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Evaporation Dynamics and NaCl Precipitation on Capillarity-Coupled Heterogeneous Porous Surfaces

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Abstract

Evaporation of saline water from porous media and associated salt deposition are important for many applications ranging from soil salinization to the protection of archaeological and civil structures. We investigate the effects of textural heterogeneity of porous media on evaporative salt precipitation patterns. Textural heterogeneity has been introduced into sand columns to form capillary-interacting porous domains comprised of different particle sizes. Several levels of textural contrasts were experimentally studied in terms of their effects on drying rates, salt precipitation patterns and drying front displacement rates. Results show that in contrast with evaporation of pure water, evaporation of saline solutions from media with textural contrasts exerted only a minor effect on evaporation rates. This difference is attributed to enhanced liquid transport within the highly porous precipitated salt on the surface. The presence of textural contrasts significantly enhanced preferential spread of salt precipitation on the surface. The enhancement is attributed to low surface water content of the coarse domain and larger spacing between remaining evaporating wet pore clusters in which greater local evaporation rates are accelerating salt precipitation. The study illustrates the interactions between the water characteristic curve, preferential drying front displacement and subsequent salt deposition patterns, as also seen in high resolution X-ray tomography used to verify the suggested mechanisms. The study provides new insights into mechanisms of saline water evaporation and the complex effects of porous media heterogeneity on salt precipitation patterns and dynamics; all critical ingredients for enhanced understanding of the salt distribution in and transport in soils.
1. Introduction

Saline water evaporation from porous media and salt precipitation affects water management, soil and groundwater salinization, and various other environmental processes [Jardine et al., 2007; Prasad et al., 2010; Xiao et al., 2011; Russo, 2013]. Enhancing our understanding of saline water evaporation processes is essential to predict locations and patterns of salt precipitation and the associated drying behaviour. Salt precipitation rates and deposition patterns are controlled by complex interactions between the transport properties of the porous medium, atmospheric conditions, and characteristics of the evaporating saline solution [Smits et al., 2012; Norouzi Rad et al., 2015; Jambhekar et al., 2015; Borgman et al., 2017; Vanderborght et al., 2017].

Evaporation from porous media has been the subject of active research in the past due to its central role in the hydrologic cycle and surface energy balance. More recently these efforts have been directed at investigating the effect of the changing climate patterns on the land-climate feedback processes [Fetzer et al., 2016; Tuttle and Salvucci 2016; Decker et al., 2017]. The evolution of evaporation, transpiration and precipitation rates in the changing climate will undoubtedly affect and be affected by the distribution and accumulation of salts in soils. Accurate formulation and prediction of evapotranspiration processes therefore relies on improved physical understanding of the mechanisms which govern salt effects on evaporation dynamics.

Basic mechanisms of pure water evaporation from initially saturated porous media are well understood and these are often described in terms of three different drying regimes. In the initial stage, the evaporation rate is relatively constant and liquid is supplied through capillary flow paths to the vaporization plane at the surface [Thiery et al., 2017] (the so-called stage-1 evaporation regime). This stage is followed by a transition period, where the drying rate
continuously decreases due to the disruption of the capillary pathways, into the final evaporation stage (stage-2 evaporation regime) where all liquid connections to the surface are disrupted and evaporation is dominated by vapour diffusion through the porous medium [Yiotis et al., 2007]. Despite loss of hydraulic connectivity to the surface, stage-2 evaporation rates from the subsurface can be as high as 50% of potential evaporation rates [Deol et al., 2014]. When salt is present in the evaporating solution, the evaporation process becomes more complex. Evaporation of saline solutions results in salt precipitation at the vaporization surface when the local solution concentration exceeds its solubility limit. Recent studies have hypothesised that due to the highly porous nature of the precipitating salt crust, the ensuing evaporation rate would be controlled by the liquid transport through the evolving salt layer [Sghaier and Prat, 2009; Eloukabi et al., 2013; Bergstad and Shokri, 2016; Shokri-Kuehni et al., 2017a]. Furthermore, during the early stages of evaporation, the rates of capillary flow through the precipitated salt layer meets the external evaporative demand and may result in a constant evaporation rate similar to the stage-1 evaporation regime in a pure water system. As evaporation proceeds transport through the salt layer is limited, possibly due to the salt precipitation gradually drying out, causing a reduction in the evaporation rate [Eloukabi et al., 2013; Shokri-Kuehni et al., 2017a]. The dynamics of the interacting salt precipitation layer and its influence on evaporation behaviour in porous media is dependent on the preferential precipitation patterns, which is governed by the presence of porous media heterogeneities. In this study, we focus on the effects of the presence of capillary-coupled textural contrasts on salt precipitation and evaporation dynamics.

