Impact of geochemical uncertainties on geothermal fluid production and scaling precipitation in geothermal plants and facilities

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ABSTRACT

Scaling is one of the most common operational challenges occurring in geothermal plants. In order to evaluate its associated risks, a proper understanding of the interaction between the geothermal brine composition, operating conditions and pipe materials is key. However, uncertainties in geothermal brine composition, due to sub-optimal sampling, added inhibitors or measurement uncertainties, can significantly impact the accuracy and precision of the prediction of geothermal fluid production, and scaling and corrosion potentials**.** In this work we have evaluated the risk of scaling in geothermal plants under composition uncertainties. For a better understanding of the impact of compositional uncertainties on fluid properties, two non-intrusive uncertainty propagation methods were employed (Monte-Carlo and Sobol sampling). A fluid composition database was used to characterize the composition uncertainty bounds for different minerals. The work was demonstrated for barite precipitation in a simplified modelled heat exchanger. At higher temperatures in the heat exchanger the width of distribution was found to be relatively small due to many samples resulting in no scaling at all, while the difference between the distribution mean and nominal value was relatively high at 46%. At lower temperatures scaling was predicted to occur for all samples considered, and while the range of possible scaling amount values increased, the distribution mean was significantly closer to the nominal value (13%). This uncertainty quantification workflow could enable operators to make a robust decision on how to control and mitigate scaling and corrosion issues in their plants.

1. INTRODUCTION

Scaling, along with corrosion, is one of the most common operational challenges occurring in geothermal plants which can complicate the efficient operation of geothermal system. The deposition of solid scales can lead to clogging of wells, reservoirs or surface facilities, reduction of flowrates within the wellbore and topside equipment, reduce the transfer of heat within heat exchanger systems, ultimately affecting the lifespan and economic viability of geothermal systems.

Based on the chemical composition of the geothermal brine, which depends on the geological formation and on the operational settings, different types and amounts of scales can form within the system. Examples include calcite ($CaCO_3$), barite ($BaSO_4$), celestite ($SrSO_4$), as well as lead-based and silica scales (Regenspurg et al. 2010). In this work, the focus was on barite scaling, although the developed methods are generally applicable to any type of scale. Barite is formed through the reversible reaction of barium and sulphate ions:

$$Ba^{2+} + SO_4^{2-} \leftrightarrow BaSO_4 \qquad [1]$$

And its solubility is primarily dependent on temperature, decreasing with decreasing temperature, with minor dependencies on salinity and pH (only above a pH of 9) (Zhen Wu 2016).

In order to evaluate the associated risks of scaling, a proper understanding of the interaction between the geothermal brine composition, operating conditions and pipe materials is key. An accurate prediction of the scaling amount and location in the geothermal systems depends heavily on characterization of the geothermal fluid which is impacted by the uncertainties in the fluid sampling, added inhibitors and measurement analysis. Most of the current analysis is limited to treating the fluid composition in a deterministic manner and does not include the uncertainties in fluid compositions and their potential impact on the flow-chemistry challenges.

The objective of this paper is to develop a workflow for evaluating the effects of uncertainties in geothermal fluid composition on the potential formation of mineral scales. In the next section (Section 2), the modelling and uncertainty quantification methodologies are explained. This is followed by the description of the case study in Section 3, and the results of applying the uncertainty quantification workflow to this case study

in Section 4. Finally, Section 5 ends with the main outcomes and conclusions of the work and plans for future developments.

2. APPROACH

In this section, the modelling and uncertainty quantification approaches used within this work are explained.

2.1 Geochemical scaling model

In order to model the scaling potential of barite within the geothermal system considered, the aqueous geochemical calculation software PHREEQC (version 3) was used (Parkhurst and Appelo 2013). Among other functionalities, PHREEQC allows for the speciation and calculation of saturation indices and precipitation amounts of a wide range of minerals and compounds, including barite (Parkhurst and Appelo 2013).

PHREEQC calculations are performed by defining an input file which contains several set-up and calculation blocks, starting with the initialization of both aqueous and gas phase compositions at reservoir conditions. Once the composition of the total fluid has been defined, an equilibrium block is added to calculate the saturation index and scaling potential of barite at a given temperature and pressure conditions. In this work, the Pitzer thermodynamic database was used as a basis for the speciation equilibrium calculations. The initial brine compositions and temperature-pressure combination at which the calculation were performed will both be given in Section 3.

2.2 Uncertainty quantification

The aim of uncertainty quantification is to quantify the effects of uncertainties arising during the modelling process on the potential variation of the model's outcome, and to determine which input parameters contribute the most to this variation.

Uncertainty is typically divided into two different sources: an epistemic (systemic) part, which stems from a lack of knowledge or data (e.g. a simplified model or model parameter for which an accurate value is lacking), and an aleatoric part, which comes from intrinsic randomness associated with the modelled system (e.g. initial conditions that are never exactly the same) (Zhang 2020).

