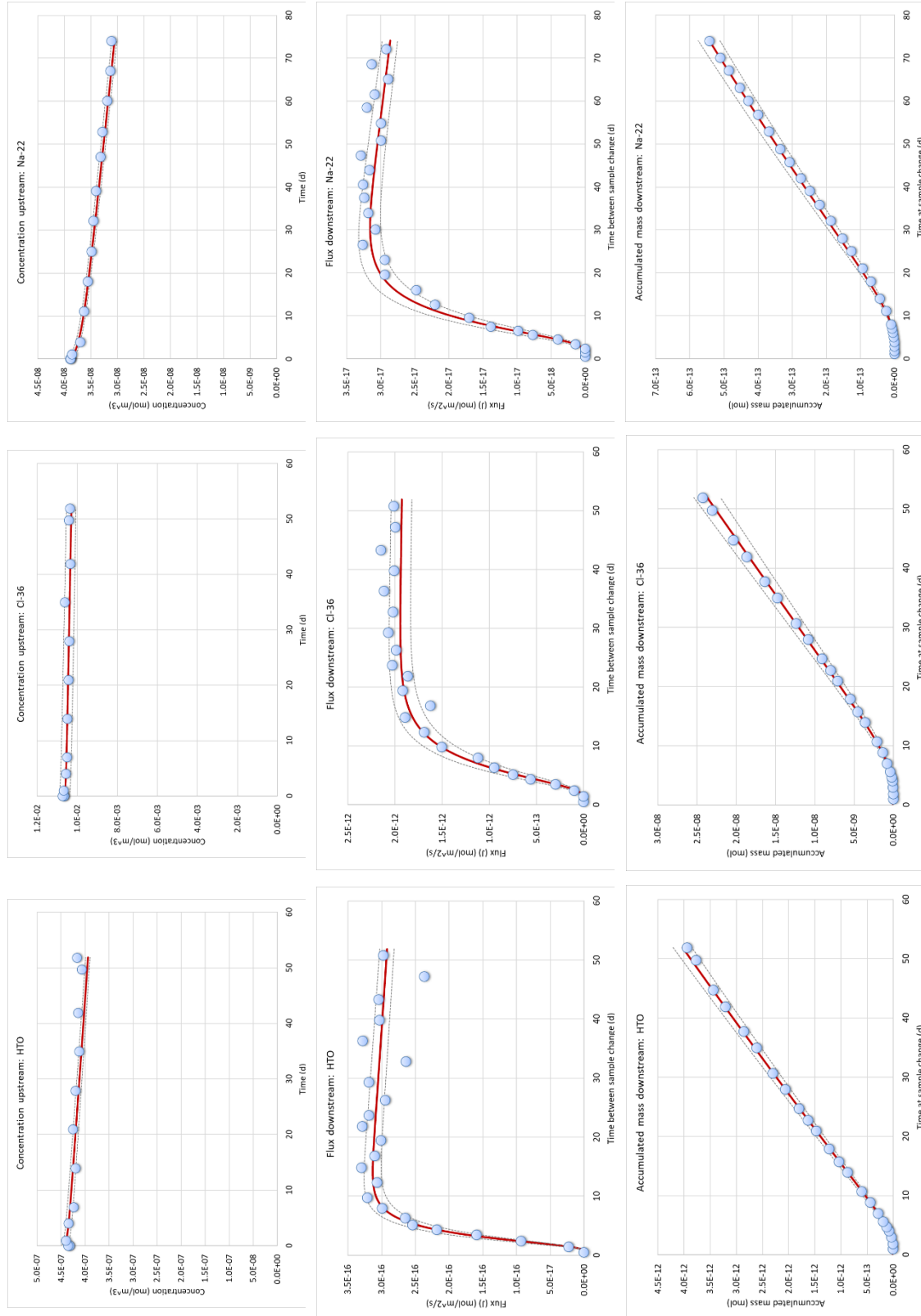
BUL1-1-955.62-DI (S2) – Opalinus Clay – SPW-BUL

Sample diameter 63.55 mm – sample thickness 12.62 mm



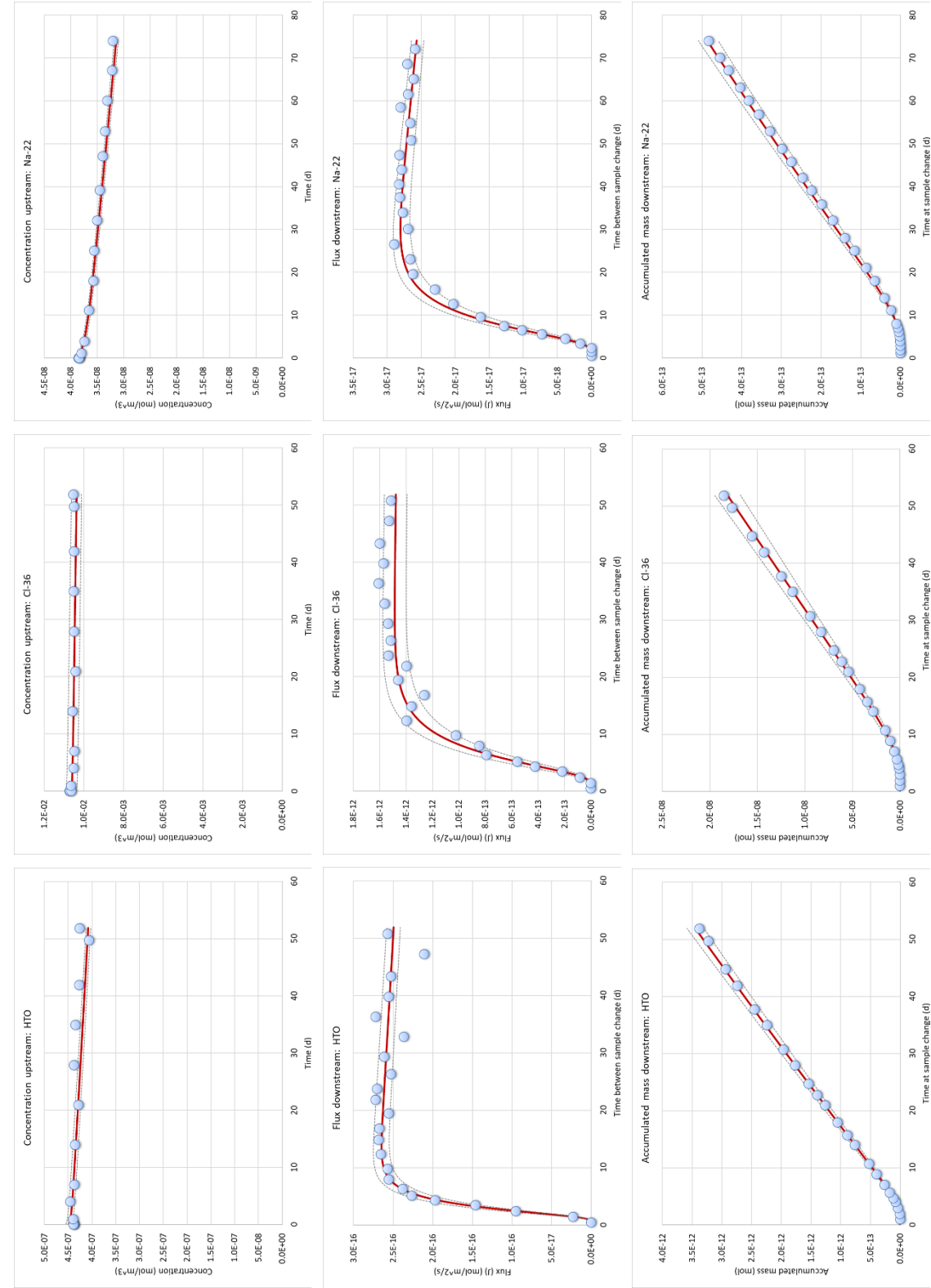
BUL1-1-965.46-DI (S1) – Opalinus Clay – SPW-BUL

Sample diameter 63.52 mm – sample thickness 11.97 mm



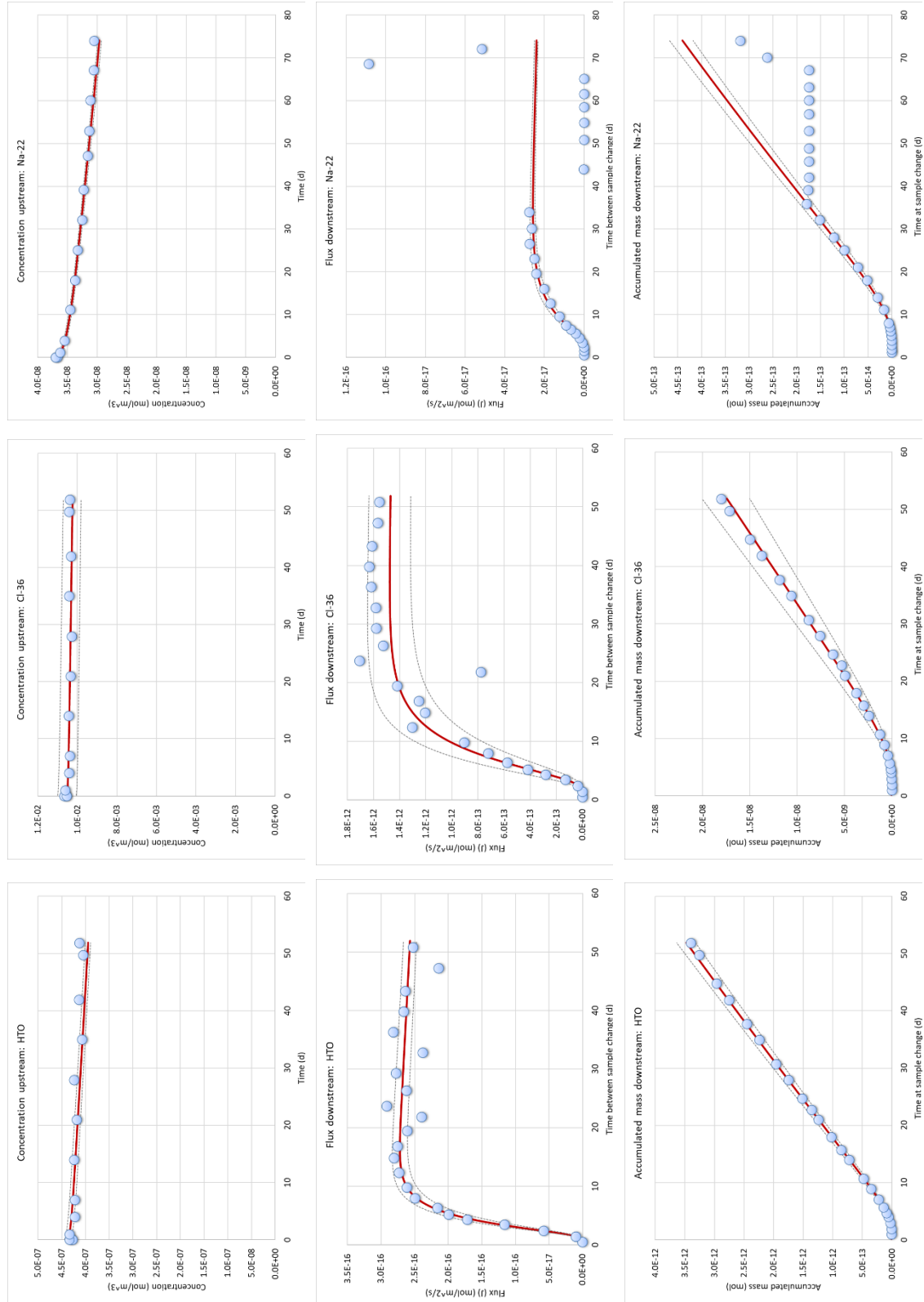
BUL1-1-969.15-DI (S2) – Opalinus Clay – SPW-BUL

Sample diameter 63.60 mm – sample thickness 13.36 mm



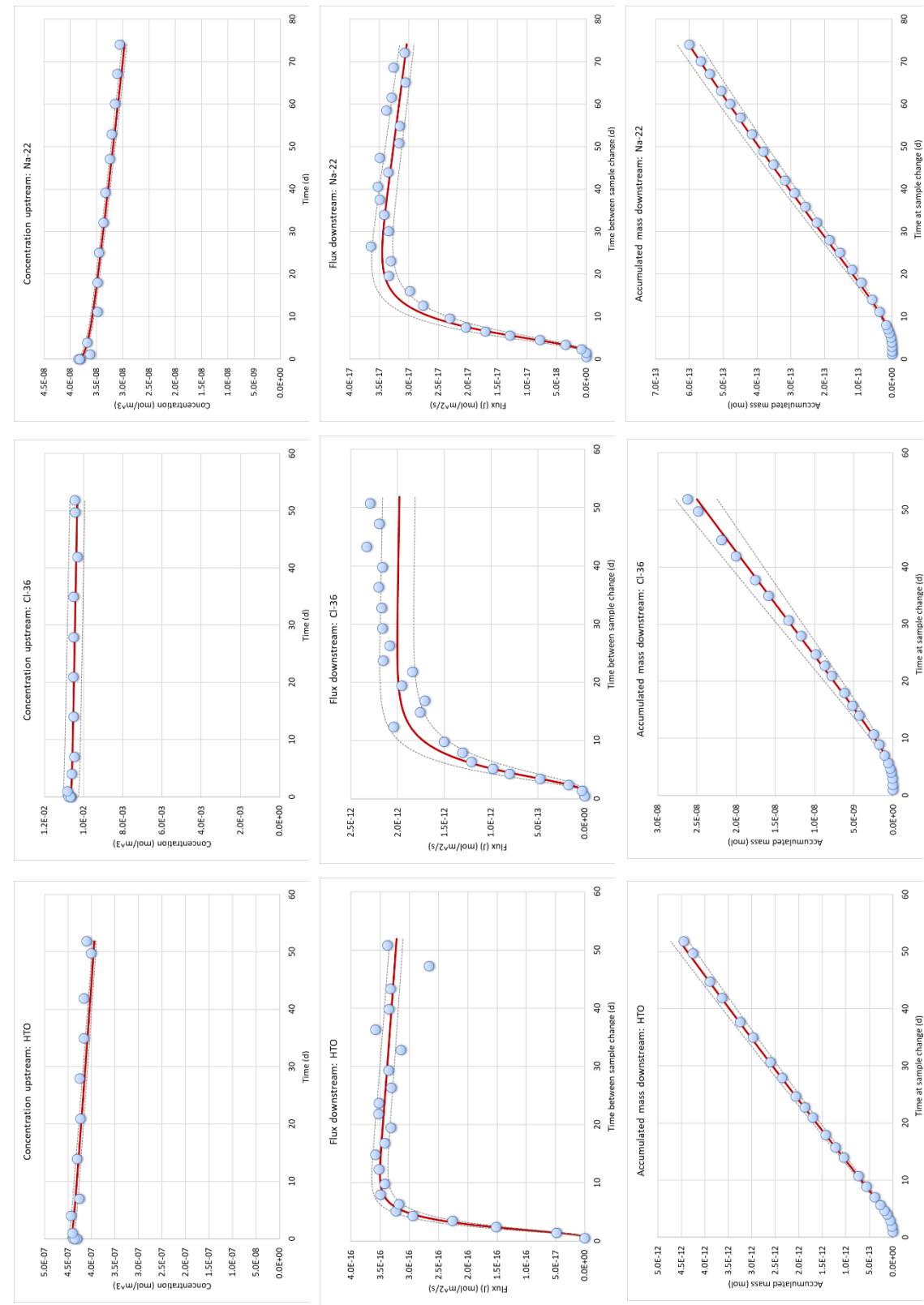
BUL1-1-975.04-DI (S2) – Opalinus Clay – SPW-BUL

Sample diameter 63.57 mm – sample thickness 12.89 mm



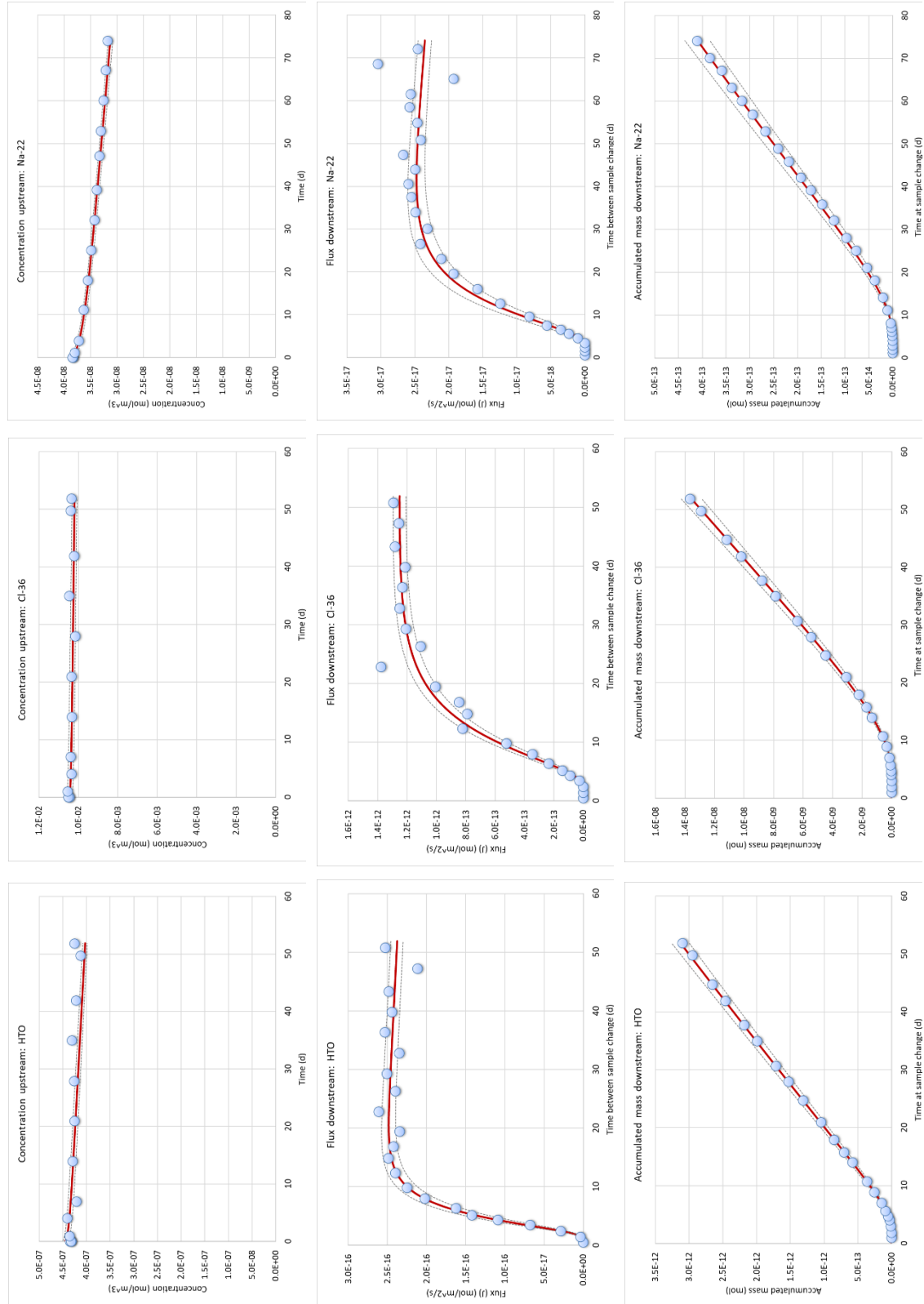
BUL1-1-984.49-DI (S1) – Opalinus Clay – SPW-BUL

Sample diameter 63.54 mm – sample thickness 10.29 mm



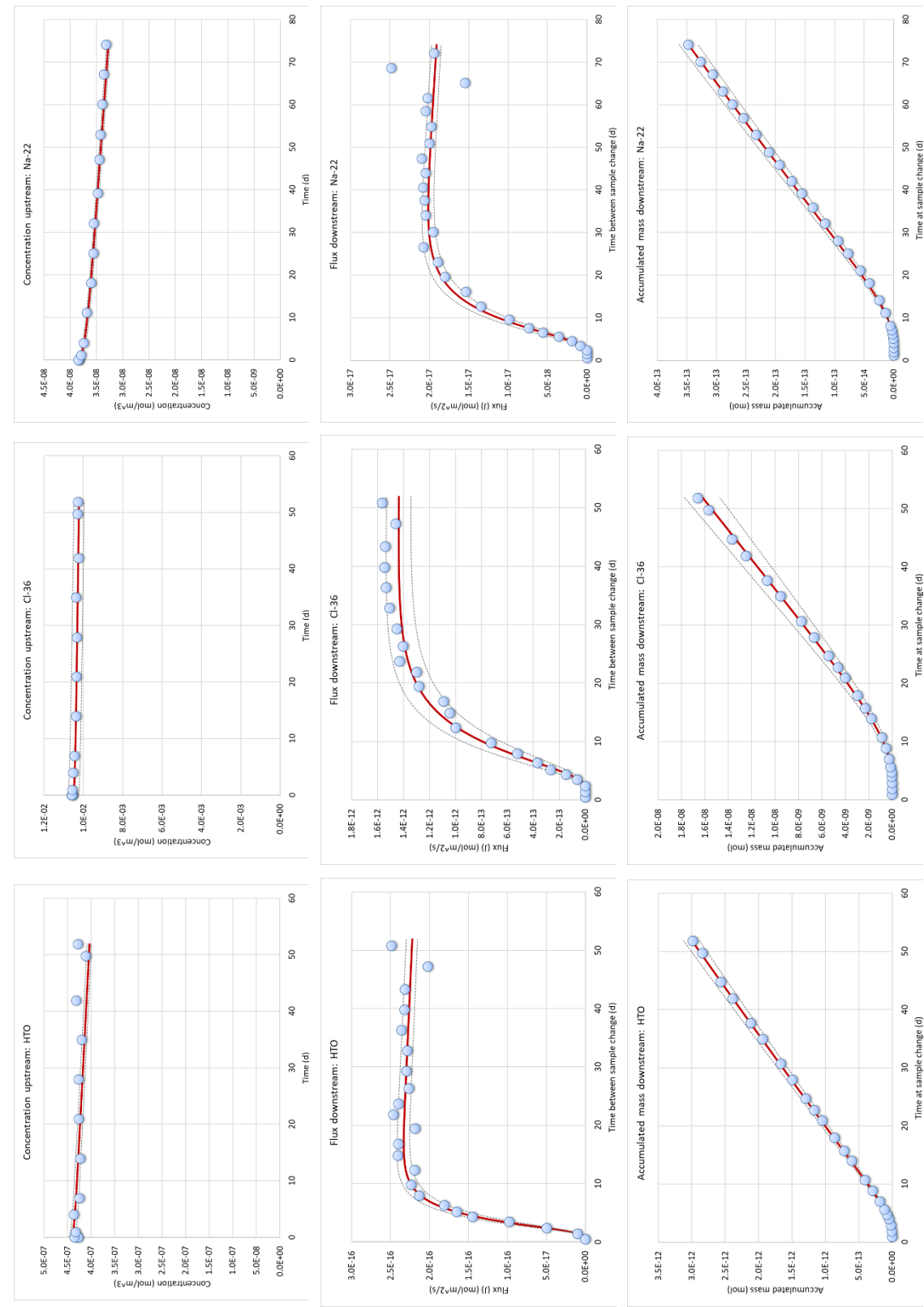
BUL1-1-995.09-DI (S2) – Opalinus Clay – SPW-BUL

Sample diameter 63.65 mm – sample thickness 13.40 mm



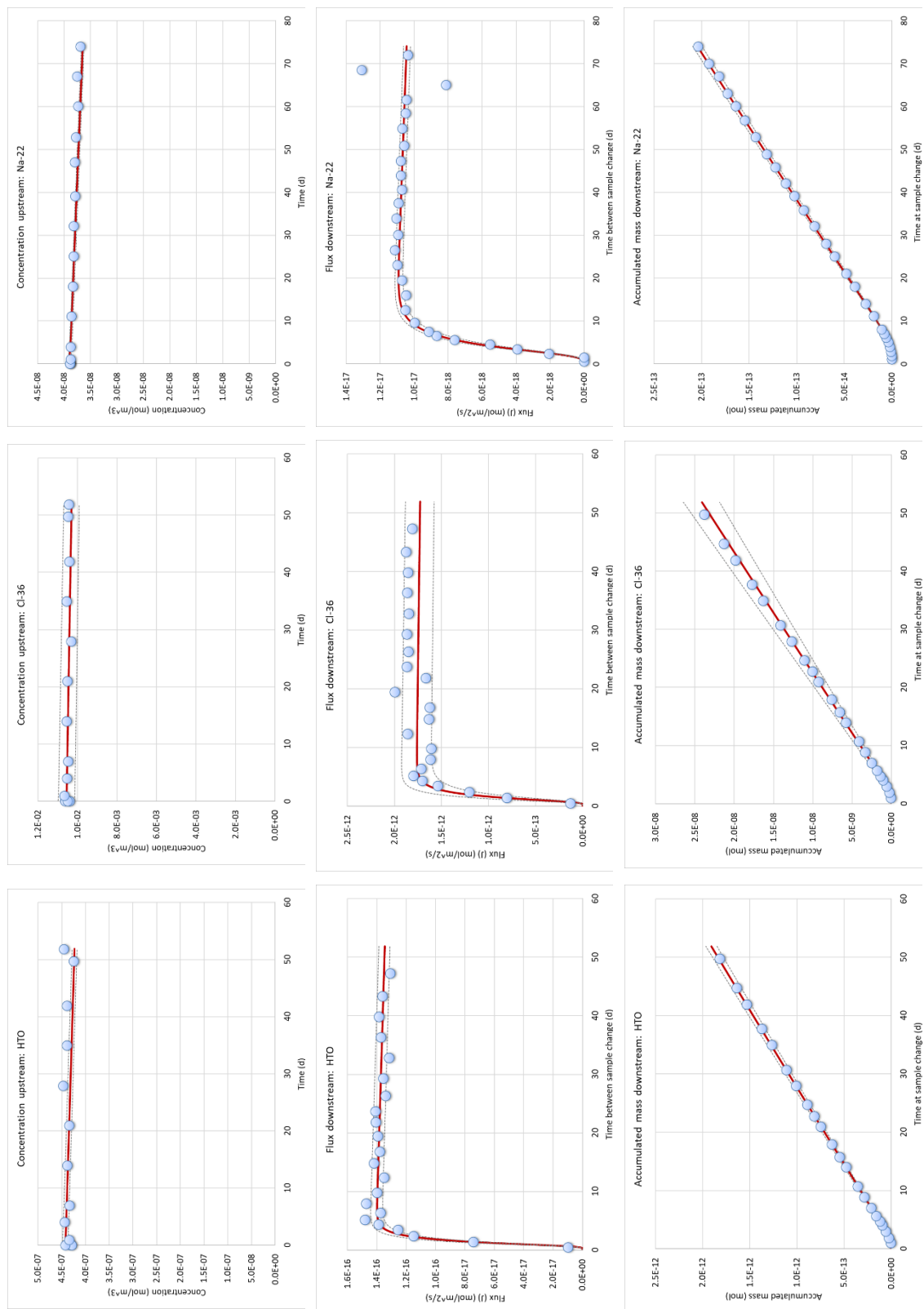
BUL1-1-1002.19-DI (S1) – Stafflegg Formation – SPW-BUL

Sample diameter 63.65 mm – sample thickness 11.50 mm



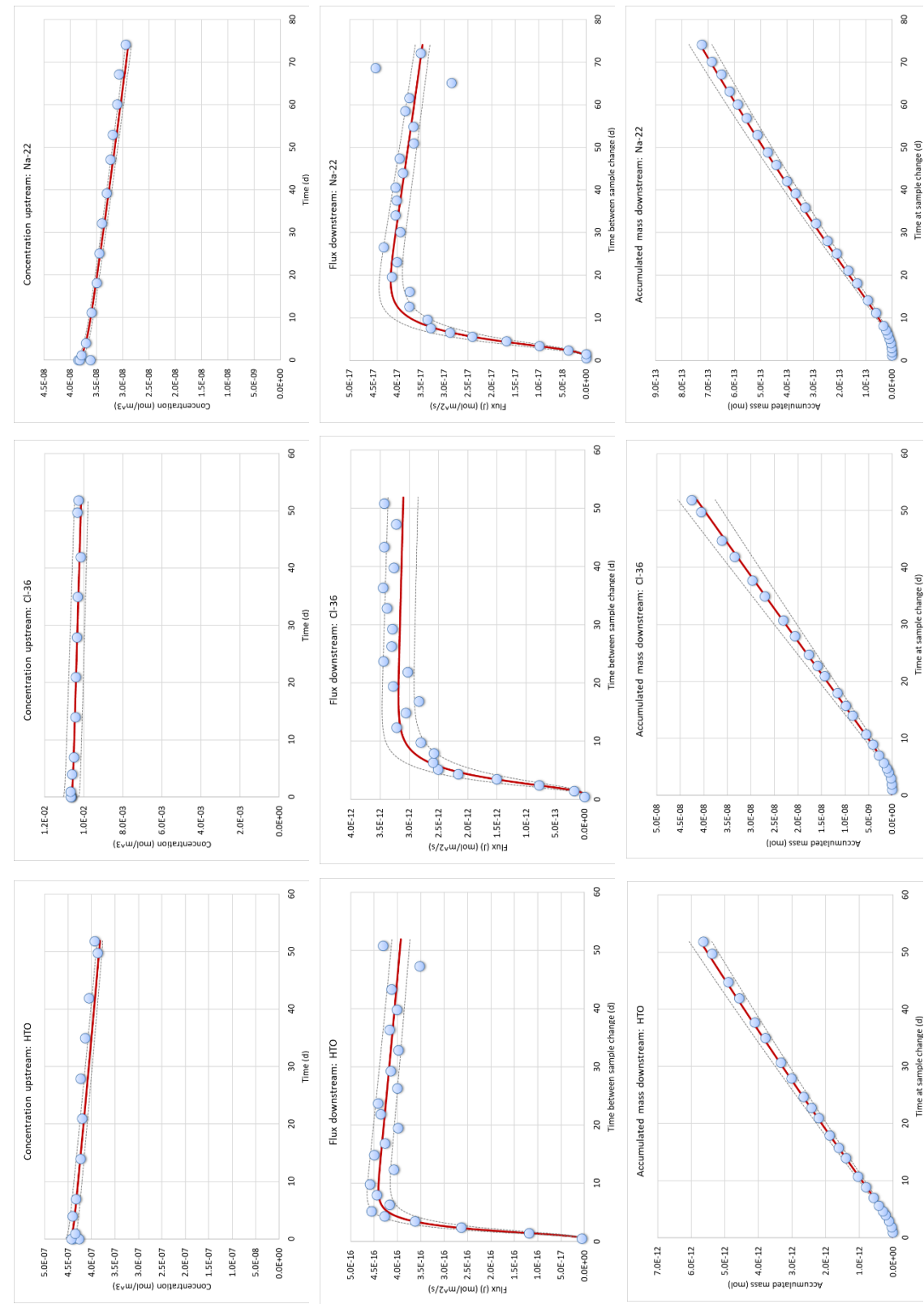
BUL1-1-1008.42-DI (S2) – Stafflegg Formation – SPW-BUL