Natural soils may contain various types of layers, textural contrasts and material interfaces that reflect depositional and scouring processes, refilling of cracks formed during freezing and thawing or swelling and shrinking cycles, and the interception of Aeolian dust and other local plant alterations. The presence of such porous media discontinuities induces spatially
abrupt changes in the capillarity and flow properties, and modify transport processes as reported in several studies [Pillai et al., 2009; Lehmann and Or, 2009; Nachshon et al., 2011; Veran-Tissoires et al., 2012; Bechtold et al., 2011, Bergstad et al., 2017]. In order to understand and describe salt transport and distribution in soils, it is essential to investigate how the presence of such heterogeneity influences the evaporative fluxes and salt precipitation dynamics. During evaporation from porous media containing abrupt textural contrasts (represented here as vertical inclusions of fine and coarse sand), the main vaporization plane remains anchored primarily at the surface of fine-textured porous media and is supplied by lateral capillary flow from the coarse domain [Lehmann and Or, 2009]. This lateral flow is driven by a capillary pressure gradient determined by differences in pore size distributions that drives liquid from the large pores in the coarse domain supplying evaporation from the surface of the fine domain (capillary pumping) [Lehmann and Or, 2009]. Consequently, the fine sand remains saturated long after the drying front (the interface between saturated and unsaturated porous media) propagates through the coarse region. A recent study by Bergstad et al. [2017] illustrated the unique role of textural heterogeneity in promoting preferential salt deposition during evaporation from porous media. Bergstad et al. [2017] have shown that despite the dominance of evaporation flux from the fine sand region, salt begins to precipitate preferentially over the coarse regions of the surface for low concentration salt solutions. This counterintuitive result is associated with preferential drying (air invasion) of coarse surface due to lower air entry pressure (as a result of larger pores in the coarse domain) that reduce the number of remaining evaporating pores. This in turn, causes locally higher per pore evaporation rates from these spatially distributed pores (see Bergstad et al. [2017] for detail).

Preferential salt precipitation patterns and salt crust expansion over surfaces of heterogeneous porous media depends on the pore size distributions and hydraulic properties of the respective
domains [Bergstad et al. 2017]. Motivated by the important impacts of the presence of heterogeneity on precipitation dynamics in porous media, we seek to quantify how capillary-coupled porous interfaces and their characteristics affect salt precipitation patterns and evaporation dynamics of saline solutions by performing a series of well-controlled drying experiments at the macro- and micro-scale.

2. Experimental Considerations

Evaporation experiments were conducted in an environmental chamber at constant temperature of 30°C and relative humidity (RH) of 30%. Following Bergstad et al. [2017], sand columns containing textural heterogeneity were packed, in cylindrical columns (70 mm in diameter and 70 mm in height) containing a radially symmetric fine-textured inclusion in the middle of a coarse-textured porous medium. The diameter of the fine inclusion (2.3 cm) was equal to a 1/3 of the column’s diameter (the lateral extent of evaporative water extraction from the coarse to the fine domain extends over the length of the coarse domain [Lehmann and Or, 2009]). Silica sand was separated by sieving into the following size ranges; 1250-1600 μm, 1000-1250 μm, 700-1000 μm, 500-700 μm and 300-500 μm. Six different textural contrasts were created by varying the particle size range of the fine (700-1000 μm, 500-700 μm and 300-500 μm) and coarse domains (1250-1600 μm and 1000-1250 μm).

All sand columns were saturated with 3 Molal solution of NaCl (note that the solubility limit of NaCl in water at 30°C is 6.14 M). Additional experiments were conducted using pure (deionized) water in sand columns containing similar textural contrasts for reference (sand grains were packed into a cylindrical column with 160 mm in height and 80 mm in diameter in this case). The water mass loss rates from the columns were recorded automatically every 5 minutes using digital balances, while an automatic imaging system photographed the surface of the columns every hour to record salt precipitation development. Customized
codes were developed in MATLAB following the procedure described in Shokri et al. [2008] to segment the recorded images into binary images where each pixel was assigned a value corresponding to either un-covered sand surface (black) or salt-covered sand surface (white) based on the grey level intensity. This enabled us to analyse the evolution of fractional surface coverage by salt precipitation at the surface of coarse and fine domain respectively.

