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Simulating Reservoir Processes of Siliciclastic ATES Reservoirs in Flanders through Empirical Experiments.

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ABSTRACT

In Flanders, siliciclastic reservoirs hold a strong potential for the exploitation of Aquifer Thermal Energy Storage (ATES) geothermal reservoirs. However, temperature changes within these reservoirs might affect both permeability of the reservoir as well as groundwater quality. In order to understand the effects of changes in fluid temperature, static reactivity tests in combination with core-flooding experiments and numerical modelling are used during this study. This paper presents the results of static reactivity tests within the Formation of Brussels, which is an important aquifer in Central Belgium and is targeted as an ATES reservoir in the Province of Flemish-Brabant. These empiric tests highlight the effect of temperature on carbonate precipitation, surface exchange reactions and dissolution of silica and feldspars. In addition, this paper shows that the geochemical behavior of different lithotypes varies in function of their mineralogy.

1. INTRODUCTION

ATES is a sustainable technology that is capable to fulfill both heating and cooling demands of residential, office and industrial buildings. Since these systems are capable of replacing traditional heating systems, powered by fossil fuels, this technology possesses a strong potential to decrease greenhouse gas emissions in Belgium. This paper focusses on the north-eastern part of Flanders, where several suitable siliciclastic reservoirs are present in the subsurface as visualized in Figure 1.

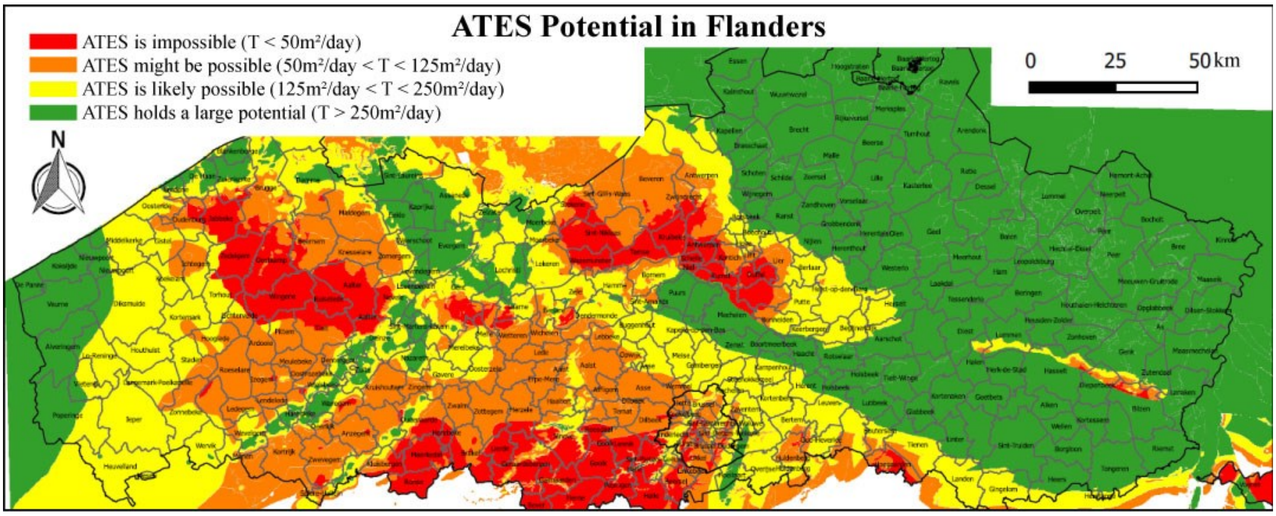

Figure 1: Potential for ATES geothermal projects in Flanders, based upon the presence of an aquifer with a suitable transmissivity (T) (modified after François and Van Lysebetten, 2017).

An inherent process associated with all geothermal installations are variations in fluid temperatures, which might affect chemical equilibrium conditions within these geothermal systems. Altering these chemical conditions might lead to water-rock interactions, which potentially trigger dissolution and precipitation reactions of minerals such as calcite, iron(hydr)oxides, feldspars etc. Reactions as such lead to the degradation of reservoir quality, by blocking pore-throats and thus decreasing permeability of the exploited reservoir system (Possemiers, 2014 and Burté et al., 2016). Moreover, due to mixing of different water types, redox conditions in ATES reservoirs might be altered, leading to the dissolution of iron sulphides, organic matter or (hydr)oxides, which is associated with the potential release of As, Ni and other heavy metals (Jesußek et al., 2012). In addition to these reservoir technical issues, ATES systems in Flanders are often located in formations that are widely used as a freshwater reservoirs, raising concerns with regard to the influence of reservoir exploitation on groundwater quality.