In this work, the focus was the latter (aleatoric) form of uncertainty, as this is the type uncertainty in brine composition are assumed to fall under. It can be argued that uncertainties in chemical composition can be reduced by several samples which can be obtained from the field but for the current study the uncertainties in the multiple samples obtained from a certain field or formation was treated as an aleatoric uncertainty. Quantification of the effects of these uncertainties was done through a non-intrusive method in which a large number of different brine compositions were randomly sampled using Monte-Carlo and Sobol methods. These samples were generated randomly from a uniform distribution within given upper and lower bounds specific to each brine component considered (the values of these bounds are given in Section 3). For each sampled composition the geochemical speciation models were executed, resulting in a range of model outputs (e.g. barite scaling potential) that could be statistically analysed to determine the output distribution and variability. Figure 1 gives a schematic overview of this uncertainty quantification workflow.

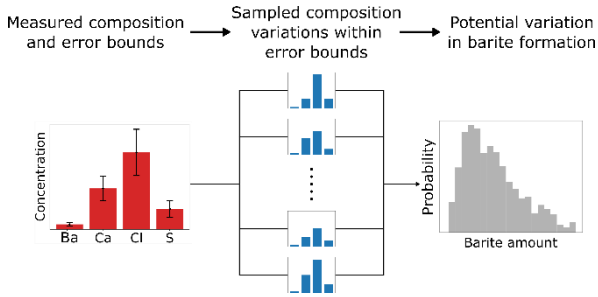

Figure 1. Schematic overview of the general uncertainty quantification workflow.

In addition to the statistical analyses of the output range, the impact each individual brine element had on the barite scaling potential was also quantified. This was done using so-called Sobol sensitivity indices, which are a variance-based approach in which the magnitude of the impact of a given input parameter on the output is determined by evaluating how (much) the output changes when the input parameter is varied (Saltelli et al. 2010). This impact is often divided into a specific set of "order" effects, e.g. first order, second order, and total order effects. First order effects look at the impact of varying a given parameter by itself while keeping the other parameters fixed. The second order effects vary the parameter together with one of the other input parameters to give an indication how their interaction affects the output and this method will be applied to characterize higher order interactions (Saltelli et al. 2010). As third or higher order effects become more and more difficult to evaluate, they are typically left out of analyses and the total order effect is calculated which summarizes all order effects of a given input parameters.

To increase the efficiency and accuracy of Sobol sensitivities, an additional sampling scheme, called Sobol-Saltelli sampling, is used to generate the different brine compositions. This method generates quasi-random samples which are more evenly distributed between the upper and lower bounds of the sampling limits, therefore giving a better indication of the model sensitivities throughout the uncertainty range, even at low number of samples (Saltelli et al. 2010). For reference, Figure 2 gives a comparison between samples generated from a random uniform distribution, and through the use of a Sobol sampling scheme.

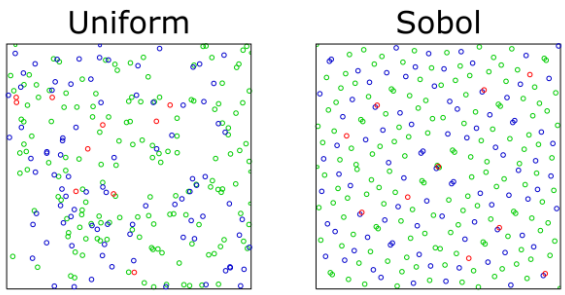

Figure 2. Example of samples generated using a (pseudo-)random uniform distribution (left) and a Sobol sampling strategy. Points in red correspond to the first 10 samples, those in blue to the first 100 samples, and green the remaining samples. As can be seen, the Sobol samples are distributed more evenly throughout the sampling bounds

3. DESCRIPTION OF THE CASE STUDY

The concentrations of the main components in the geothermal brine that were included in this case study are given in Table 1. These elements were chosen as they were expected to have the largest impact on barite scale formation. In addition, this table also lists the uncertainty bounds for each component, which were generated from a fluid composition database containing different measurements for a geothermal fluid from a single site and several sites within the same formation (Wasch et al. 2019). In addition, Table 2 gives the mole fractions of the gas phase of the total geothermal fluid. As the gas phase was not included in the uncertainty analysis, no variation bounds are given.

Table 1. Nominal concentrations of components included in the uncertainty analysis and associated uncertainties based on variations of the samples from a single well or group of wells in the same formation.