Sample diameter 63.70 mm – sample thickness 12.57 mm

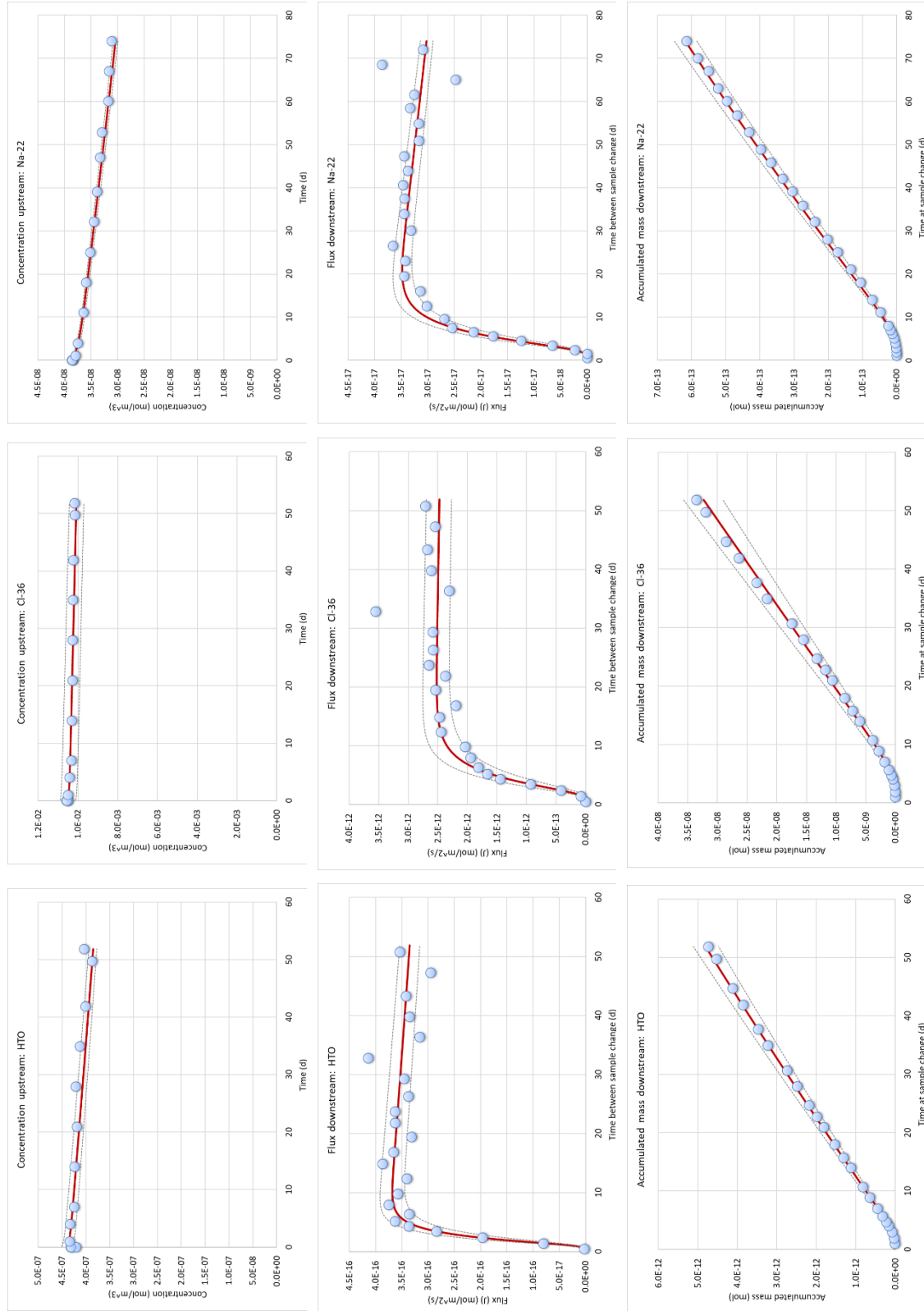


BUL1-1-1014.23-DI (S1) – Stafflegg Formation – SPW-BUL

Sample diameter 63.70 mm – sample thickness 12.77 mm

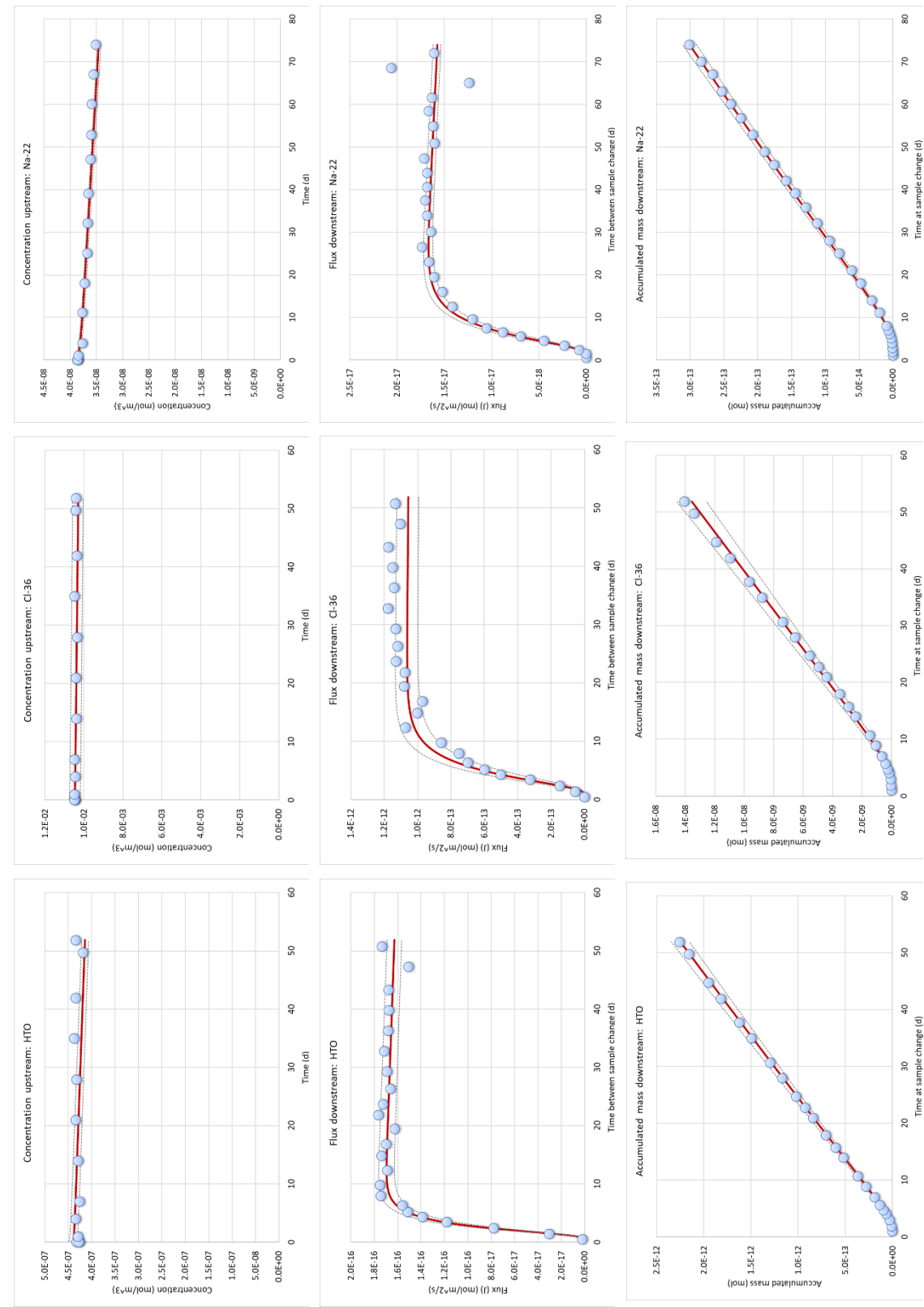


BUL1-1-1017.33-DI (S1) – Stafflegg Formation – SPW-BUL
 Sample diameter 63.65 mm – sample thickness 12.51 mm



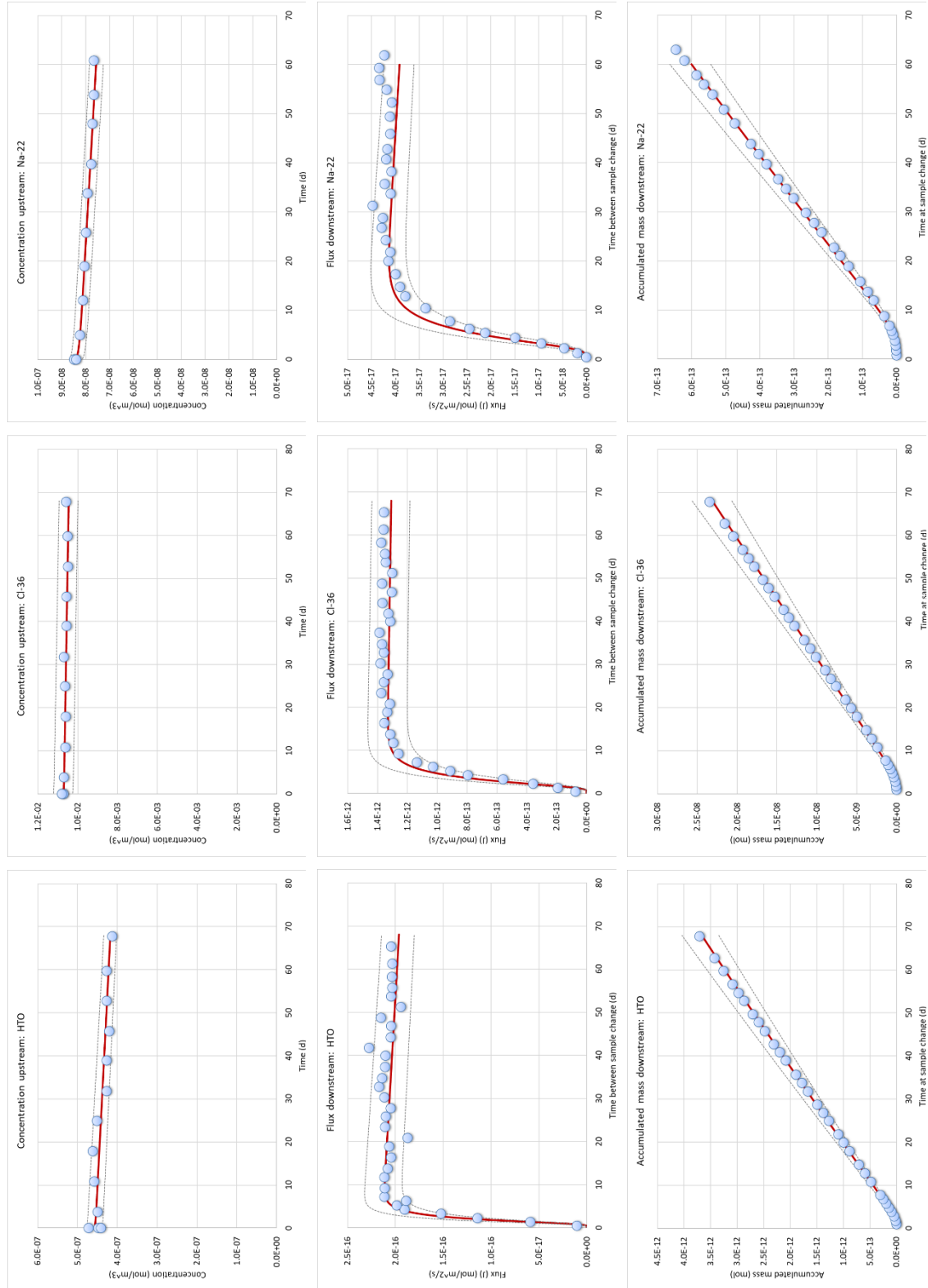
BUL1-1-1025.96-DI (S1) – Staffelegg Formation – SPW-BUL

Sample diameter 63.62 mm – sample thickness 13.36 mm



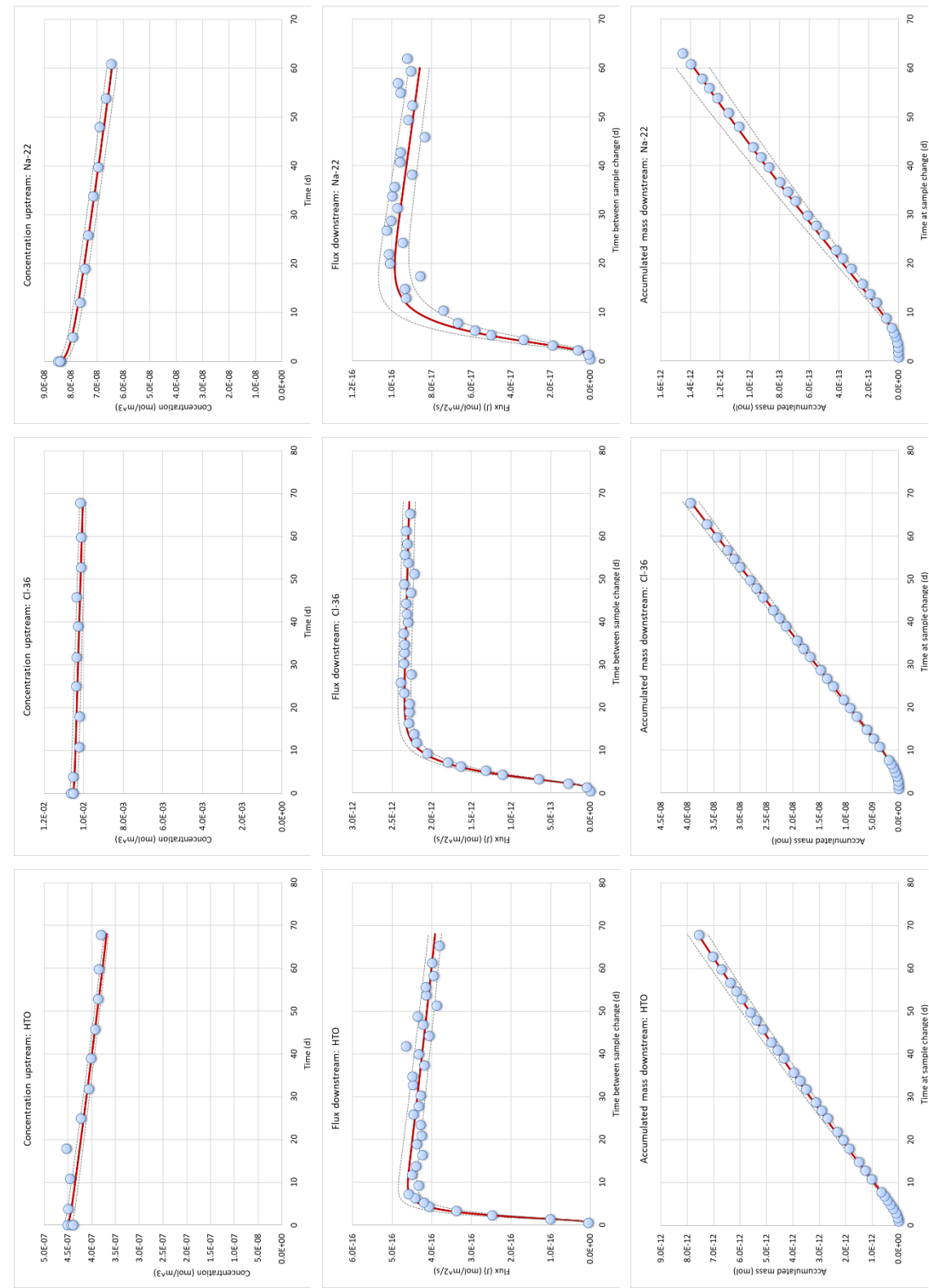
STA2-1-633.23-DI (S1) – Villigen Formation – SPW-STA

Sample diameter 63.48 mm – sample thickness 12.27 mm



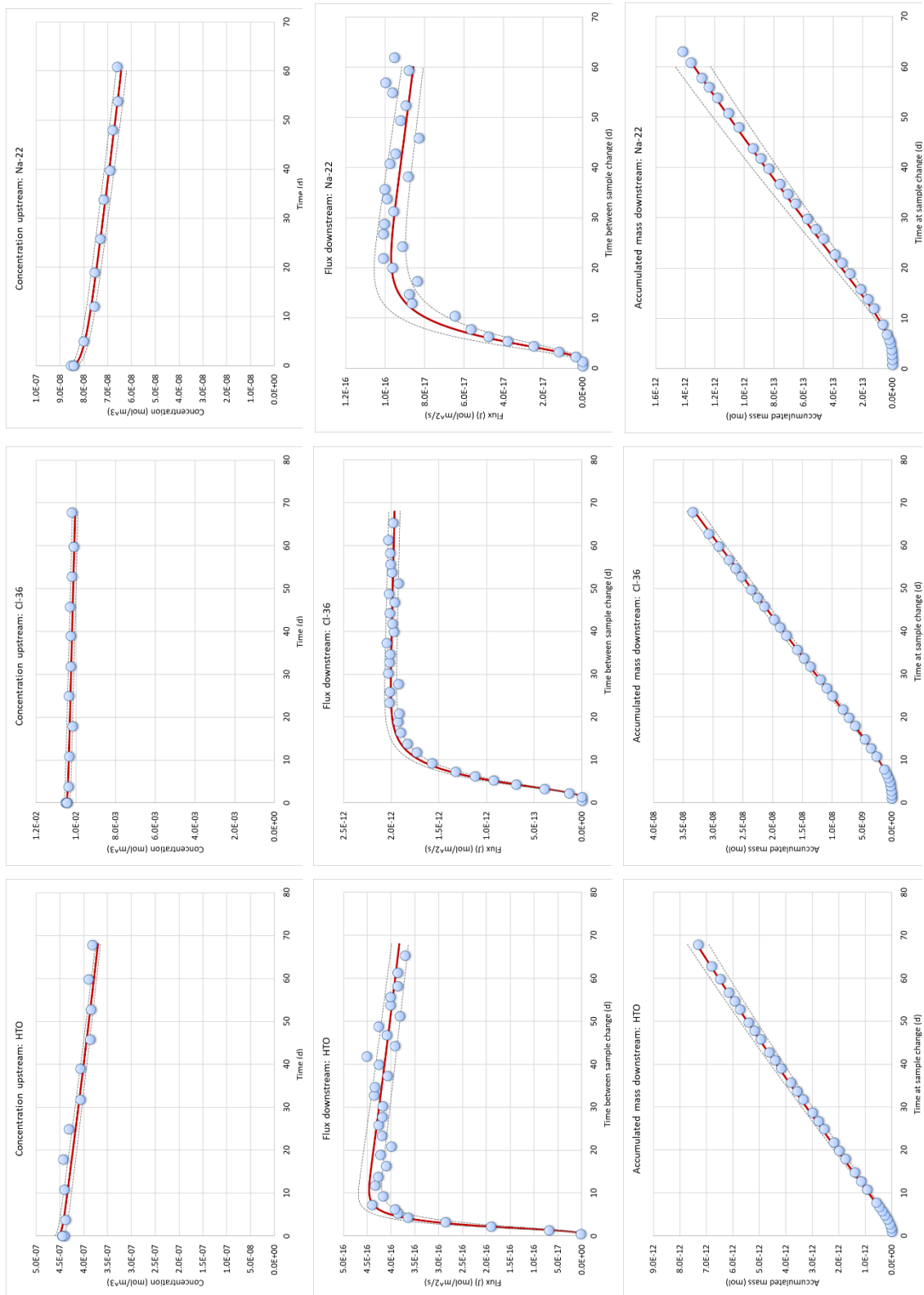
STA2-1-752.73-DI (S2) – Parkinsoni-Württembergica-Schichten – SPW-STA

Sample diameter 63.42 mm – sample thickness 11.60 mm



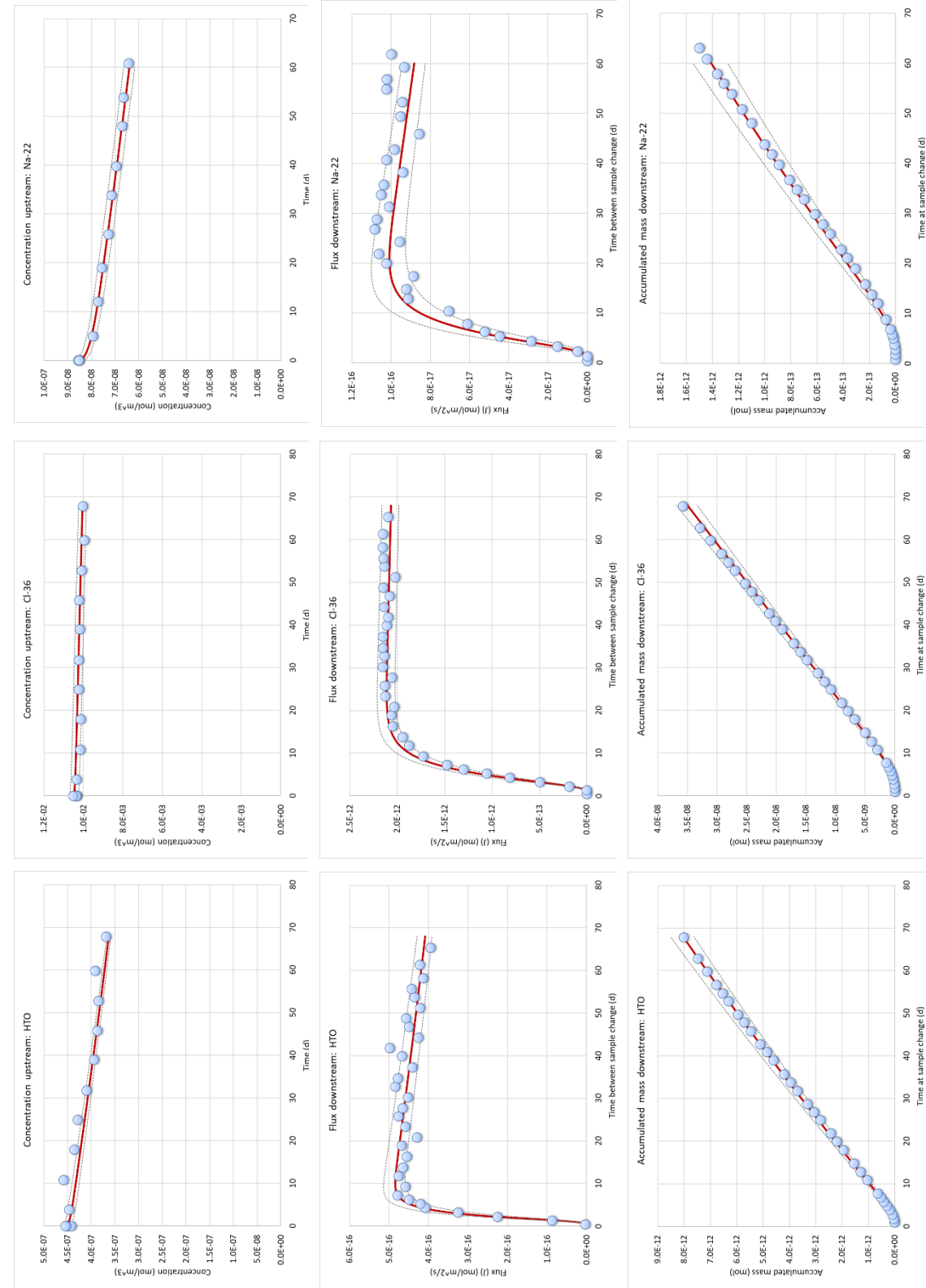
STA2-1-758.02-DI (S1) – Parkinsoni-Württembergica-Schichten – SPW-STA

Sample diameter 63.58 mm – sample thickness 12.16 mm



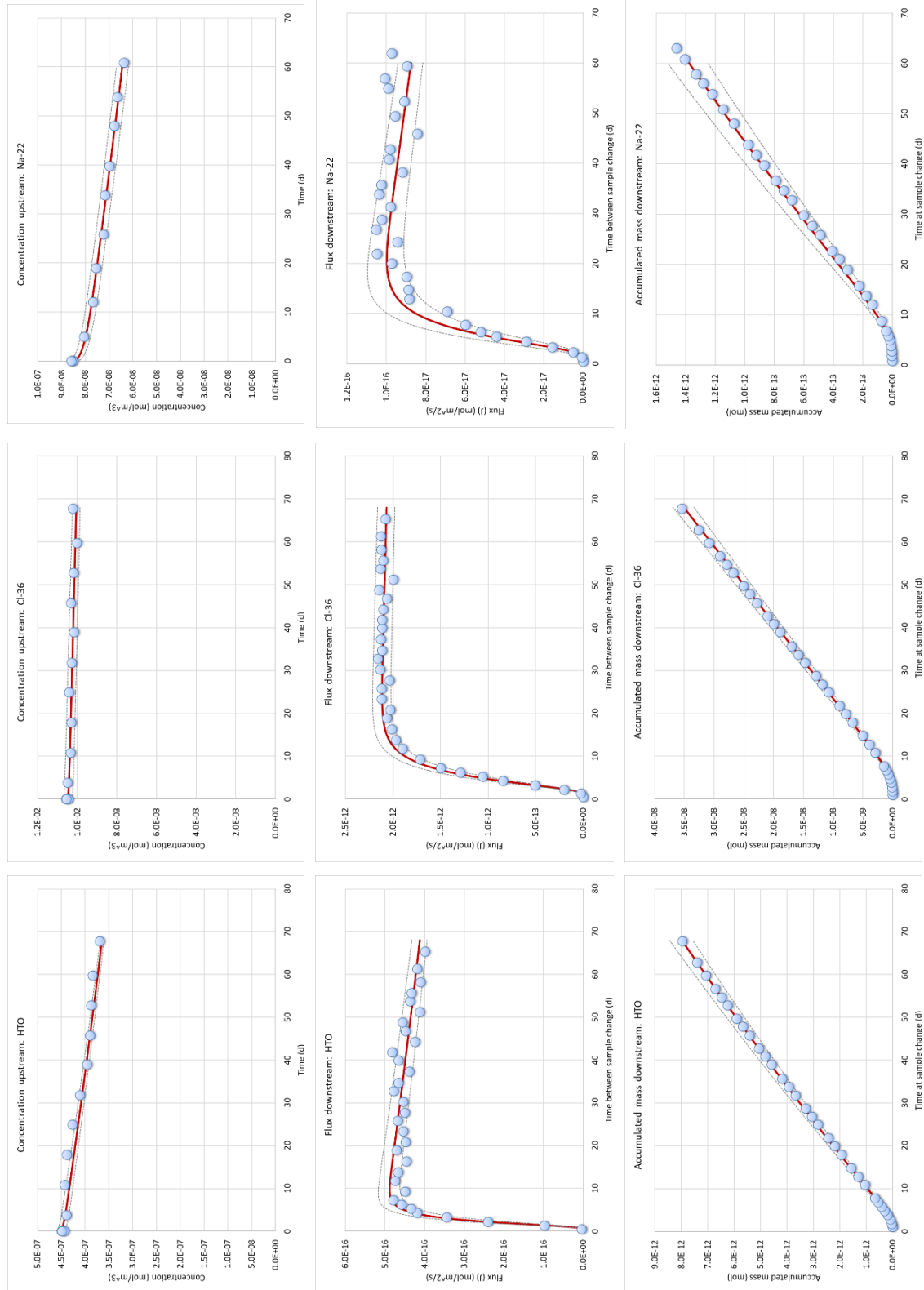
STA2-1-758.23-DI (S2) – Parkinsoni-Württembergica-Schichten – SPW-STA

Sample diameter 63.64 mm – sample thickness 11.85 mm



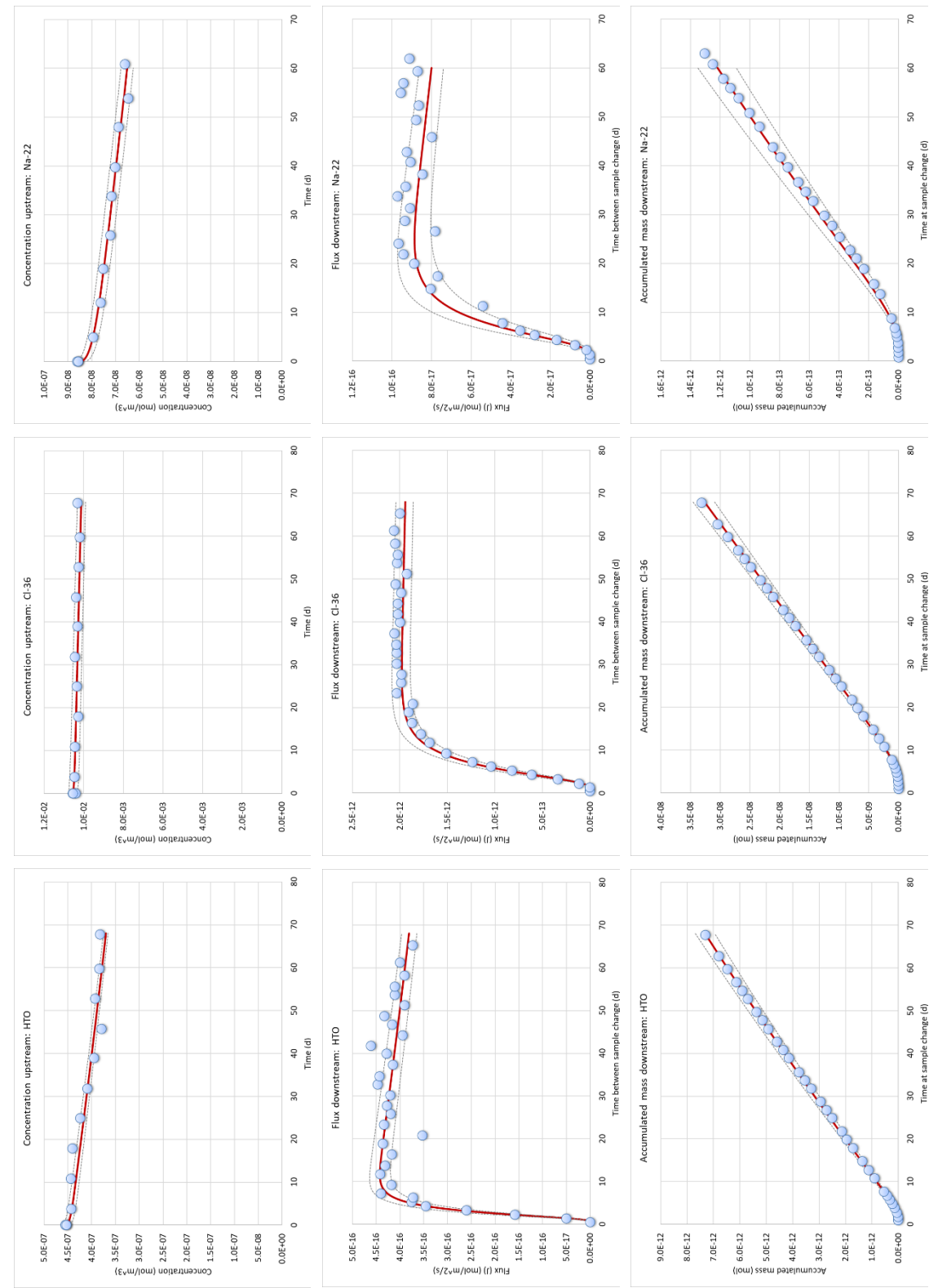
STA2-1-765.21-DI (S1) – Parkinsoni-Württembergica-Schichten – SPW-STA