In addition to the sand column experiments above, we studied details of the liquid phase distribution in the hydraulically coupled coarse and fine sand domains during evaporation using glass Hele-Shaw cells (100 mm height, 50 mm width and 4 mm thickness) with half of the cell filled with sand with particle size in the range of 1000-1250 μm, and the other half packed with fine sand (either 300-500 μm or 700-1000 μm). The experiments with the Hele-Shaw cells were conducted at the same environmental conditions and NaCl concentration as before. Images of the cells were recorded and segmented in MATLAB which was used to quantify the dynamics of the drying front displacement in sand packs in the presence of vertical textural discontinuity.

Direct and detailed observations of textural contrasts effects on pore-scale dynamics of liquid phase distribution and salt precipitation during evaporation were obtained from time-lapse X-ray micro computed tomography imaging. Two columns with 10 mm diameter and 20 mm height were packed with different textural contrast in the same arrangement as the macro-scale columns (and saturated with the same NaCl concentration), where the fine sand was 300-500μm in both cases and the coarse sand varied from 1000-1250μm and 1250-1600μm in the two cases. The experiments were carried out within a Nikon X-Tek 225kV/320kV customized bay (operating at 80 kV and 250 μA) housed in the Henry Moseley X-ray Imaging Facility of The University of Manchester. Each 3-D scan (24 scans taken every 60 minutes) was computationally reconstructed from 2000 radiographs. The evaporation experiments and image analysis were performed following the procedures detailed in
Bergstad et al [2017]. Additionally, Avizo Fire 9.2 (FEI, 2017) was used to visualize the segmented X-ray scans in 3D.

The capillary pressure-saturation relationship (also denoted as the soil water characteristic curve) of each sand domain with a well-defined particle size range was measured using a HYPROP device (Decagon Devices, USA). The parametric van Genuchten (VG) model [van Genuchten, 1980] was used to describe the experimental results with the VG parameters presented in Table 1. The obtained water retention curves are presented in Figure 1. The results presented are in good agreement with values reported in literature for sands of similar particle size distributions and porosity [Lehmann et al., 2008; Yanful et al., 2003].

The air entry value, $h_b$, was calculated using the equation given in Shokri and Salvucci [2011]:

$$h_b = \frac{1}{\alpha} \left[ \left( \frac{n}{n-1} \right)^{\frac{1}{n}} - \frac{1}{n} \left( 1 + \frac{n}{n-1} \right)^{\frac{1}{n}} \right]$$

(1)

where $\alpha$ and $n$ are the VG parameters of each sand. In this work we quantify the magnitude of the textural contrast between fine and coarse sand as $h_{bF}/h_{bC}$, where $h_{bF}$ and $h_{bC}$ are the air entry values of the fine and coarse sand, respectively. Consequently, as $h_{bF}/h_{bC}$ increases, the degree of textural contrast increases.
Figure 1. Relationships between capillary pressure and water content measured under hydrostatic conditions. The curves were fitted according to the van Genuchten model [1980] for the fine and coarse sands used in the evaporation experiments.

Table 1. Air entry pressure $h_b$, saturated and residual water content ($\theta_s$ and $\theta_r$, respectively) and shape parameters $n$ and $\alpha$, fitted to the van Genuchten model [1980] to describe the water retention curves of sand domains used in this study.

| Particle size range (μm) | $|h_b|$ (mm) | $\alpha$ (mm$^{-1}$) | $n$ (--) | $\theta_r$ (--) | $\theta_s$ (--) |
|--------------------------|-------------|----------------------|---------|----------------|----------------|
| 300-500                  | 165         | 0.00526              | 14.31   | 0.073          | 0.404          |
| 500-700                  | 123         | 0.00608              | 9.50    | 0.085          | 0.390          |
| 700-1000                 | 80          | 0.00870              | 8.86    | 0.106          | 0.438          |
| 1000-1250                | 60          | 0.01167              | 8.15    | 0.103          | 0.430          |
| 1250-1600                | 50          | 0.01340              | 7.55    | 0.085          | 0.370          |
3. Results and Discussions