This paper presents preliminary results based on static reactivity tests in combination with core-flooding experiments in order to empirically assess and quantify the reactivity of different siliciclastic reservoirs. In total, these experiments focus on five of the major siliciclastic sediment systems in Flanders, being carbonate cemented sand(stone), organic-rich sand(stone), arkosic sand(stone), glauconitic sand(stone) and iron-rich sand(stone). With these empirical tests ATES reservoir conditions can be mimicked within a lab-environment, allowing to control and monitor relevant parameters, such as temperature, flow-rate, geochemical composition of the fluid, heat-exchange, differential pressure, etc.

2. GEOLOGICAL SETTING

The samples studied by experiments in this paper are part of the Formation of Brussels, which consist of marine sands deposited during the early Middle-Eocene in Central Belgium (Houthuys, 2011). This formation is characterized by good reservoir properties with hydraulic conductivities in the order of 1 to 8 10^{-4} m/s (Possemiers et al., 2012). Therefore, it acts as a major groundwater source for the region and is currently widely targeted as an ATES reservoir in the Province of Flemish-Brabant and the Region of Brussels. Furthermore, the Brussels Sands are characterized by a rather complex depositional setting, leading to an heterogenous deposit with an anisotropy that complicates fluid flow modeling (Huysmans et al., 2008). In general, the Brussels Sands were deposited as a tidal sandbar system at the beginning of an important transgression. The base of this formation is erosive in nature, possible related to strong SSW-NNE tidal currents, explaining the distribution of the Brussels Sands through Belgium (Figure 2) (Huysmans et al. 2008 and Houthuys, 2011).

Figure 2: Map of Belgium highlighting the occurrence of the Formation of Brussels, with the location of the studied Mont-Saint-Guibert quarry (modified after Houthuys, 2011).

The depositional setting of the Formation of Brussels is described in detail by Houthuys (1990) and Houthuys (2011), based on an extensive study combining outcrops and borehole data. The Brussels Sands are deposited in two different stages, of which the first stage is dominated by coarse clastic sands which contain coarse glauconite and are deposited as thick cross beds, filling tide-parallel channels with dimensions of several kilometers in width and tens of meters in depth. This contrasts with the later depositional stage, which is more dominantly made of finer, carbonate-rich sands (Houthuys, 1990). Overall, the Brussels Sands thus consist of unconsolidated fine to coarse grained quartz sands that contain feldspars, silex, glauconite, carbonates and heavy minerals (Huysmans et al., 2008). Due to its complex depositional setting, reservoir heterogeneity of the Brussels Sands is a multi-scaled problem of sub-horizontal tabular sets (meter-scale) containing laminae and mud-drapes (millimeter-scale) (Huysmans et al., 2011). Since this study aims to understand differences in reactivity between different lithologies of the Brussels Sands, these heterogeneities in mineralogy, grain size, porosity and permeability have to be taken into account. Therefore, sampling tubes as illustrated in Figure 3 were used to collect undisturbed samples of the Brussels Sands in the quarry of Mont-Saint-Guibert, located South of Wavre (Figure 2). These samples provide useful information on the state of the undisturbed, in-situ pore-network and grain size distribution, which are critical input parameters for core-flooding experiments.

Figure 3: Sampling tube for undisturbed samples (left), allowing to gather samples of unconsolidated sands whilst keeping sedimentary structures intact (right).

2.1 The Formation of Brussels as an ATES reservoir

Within the Brussels Sands, several ATES projects are currently under development or being exploited, such as the ATES system of the Department of Environment in Brussels, Brussels Airport and several other buildings in the region (Vancauwenberghe, 2018). Several of these projects experienced problems related to well-clogging caused by precipitation of iron(hydr)oxides, hampering productivity of these systems within this reservoir. These problems are most extensively described and studied for the KBC building near Leuven (Figure 2). The latter experienced severe productivity problems during the first years of operation and for that reason the flow rate throughout these wells had to be decreased (Van Bael et al., 2001 and Possemiers et al., 2016). Possemiers et al. (2016) assessed the geochemical evolution of the studied fluids by numerical modelling this case-specific situation with PHT3D, which combines both the groundwater flow and solute transport code from MODFLOW with the chemical reactions code from PHREEQC-2. Based on these models Possemiers et al. (2016) concluded $Fe(OH)_3$ precipitation is influenced by two different mechanisms, being mixing of water types (1) and the temperature of the injected fluids (2). Mixing water types with a different geochemistry, being either oxic or reducing groundwater rich in dissolved Fe(II), accounted for the bulk of the initial precipitation. In addition to this mixing effect, Possemiers et al. (2016) also concluded that precipitation rates of $Fe(OH)_3$ was influenced by the temperature of injected fluids. The static reactivity tests, discussed in the next section, thus mainly aim to understand and quantify the effect of the fluid temperature on the reactivity and leaching behavior of these sands as such.