Sample diameter 63.54 mm – sample thickness 10.96 mm



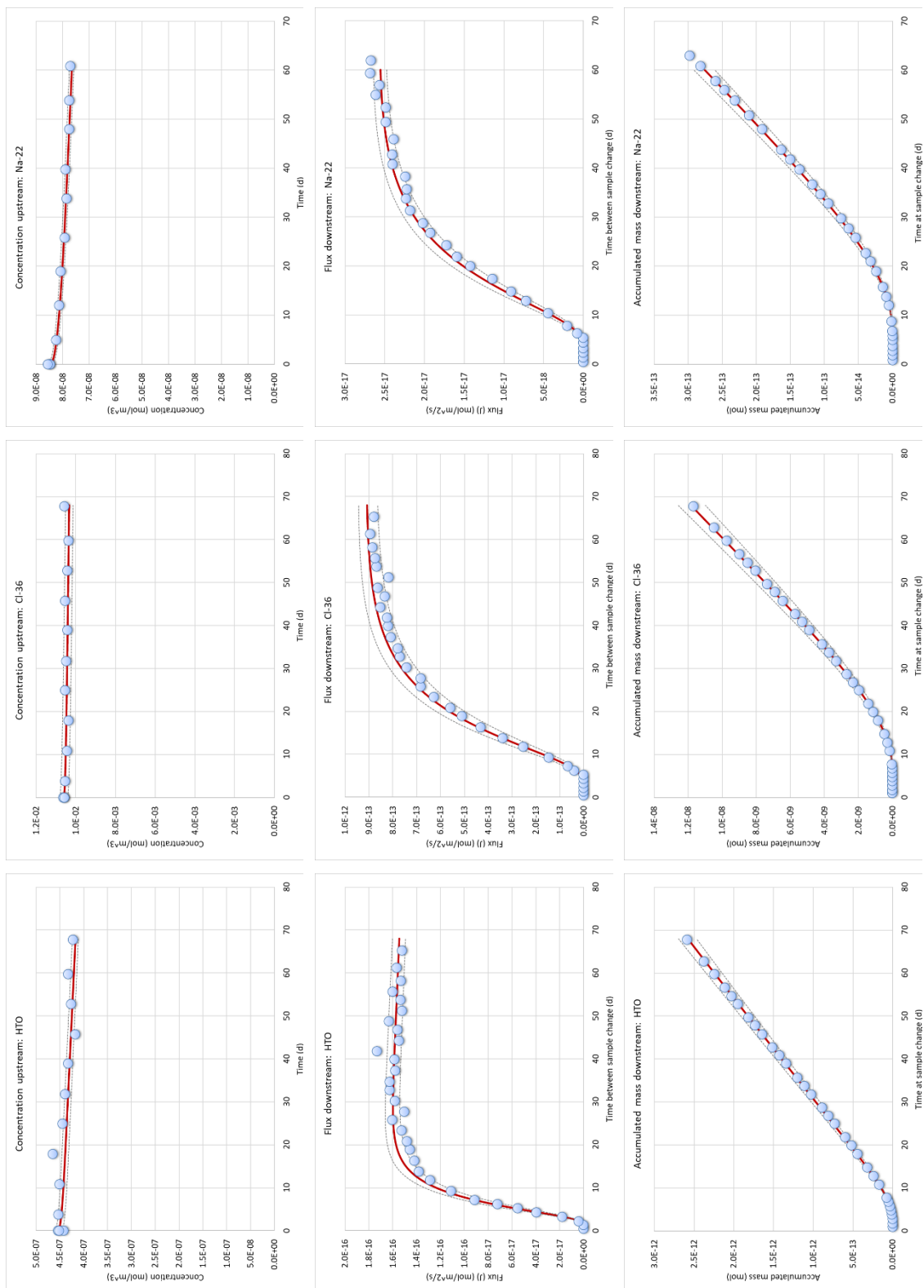
STA2-1-779.96-DI (S2) – Wedelsandstein Formation – SPW-STA

Sample diameter 63.35 mm – sample thickness 11.78 mm



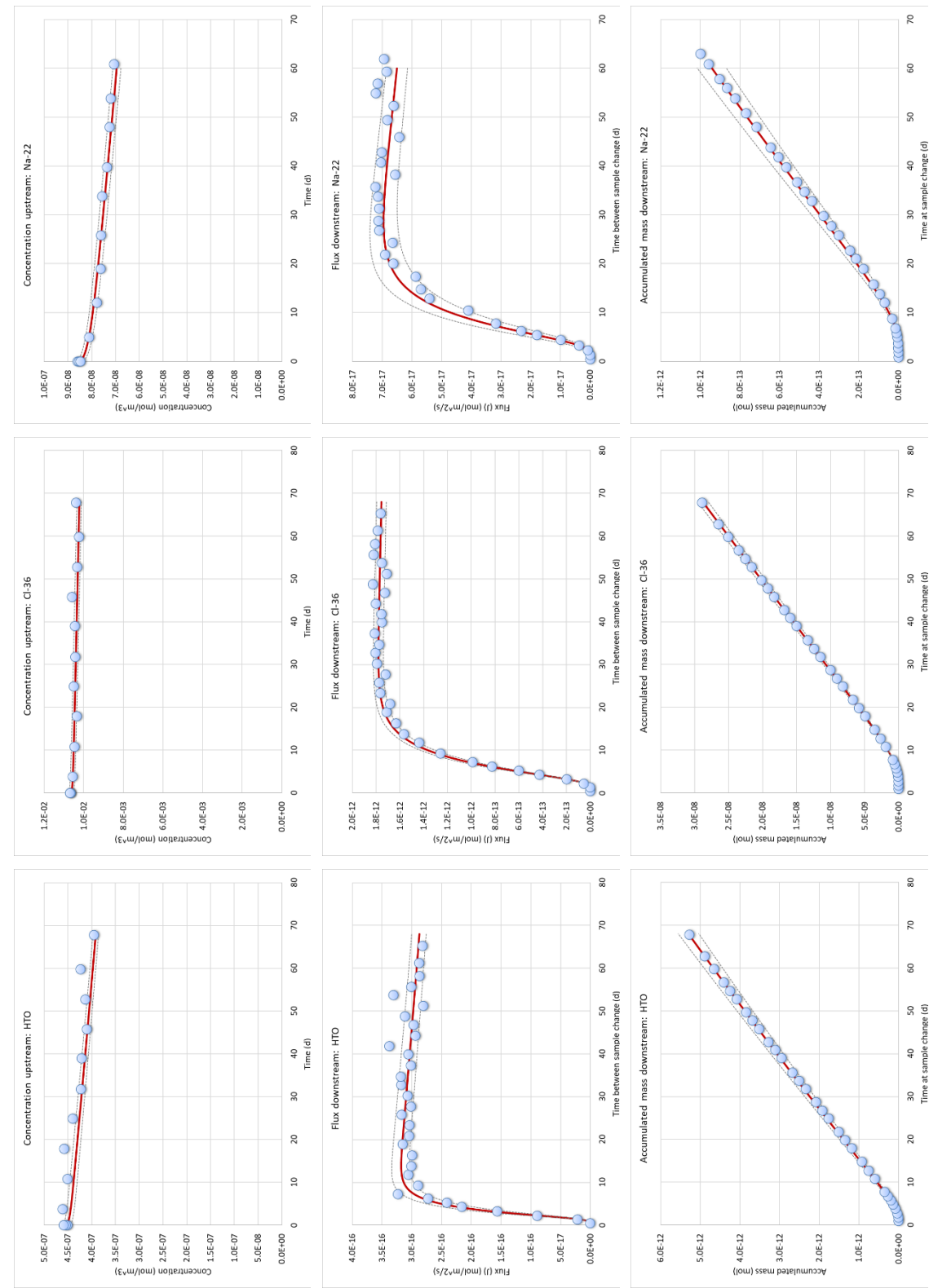
STA2-1-912.69-DI (S2) – Staffelegg Formation – SPW-STA

Sample diameter 63.74 mm – sample thickness 12.96 mm



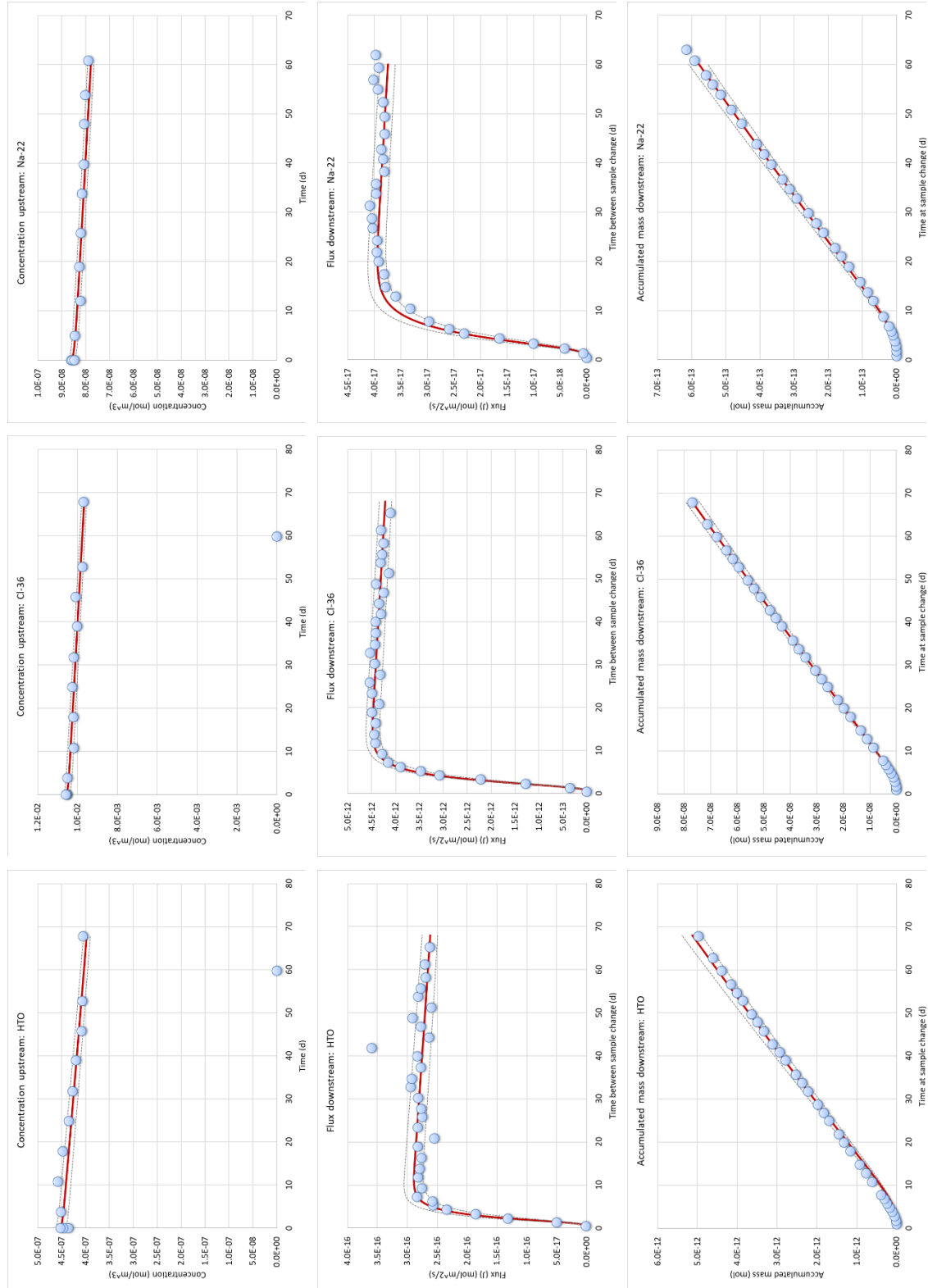
STA2-1-925.51-DI (S1) – Staffelegg Formation – SPW-STA

Sample diameter 63.30 mm – sample thickness 13.70 mm



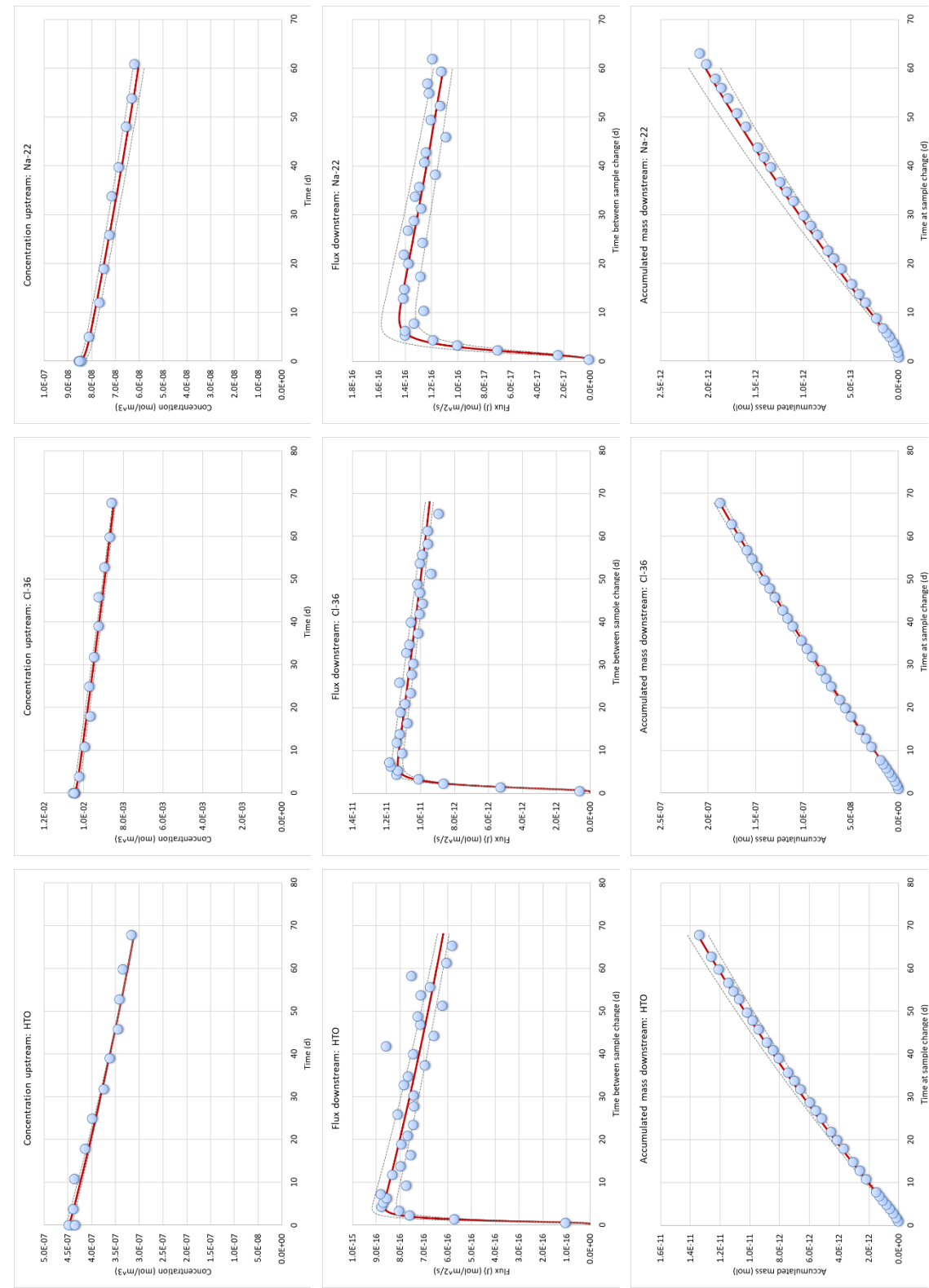
STA2-1-930.53-DI (S1) – Staffelegg Formation – SPW-STA

Sample diameter 63.68 mm – sample thickness 12.13 mm



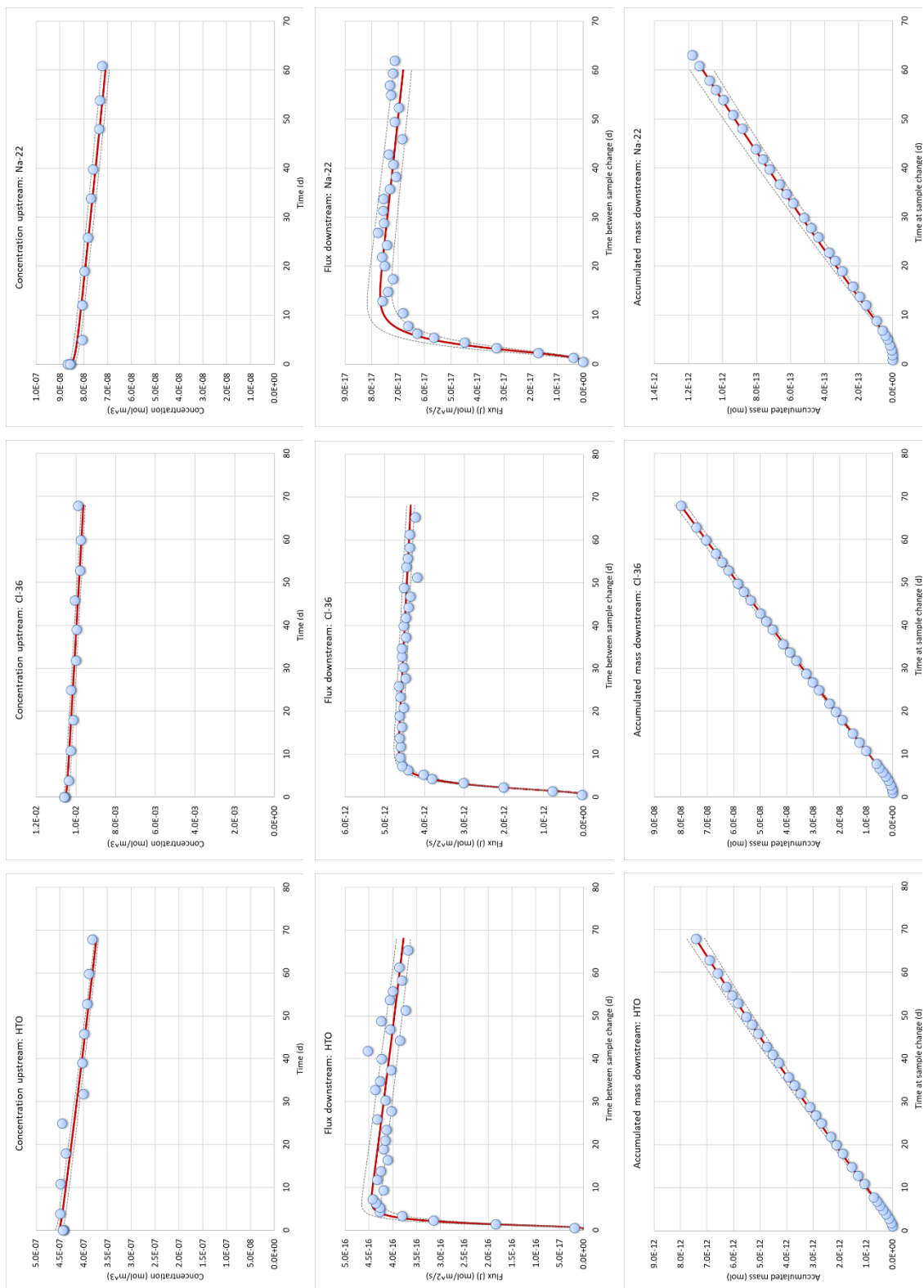
STA2-1-955.40-DI (S1) – Klettgau Formation – SPW-STA

Sample diameter 63.46 mm – sample thickness 12.58 mm



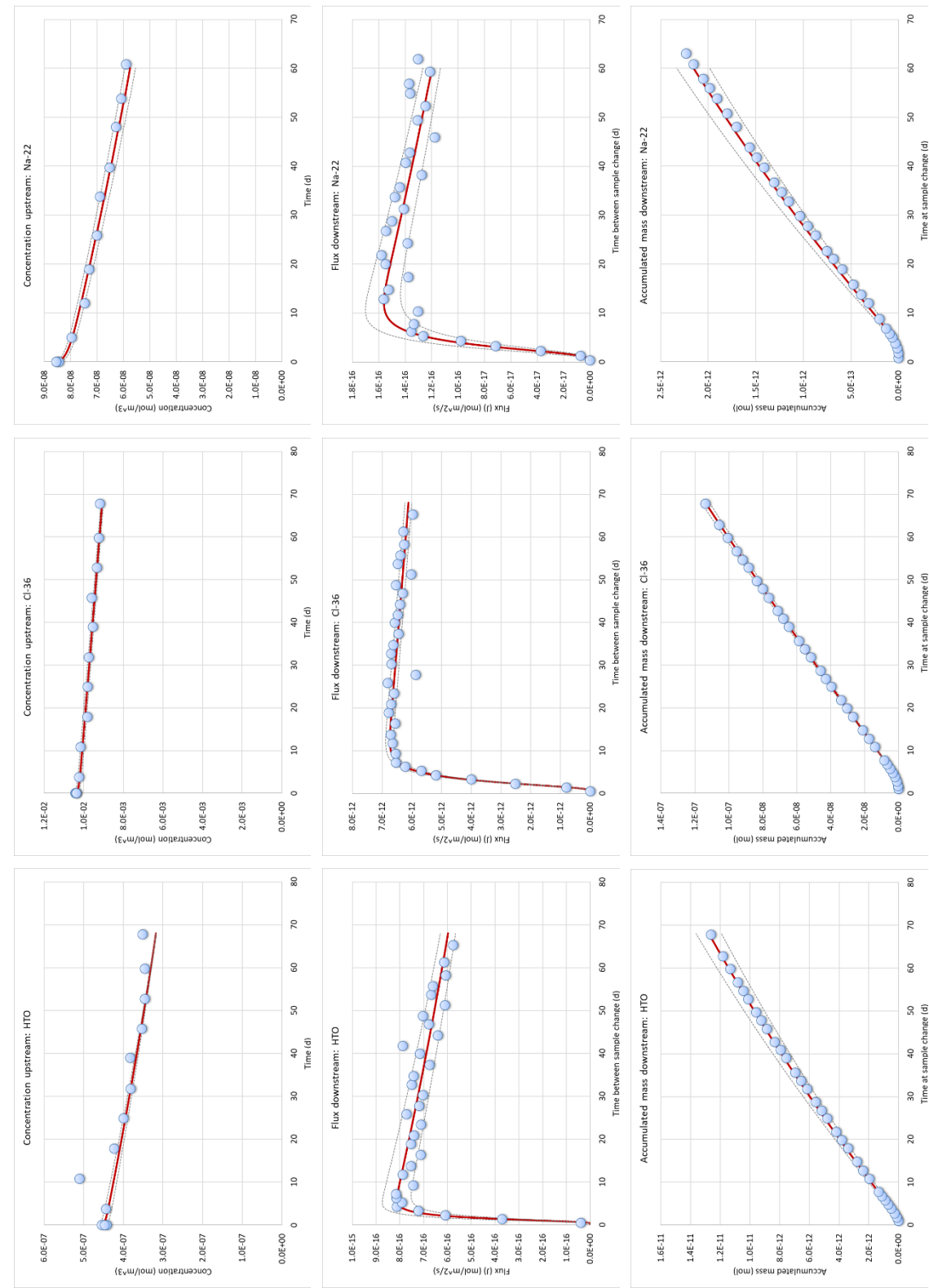
STA2-1-957.28-DI (S1) – Klettgau Formation – SPW-STA

Sample diameter 63.64 mm – sample thickness 13.11 mm



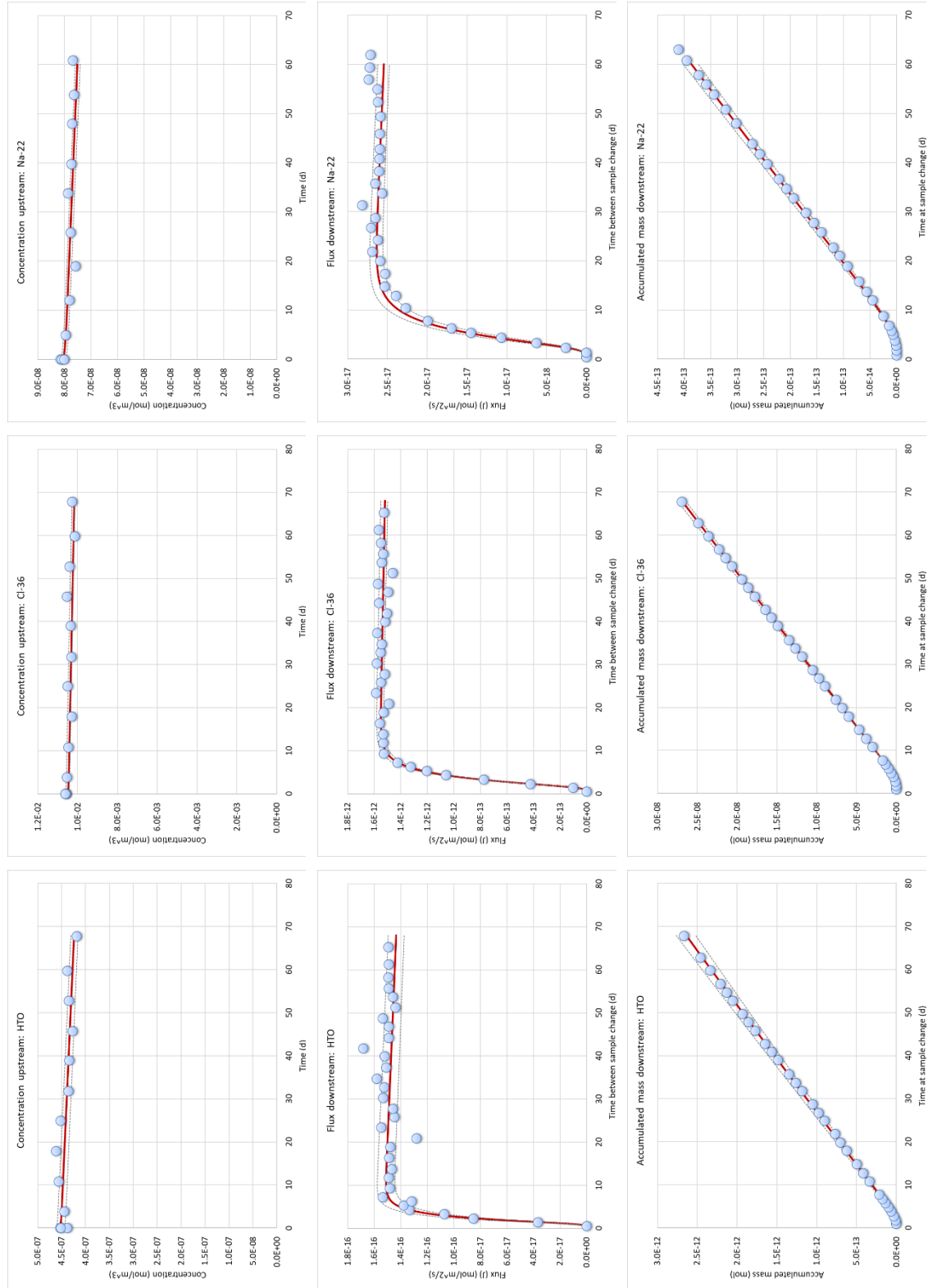
STA2-1-963.35-DI (S2) – Klettgau Formation – SPW-STA

Sample diameter 63.41 mm – sample thickness 12.14 mm



STA3-1-650.49-DI (S2) – Wildegge Formation – SPW-STA

Sample diameter 63.45 mm – sample thickness 12.05 mm



STA3-1-686.50-DI (S2) – Wildegg Formation – SPW-STA

Sample diameter 63.52 mm – sample thickness 12.04 mm

Experiment terminated

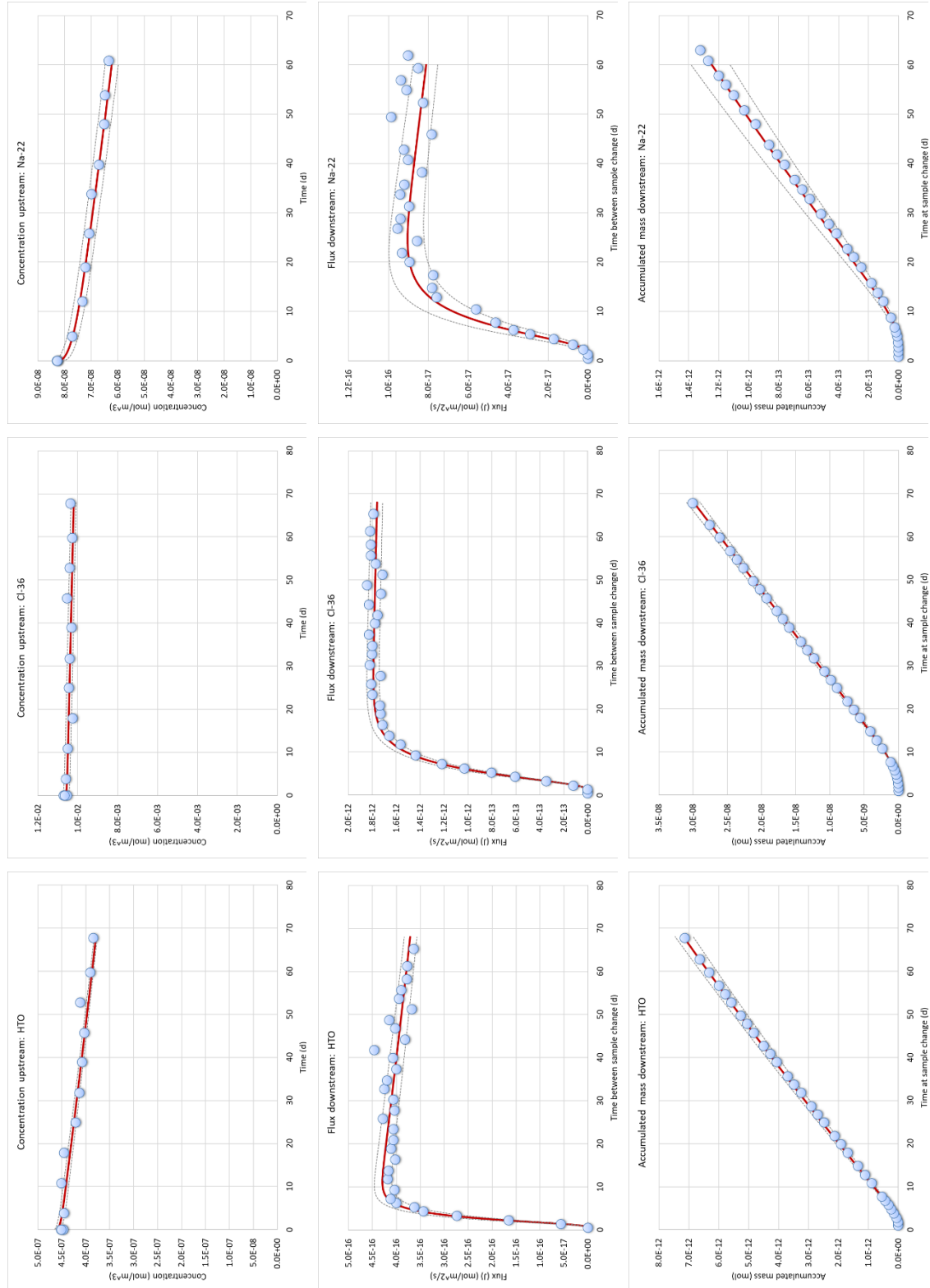
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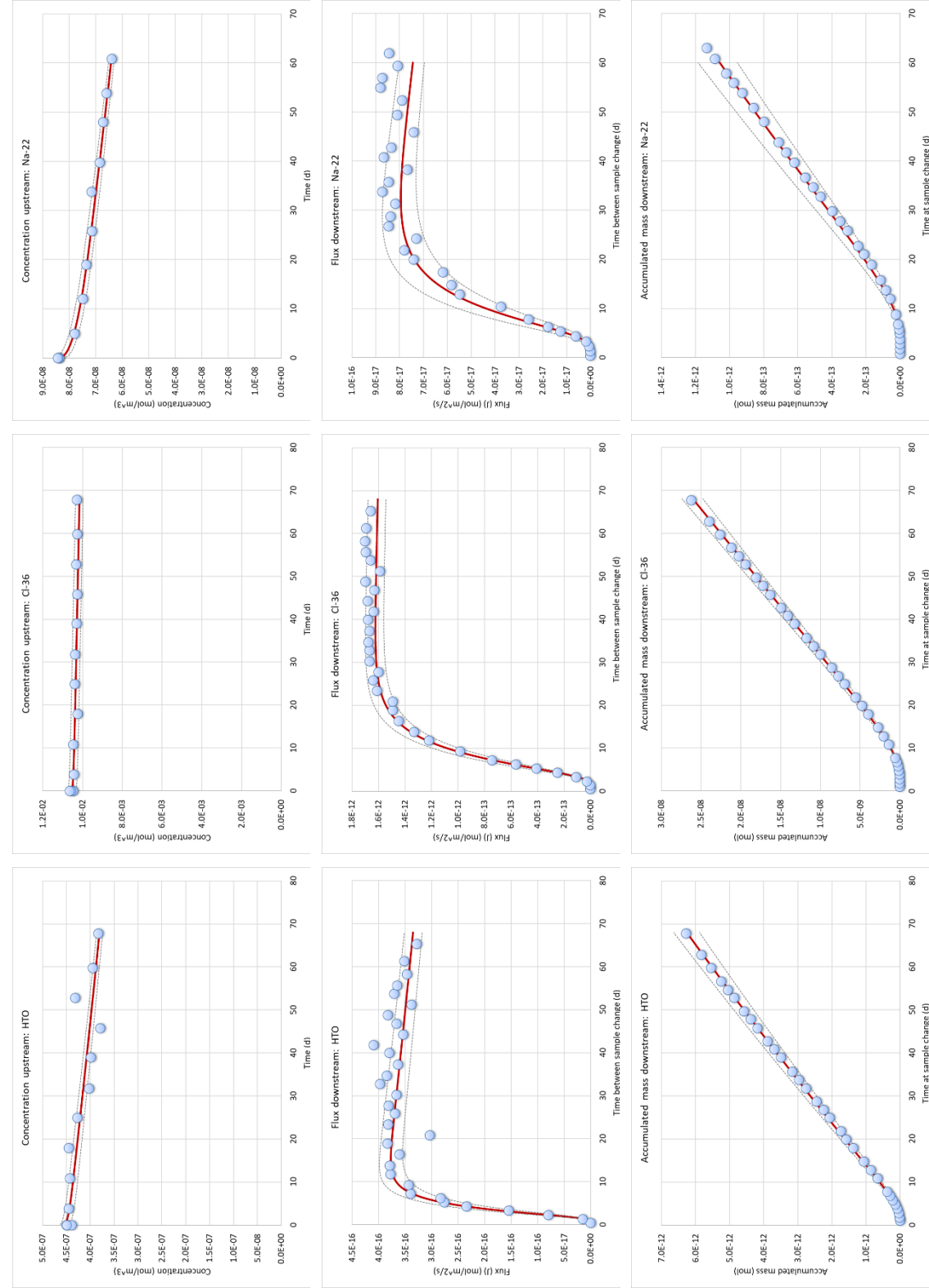
STA3-1-748.38-DI (S2) – Wedelsandstein Formation – SPW-STA