3.1 Effect of textural contrasts on evaporation from porous media

Figure 2(a) illustrates that for the evaporation of water, the duration of stage-1 evaporation from heterogeneous sand packs increased with increasing levels of textural contrast (expressed in terms of the ratio of the fine to coarse air entry values: $h_b^F/h_b^C$). This result is in-line with the analysis presented in Lehmann and Or [2009]. Surprisingly, this correlation appears not to hold for saline water, as shown in Fig. 2(b). Our experimental data show no significant change in the duration of the constant evaporation rate period for all textural contrasts studied. This would appear to be due to the presence of salt precipitation at the surface. This is a counter-intuitive result, since the presence of such heterogeneities has momentous effects on the transport processes and phase distributions during evaporation [Lehmann and Or, 2009; Bechtold et al., 2011]. However, our results clearly show that despite the presence of increasingly pronounced heterogeneity, the evaporation dynamics is controlled by the properties of the precipitated salt at the evaporating surface.

In the initial stages of evaporation, the presence of salt in the evaporating solution causes the suppression of the saturated vapour pressure resulting in a lower evaporation rate compared to evaporation of pure water under similar conditions [Shokri-Kuehni et al., 2017b] (Figure 2). As the salt begins to precipitate and expand over the surface (active evaporation areas are converted to precipitation areas), the active evaporation surface is extended to the exterior of the precipitation layer and the evaporation rate is controlled by the pore-scale dynamics of the salt precipitation, which is considered next.
Figure 2. Evaporation rate from sand packs with different textural contrasts. The legend indicates the value of $h_b^F/h_b^C$ (indicating the ratio of air entry pressure for fine and coarse sand) saturated with (a) pure water and (b) 3M NaCl solution (inset shows the corresponding evaporative mass loss curves).

3.2 Pore-scale salt precipitation dynamics

Investigation of the formation and growth of the precipitated salt at the surface of evaporating sand columns at high spatial and temporal resolution was made possible by means of time-lapse X-ray micro computed tomography (CT). The X-ray images reveal that the precipitated salt at the surface goes through a complex dynamics and evolution which may influence the evaporation behaviour.
Figure 3(a) shows an example of the evolution of the precipitated salt around a sand grain at the surface of the coarse domain in a heterogeneous sand pack with $h^F_0/h^C_0=3.30$. The time sequence illustrates how the growth of the salt results in contact with neighbouring precipitation (6 hours) which ultimately result in significant deformation of the shape and distribution of the precipitated salt (8 hours). This suggests that solute redistribution within the precipitated salt has caused dissolution and subsequent re-growth of crystals. Such deformations, or re-structuring, of precipitated salt was observed regularly throughout the X-ray experiment (for both columns with different textural contrast). This direct observation confirms that the salt precipitation comprises liquid phase which is regularly transported within the porous precipitated salt network at the surface, causing dissolution and re-crystallization. Figure 3 (b) shows salt precipitation formed around another grain at the same surface, and illustrates the highly porous structure of the salt, and that the porosity of the precipitated salt continuously changes during drying. Shokri-Kuehni et al. [2017a] recently illustrated that precipitated salt at the surface is a highly evolving porous structure, which facilitates a capillary supply of the evaporating solution from the porous medium to the external surface (acting as a “capillary conductor”). The pore-scale results (Figure 3) verify the evolving porous structure of precipitated salt during evaporation from porous media.

The pore-scale observations presented in Figure 3 could be considered as a first step toward further pore-scale quantifications specifically targeting the link between the complex evolving geometry of precipitated salt and the dynamics of evaporation. In order to describe the drying process in the presence of salt (Figure 2b), it is necessary to take into account such dynamics. Bergstad and Shokri [2016] reported similar evaporation curves to Figure 2 from sand packs with different mixed wettability condition saturated with NaCl solution. They showed that while the presence of hydrophobic grains does reduce capillarity (and subsequently the duration of stage-1 evaporation of pure water), the capillary pumping effect
of wet salt precipitation at active surface sites support the evaporative demand which results in negligible effect of mixed wettability on the overall evaporative mass loss and duration of stage-1. These results suggest that the formation of precipitated porous salt on the surface of heterogeneous sand packs supplying the evaporative demand introduces an additional level of control on the resulting evaporation behaviour and that in so doing reduces the dependency of drying dynamics on the porous media heterogeneities. This is an important consideration for future modelling approaches and predictions of saline water evaporation dynamics, with significant impacts on hydrological applications and water management.

