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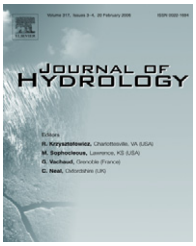

Influence of transport parameters and chemical properties of the sediment in experiments to measure reactive transport in seawater intrusion

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Summary A study of seawater intrusion under controlled laboratory conditions using multicomponent heterovalent chromatography is presented. The aim was to understand the influence of several variables on hydrogeochemical transport, especially on gypsum precipitation and cationic exchange. In addition, the study aimed to provide experimental data on how water composition changes during simulated seawater intrusion.

The experimental results show that dispersion modifies the shape of the elution curves for the different solutes for processes such as cation exchange and precipitation–dissolution. The maxima and minima of these curves are very smooth, and entirely absent in several instances. Altering the cation exchange capacity of the sediment produced changes in the height of the calcium peak and in the maxima and minima of magnesium. In several experiments the high concentrations of calcium and sulfate during the first stages of the intrusion induced gypsum precipitation. The subsequent dissolution of the gypsum raised the concentration of sulfate higher than that in seawater. Saturation indices (SI) for gypsum in the samples collected were calculated with PHREEQC (version 2). Gypsum SI values are in agreement with experimental observations.

Piper diagrams demonstrate that the experimental variables of transport parameters and cation exchange capacity (CEC) strongly influence the hydrochemistry of seawater intrusion. The experimental data deviate substantially from the theoretical freshwater–seawater mixing line, and the shape of the pathway between the end members depends on the experimental conditions.

The experimental data obtained during column experiments and the physical and chemical parameters determined in each experiment can be used in the validation of

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multicomponent transport models. These hydrogeochemical models may aid in the interpretation of field data.

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Introduction

Several studies have used multicomponent cation exchange approaches to describe reactive transport in porous media (Callahan et al., 2000 provides a detailed listing). Of these, Valocchi et al. (1981a, 1981b) developed a fundamental analysis of cation exchange process, or ion chromatography, in which treated wastewater is injected into a salinized aquifer. They subsequently proposed a model for this process. Later, Appelo and Willemsen (1987) and Appelo (1994, 1996) discussed the cation exchange principles involved in salinization and derived specific equations for multicomponent problems in this context.

Seawater intrusion is a particular case of multicomponent reactive transport requiring a more complex analysis because of the large number of solutes at very high concentration (ionic strength around 0.7) and the different types of reactions involved, such as cation exchange and precipitation—dissolution. However, a chromatographic sequence of salinization has not been clearly observed in seawater intrusion field-scale experiments, even though the $CaCl_2$—water type is an ubiquitous indicator of salt water upconing in coastal areas (Appelo and Postma, 2005). Instead, information on the sequence of compositions for saltwater intrusions is obtained from columns or small-scale field experiments (Appelo et al., 1990; Gomis et al., 1997; Van Breukelen et al., 1998; Andersen et al., 2005), or from multicomponent models of reactive transport (Gomis et al., 1996; Parkhurst and Appelo, 1999).

With regard to laboratory research, Beekman and Appelo (1990) performed experiments in columns 7.5 cm long in which they displaced fresh water with seawater that had been diluted 1:1 with distilled water. The authors modeled the chromatographic results by taking into account mainly the cationic exchange processes.

Gomis et al. (1997) carried out displacement experiments using seawater in a sediment column 100 cm in length. In these experiments, some of the samples taken at the exit of the column during the initial stages of the intrusion were supersaturated with gypsum, which precipitated into the sampling vials, even though the sediment did not initially contain gypsum. The chromatographic experimental results showed the signs of gypsum precipitation in the sediment column: comparison of the sulfate curve to that of chloride (a conservative solute) revealed a considerable delay between the two solutes. The high concentrations of calcium and sulfate induce the precipitation of gypsum, and its subsequent dissolution increases the concentration of sulfate to a level higher than its concentration in seawater, as seen in the experimental results.

Moreover, applying a multicomponent reactive model to the experimental data (Gomis-Yagües et al., 2000) has shown that gypsum precipitation during the early stages of the advance of the seawater front causes a decrease in sulfate concentration with respect to the conservative mixture of freshwater and seawater under conditions that are unfavorable to sulfate reduction by bacteria. Thus, gypsum precipitation may explain the non-conservative behavior of sulfate during seawater intrusion, in addition to bacterial sulfate reduction by organic matter.

Although an obvious decrease in sulfate concentration has been observed in previous field studies (Stuyfzand, 1992; Bocanegra et al., 1992; Custodio, 1992; Fidelibus and Tulipano, 1996), authors have either given no explanation for the decrease or have attributed it to degradation of organic matter by sulfate-reducing bacteria, because field samples were not saturated with respect to gypsum. In recent years, the hypothesis of gypsum precipitation during seawater intrusion, first described in Gomis-Yagües et al. (2000), has increasingly been cited (Slomp and Van Cappellen, 2004). It is also discussed as a possibility in field studies on seawater intrusion phenomena (Pulido-Leboeuf, 2004). This author has argued that gypsum saturation indices are generally higher than the theoretical conservative mixing line (especially for mixtures containing less than 40% seawater). This suggests the existence of a sulfate source other than seawater. Such a source could be the dissolution of gypsum present in the metapelitic strata; in this case, gypsum need not be very abundant because saturation is not reached. The possibility of sulfate reduction or precipitation in samples containing less than 5% seawater or more than 40% seawater cannot be ignored, since these samples have saturation indices well below those calculated for conservative mixing.

Therefore, it is important to understand the chemical reactions such as gypsum precipitation and cationic exchange that affect groundwater quality during seawater intrusion. In these processes, the behavior of the chemical species depends on several factors, including differences in selectivity coefficients, the value of the cation exchange capacity of the sediment, contrasts between the chemical composition of the injected fluid and the fluid initially resident in the pores of the sediment, and dispersion (Lambrakis, 2006).

In several studies the results have proven insensitive to the dispersion characteristics of the column — i.e., the precise value of the Péclet number — and the shape of fronts was found to be essentially unaffected by dispersion (Vulava et al., 2002). Similarly Petales and Lambrakis (2006) found that small variations in dispersion did not significantly affect the chemical composition of the groundwater across the simulated flow path, as shown in related studies (Lambrakis and Kallergis, 2000).

Therefore, the aim of this article was to investigate multicomponent heterovalent chromatography for the process of seawater intrusion as modeled under controlled laboratory conditions. We sought to understand the influence of several variables, such as dispersion and CEC, on hydrogeochemical transport, especially on gypsum precipitation and cationic exchange. We also wished to collect experimental data on how the composition of seawater changes during the intrusion process. These data and the physical and chemical parameters determined in each experiment can be used in the validation of multicomponent transport models.

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