Sample diameter 63.63 mm – sample thickness 9.44 mm



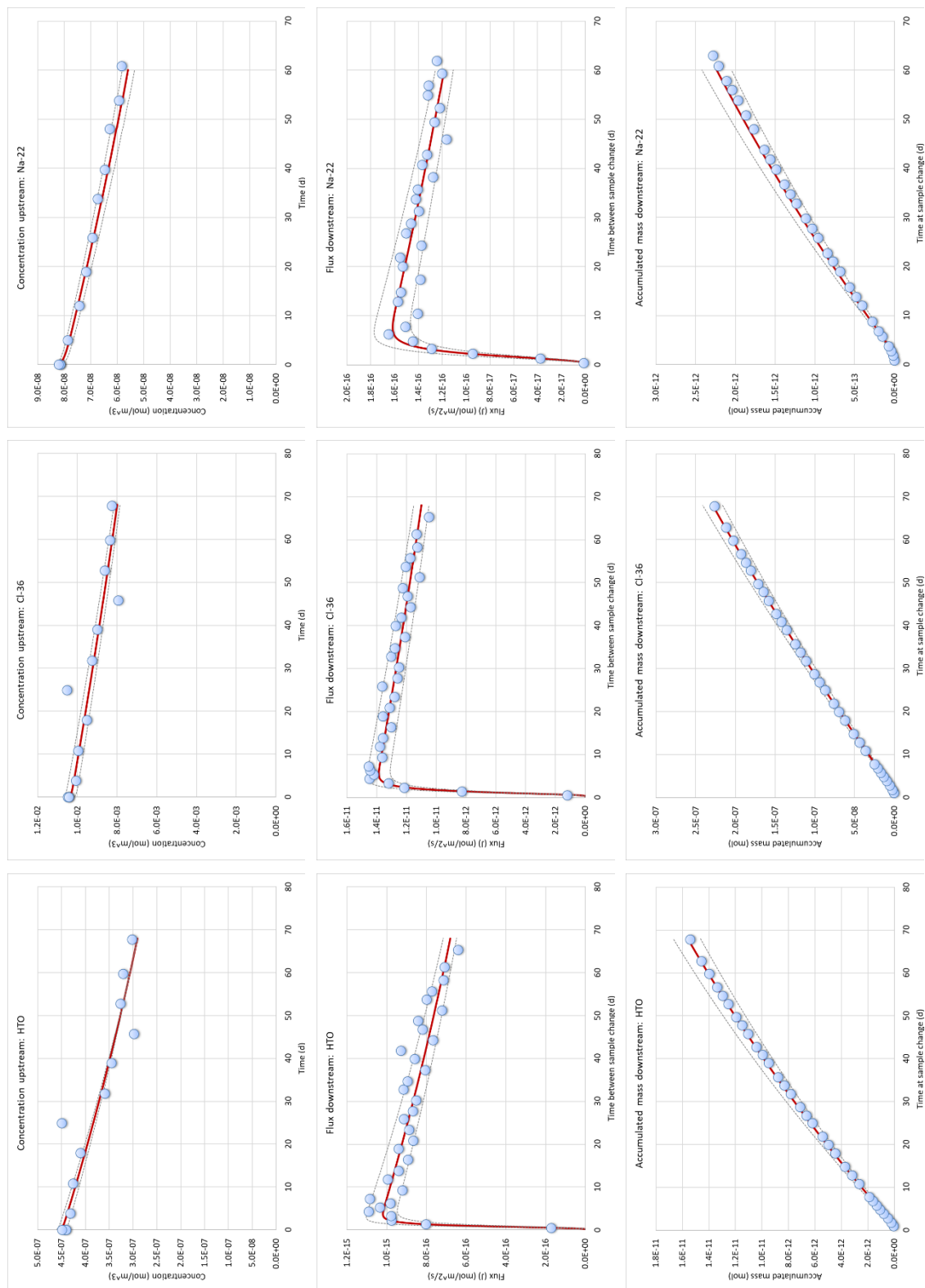
STA3-1-758.75-DI (S1) – Wedelsandstein Formation – SPW-STA

Sample diameter 63.21 mm – sample thickness 12.84 mm



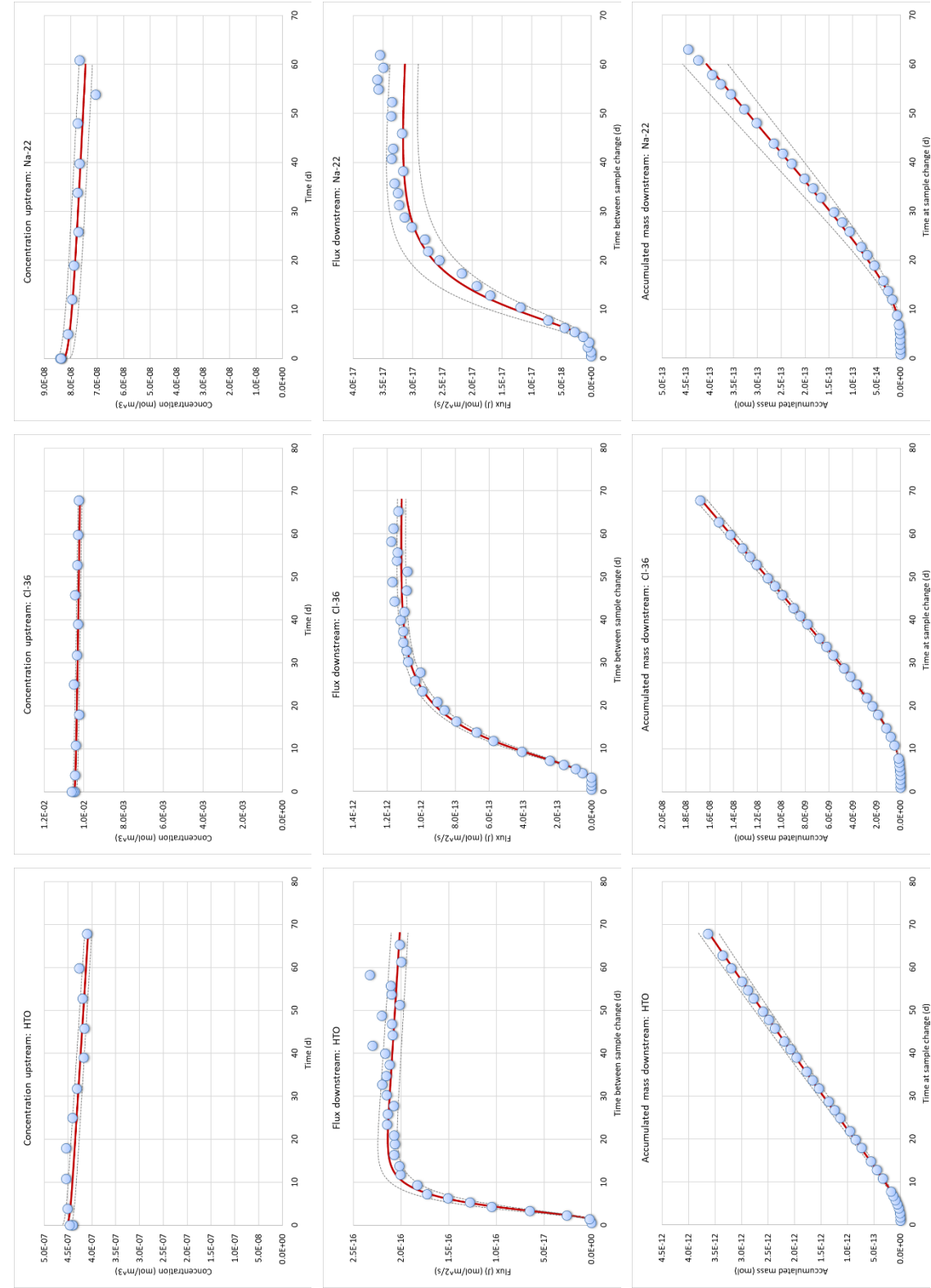
STA3-1-768.34-DI (S1) – Murchisonae-Oolith Formation – SPW-STA

Sample diameter 63.38 mm – sample thickness 11.65 mm



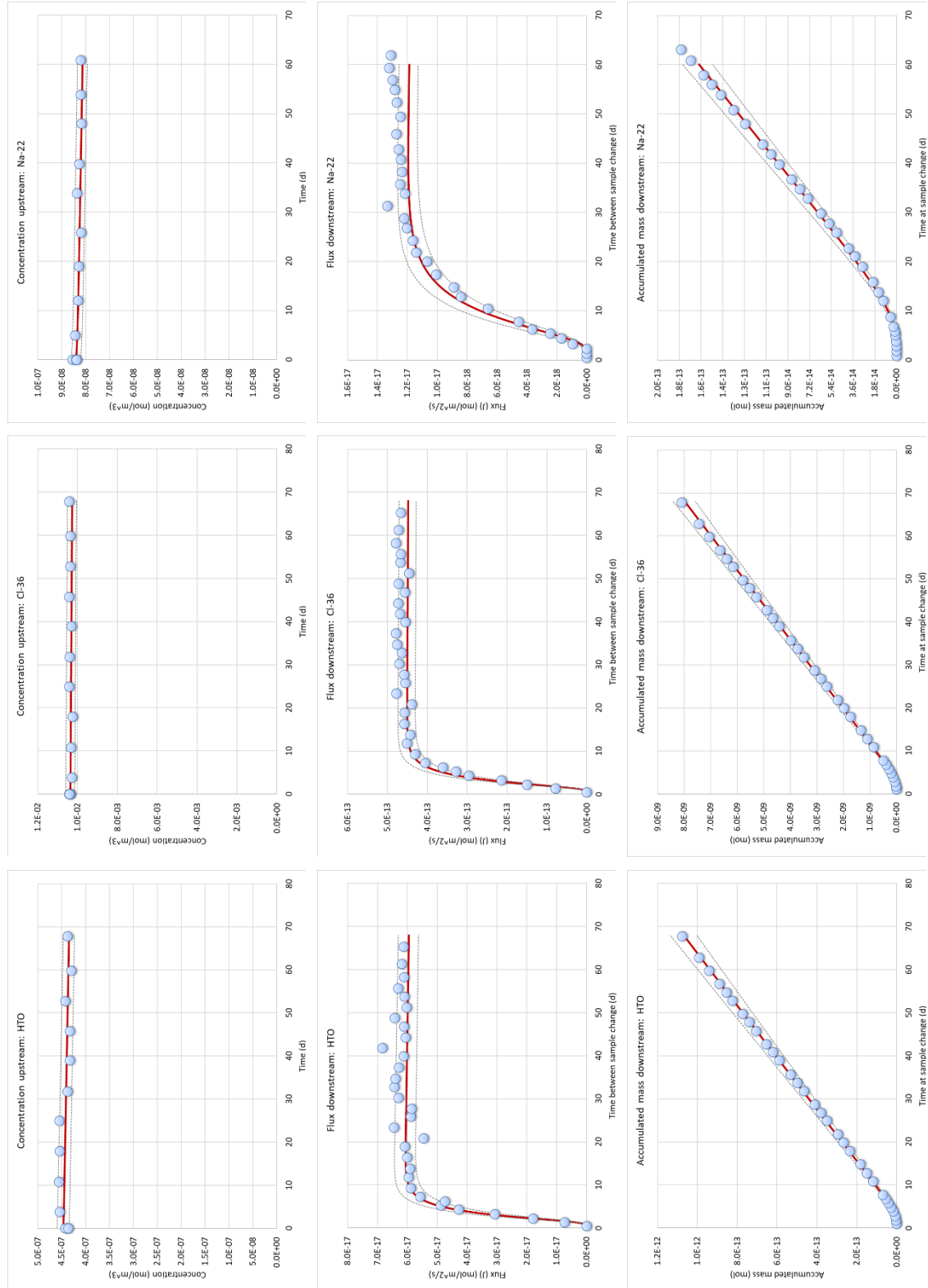
STA3-1-896.82-DI (S2) – Staffelegg Formation – SPW-STA

Sample diameter 63.67 mm – sample thickness 12.13 mm



STA3-1-919.66-DI (S2) – Staffelegg Formation – SPW-STA

Sample diameter 63.47 mm – sample thickness 12.27 mm

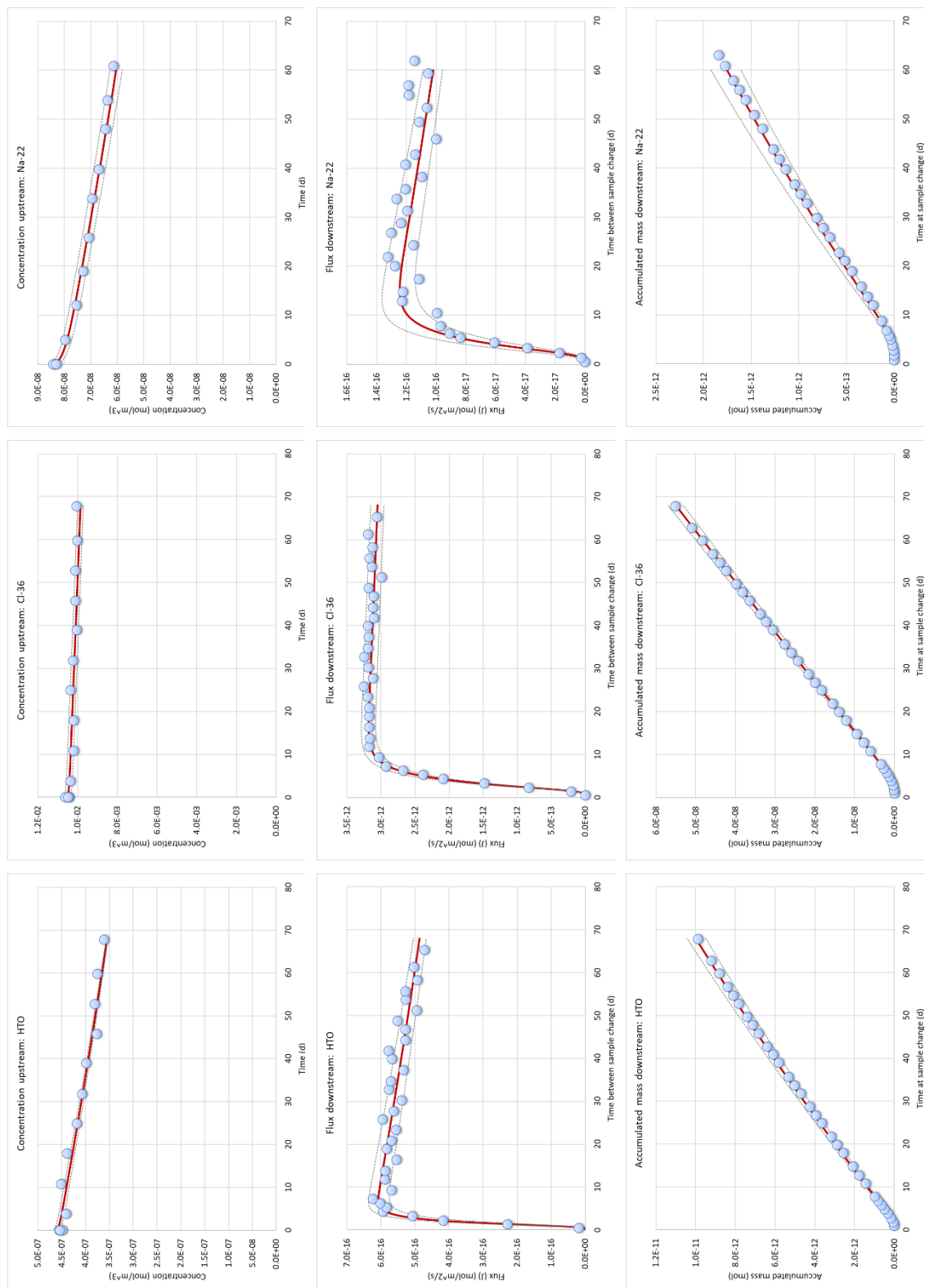


I.3 Samples from Zürich Nordost

- MAR1-1
- TRU1-1

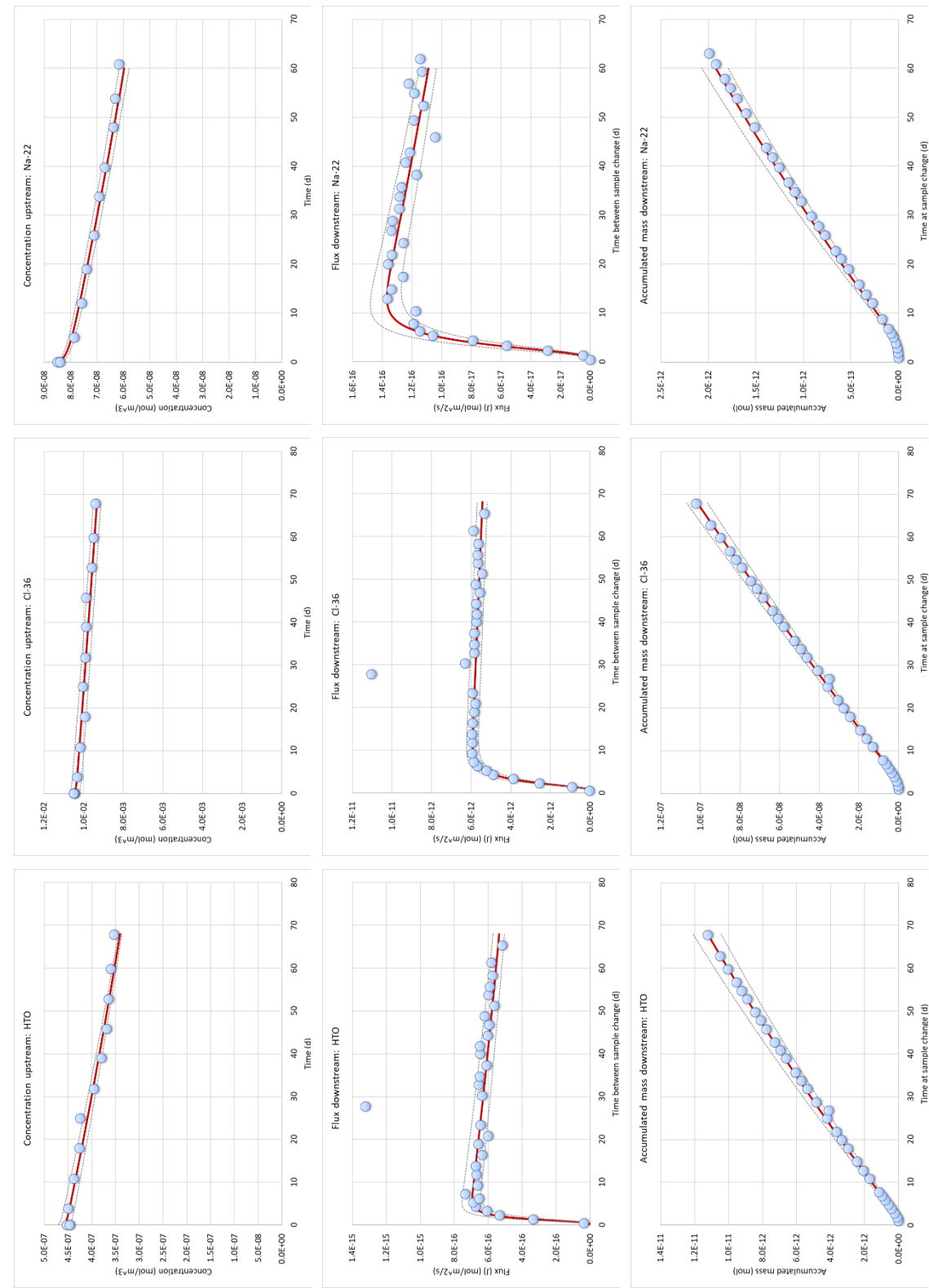
MAR1-1-494.63-DI (S2) – Wildegg Formation – SPW-TRU

Sample diameter 63.57 mm – sample thickness 11.82 mm



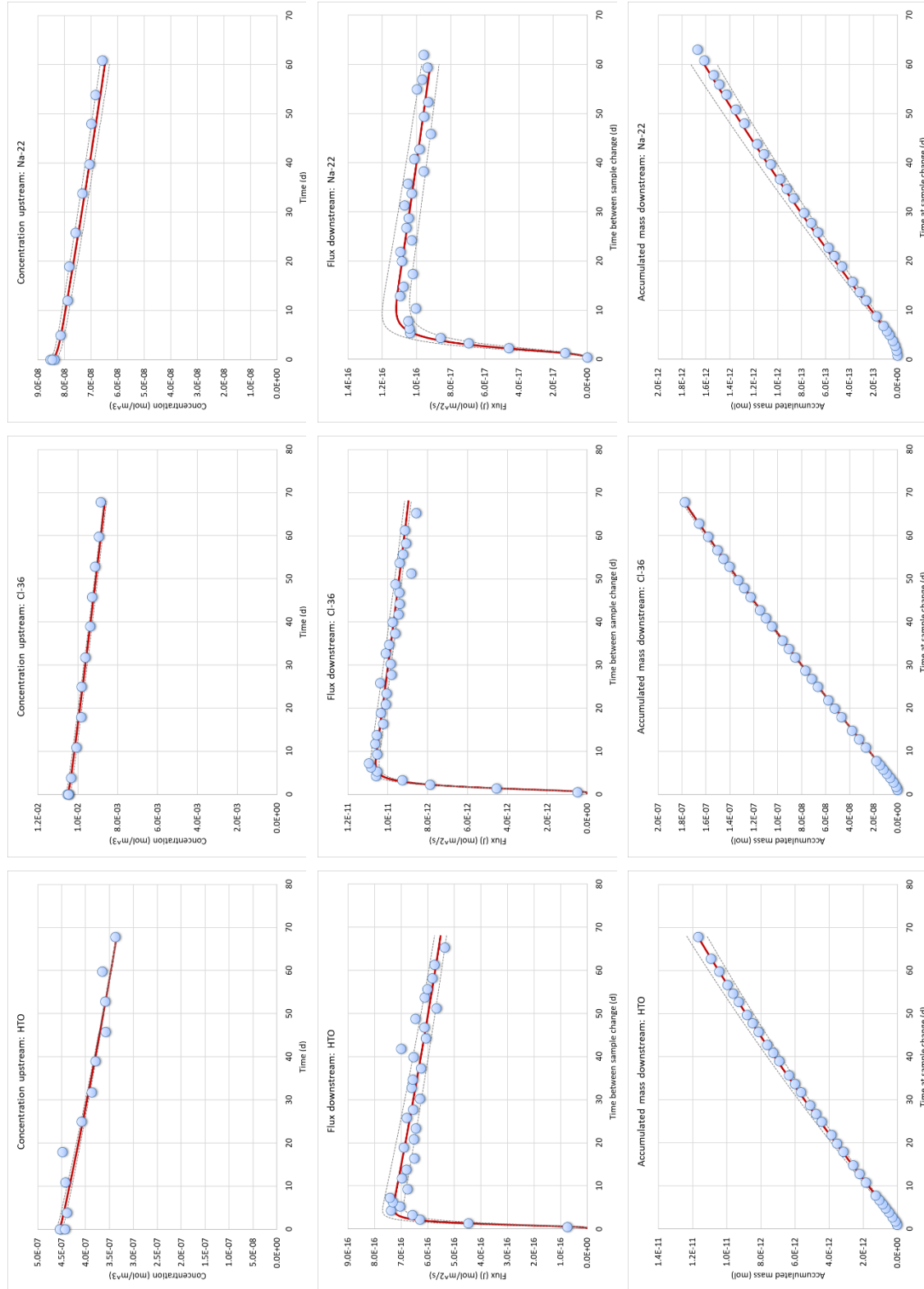
MAR1-1-566.33-DI (S1) – Wedelsandstein Formation – SPW-TRU

Sample diameter 63.54 mm – sample thickness 12.18 mm



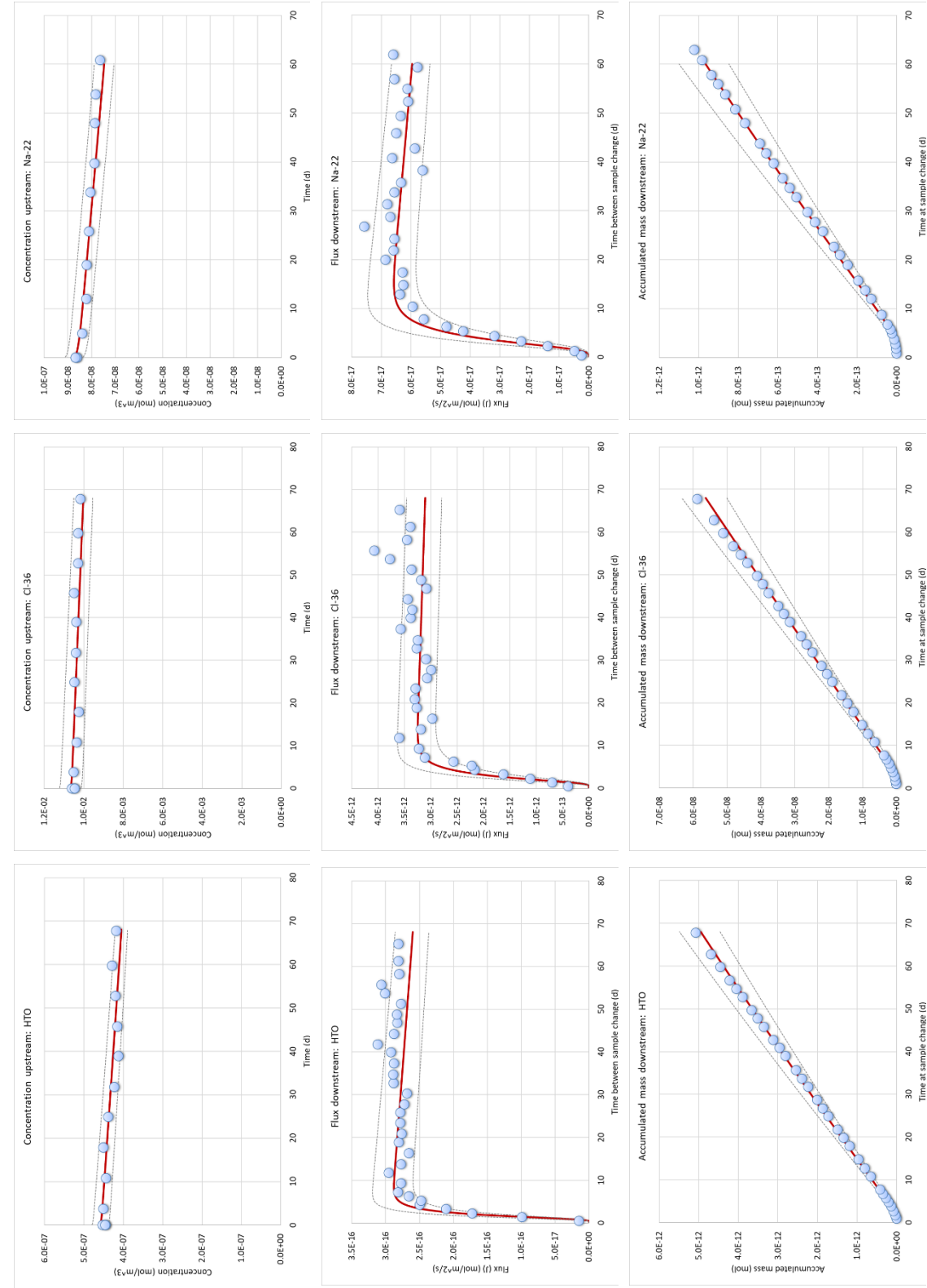
MAR1-1-570.12-DI (S1) – Wedelsandstein Formation – SPW-TRU

Sample diameter 63.70 mm – sample thickness 12.48 mm



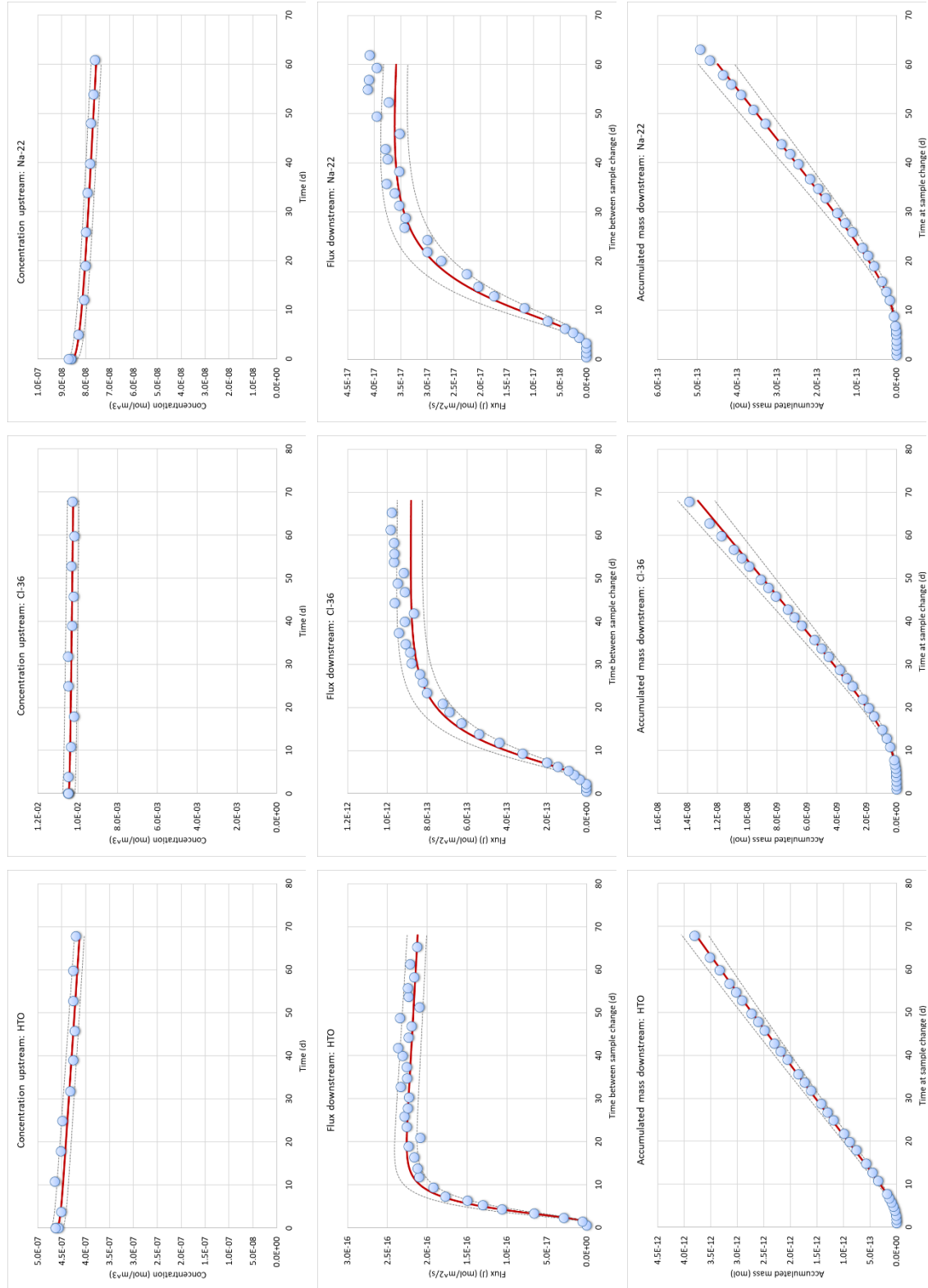
MAR1-1-588.40-DI (S2) – Wedelsandstein Formation – SPW-TRU