3. METHODOLOGY

During this study both static reactivity tests and core-flooding experiments are used in order to assess mineralogical reactivity of the studied ATES reservoirs. However, this paper only presents results from static reactivity tests in further detail, since core-flooding experiments are ongoing.

3.1 Static Reactivity Tests

Static reactivity tests provide initial information regarding the mineralogical reactivity of the studied aquifers. The main purpose of these tests is to assess the influence of reservoir temperature on mineral solubility and cation-exchange reactions of sandy lithologies of the Brussels Sands. In contrast to core-flooding experiments, which are also part of this study and which are currently ongoing, fluids are thus not actively flushed through the sands and there is no temperature gradient present between the sands and fluids.

A liquid-to-solid ratio of 2.5 was used for these tests, combining 300 grams of oven-dried (40°C) sand with 750 grams of formation water from the Formation of Brussels. These mixtures were transferred to closed jars, after which they were placed in an oven, allowing to control the temperature during the experiment. During these experiments, sands reacted statically for 72 hours at a constant temperature, starting at 10°C and ending at 90°C, with every timestep a temperature increase of 10°C. During these tests, fluid samples of 5 mL were gathered every 1st, 3rd, 24th, 48th and 72th hour after the temperature of the experiment was increased. The geochemistry of these fluids was analyzed by ICP-OES.

Next to the studied sand samples, the static reactivity tests consisted of a blank solution made of pure formation water, yielding information on pure chemical solubility of elements within the fluid itself. Furthermore, one duplicate sample (M1A & M1B) was used as well in order to verify the reproducibility of the static reactivity tests as such.

4. RESULTS

This contribution only presents data of static reactivity tests which were performed on two lithotypes of the Brussels Sands.

4.1 Litholog and grain size variation

The quarry of Mont-Saint-Guibert generally consists of either coarse grained sands, rich in iron(hydr)oxides or of fine grained sands rich in feldspars and carbonates. The heterogeneity within this reservoir system translates itself in tabular-sets of several meters in thickness with fine internal laminae and mud-drapes (Figure 4). Bioturbations are often observed near the erosive contact between two successive tabular beds and often lead to formation of cemented nodules.