Element	Nominal Concentration (mg/l)	Minimum/maximum percentage deviation	
		Variation in well	Variation in formation
Ba	5.5	±3%	±30%
C	0.001	±3%	±30%
Ca	7450	±3%	±30%
Cl	145000	±3%	±30%
K	2200	±0.5%	±5%
Mg	1150	±4%	±40%
Na	85000	±2%	±20%
S	585	±4%	±40%

Table 2. Mole fraction of the three main components in the gas phase of the fluid considered. As no uncertainty quantification was applied to the gas phase, no deviation bounds are given.

Gas	Mole fraction
CO_2	0.257
Methane	0.572
Nitrogen	0.101
Others	0.07

Before the uncertainty quantification was applied, a number of simulations were performed to investigate the effect some of the brine component might have on barite scaling amounts. This was done to provide a basis for comparing the uncertainty quantification results against. The main components from Table 1 expected to have an impact on barite formation (apart from barium and sulphur which are the main constituents of barite) were calcium, which was identified as a enabling factor by Dai et al. (2021) (due to a decrease in ion pair stability increasing sulphate concentration in calcium rich solutions), but an inhibiting factor by Graham et al. (2003), and Azaza (2017) (due to calcium ions adsorbing on barite nuclei impeding scale growth and competition from calcium sulphate formation), and magnesium, which affects barite precipitation according to Graham et al. (2003).

Figure 3 and Figure 4 show the predicted amount of barite scaling at different temperatures for increasing amounts of calcium and magnesium concentrations in the geothermal brine respectively. As can be seen, increasing calcium concentrations show a clear and significant inhibiting effect on barite precipitation (therefore agreeing with Graham et al. (2003) and Azaza (2017)), while increasing magnesium concentration only show a very minor inhibiting affect.

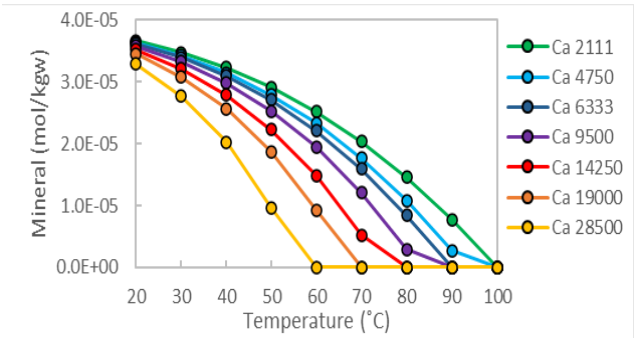

Figure 3. The effect of different initial calcium concentrations on barite precipitation, simulated for cooling from 105 to 20 °C.

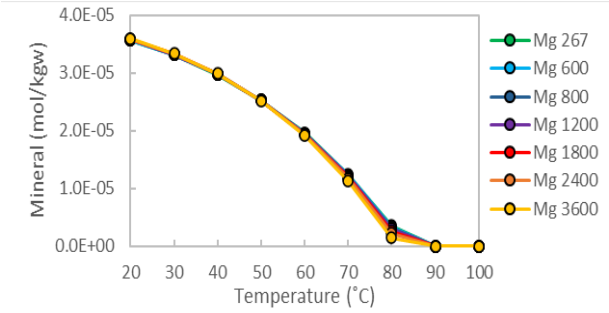

Figure 4. The effect of different initial calcium concentrations on barite precipitation, simulated for cooling from 105 to 20 °C.

The uncertainty quantification was applied to a simplified modelled heat exchanger to simulate the cooling of a geothermal brine. The heat exchanger contained a pre-defined linear temperature and pressure profile starting at 100 °C and 10 bar at the inlet, and ending at 40° and 5 bar at the outlet. At five equally spaced points along the length of the heat exchanger, the uncertainty quantification of barite scaling was applied.

4. RESULTS & DISCUSSION

In this section, the results of the uncertainty quantification of barite scaling are given, starting with a comparison between the uncertainties of composition on the well level and formation level at a single temperature-pressure condition. After this, the results of the uncertainty quantification applied to the simplified heat exchanger are given and discussed.

4.1 Initial results at single operating condition

In order to compare the differences between the smaller well variation and larger formation variations, the uncertainty quantification workflow was first applied to a single temperature-pressure combination of 70 °C and 10 bars, respectively.

In total, around 10,000 brine samples were generated for both the well and formation variations. Figure 5 gives the calculated barite scaling amounts as calculated using PHREEQC for all of these samples, both as a box plot and histogram plot.

As can be seen, the most noticeable difference between the well and formation variations is the width of the respective distributions (bottom plots), which, as expected, is very narrow for the smaller variations, and broad for the larger variations. For the uncertainties based on well variations, the mean value for the barite precipitation is significantly higher than that for the formation variations and is roughly comparable to the nominal value, being only 0.7% lower than the nominal value. On the other hand, the mean of the formation variations is considerably lower than the nominal value (around 20.7%), and lies about one standard deviation away from the mean of the well variations. This is likely due to the fact that for the larger variations, there is a considerable number of brine compositions for which there is very little to no barite scaling at all predicted. This leads to a large "tail" towards the lower amounts of barite precipitation in the formation variations and skews both the distribution itself as well as its mean towards lower values.