Sample diameter 63.54 mm – sample thickness 11.79 mm



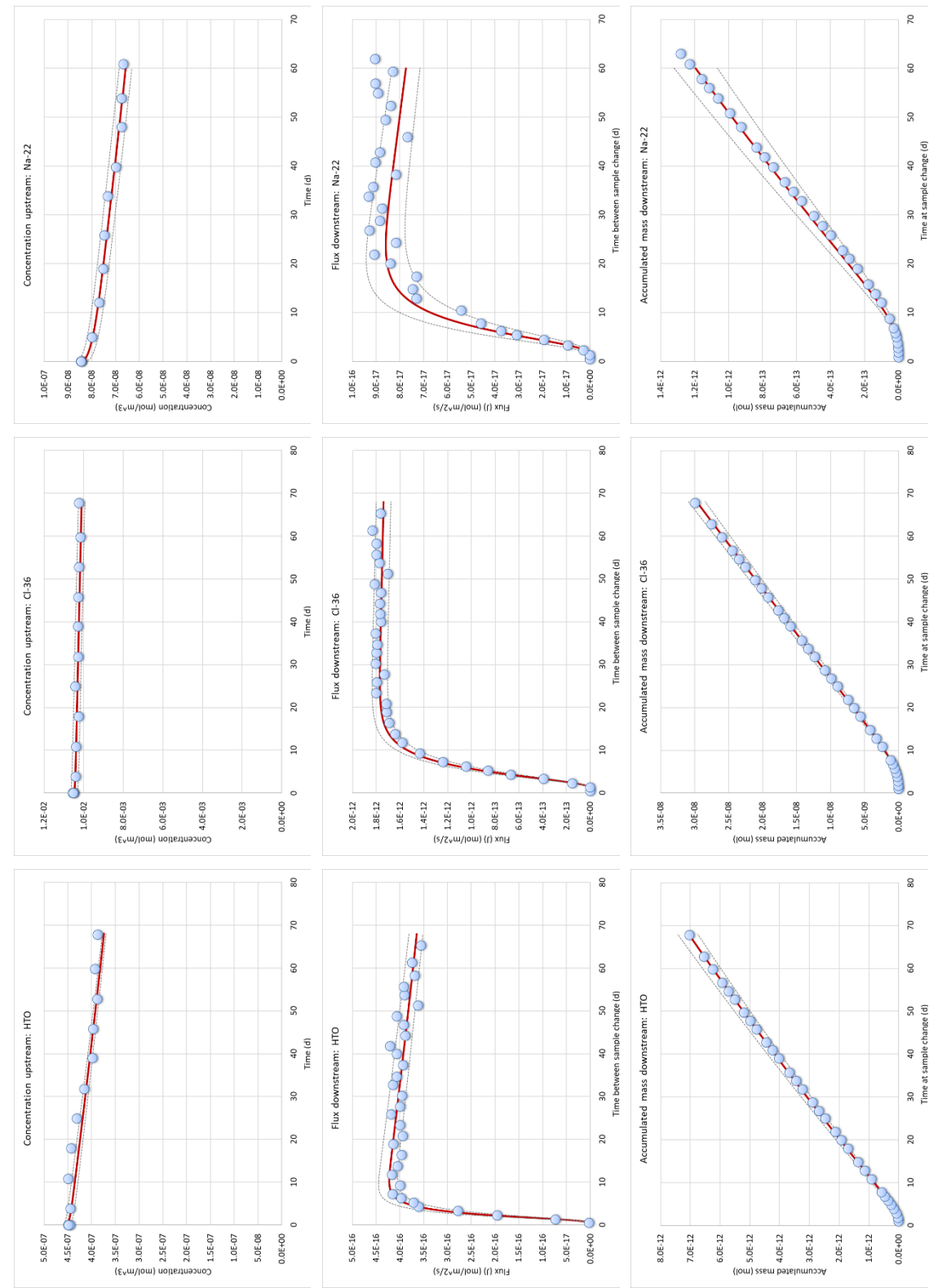
MAR1-1-717.76-DI (S1) – Staffelegg Formation – SPW-TRU

Sample diameter 63.64 mm – sample thickness 12.00 mm



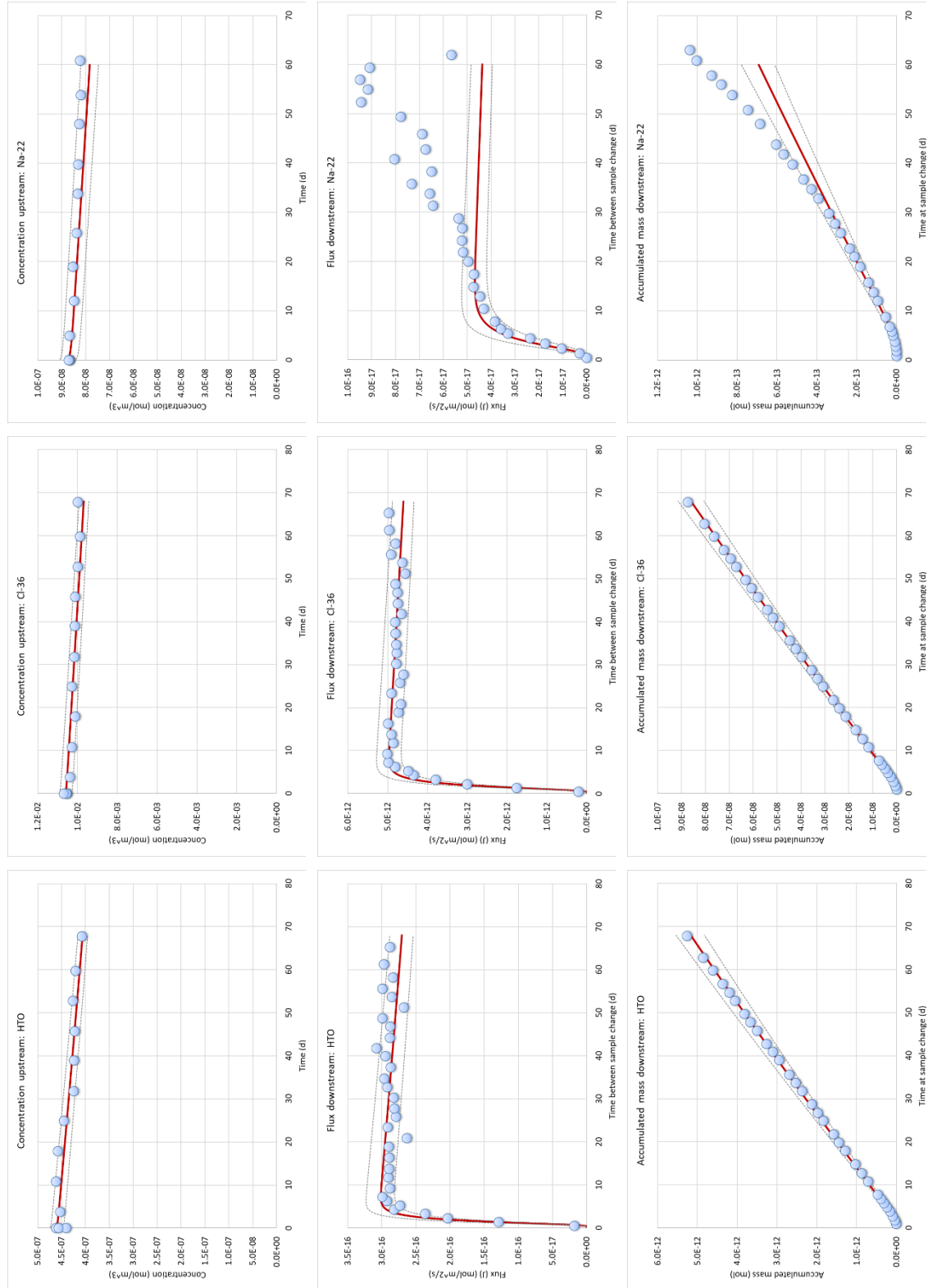
MAR1-1-729.30-DI (S1) – Staffelegg Formation – SPW-TRU

Sample diameter 63.60 mm – sample thickness 11.24 mm



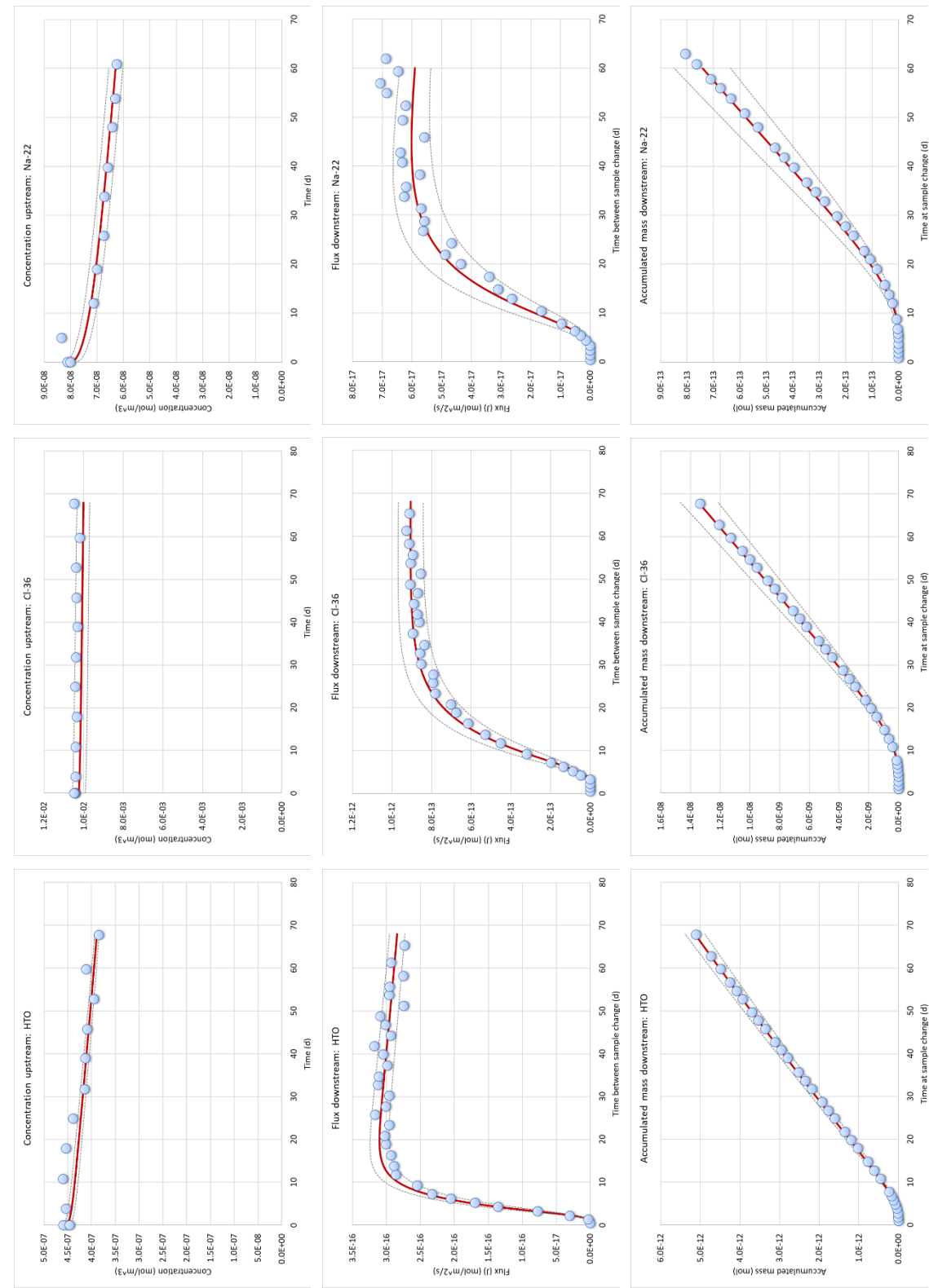
MAR1-1-738.21-DI (S1) – Staffelegg Formation – SPW-TRU

Sample diameter 63.86 mm – sample thickness 12.98 mm



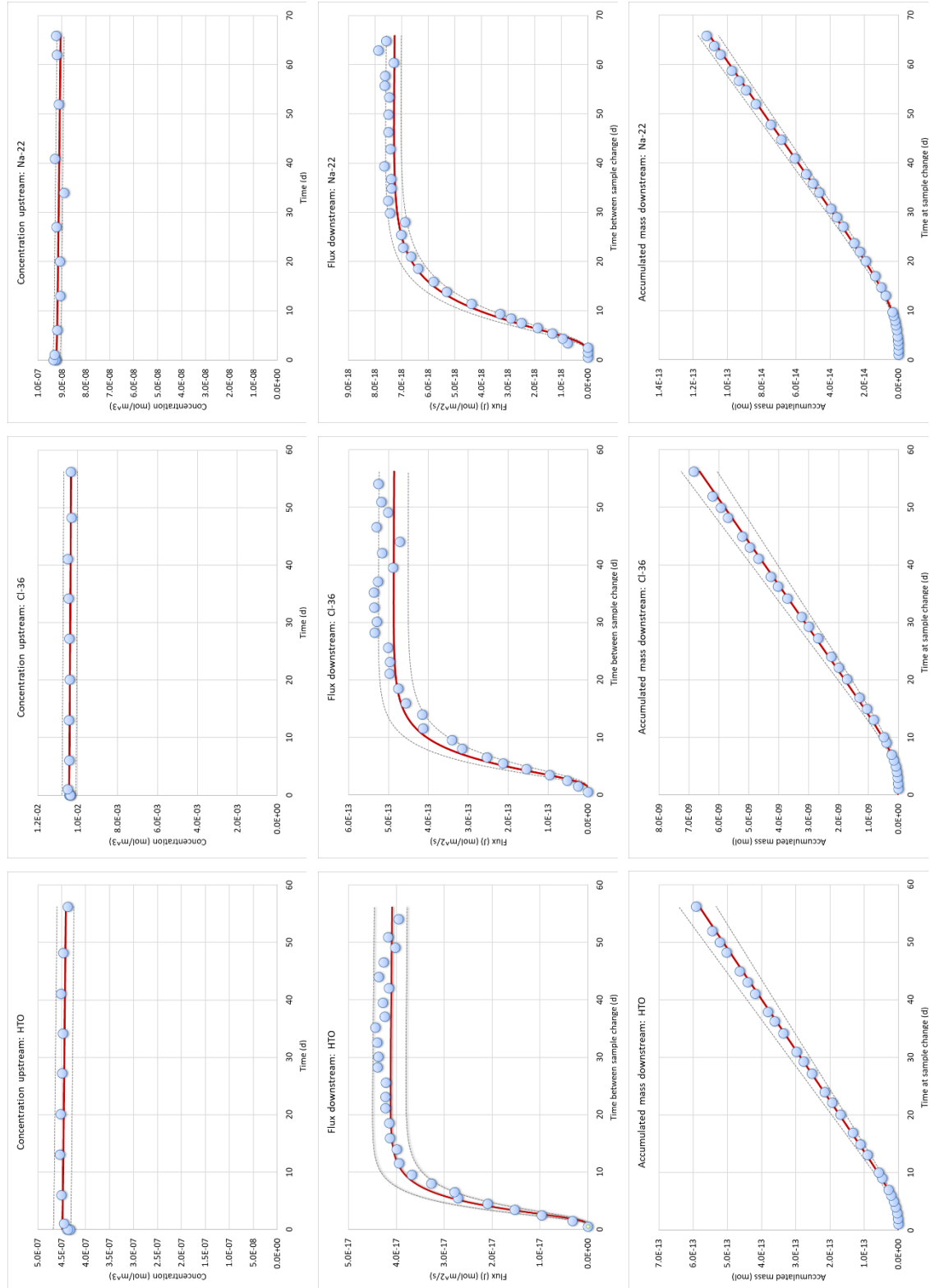
MAR1-1-742.25-DI (S1) – Staffelegg Formation – SPW-TRU

Sample diameter 63.92 mm – sample thickness 13.33 mm



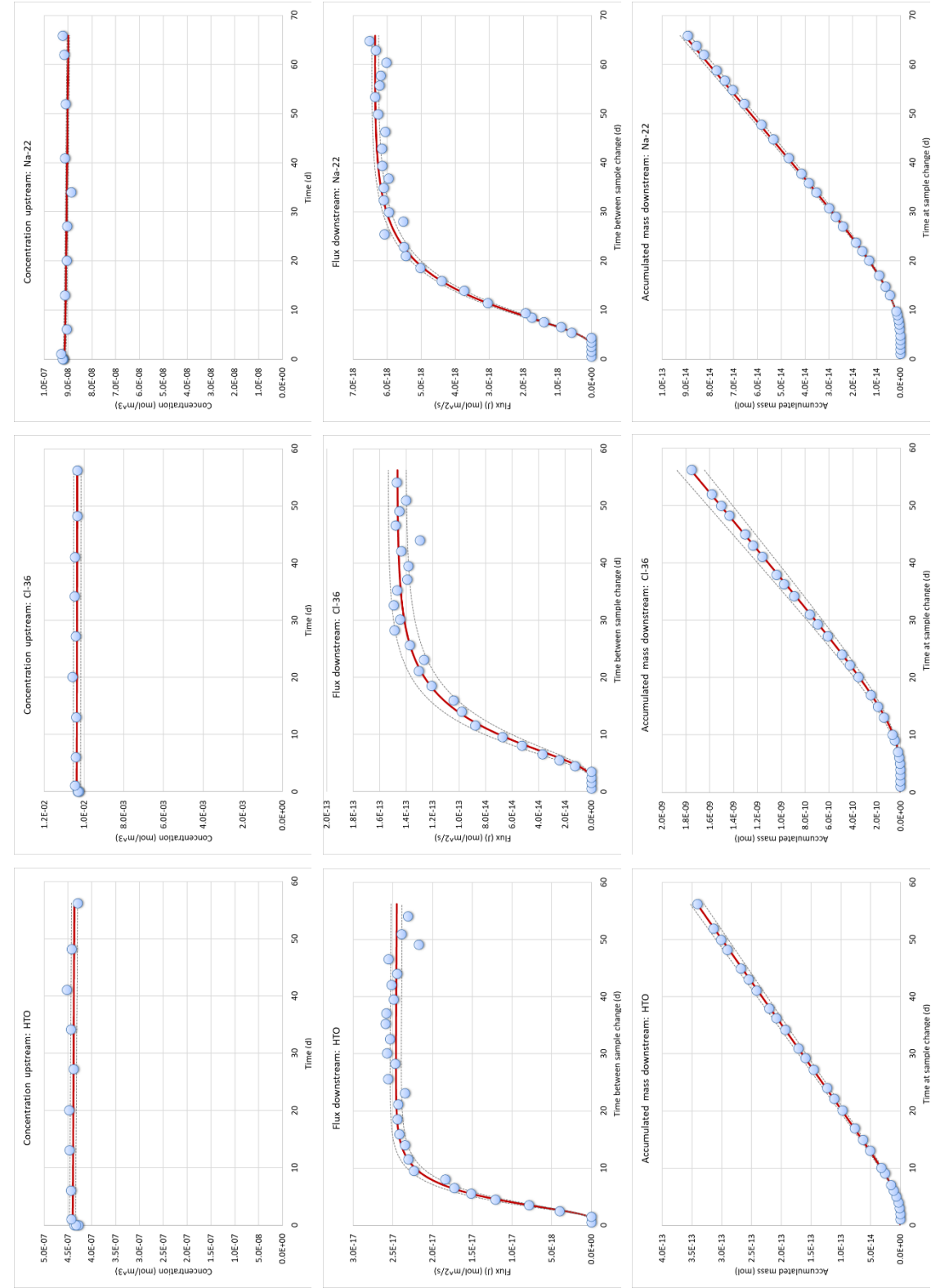
TRU1-1-620.33-DI (S1) – Felsenkalke/Massenkalk – SPW-TRU

Sample diameter 63.72 mm – sample thickness 13.59 mm



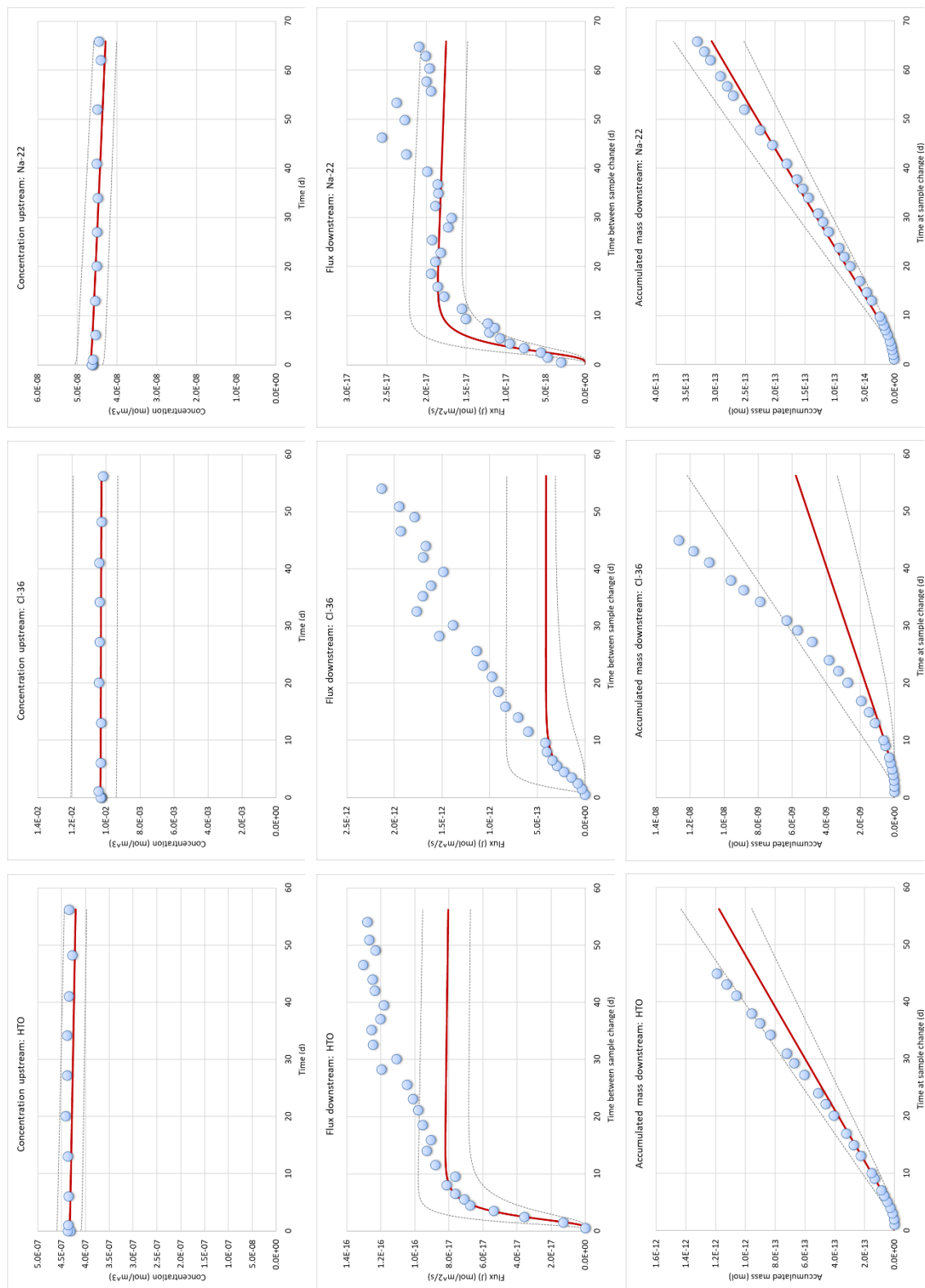
TRU1-1-660.04-DI (S2) – Villigen Formation – SPW-TRU

Sample diameter 63.51 mm – sample thickness 11.71 mm



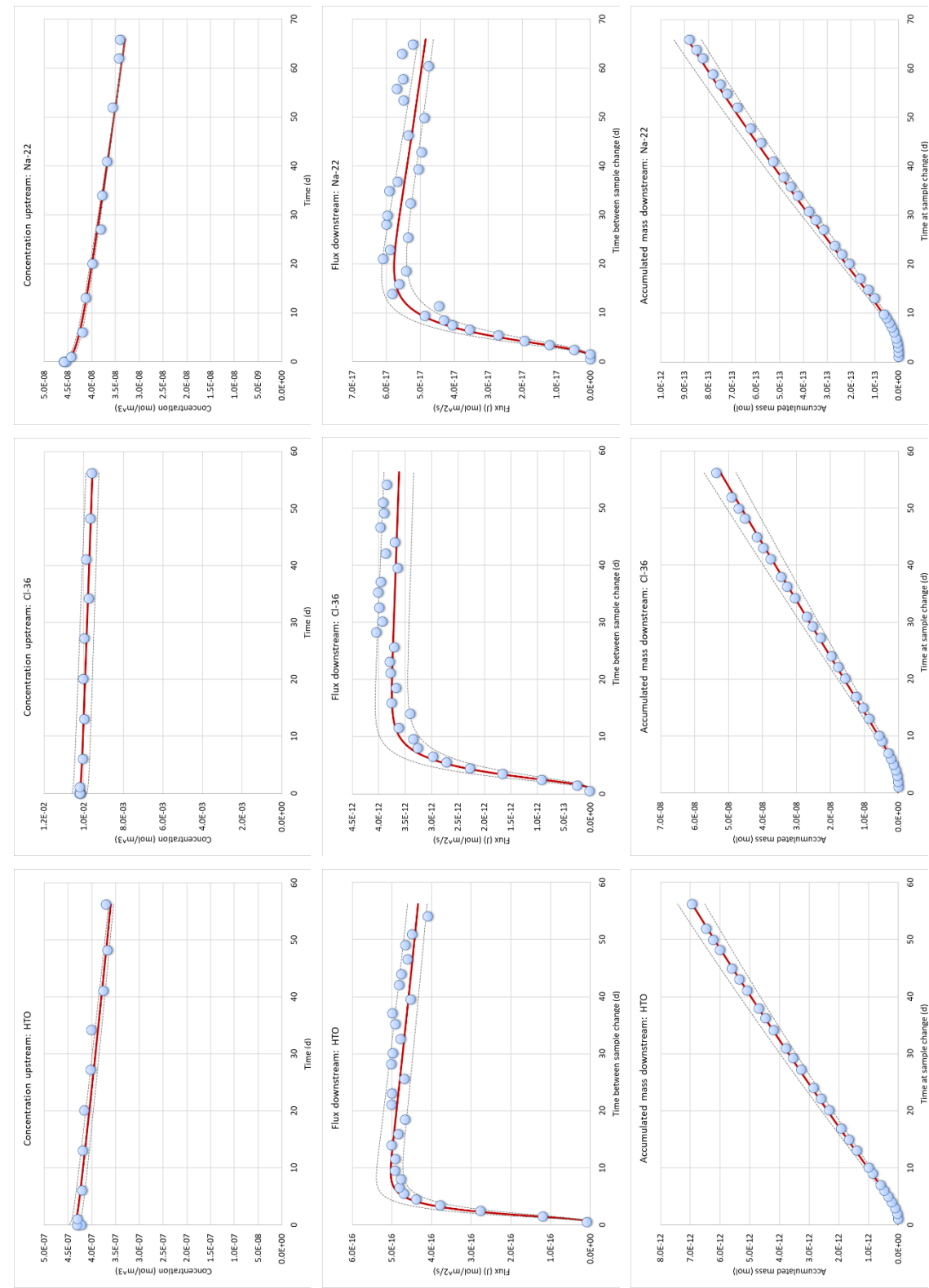
TRU1-1-710.95-DI (S2) – Villigen Formation – SPW-TRU

Sample diameter 63.50 mm – sample thickness 12.52 mm



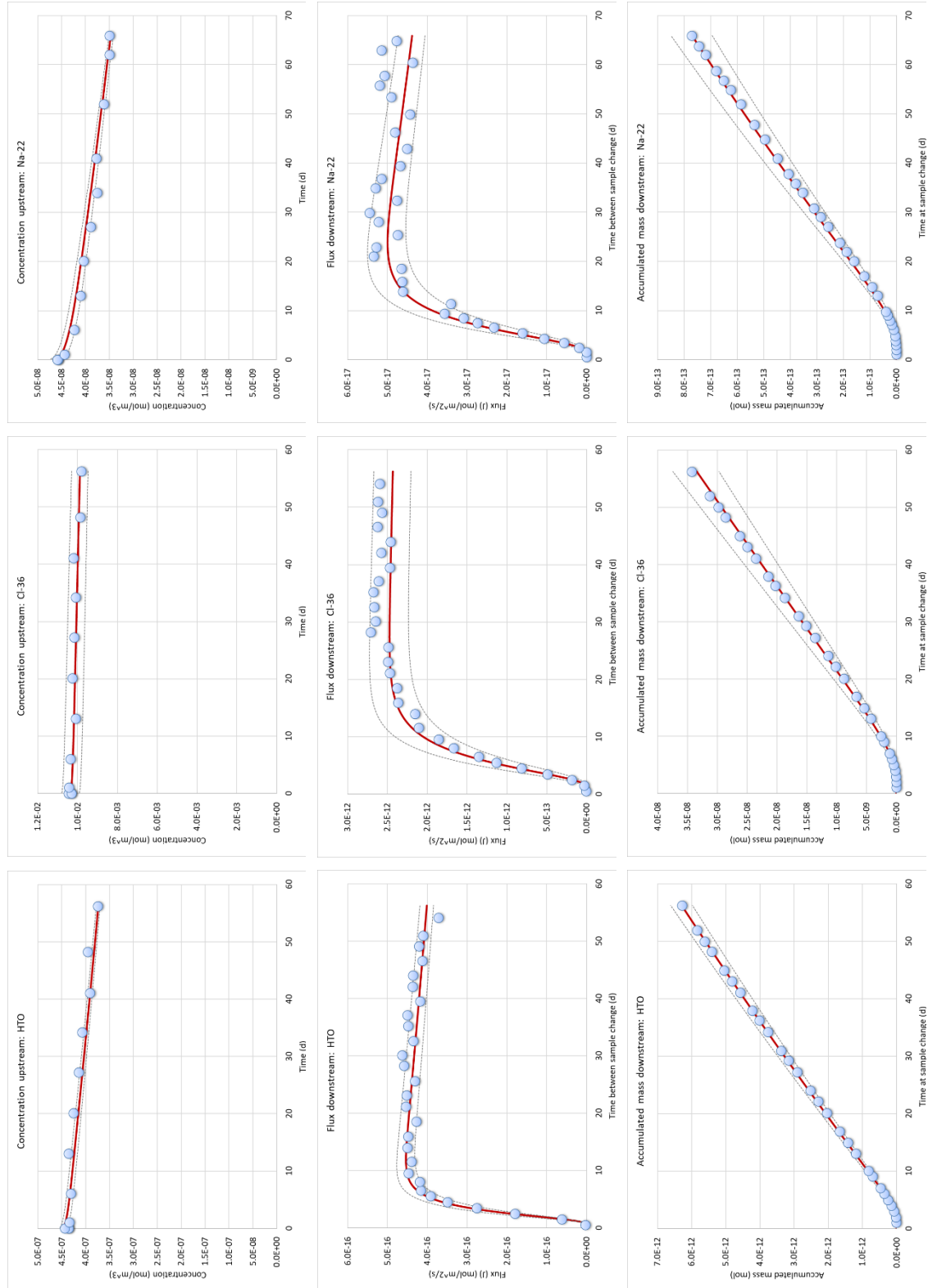
TRU1-1-728.55-DI (S1) – Variansmergel Formation – SPW-TRU