Figure 3. Sequences showing two regions of interest taken from virtual horizontal X-ray CT cross sections recorded over time illustrating examples of typical salt precipitation formed around two separate sand grains at different locations on the surface of the coarse sand after
(a) 5, 6 and 8 hours of evaporation and (b) 10 and 24 hours of evaporation, from the evaporating column with $h^F_b / h^C_b = 3.30$.

### 3.3 Macro-scale salt precipitation patterns influenced by the textural contrast

The recorded images of evaporating heterogeneous sand surfaces were segmented to quantify the surface fraction covered by precipitated salt for the coarse and fine domains. This information enabled the assessment of the role of textural discontinuity on salt precipitation patterns, as shown in Figure 4. The results in Figure 4 correspond to salt precipitation patterns observed 1 and 6 days from the onset of the evaporation experiment. Results suggest that with the decrease in the strength of the textural contrast between fine and coarse sand (i.e. lower ratios of $h^F_b / h^C_b$), the salt precipitation on the coarse surface is reduced for the same initial salt concentration (similar in all sand columns). In contrast, a larger fraction of the fine domain surface was covered by the precipitating salt for lower strength of textural contrasts.

Furthermore, the results show that the onset of salt precipitation on the fine surface was strongly influenced by the difference between the air entry pressure of fine and coarse sand $\Delta h_b = h^F_b - h^C_b$. When the textural contrast between fine and coarse domain increases, the fine surface remains saturated for a longer elapsed time, reflected in larger values of $\Delta h_b$ [Lehmann and Or, 2009]. These considerations may suggest that the onset of precipitation on the fine surface is strongly correlated to the air entry pressure of this domain, which would explain the observed delay in the onset of salt precipitation for high textural contrasts (Figure 4 (b)).

For a given initial salt solution concentration, and for similar external conditions, salt precipitates first where the local concentration at the phase change plane exceeds the solubility limit. It follows that preferential salt precipitation is strongly influenced by higher
local evaporation flux for a given porous medium surface. In a heterogeneous sand column, the coarse domain is preferentially invaded by air, resulting in formation of disconnected liquid clusters separated by dry sand on the surface. The evaporation flux from the few remaining liquid filled pores is expected to be higher than for a same sized pore on the nearly saturated fine surface [Shahraeeni et al., 2012]. Consequently, we expect early onset of salt precipitation over the coarse surface [Bergstad et al., 2017]. The fewer of these pores on the surface (i.e. the greater distance between disconnected liquid clusters on a surface), the higher the evaporation flux is per pore [Fabrikant, 1985; Shahraeeni et al., 2012; Aminzadeh and Or, 2017]. The observed precipitation patterns (Figure 4) suggests that, since a higher textural contrast results in earlier and faster salt precipitation on coarse surface, reflecting a higher evaporation flux, the spacing between (or number of) wet pores after air invasion into the coarse surface must have increased with textural contrast (which is discussed next).
Figure 4. (a) The fraction of surface covered by precipitated salt in coarse and fine sand after 1 and 6 days from the start of the experiments, respectively. The inset shows the onset of precipitation on the surface of fine sand as a function of the difference between air entry pressure of fine and coarse sand $\Delta h_b$. (b) The surface of columns with $h_b^F/h_b^C$ equal to 1.33 and 3.30 after 1 and 6 days after the onset of the evaporation experiments.
3.4 Textural contrast effects on preferential capillary flow and salt precipitation

Two sets of evaporation experiments were conducted using Hele-Shaw cells to investigate details of the drying front (defined as the interface between saturated and partially wet zones) depth and velocity as it recedes through the fine and coarse sand domains. The magnitudes of the textural contrasts in these two sand packs were \( \frac{h_b^F}{h_b^C} = 2.75 \) and 1.33 (note that the coarse sand domain was identical in both sand packs and the textural contrasts were varied by changing the particle size of fine sand domain). The results are presented in Figure 5. As illustrated in other studies [Lehmann and Or, 2009; Bergstad et al., 2017], the drying front is formed preferentially in the coarse domain first due to the presence of larger pores (lower air entry pressure). During pure water evaporation, the fine domain remains saturated until the drying front depth in the coarse domain exceeds the characteristic length \( \Delta h_b = h_b^F - h_b^C \) [Lehmann and Or, 2009], marking the onset of air invasion and formation of a front in the fine domain.