These results also show that the distribution in barite scaling amount is not simply linear in magnitude of uncertainty. I.e., even though the formation variations were 10 times larger than the well variations, the resulting barite distribution is not simply 10 times wider around the same mean. Instead, both the mean and shape of the distribution change significantly in a non-linear fashion. Thus, when investigating uncertainty, it is important to have a good estimate of the magnitude of variation in the uncertain inputs in order to be able to create a reliable estimate of output uncertainties.

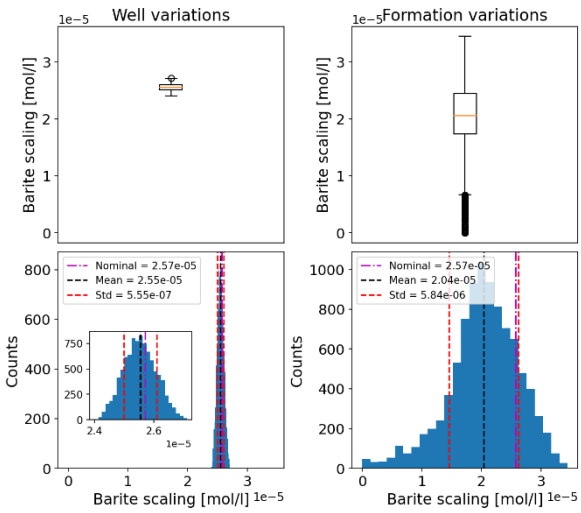

Figure 5. Barite scaling amounts calculated using PHREEQC for the smaller well variations (left), and larger formation variations (right). Results are plotted both as a box plots (top) and histogram plots (bottom). For both cases, roughly 10,000 brine samples were generated.

In addition to analyzing the barite distribution itself, the sampling results were also used to evaluate the impact each of the varied brine components has on the scaling uncertainty of barite. In this step, the importance of each component to the amount of barite precipitation is calculated. This importance can both be due to their relevance to the chemistry of barite precipitation, or due to the component's large uncertainty bounds themselves. The component importance analysis was done using Sobol sensitivity indices (see Section 2.2). Again, this analysis was done for both well and formation variations at 100 °C and 10 bars using 10,000 samples for each case (this time generated using the Sobol sampling scheme).

Figure 6 show the first, second, and total order effects that each of the eight brine components included in the analysis have on the formation of barite. As can be seen, for both variation magnitudes, four of the elements

stand out as having a significant impact on the outcome (barium, sulphur, calcium, and chlorine), while the other four (carbon, potassium, magnesium, and sodium) have negligible to no importance. These findings, especially the high second order effect of the combination of barium and sulphur, align closely with what would be expected from the chemistry of barite precipitation. Barite itself is a compound of barium and sulphate, and thus its formation primarily depends on the presence of the elements making up these two ions. According to Tranter et al. (2020), the ratio between sulphate and barium is an important control on the amount of scaling that can be expected, with less precipitation if the initial ratio deviates more from unity. This could also explain why the second order effects between barium and sulphur increase for the larger formation variations: as the variations get bigger, so does the variability in barium to sulphate ratio, increasing the sensitivity of barite to the combination of these elements. Furthermore, calcium also shows a minor impact on barite scaling according to its Sobol index, which is in line with the investigation discussed in Section 3. In addition, chlorine also seems to a have significant contribution to the uncertainty in barite formation. However, as this was not identified as a contributing factor in the literature, thus the reason for this significant contribution is not fully known. Finally, as was discussed in Section 3, while magnesium was identified in literature as a moderate contributing factor, model results (Figure 4) found it to be very minor, which is also reflected in the Sobol index being (close to) zero.

Another observation made by comparing the smaller well variations to the larger formation variations was that while the four most important elements remain the same, the magnitudes of their sensitivities change considerably, especially those of barium and sulphur. For the smaller variations (well level) sulphur had by far the highest impact on the outcomes, but for the larger variations (formation level) barium becomes the most significant contributor. This analysis was further pursued by looking at the correlations between the four most significant brine components for the well and formation variations, shown in Figure 7 and Figure 8 respectively. For the well variation there is a relatively strong (negative) correlation between sulphur and barite formation, (bottom right), while barium (top left) only shows a moderate (positive) correlation. However, when the larger formation variations are considered, the correlation between sulphur and barite becomes much less pronounced, and takes on a less linear nature, corresponding to a similarly large decrease in first order effects (also note that the decrease in first order effects is much larger than that of total order effects, which is reflected in the fact that the relation between sulphur and barite goes from being very linear, to a more complex dependency). On the other hand, when going from well to formation variations, the correlation between barium and barite becomes more pronounced, resulting in higher first (and total) order effects. Finally, the change in correlation also seems to explain why the first order effects of the other two components, calcium and chlorine, decrease when moving from well to formation variations: while for well variations they have minor to moderate correlation with barite, this correlation largely disappears at formation variations. These changes could also be due to the fact that uncertainties of other components start to play a larger role in the total solution equilibrium, and the interaction between components becomes more significant than simple first order (linear) relations when considering larger formation variation.