Sample diameter 63.40 mm – sample thickness 13.17 mm



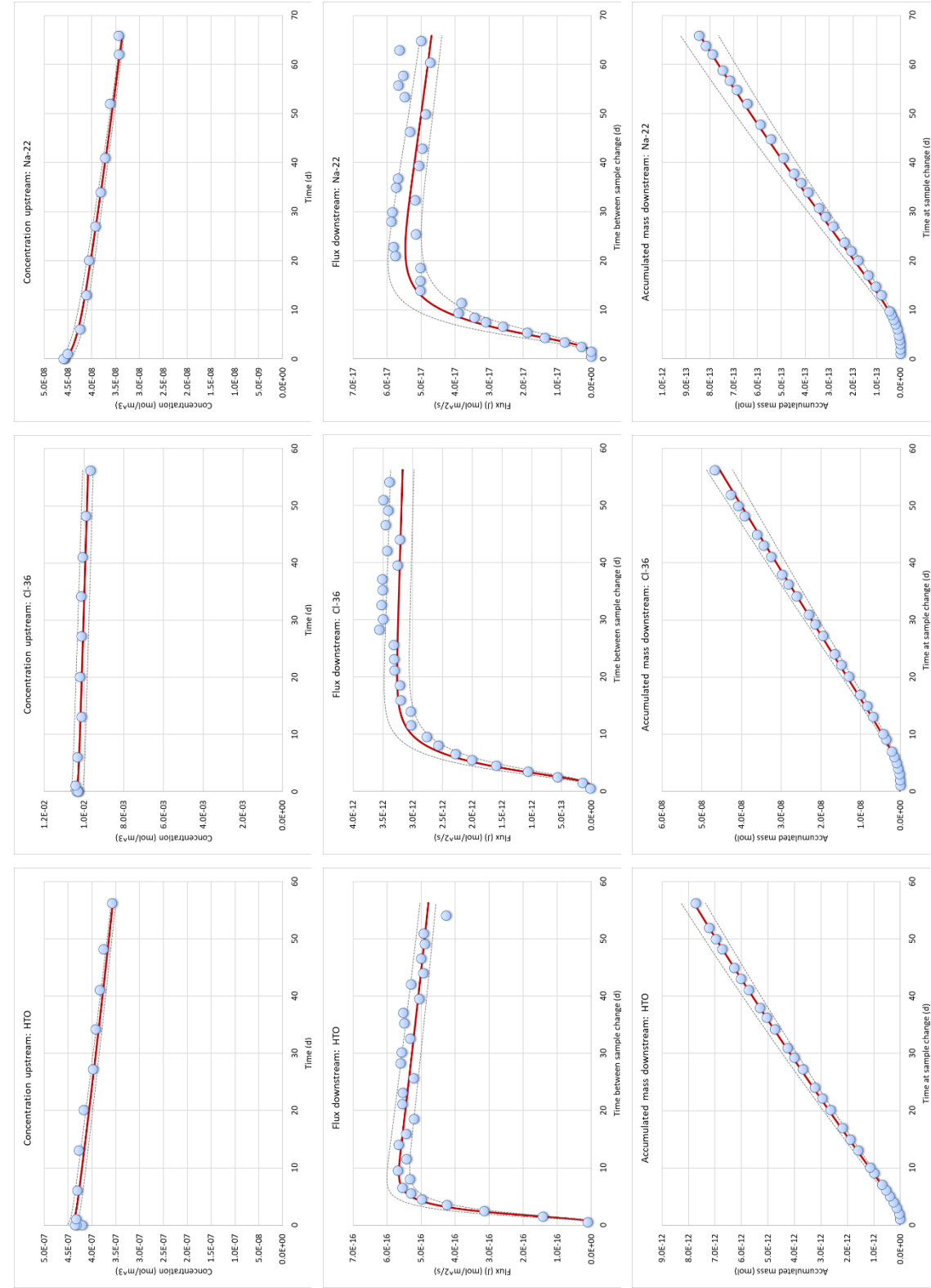
TRU1-1-735.62-DI (S1) – Variansmergel Formation – SPW-TRU

Sample diameter 63.60 mm – sample thickness 14.13 mm



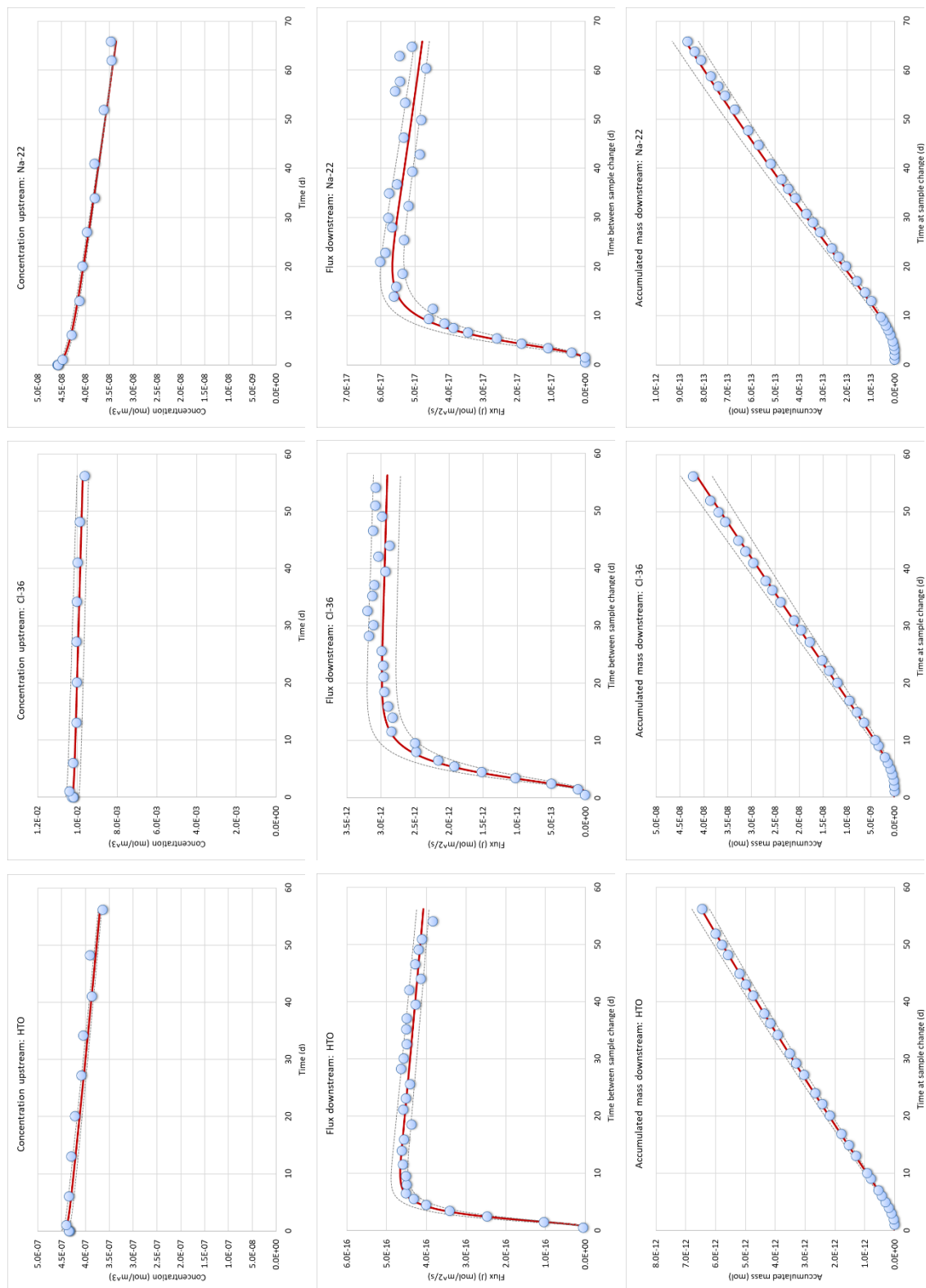
TRU1-1-748.98-DI (S2) – Parkinsoni-Württembergica Schichten – SPW-TRU

Sample diameter 63.62 mm – sample thickness 13.09 mm



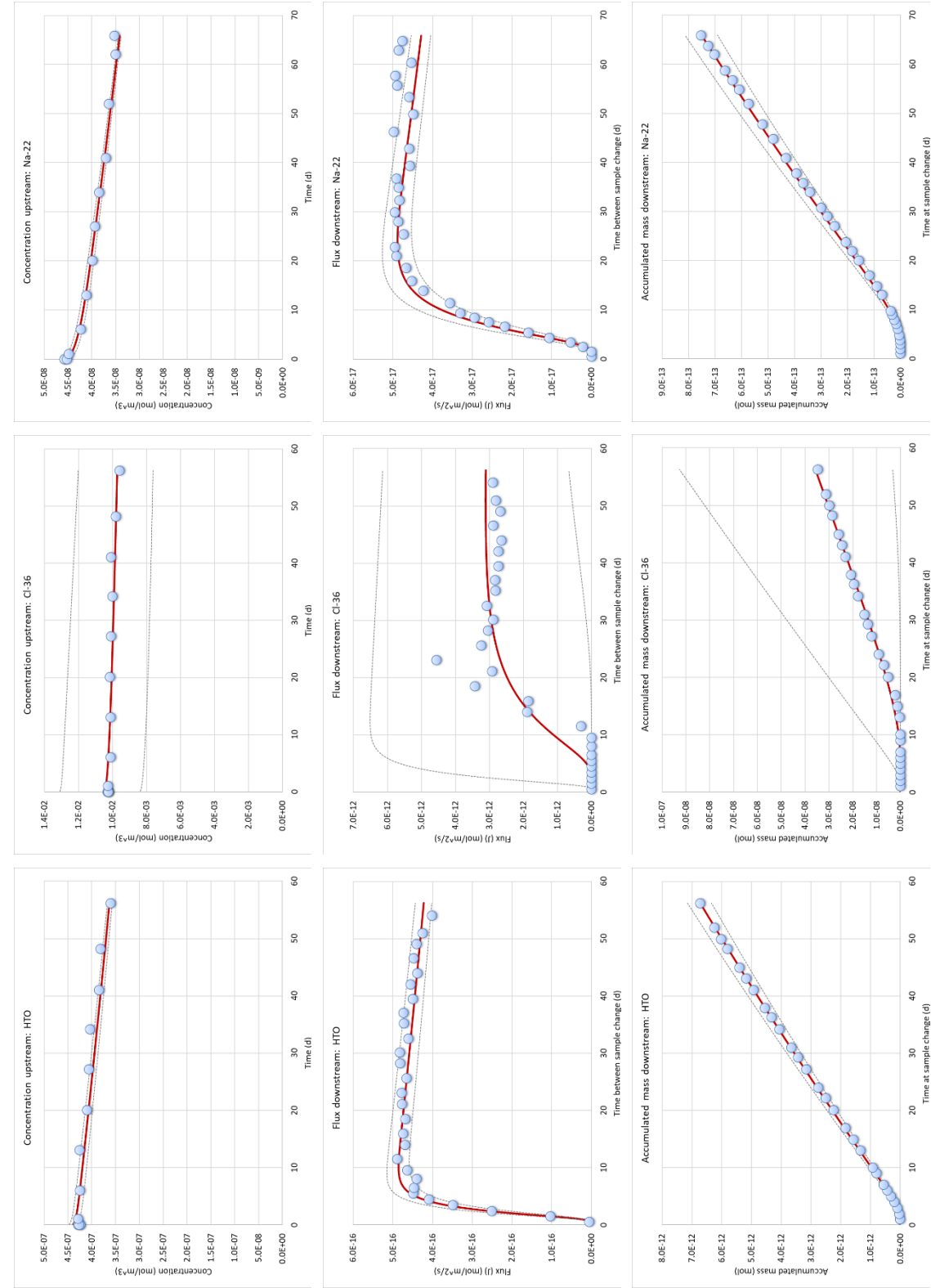
TRU1-1-750.88-DI (S1) – Parkinsoni-Württembergica Schichten – SPW-TRU

Sample diameter 63.57 mm – sample thickness 13.26 mm



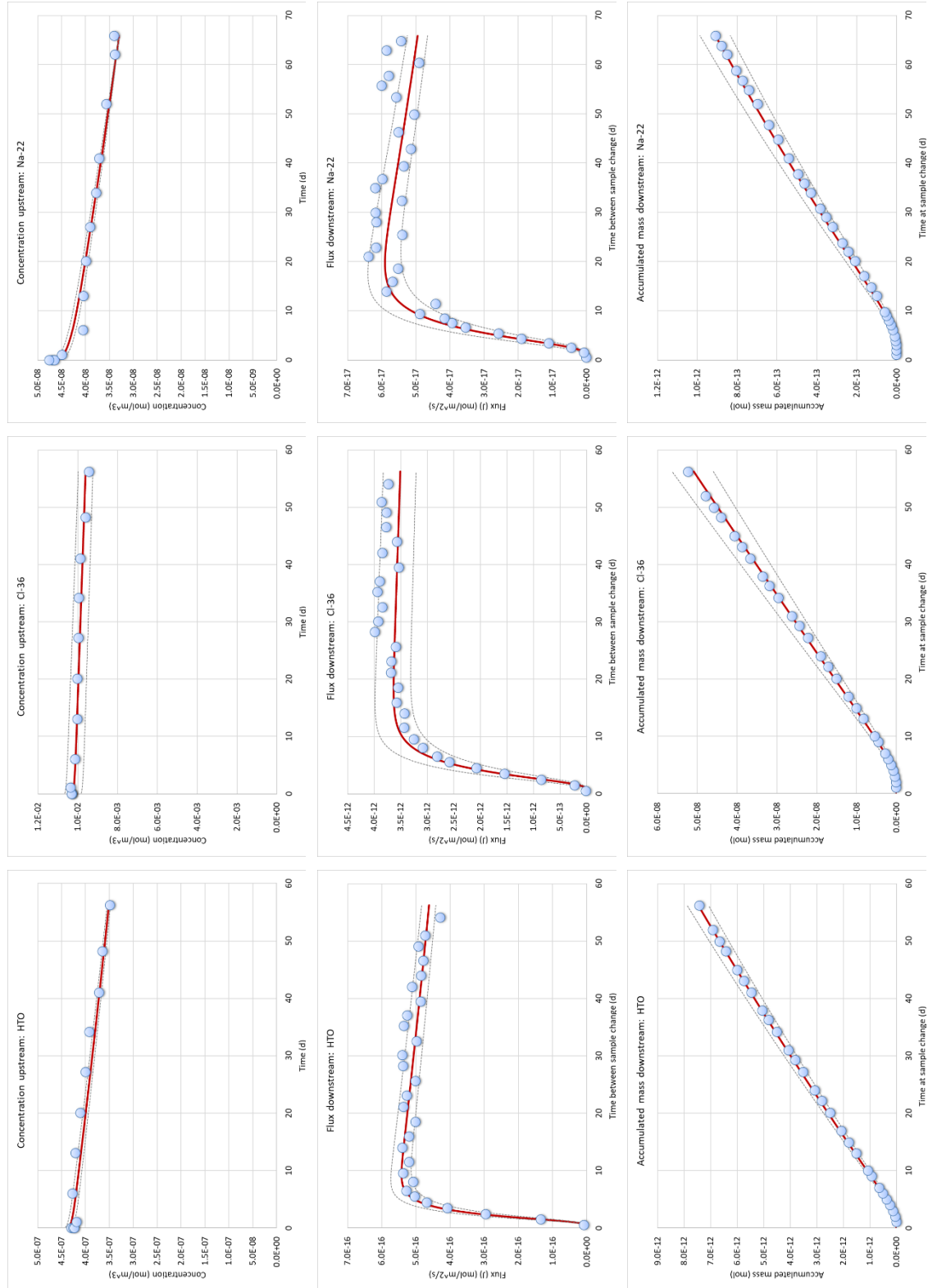
TRU1-1-756.39-DI (S1) – Parkinsoni-Württembergica Schichten – SPW-TRU

Sample diameter 63.55 mm – sample thickness 12.32 mm



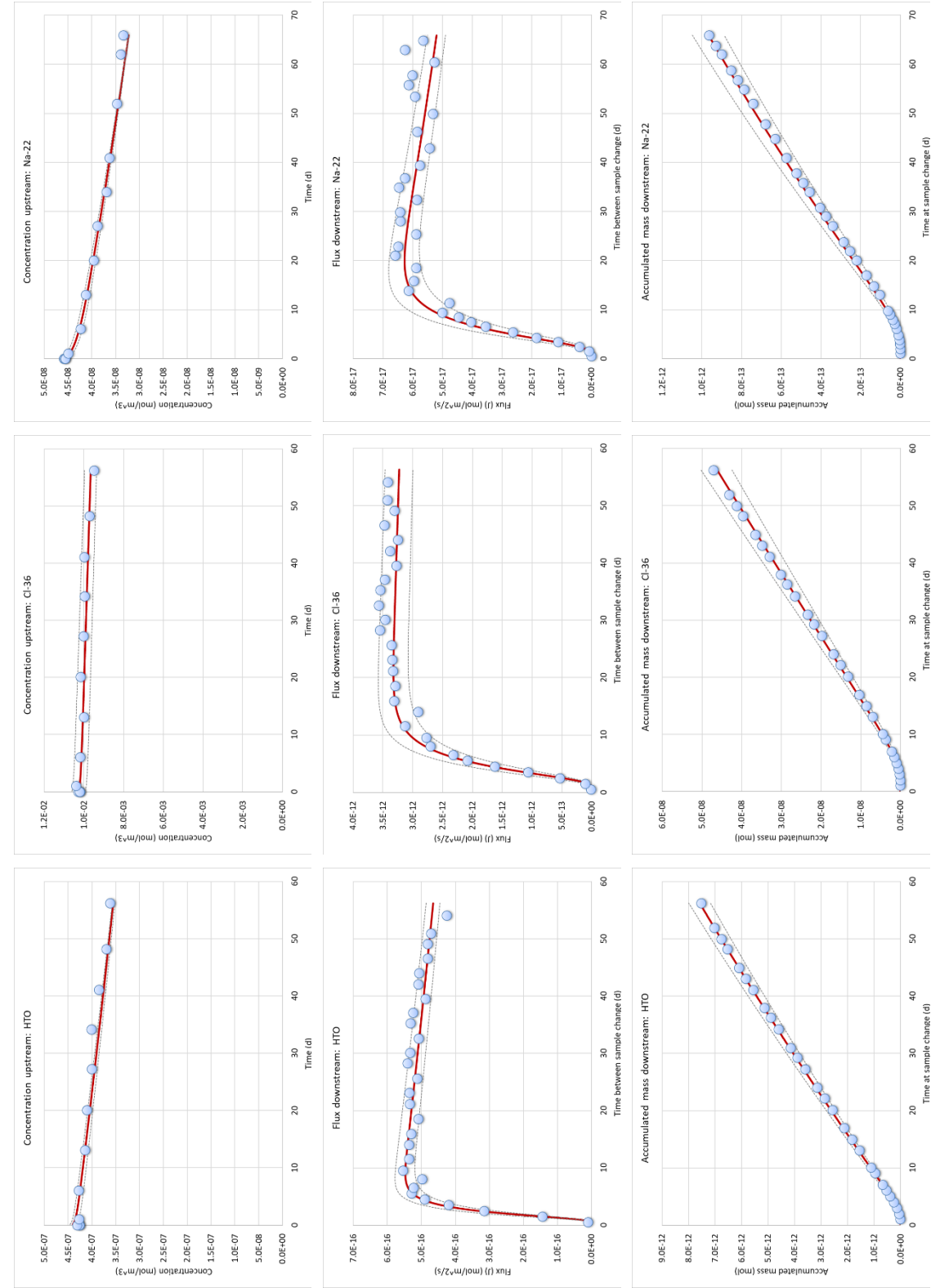
TRU1-1-756.39-DI (S2) – Parkinsoni-Württembergica Schichten – SPW-TRU

Sample diameter 63.55 mm – sample thickness 13.10 mm



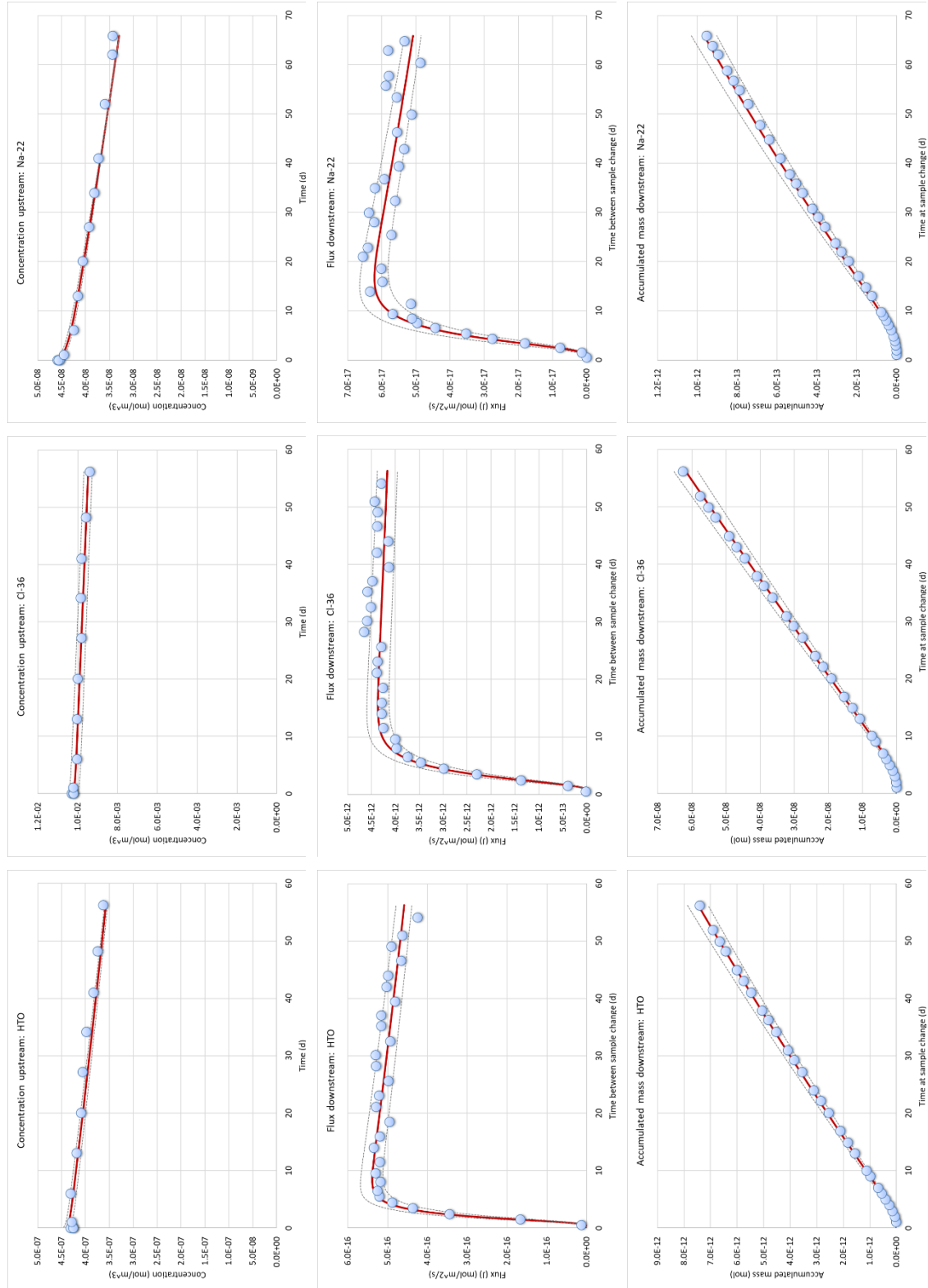
TRU1-1-778.32-DI (S2) – Humphriesoolith Formation – SPW-TRU

Sample diameter 63.70 mm – sample thickness 12.86 mm



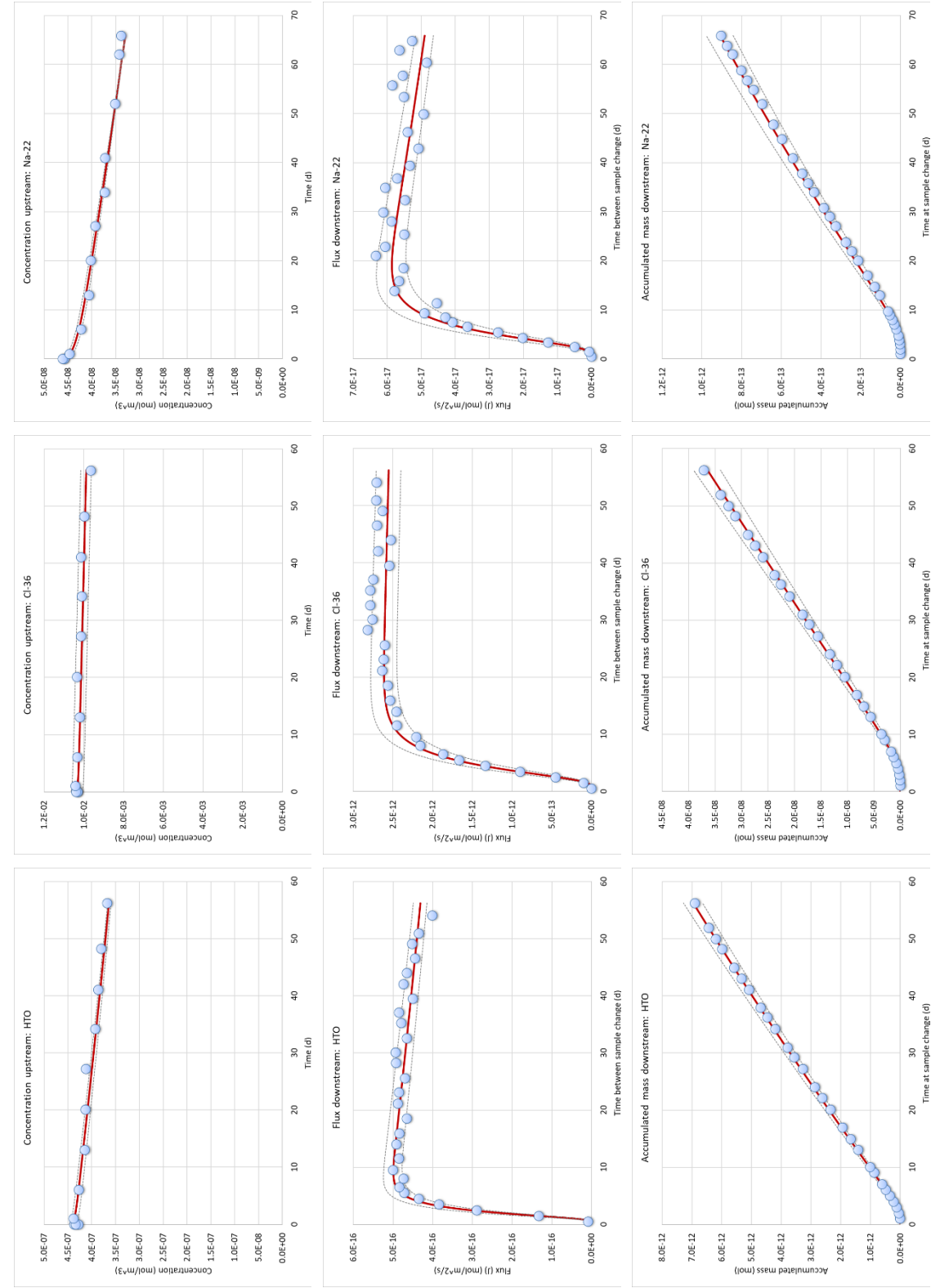
TRU1-1-788.76-DI (S1) – Wedelsandstein Formation – SPW-TRU

Sample diameter 63.63 mm – sample thickness 13.50 mm



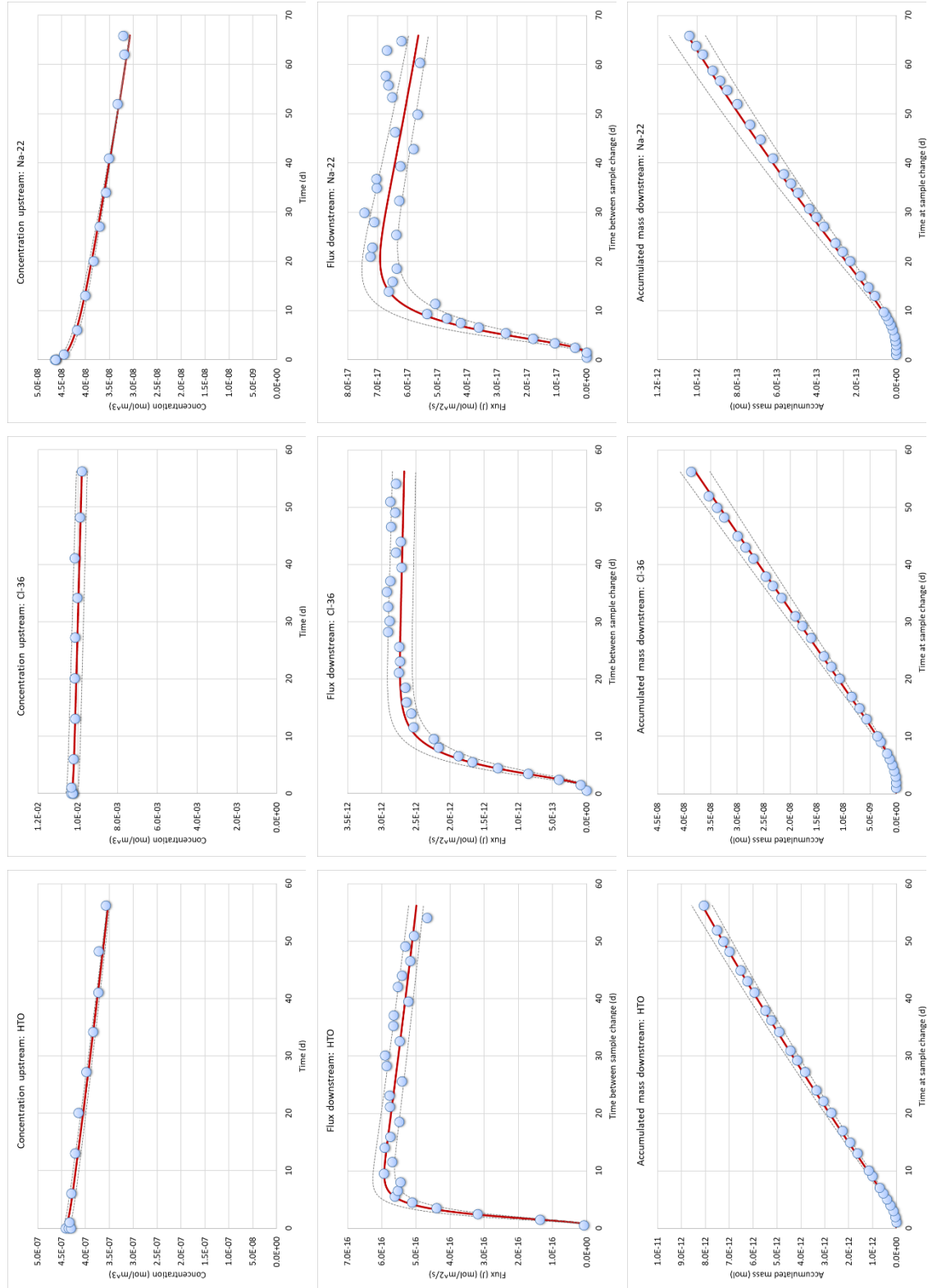
TRU1-1-794.56-DI (S1) – Wedelsandstein Formation – SPW-TRU