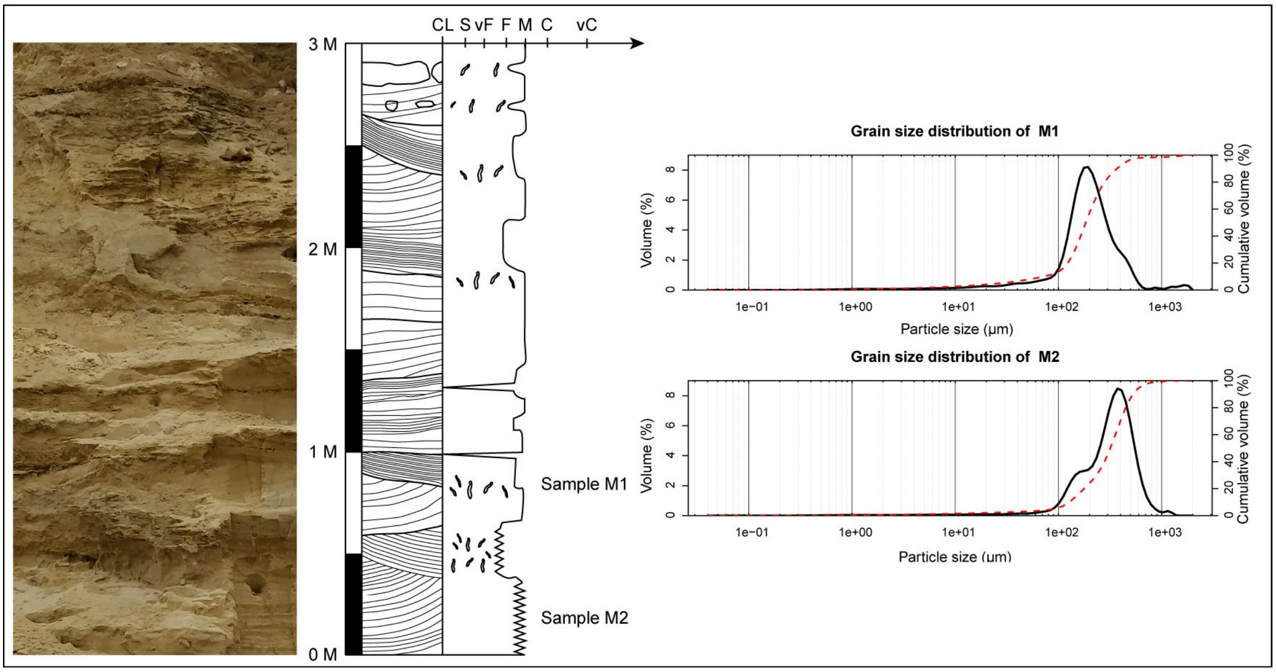

Figure 4: Outcrop with corresponding litholog of the Brussels Sands in the quarry of Mont-Saint-Guibert, highlighting the presence of meter-scaled tabular-sets with finer internal laminae. Sample M1 represents a fine-grained, carbonate-rich lithology, while sample M2 represents a coarse grained lithology.

4.2 Static Reactivity Tests

Results from these tests indicate a clear correlation between concentrations of specific chemical elements and the temperature of the experiment. A sudden and exponential evolution in concentrations of certain chemical elements, such as Ba, Ca, Cr, K, Li, Mg, Mn, Si and V, was observed in the initial three hours after raising the temperature of the experiment by 10°C. However, in the following hours, whilst the temperature of the experiment remained constant, geochemical concentrations evolved in a linear way (Figure 5A). Untreated formation water of the Brussels Formation was used as a reference to assess the influence on changing temperatures and redox conditions without any interaction with sandy lithologies (blue line, Figure 5). For most chemical elements this reference was characterized by a stable geochemistry, with as exception Ca (and to a minor degree Sr). Sample M1 represents a fine-grained, carbonate and feldspar rich lithotype occurring in the Formation of Brussels and was analyzed as a duplicate (M1A & M1B). Sample M2 corresponds to a coarse grained sand of which the grains are coated by iron(hydr)oxides.

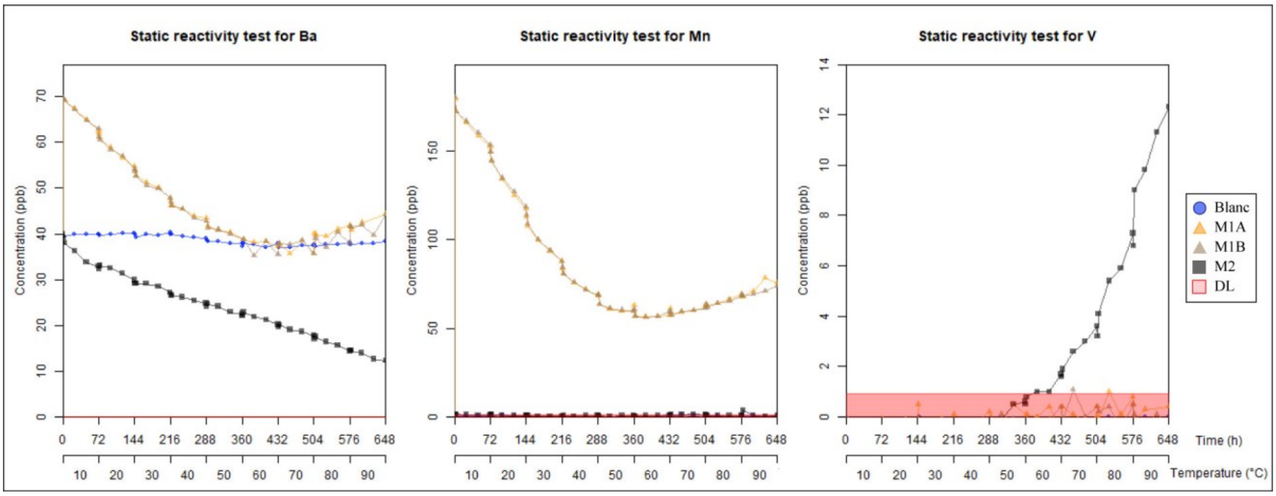

Figure 5: Evolution of the concentration of Barium, Manganese and Vanadium, with detection limits visualized as a red box. As a reference, untreated formation water of the Brussels Formation was used. Samples M1A and M1B correspond to fine-grained carbonate and feldspar rich lithologies, while sample M2 corresponds to medium-grained sands rich in iron(hydr)oxides.