These results again show the importance of having a good estimate of the uncertainty bounds when performing uncertainty quantification, as the outcomes and potential conclusions drawn from them can change. More specifically, the results seems to indicate that when looking at small potential measurement errors, it is more important to limit the uncertainty in sulphur, as at small variation this seems to be the main contributor to uncertainty in barite formation, while for large possible uncertainties, it is better to limit the potential error in barium. Thus, when taking and analyzing samples, in the case of barite improving the accuracy of sulphur and barium (and to a lesser extend calcium and chlorine) should take precedence over other components.

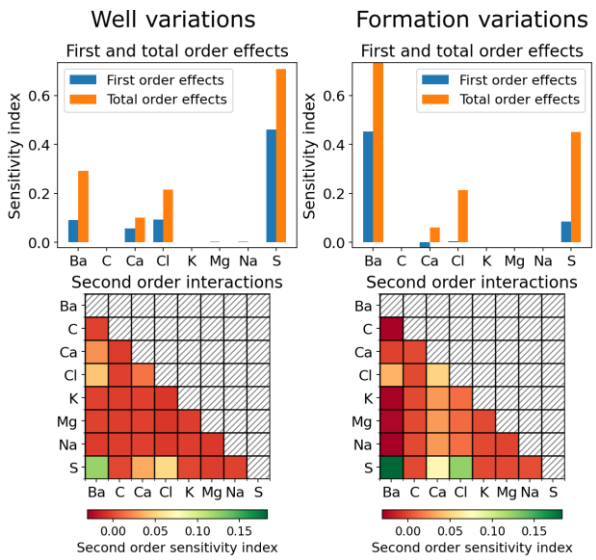

Figure 6. First and total order Sobol sensitivities (top) as well as second order sensitivities (bottom) for the eight elements included in the brine uncertainty quantification for the smaller well variations (left) and larger formation variations (right).

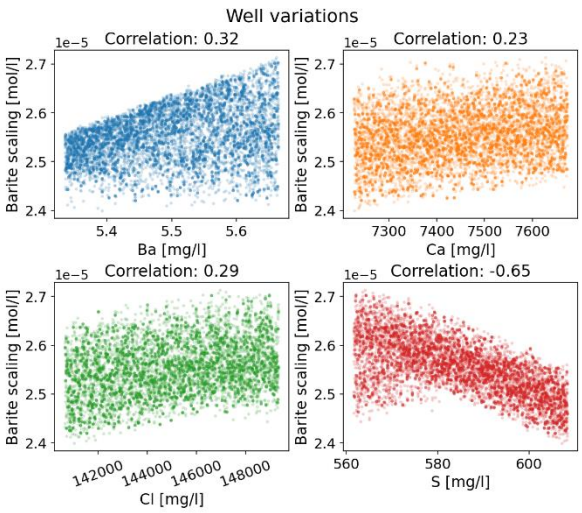

Figure 7. Correlation between the barite formation and component concentration of the four brine components with highest impact on barite formation for the well variations.

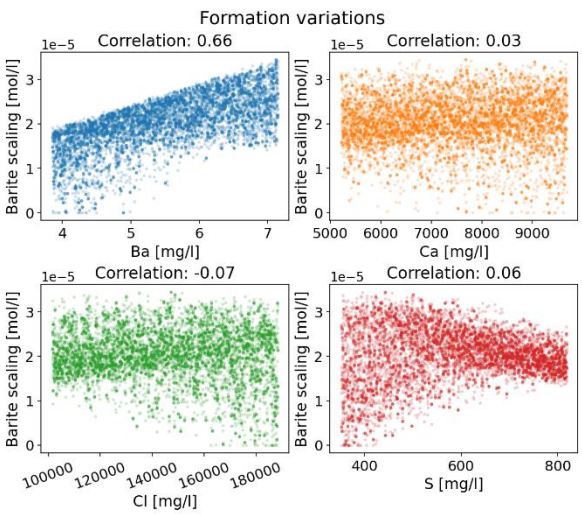

Figure 8. Correlation between the barite formation and component concentration of the four brine components with highest impact on barite formation for the well variations.