Sample diameter 63.65 mm – sample thickness 12.15 mm



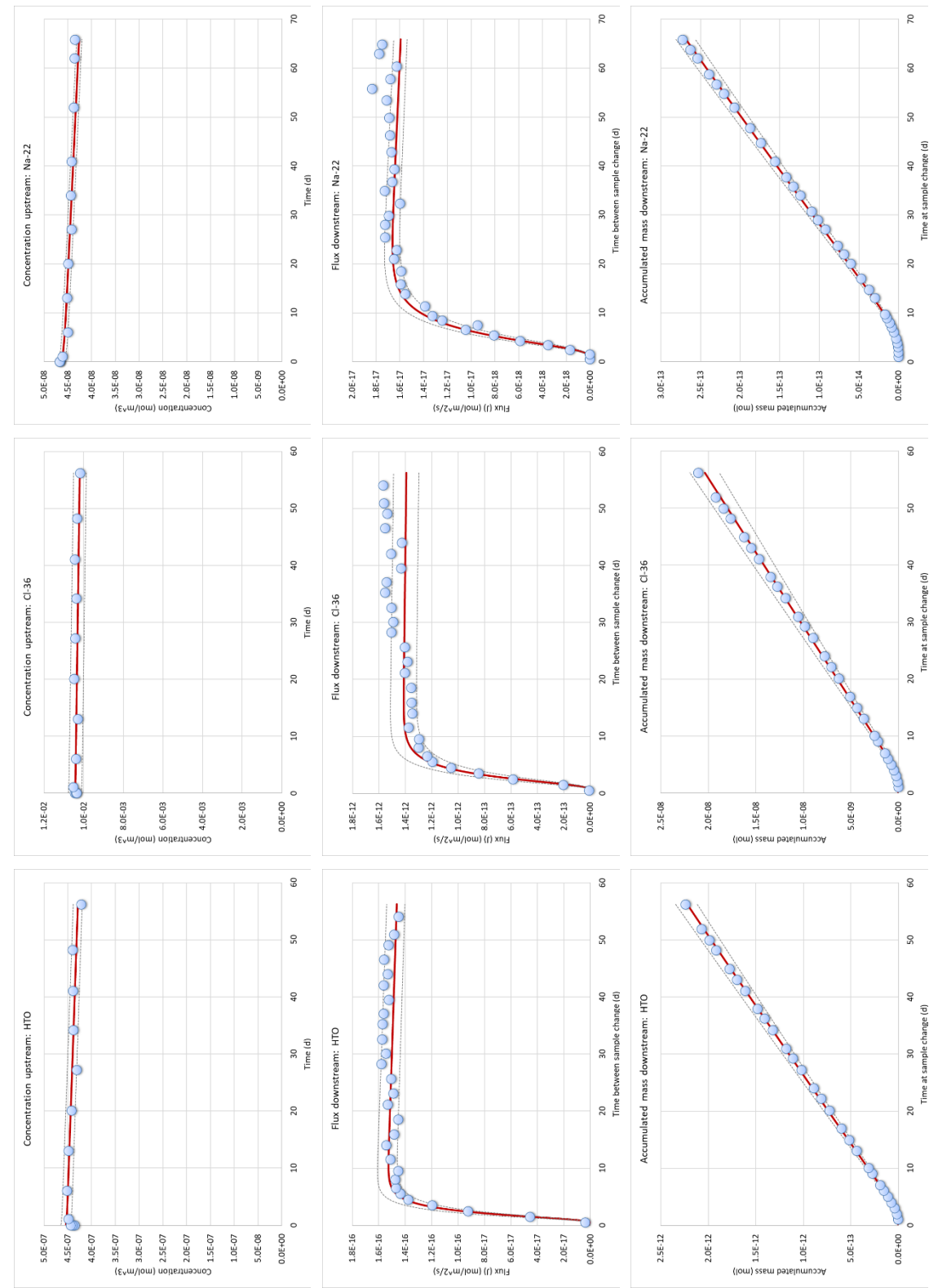
TRU1-1-802.48-DI (S1) – Wedelsandstein Formation – SPW-TRU

Sample diameter 63.60 mm – sample thickness 11.88 mm



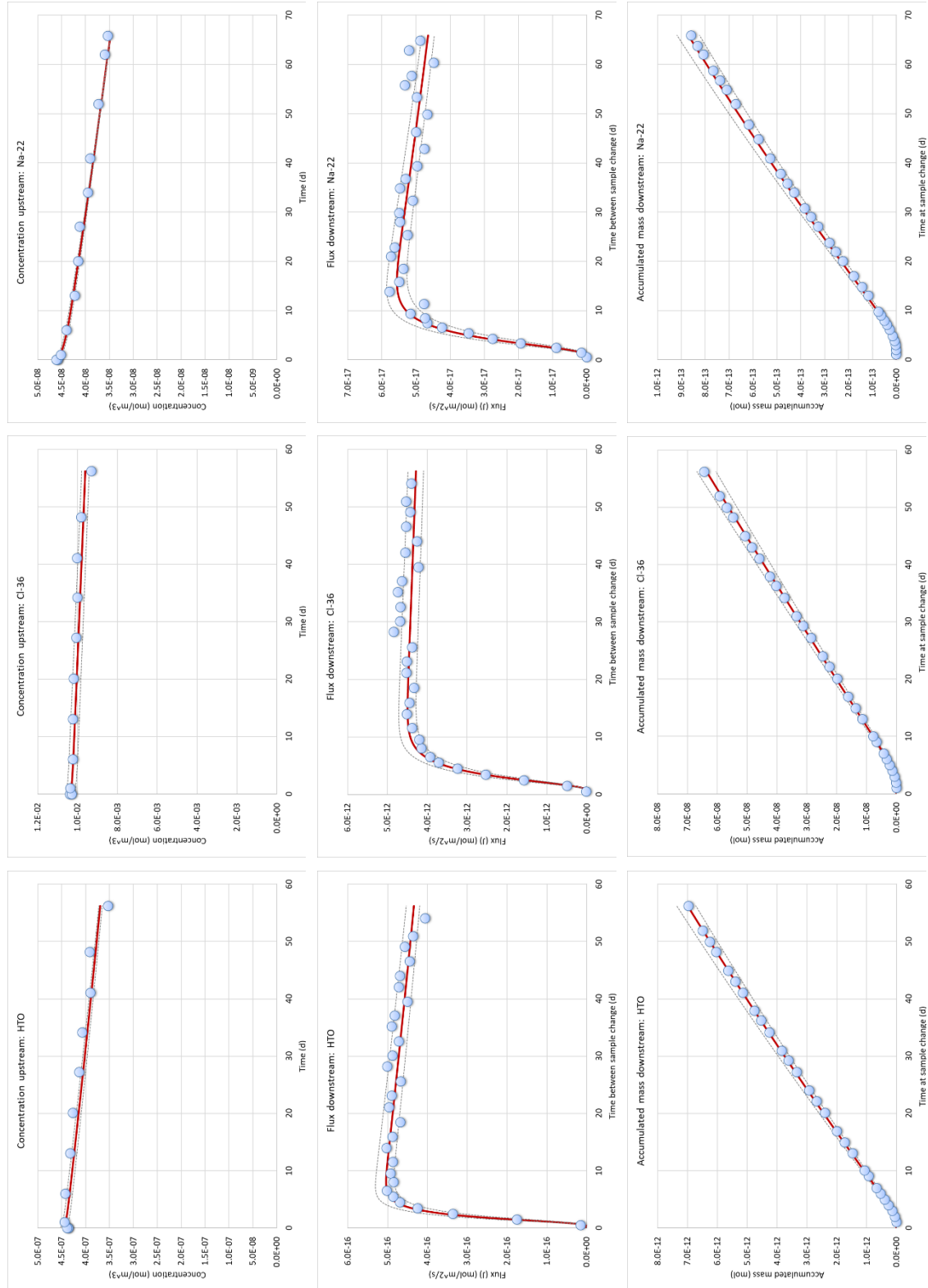
TRU1-1-807.74-DI (S1) – Wedelsandstein Formation – SPW-TRU

Sample diameter 63.60 mm – sample thickness 13.45 mm



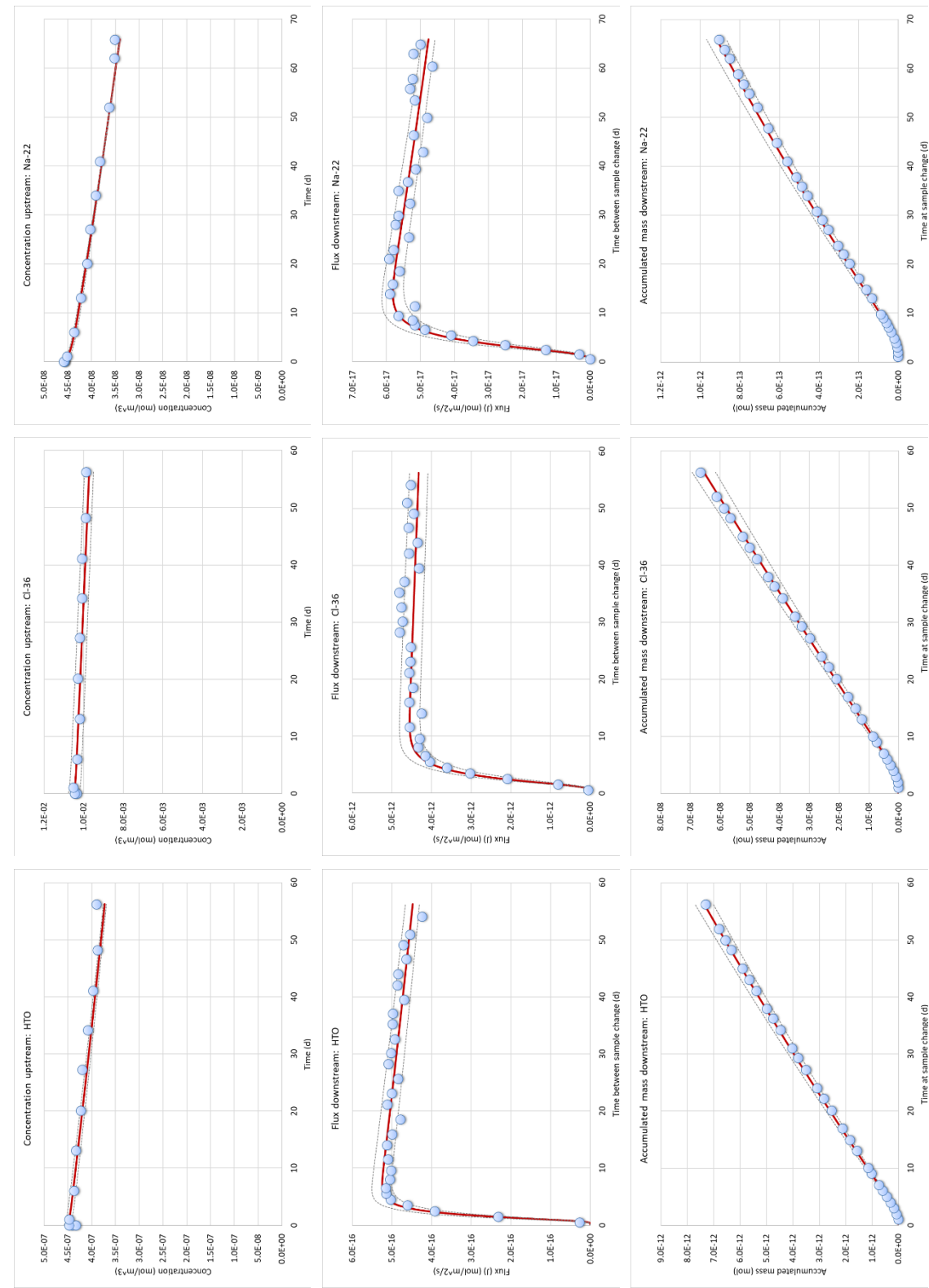
TRU1-1-814.82-OD (S1) – Wedelsandstein Formation – SPW-TRU

Sample diameter 63.51 mm – sample thickness 13.16 mm



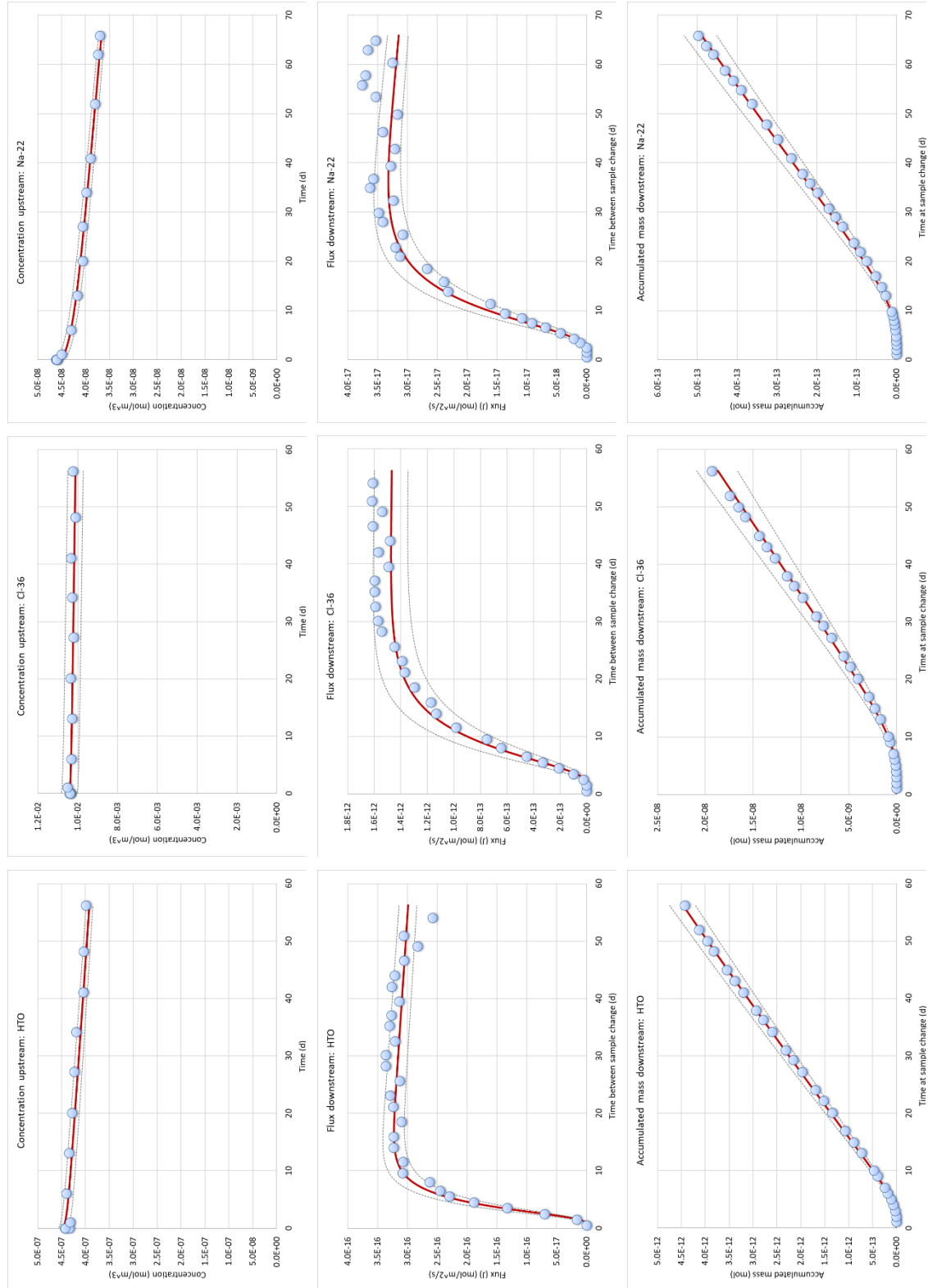
TRU1-1-815.28-DI (S2) – Wedelsandstein Formation – SPW-TRU

Sample diameter 63.75 mm – sample thickness 12.26 mm



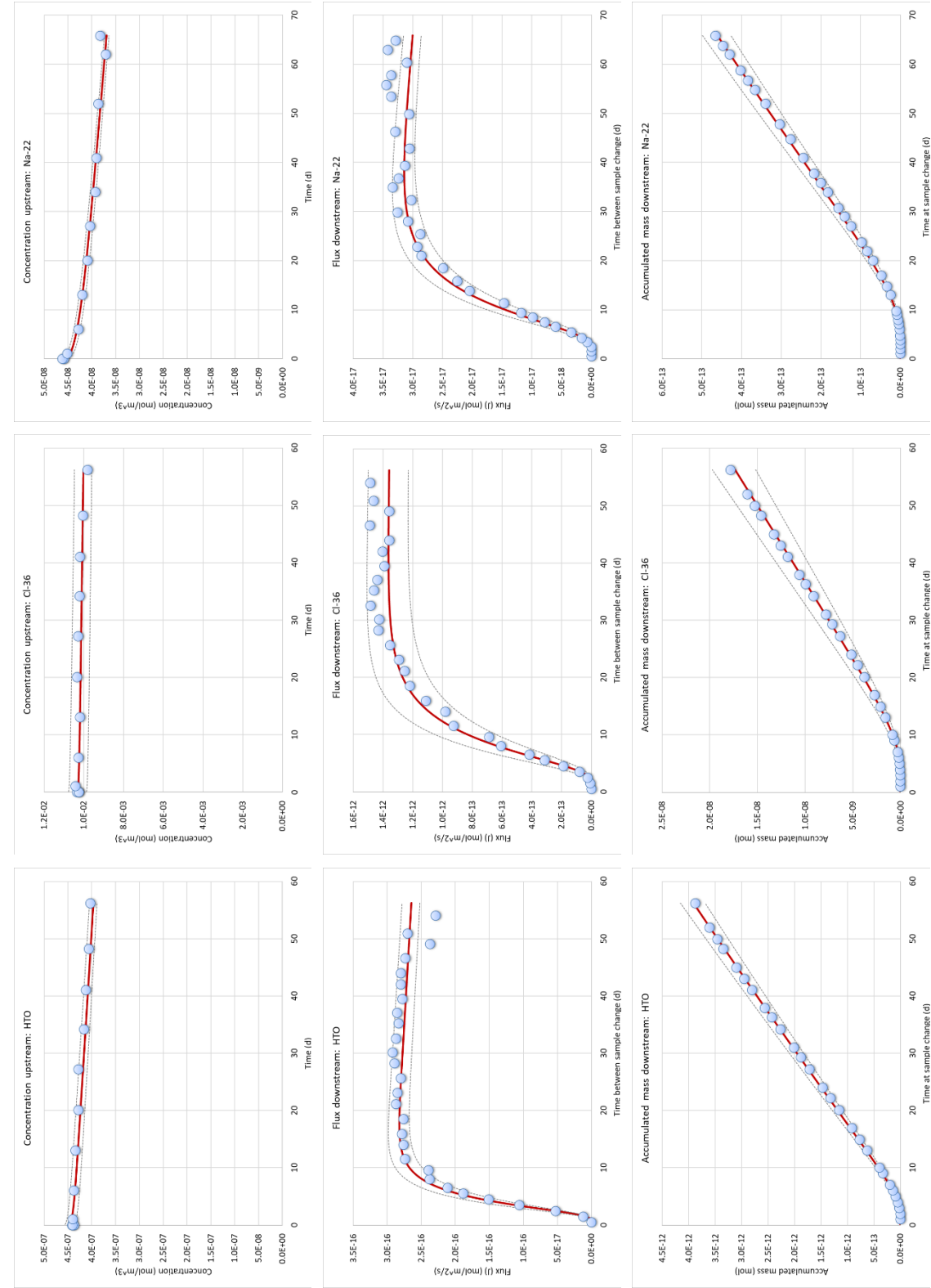
TRU1-1-831.37-DI (S1) – Opalinus Clay – SPW-TRU

Sample diameter 63.60 mm – sample thickness 12.45 mm



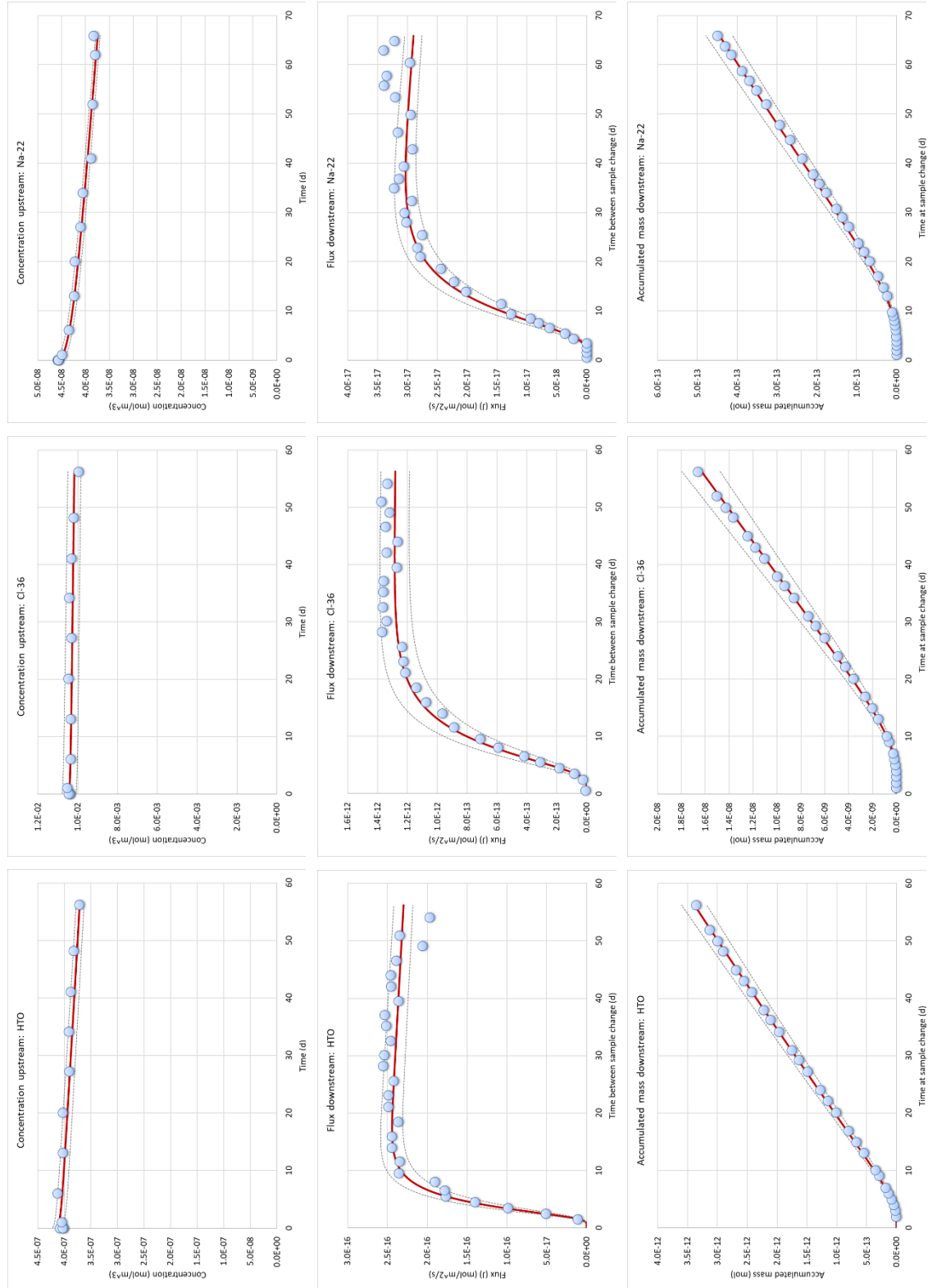
TRU1-1-839.97-DI S1 – Opalinus Clay – SPW-TRU

Sample diameter 63.67 mm – sample thickness 13.40 mm



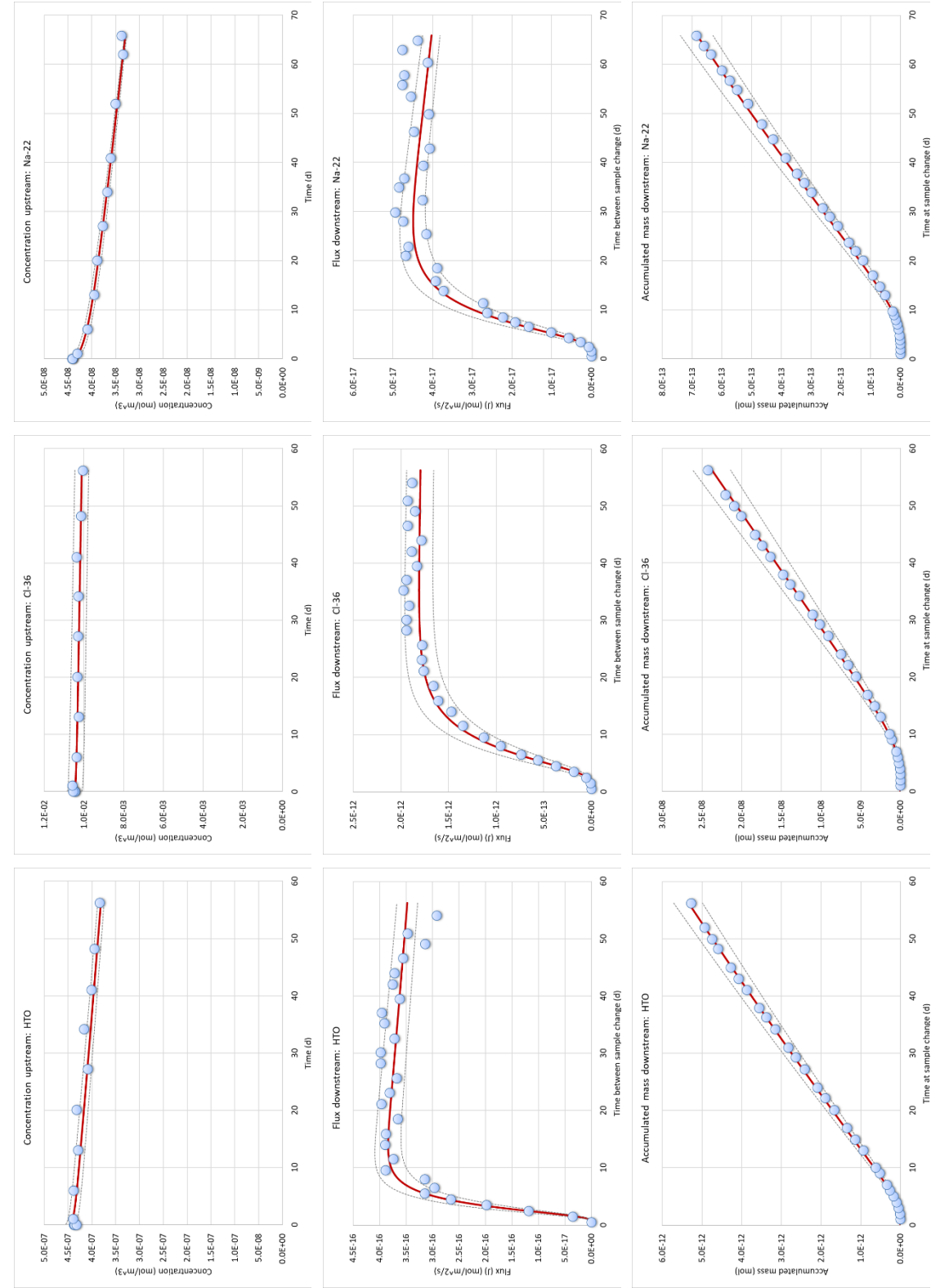
TRU1-1-870.23-DI (S1) – Opalinus Clay – SPW-TRU

Sample diameter 63.50 mm – sample thickness 12.23 mm



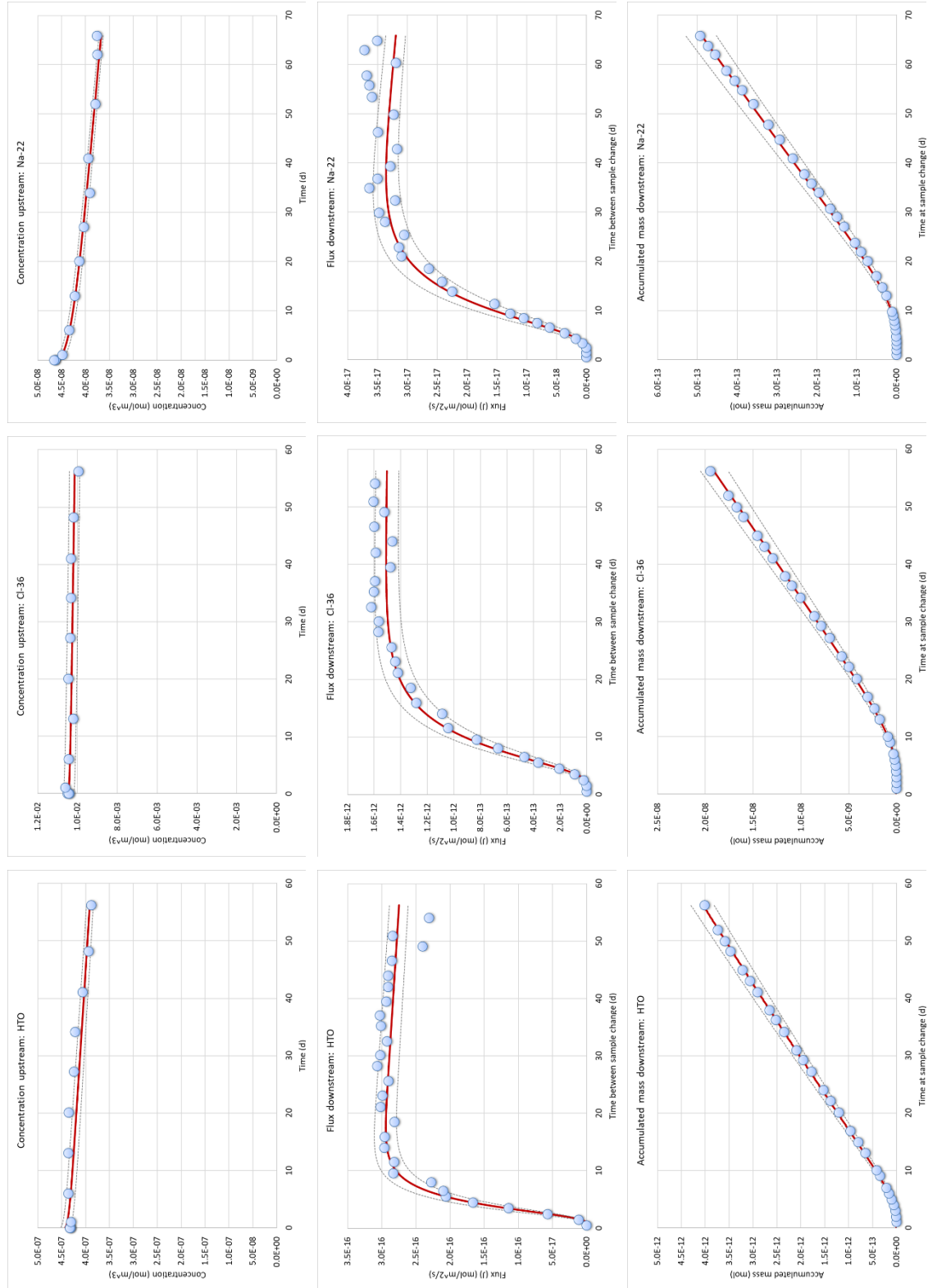
TRU1-1-882.36-DI (S1) – Opalinus Clay – SPW-TRU