Our results confirm that for a large textural contrast, the fine domain remains saturated for a longer time due to the greater capillary pressure differences between the fine and coarse domain (and the ability to extract water from larger depths in the coarse domain). Based on the air entry values obtained from the water retention curves (Figure 1), the characteristic length for the two systems was found to be 105 mm and 20 mm for \( \frac{h_b^F}{h_b^C} \) equal to 2.75 and 1.33, respectively. However, Figure 5 (a) indicates that desaturation of the fine surface starts when the depth of the drying front in the corresponding coarse domain was \( \sim 70 \) mm and 17 mm (33% and 15% shorter than the characteristic length for pure water). This is due to the effects of the de-saturating salt precipitation layer on the continuity of the liquid phase to the surface, and the corresponding evaporation dynamics discussed above, resulting in premature decoupling of the drying front. This finding is relevant to practical applications because it
suggests that the presence of salt may cause the disruption of the essential liquid supply from saturated zone to the soil surface affecting vegetation.

**Figure 5.** (a) The evolution of mean drying front depth; (b) the velocity of the drying front in the coarse and fine sand domains for two columns with different textural contrasts with values of $h_b^f/h_b^c = 2.75$ and $1.33$. The x-axis in (b), $M (-)$, represents the cumulative water mass loss by evaporation as a fraction of the final mass loss measured; (c) the segmented images (black = saturated region, white = unsaturated region) of the heterogeneous Hele-Shaw cells 2.5 days from the onset of the evaporation experiments. The coarse sand was identical in both experiments with the particle size range of 1000-1250 μm, however, the fine sands with
particle size ranges of 300-500 μm was used in $h_b^{F}/h_b^{C} = 2.75$ and 700-1000 μm in $h_b^{F}/h_b^{C} = 1.33$. The estimated pressures at the column surfaces $P_{\text{High}}$ and $P_{\text{Low}}$ with $h_b$ the air entry pressure and $L$ the drying front depth; and (d) the water retention curve for the coarse sand which confirms the presence of lower water content at the surface of the coarse domain in the column with the higher textural contrast (due to higher capillary pressure).

The preferential displacement of the drying front in the coarse domain, influenced by the degree of textural contrast, also affects the preferential deposition of salt at the surface. The pressure at the drying front in the coarse domain might be approximated as the air entry value of the coarse sand $h_b^{C}$ (assuming near-hydrostatic conditions as described in Shokri et al., 2008). Therefore, the pressure head at the surface can be estimated as the summation of the air entry value and gravitational head (i.e. the depth of the drying front). Under the same cumulative mass loss, the drying front is deeper in the coarse domain for higher textural contrast as a result of the faster moving front (greater capillary gradient), and the pressure at the surface is higher (more negative) (Figure 5). According to the water retention curve, higher surface pressure corresponds to the lower water content at the surface (Figure 5d).

Less water at the surface may suggest the presence of fewer liquid patches with greater spacing supporting the evaporative demand. As already shown in Bergstad et al. [2017], greater spacing between liquid patches at the surface increases the evaporation per pore which eventually results in earlier onset of salt precipitation at the surface and consequently faster spread of salt precipitation on the coarse surface as the textural contrast increases. This explains the result observed in Figure 4. Understanding the detailed mechanisms which govern preferential salt deposition in the more representative heterogeneous porous media may aid the development of effective strategies for removing salts from the critical root zones and reclaiming salinized fields [Berezniak et al., 2017].
A closer look at the effect of textural contrast strength on preferential salt precipitation patterns was obtained from time-lapse X-ray micro-tomography. This enabled us to directly observe and verify some of the mechanisms proposed above.