It should be noted here that Figure 7 shows a positive correlation between calcium and barite (top right plot), whereas the results in Section 3 indicated that calcium has a negative affect on barite scaling. While these results (which use the same model) seem to be conflicting, the discrepancy could be due to the uncertainty range of calcium, which is much larger in Figure 3 (2100-28000 mg/l) than in Figure 7 (7200-7700). Figure 8 already shows that going from a smaller range to a larger range can significantly impact the correlation, thus it could be the case that the when going to the even larger range of 2100-28000, the correlation could change further to become negative. In addition, the correlations shown in Figure 7 and Figure 8 include samples with variation in all eight components considered, while Figure 3 looks purely at variations of calcium itself. Thus, the effects of changes in other components could to an extent have obscured or skewed the individual effects of calcium concentrations on barite scaling.

4.2 Simplified heat exchanger results

In order to evaluate the effect of different temperatures on the formation amounts, distributions, and brine component sensitivities of barite scaling, the uncertainty quantification methods were applied to a simplified heat exchanger (see Section 3 for more details on this case). At five points in the heat exchanger, 10,000 samples were generated and the previous analyses were performed. In this case only the larger formation variation were considered. It should be noted that the five points considered were not coupled to each other when it comes to mass flow, meaning that any barite predicted to precipitate in point one is not removed from the input composition of point two. Thus, each point has the same exact input concentrations as the others.

Figure 9 shows the barite distributions for the five temperature-pressure combinations considered both as box plots and histograms. As can be seen, as the temperature changes, so do the barite distributions. While at higher temperatures, where the solubility of barite is high, there is a large amount of possible brine compositions that result in no precipitation, this slowly changes as the temperature drops and solubility decreases. Eventually, at the lower temperatures (50 degrees and below), barite is expected to precipitate regardless of the brine composition, although there is still a lot of variance in the potential amount of barite that precipitates. Looking at the nominal values for each points, we see that all of them predict barite scaling to occur. Comparing them to the mean of the distribution, the largest differences are found at the higher temperatures, with a maximum difference of 46% at the first point, which reduces at the last point to a difference of around 13%. These result are due to the large number of samples for which no barite scaling is predicted, which lowers the mean of the distribution significantly.

From the analysis it can also be seen that 70 °C seems to be the point around which the possibility of barite precipitation goes from a small chance of no precipitation at all, to certain precipitation no matter the brine composition. Thus, while the largest effects of uncertainties in absolute terms are found at lower temperatures, since they have broader ranges of potential scaling amount values associated with them, the uncertainty in whether scaling occurs or does not occur are found at higher temperatures (70 °C and above). When it comes to heat exchanger performance, these latter uncertainties are expected to play a larger role, as the difference between no precipitation at all to some precipitation has quite a large impact on the flow and heat transfer within the system (mainly due to a change in wall roughness and the addition of thermal resistance due to the barite deposition layer).

This latter uncertainty is also reflected in the Sobol sensitivities shown in Figure 10, where it can be seen that the largest changes in first, second, and total order effects occur between the points at which the temperatures are highest (although the relative order of contribution of the components remains constant throughout the entire heat exchanger). On the other hand, once the temperature is below 70 °C and barite is guaranteed to precipitate, the Sobol sensitivities are mostly constant. In addition, when looking at the correlations between the four components with highest impact on barite formation (Figure 11), the largest differences are also mostly seen in the first three points.