In general, concentrations of different chemical elements follow a unique path through time, either way increasing or decreasing from certain temperatures onwards. The latter indicates that certain elements leach out and/or dissolve from the Brussels sands (e.g. Vanadium & Manganese from 60°C onwards, Figure 5) or get absorbed and/or precipitate in the Brussels Sands (e.g. Barium, Figure 5A & Manganese up to 60°C, Figure 5). However due to high detection limits of X-Ray Diffraction measurements (approximately 1 wt. %), this technique was not able to confirm a significant change in mineralogy.

5. DISCUSSION

Correlations between concentrations of different elements have been analysed, highlighting potential associations between the behavior of different elements. Concentrations of elements such as Ca, Ba, Mg & Sr are characterized by strong and positive correlation coefficients between one another, indicating that concentrations of these elements are related to relative partitioning in similar mineralogical phases (such as carbonates) or to increased surface complexation of these cations (Angove et al., 1999; Bonte et al. 2011; Bonte, 2015 and Fernandes & Baeyens, 2019). Precipitation of carbonates mainly affects concentrations of Ca (and to a minor degree Sr), which is related to a decreased solubility of carbonates with increasing temperature (Weyl, 1959). However, concentration differences of these elements cannot solely be explained by the formation of carbonate precipitates, since a different evolution was observed for the blanc reference and the studied lithotypes (Figure 6). Surface complexation of cations is an endothermic process, which thus accelerates with increasing temperatures (Angove et al., 1999; Bonte et al. 2011 and Bonte, 2015). This complexation can take place both on clay minerals, which are present in both M1 and M2, and on iron-oxide surfaces (dominantly present within M2). Therefore, concentration plots for sample M2 are characterized by a stronger decrease as compared to sample M1.

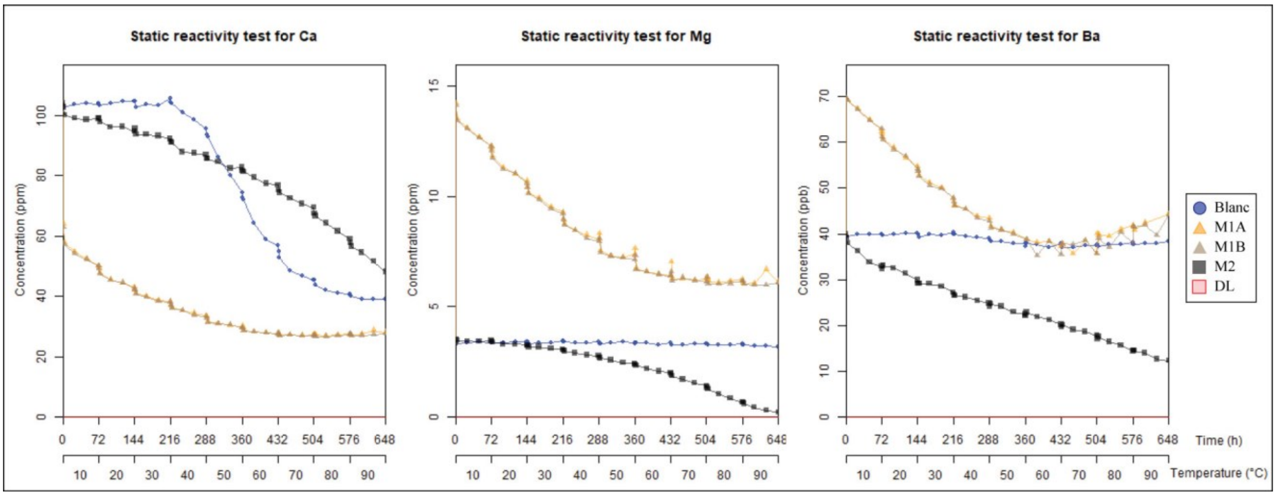

Figure 6: Evolution of the concentration for Calcium, Magnesium and Barium for the Brussels Formation. These graphs highlight the effect of carbonate precipitation on Calcium, while as this does not affect Magnesium or Barium. The decrease of the latter is associated with surface complexation reactions.