3.5. Pore-scale quantification of salt precipitation in heterogeneous porous media

The pore-scale images obtained from the X-ray tomography experiments were analysed in order to obtain the evolution of the mass of salt precipitation at the surface of coarse and fine domain for two textural contrasts, $h_b^{\text{F}}/h_b^{\text{C}} = 3.30$ and 2.75. As observed in the Hele-Shaw experiments, the majority of the evaporative mass loss is supplied from the coarse region due to the lower resistance for air invasion in larger pores (Figure S1) [Bergstad et al., 2017]. Our X-ray results further illustrate the same development of the salt precipitation patterns as observed in the macro-scale evaporation experiments (Figure 6a). Salt precipitates first on the coarse surface of the column with the higher textural contrast, and obtains a much larger amount of salt precipitation throughout the evaporation process. Conversely, the onset of precipitation on the fine surface is delayed compared to the column with the lower textural contrast. In both X-ray experiments the majority of the total cumulative salt precipitated above the coarse domain of the heterogeneous surface.

By quantifying the saturation of the liquid phase in each horizontal 2D slice of the X-ray CT scans, we could determine the evolution of the average water content at the coarse surface as influenced by textural contrast. Figure 6 (b) shows that, for the same mass loss from the coarse domain, the surface water content was consistently lower for the column with the greater textural contrast. This result confirms the analysis presented in Figure 5, and supports the proposed mechanism responsible for the observed salt precipitation patterns; the magnitude of the textural contrasts influences the preferential movement of the drying front in the coarse domain which affects the pressure at the surface. This results in a lower surface
water content which corresponds to an increase in the evaporative flux from individual active pores supplying the evaporative demand. Ultimately this causes earlier onset and accelerated growth of salt precipitation (Figure 6 (c) and Figure 4).

**Figure 6.** Evolution of (a) the mass of salt precipitation at the surface of coarse and fine domain and (b) surface water content of the coarse domain, in columns with different textural contrast of $h_b^{F}/h_b^{C} = 3.30$ and 2.75 obtained from X-ray CT experiments. (c) 3D volume rendering of the heterogeneous sand packs from Avizo Fire 9.2 (FEI, 2017) after 3 and 15 hrs of evaporation showing the salt distribution (white) at the surface.

**4. Summary and Conclusions**
Evaporation from texturally heterogeneous porous media saturated with NaCl solution with different degrees of textural contrast was investigated under constant external conditions. The evaporation curves and precipitation patterns were recorded for six different textural contrasts between coarse and fine sand distinguished by their air entry pressure ratios $h_b^F/h_b^C$. An additional set of evaporation experiments was conducted using Hele-Shaw cells to investigate details of preferential front displacement and potential consequences on salt precipitation patterns. Finally time lapse X-ray computed tomography was used to confirm the obtained results at the pore-scale. Results show that, for pure water, an increase in textural contrast in porous media results in prolongation of the stage-1 of evaporation. However, if saturated with NaCl solution, the textural contrast had no noticeable effect on the duration of the constant evaporation period. We attribute this unintuitive result to the formation of precipitated porous salt on the surface, which controls the evaporation process [Sghaier and Prat, 2009; Eloukabi et al., 2013; Bergstad and Shokri, 2016; Shokri-Kuehni et al., 2017a,b]. By means of X-ray tomography we have demonstrated the highly porous and dynamic nature of salt precipitation through direct pore-scale observations.

The contrast between the hydraulic properties of the fine and coarse sand domain influence the velocity of the drying front in the coarse domain, which in turn determines the pressure and water distribution at the coarse surface. The local distribution of wet pores impacts the evaporation flux and subsequently the salt concentration in the solution. We illustrate that increasing the textural contrast between interacting coarse and fine domains results in earlier and faster salt precipitation on the coarse domain, but delayed precipitation on the fine surface. This behaviour was attributed to the higher negative pressure at the surface of the coarse domain (the presence of less water at the surface) due to the faster receding drying front in the coarse domain of the heterogeneous sand pack as the textural contrast increases. This process leads to more evaporation per pore on the surface of the coarse domain.
Shahraeeni et al., 2012; Aminzadeh and Or, 2017] thus earlier and faster precipitation
[ Bergstad et al, 2017]. Furthermore, we show that the onset of precipitation on fine surface is
related to the magnitude of the difference between the air entry pressure of the coarse and
fine sand domain $\Delta h_b$.

The results reported here are important for improving physical understanding and accuracy of
modelling approaches for management of saline soils, as in most natural and agricultural
cases there exists domains with textural contrasts of different magnitudes, and the findings
presented here will be instrumental to identify and describe the areas where precipitation
occurs with the associated evaporation behaviour.

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