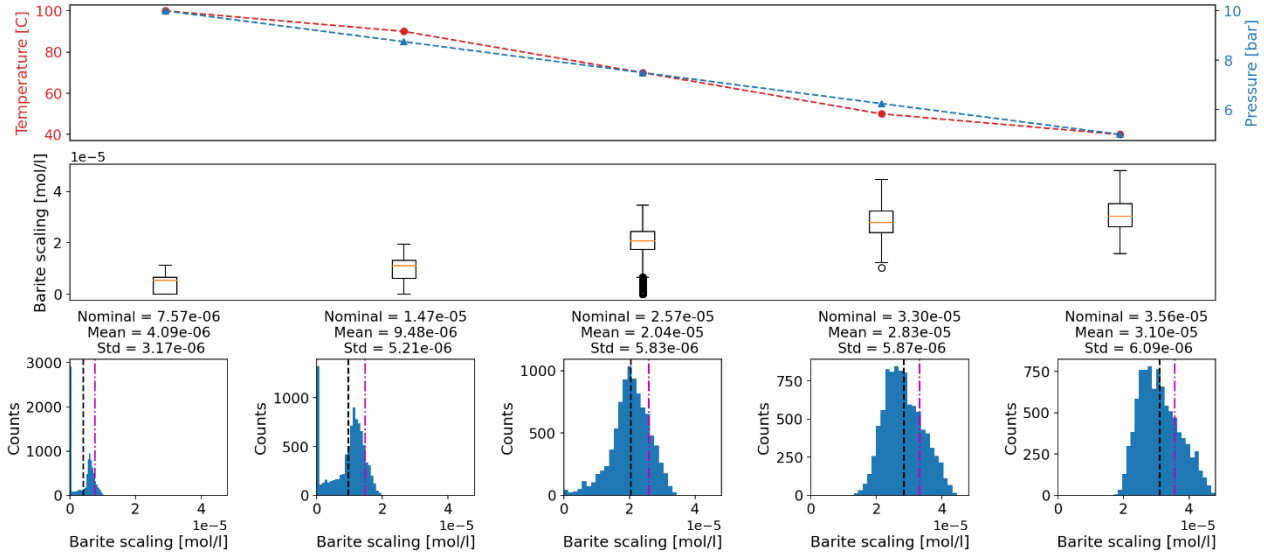

Figure 9. Barite scaling amounts calculated using PHREEQC at five temperature-pressure combination (top plot) plotted as box plots (middle plot), and histograms (bottom plots). Each combination contained the same amount of roughly 10,000 samples. Results plotted are those corresponding to the larger formation variations.

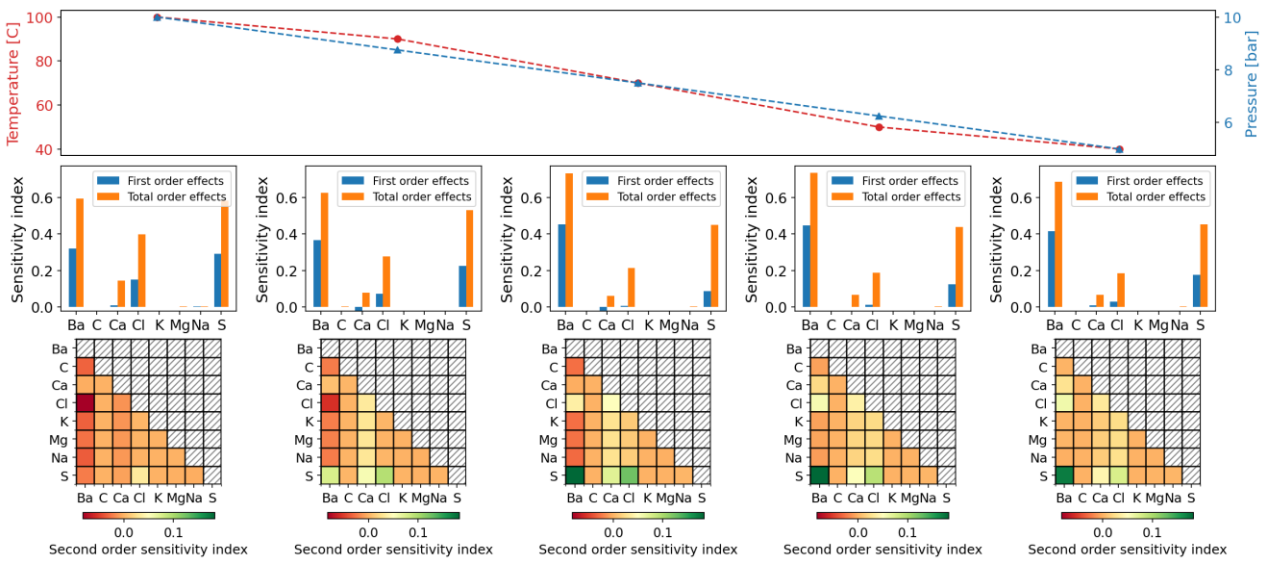

Figure 10. First and total order Sobol sensitivities (middle plots) as well as second order sensitivities (bottom plots) for the eight brine components included in the analysis at five different temperature-pressure combinations (top plot). Results plotted are those corresponding to the larger formation variations.