From 40°C onwards, Figure 7 shows an increase in silicon concentrations, which partially can be explained by an increasing solubility of silica with increasing temperatures (Van Capepellen et al., 2002). However, feldspar-rich lithologies (M1) are characterized by a more rapid increase of Si concentrations, especially from 60°C onwards. In addition, these lithologies show a similar evolution for elements such as Ba, Ca, K, Li and Mg with respect to Si. These elements are characterized by gradually decreasing concentrations up to 60°C, after which concentrations stabilize or increase. Due to the presence of feldspars in these lithologies, and the relative absence of feldspars in sample M2, this evolution is thus interpreted as a gradual dissolution of feldspars from 60°C onwards. The latter also explains the accelerated increase in Si for sample M1 in contrast to M2.

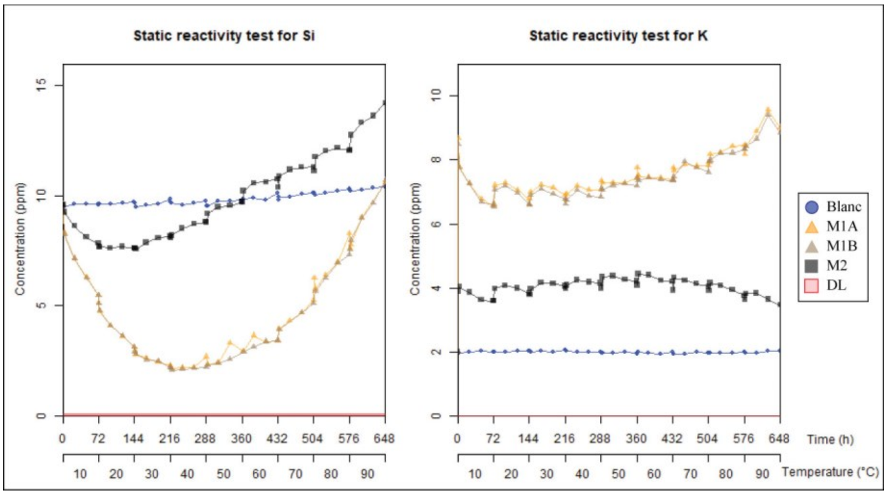

Figure 7: Evolution of Silicon and Potassium concentrations during the experiments. Solubility of Silica increases with rising temperatures, leading to higher concentrations of Silicon for both sample M1 and M2. Feldspars dissolve from 60°C onwards, explaining the stronger increase in Silicon for M1, as well as the increase in Potassium.

6. CONCLUSION

Static reactivity tests confirm the temperature-dependence of concentrations of certain elements within fluids in equilibrium with the Formation of Brussels. In general, three different trends were observed during the static reactivity tests. A first observation (1) corresponds to the expected precipitation of carbonate phases with increasing temperature. The latter leads to a decrease in concentrations of Ca (and to a minor degree Sr). A second observation is the general decrease in the concentration of cations with rising temperatures (2). This is due to an increased amount of surface-complexations taking place, binding these cations to either clay minerals or iron(hydr)oxides. The effect of these surface-complexations differ between both lithotypes. The fine-grained lithotype only contains a minor amount of clay minerals, while as the coarse grained lithotype contains both clay minerals and iron(hydr)oxides. Fluids in equilibrium with these coarse grained sands thus become generally more depleted in cations with rising temperatures. A final observation is the increased solubility of silica and dissolution of feldspar at higher temperatures (3). Biogenic Silica, present throughout the Brussels Sands, start to dissolve from 40°C onwards, while Potassium-rich feldspars dissolve from 60°C onwards. Both dissolution reactions lead to an increase in Silicon and Potassium within the fluid.

In addition to these static reactivity tests, this study also uses core-flooding experiments which are currently ongoing and therefore data are not yet presented within this paper. The latter allows to study dissolution and precipitation of mineralogical phases within a sediment core, as well as the formation of a reaction front.. In addition, numerical models in PHREEQC will be used in order to validate the dissolution of mineralogical changes, as well as verifying the surface-complexation reactions as such.

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