Sample diameter 63.72 mm – sample thickness 13.33 mm



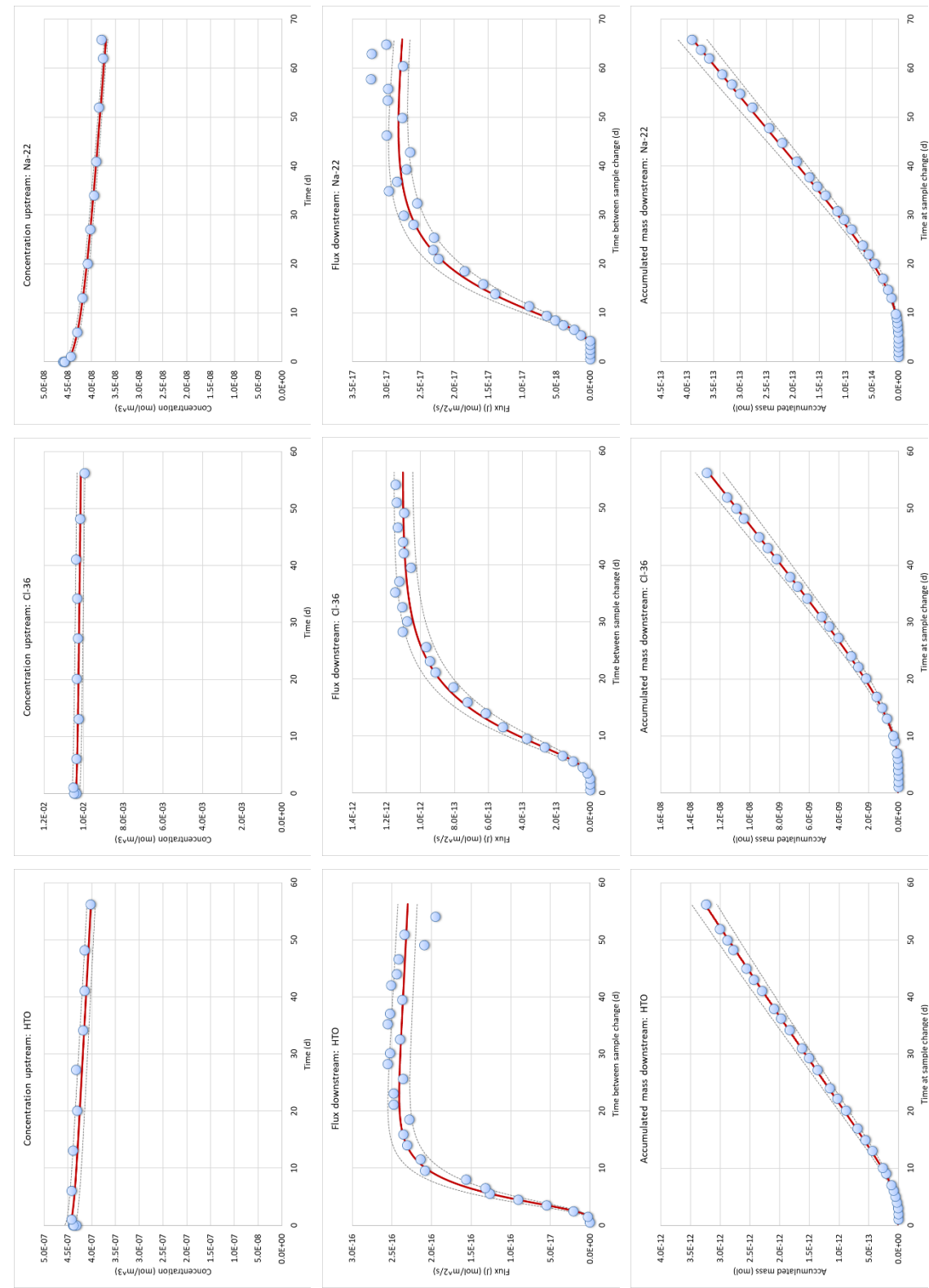
TRU1-1-890.39-DI (S1) – Opalinus Clay – SPW-TRU

Sample diameter 63.50 mm – sample thickness 13.36 mm



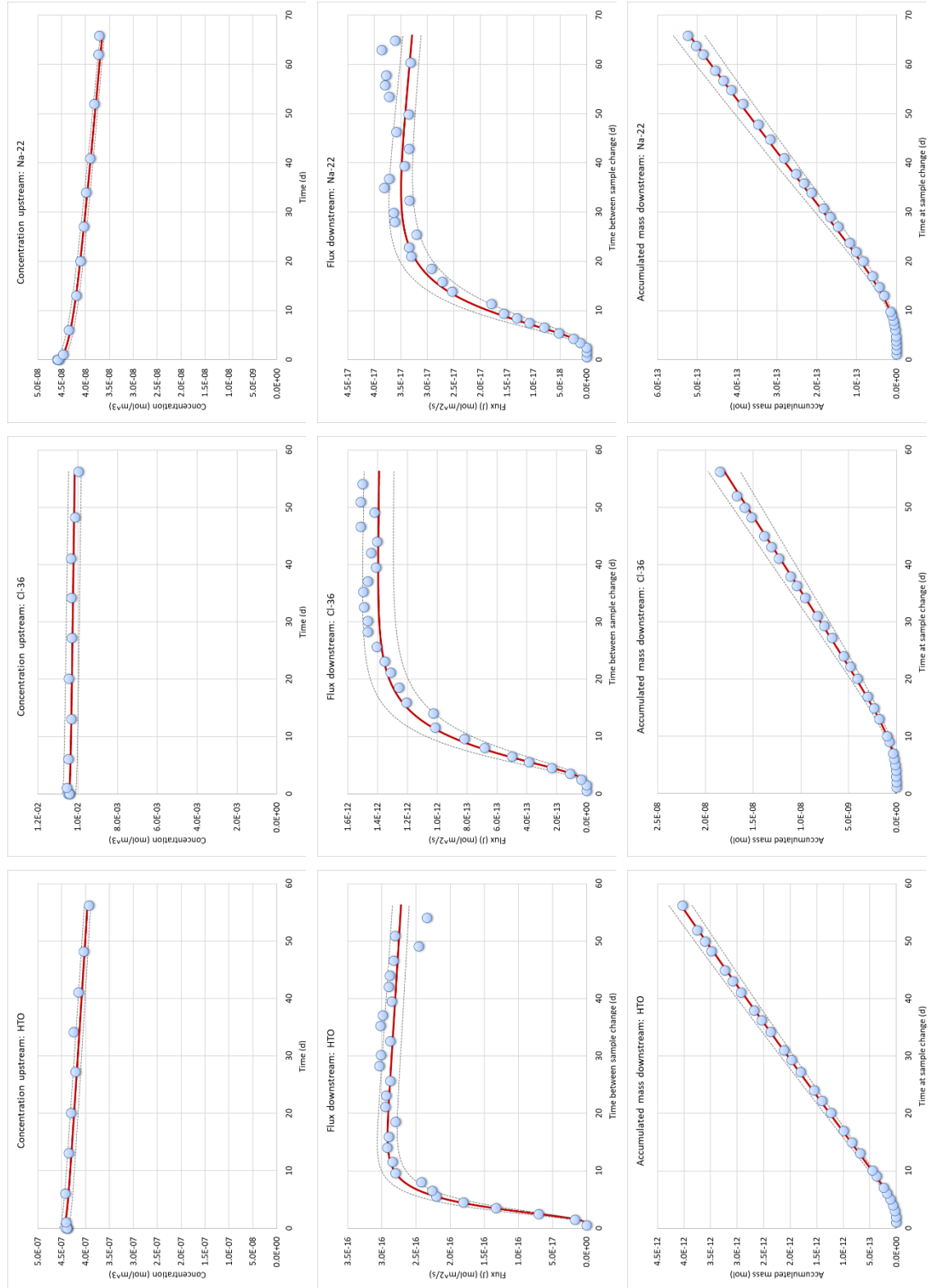
TRU1-1-902.85-DI (S1) – Opalinus Clay – SPW-TRU

Sample diameter 63.62 mm – sample thickness 14.06 mm



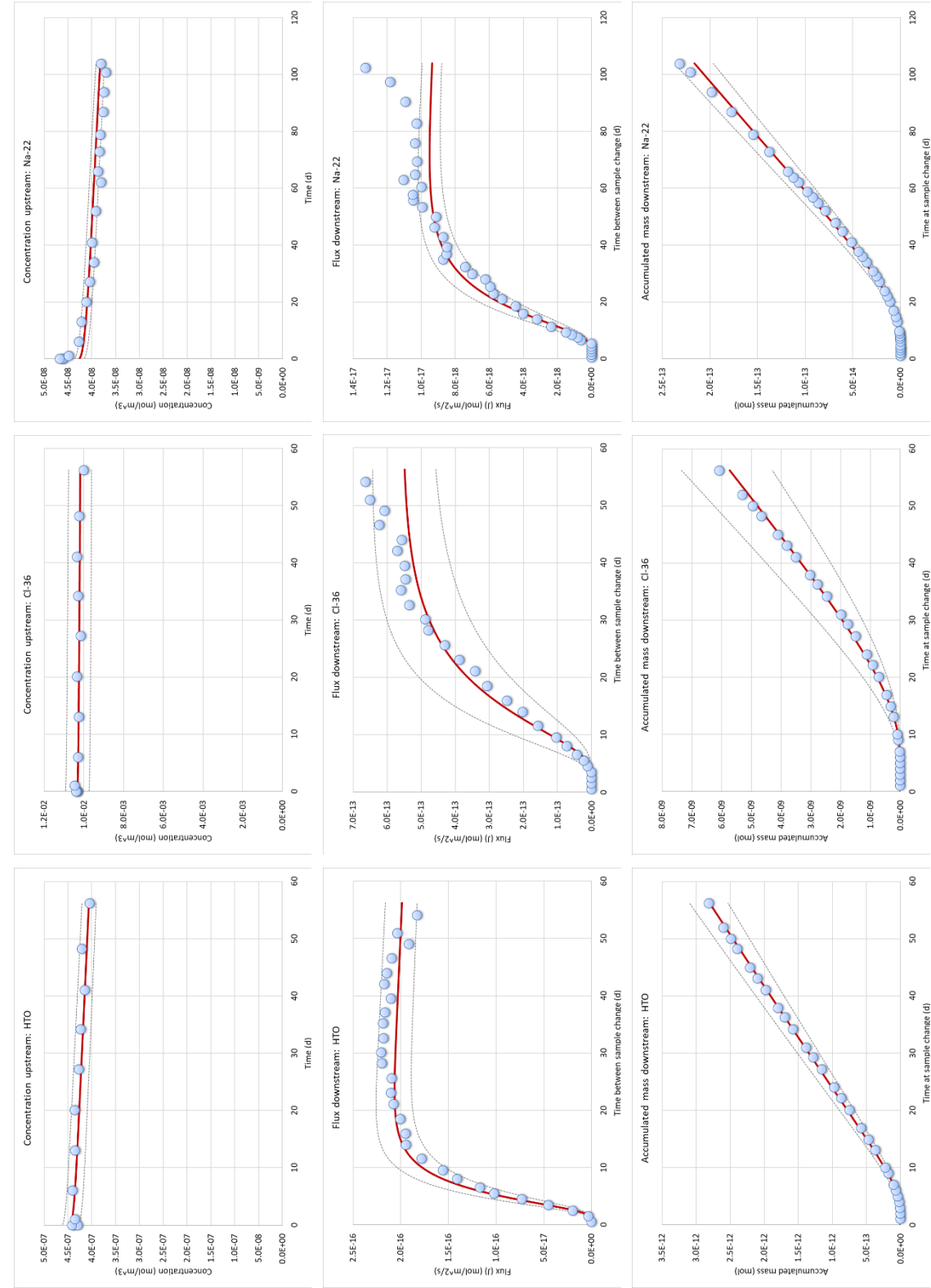
TRU1-1-913.07-DI (S1) – Opalinus Clay – SPW-TRU

Sample diameter 63.60 mm – sample thickness 12.63 mm



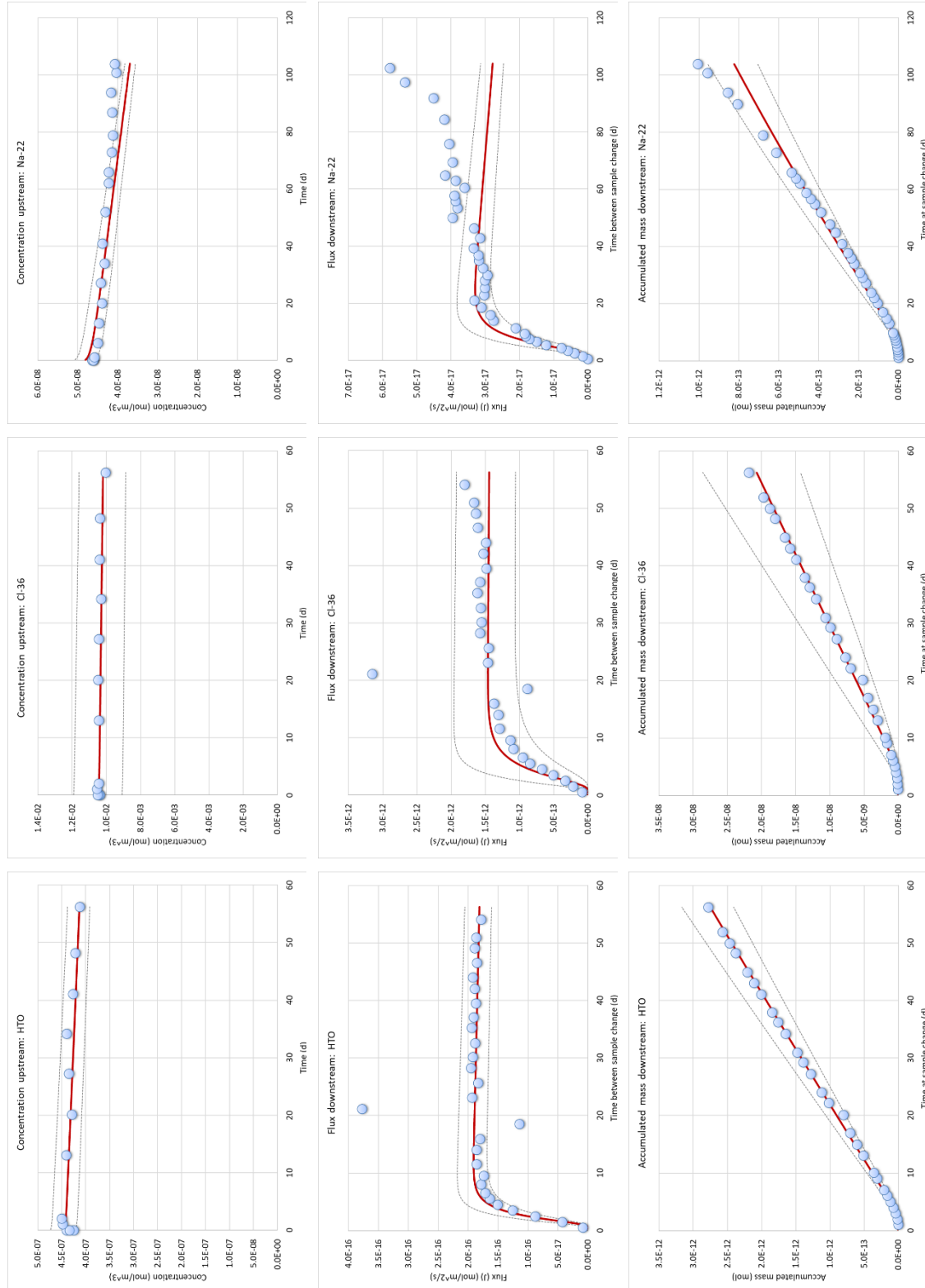
TRU1-1-923.34-DI (S1) – Opalinus Clay – SPW-TRU

Sample diameter 63.57 mm – sample thickness 12.57 mm



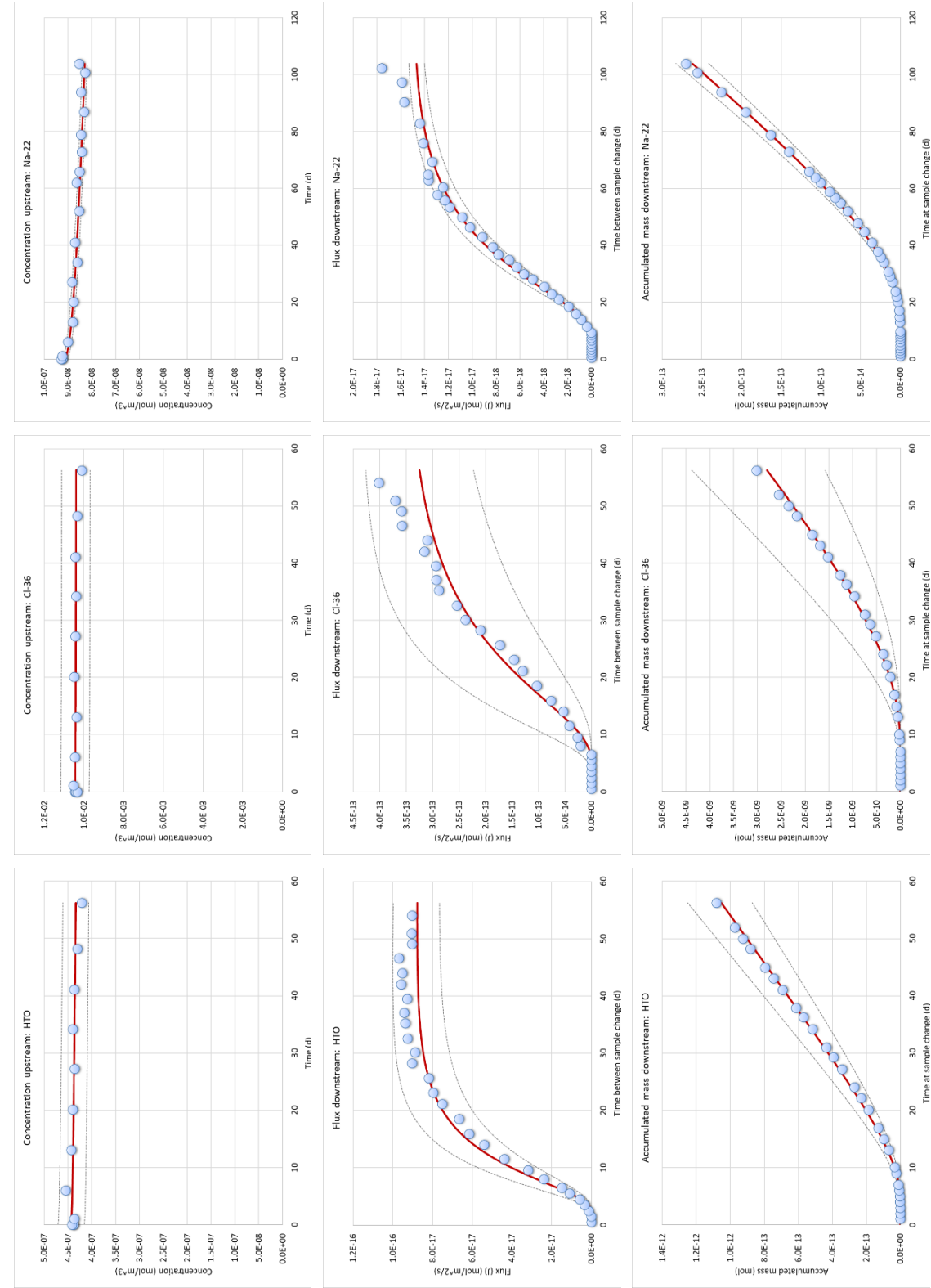
TRU1-1-930.99-DI (S1) – Staffelegg Formation – SPW-TRU

Sample diameter 63.71 mm – sample thickness 13.02 mm



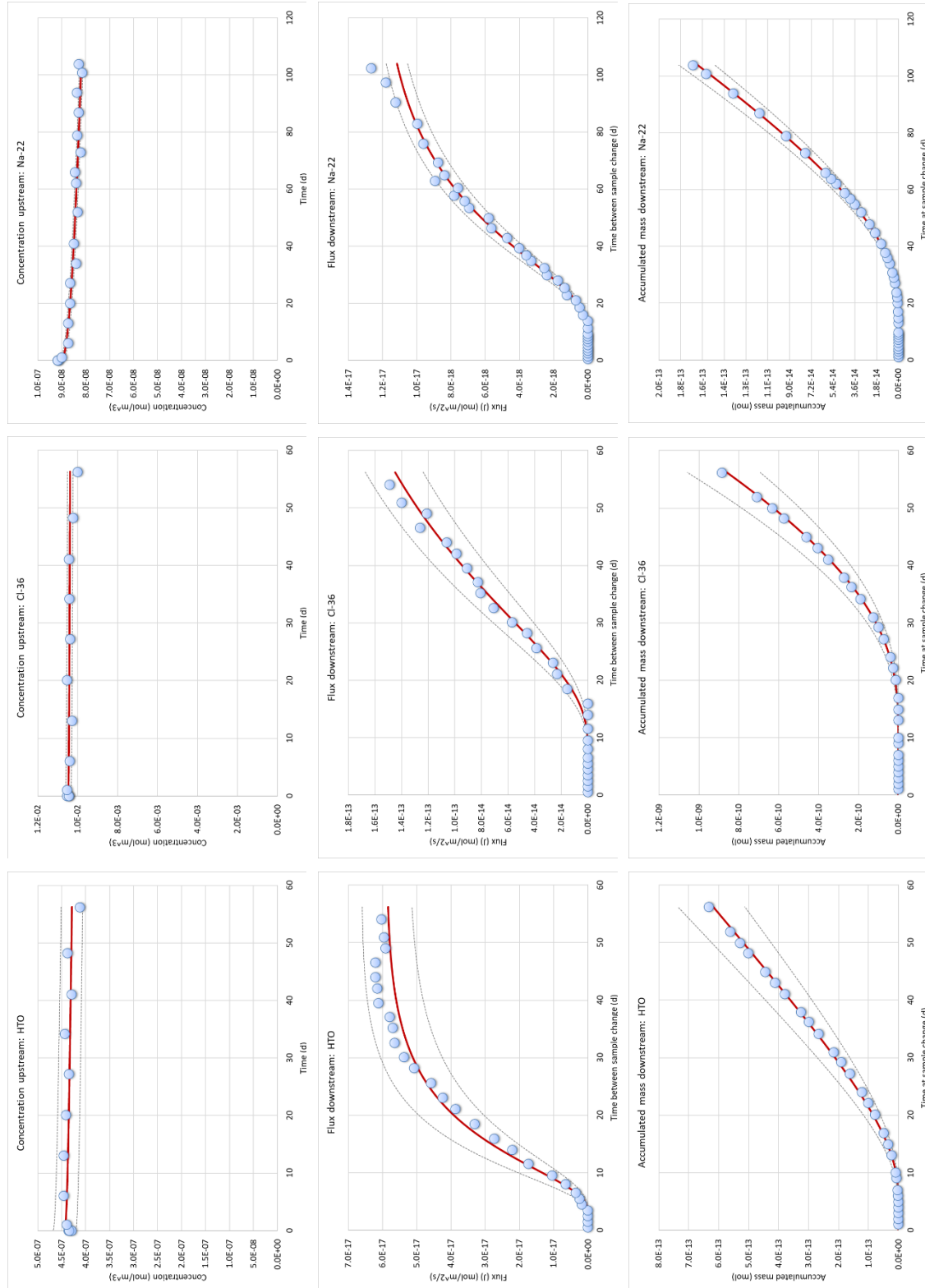
TRU1-1-938.90-DI (S1) – Staffelegg Formation – SPW-TRU

Sample diameter 63.60 mm – sample thickness 14.53 mm



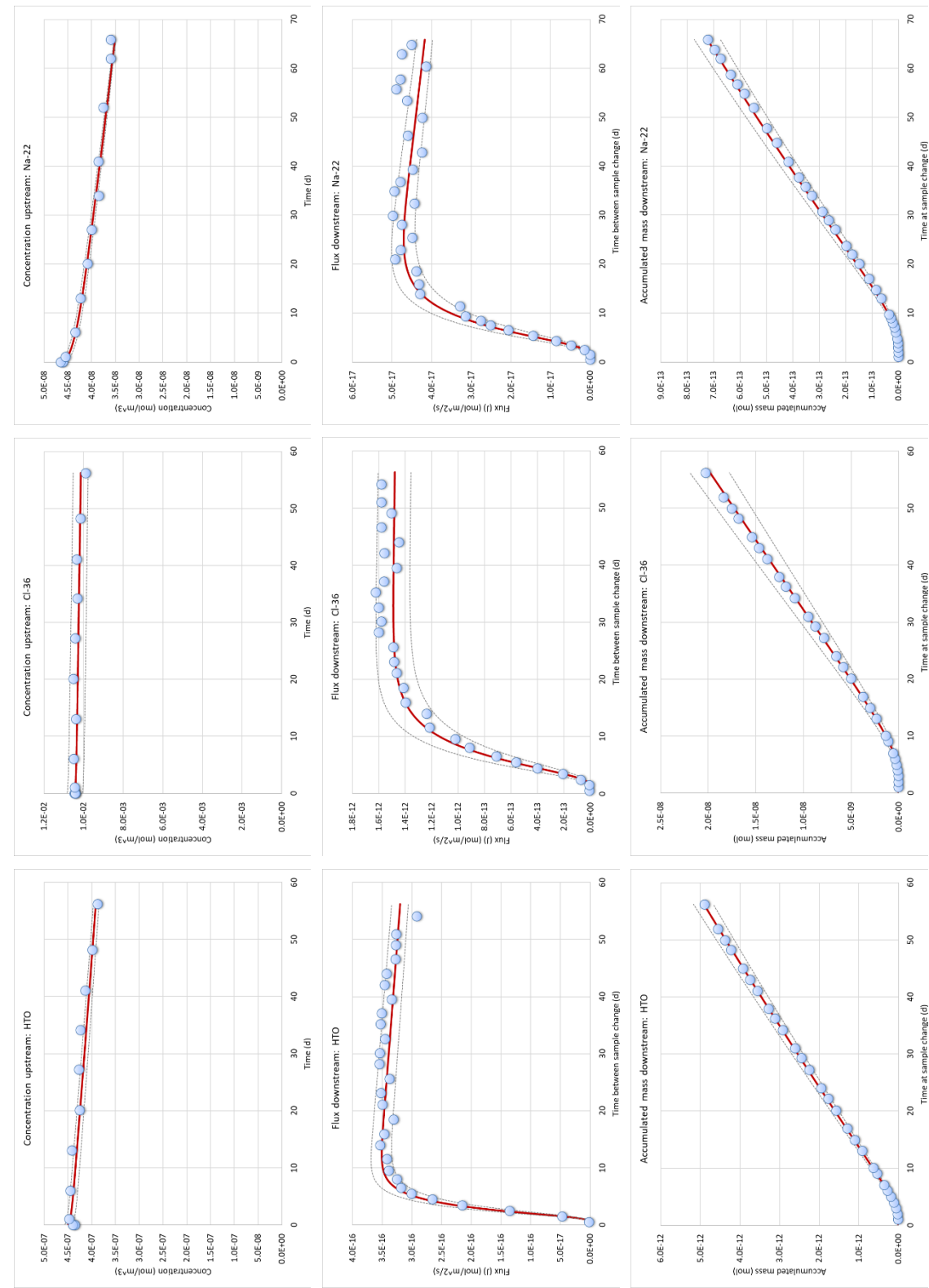
TRU1-1-943.21-DI (S1) – Staffelegg Formation – SPW-TRU

Sample diameter 63.74 mm – sample thickness 13.51 mm



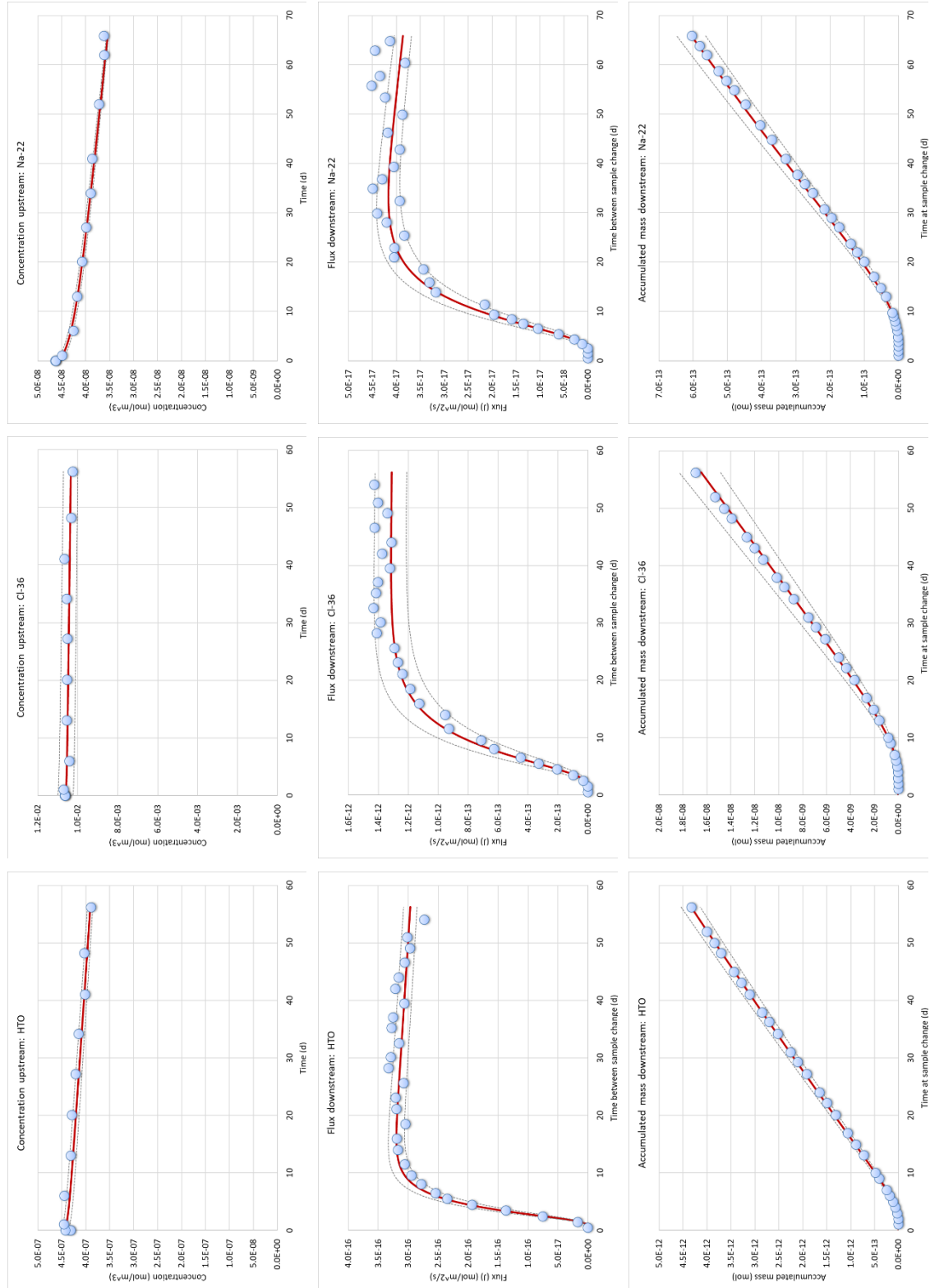
TRU1-1-955.02-DI (S1) – Staffelegg Formation – SPW-TRU

Sample diameter 63.58 mm – sample thickness 11.53 mm



TRU1-1-961.72-DI (S1) – Staffelegg Formation – SPW-TRU

Sample diameter 62.90 mm – sample thickness 12.87 mm



TRU1-1-1026.55-DI (S1) – Klettgau Formation – SPW-TRU

Sample diameter 63.72 mm – sample thickness 12.20 mm