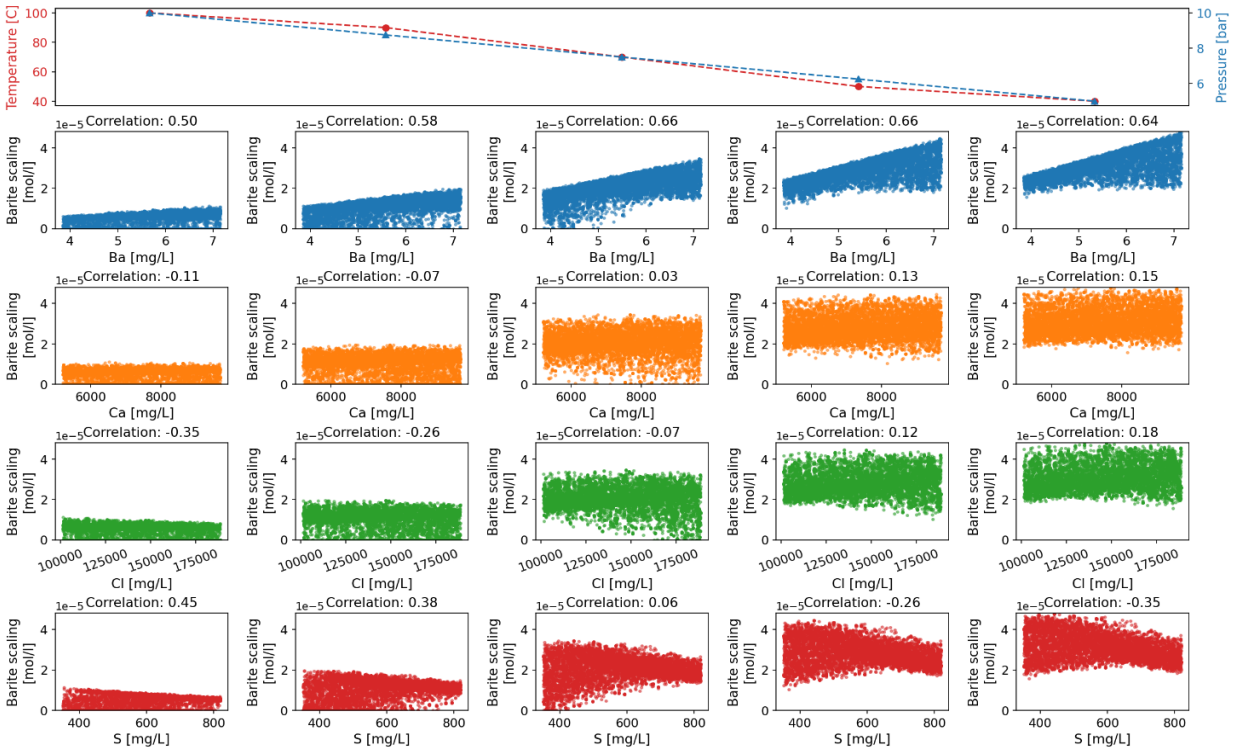

Figure 11. Correlation between the barite formation and component concentration of the four brine components with highest impact on barite formation (bottom four rows of plots; from top to bottom: barium, calcium, chlorine, and sulphur) at five different temperature-pressure combinations (top plot). Results plotted are those corresponding to the larger formation variations.

5. CONCLUSIONS

An automated workflow was made to perform uncertainty quantification analysis to assess the impact of fluid composition uncertainties on the mineral precipitation and scaling in geothermal systems. The workflow was initially demonstrated for barite precipitation based on two levels of uncertainties in chemical composition, smaller well level variations (obtaining multiple samples from a single well) and larger formation level variations (multiple samples from different wells in the same reservoir), and later applied to five points along a heat exchanger (only at formation variations). It was found that the mean barite amount considering uncertainties on the well level was higher (with a lower standard deviation) than the mean value in the formation level. In addition, the distribution of barite scaling amounts at the formation level was significantly more skewed towards lower values, even including a number of composition samples that lead to no scaling at all, while at the well level all samples predicted scaling to occur.

Based on the Sobol sensitivity analysis, the components most contributing to the variations in the saturation index were found to be sulphur and barium, where sulphur had the highest contribution at the well level, while barium had higher contributions on the formation level. In addition, for both levels, calcium and chlorine also had minor contributions, while the other four components considered (carbon, potassium, magnesium, sodium) were found to have no impact at all on the uncertainty of barite scaling based on the Sobol sensitivities.

In the simplified heat exchanger, the uncertainty distribution of barite precipitation at high temperatures were narrower (due to a lower probability of the precipitation to occur at higher temperature), but showed a greater difference between the distribution mean and nominal value (46% at the highest temperature versus 13% at the lowest temperature). As the temperature (and solubility of barite) decreases, the uncertainty bounds were increased and the skewness shifted towards a higher precipitation amount and at temperatures below 70 °C barite was predicted to precipitate no matter the brine composition. For the same heat exchanger case, Sobol sensitivity values changed mostly in the hot side of heat exchanger when uncertainty in brine composition still lead to samples with no barite precipitation. At lower temperatures, when barite scaling was predicted to occur for all samples, Sobol sensitivities remained fairly constant. Furthermore, throughout the heat exchanger, barium and sulphur always contributed most to the variations in barite precipitation amounts. Therefore, for the purpose of barite scaling prediction, it is important to improve the accuracy of these components when sampling and characterizing geothermal brines.

A generic workflow was developed which can also be applied to other minerals precipitation. The developed precipitation uncertainty quantification workflow will be calibrated with experimental measurement data and further integrated in an optimization toolbox with the

goal of finding the best filter design and operational conditions to minimize scaling potentials considering brine composition uncertainties.

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