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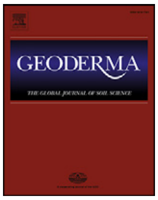

To which extent do rain interruption periods affect colloid retention in macroporous soils?

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ABSTRACT

Colloid retention in soils has mainly been studied in model homogeneous porous media, and during single irrigation events. However, in the field, colloidal suspensions flow through a small number of preferential flow paths, and the soil experiences succession of rainfalls, interrupted by dry periods. We therefore performed series of successive rainfalls (21 mm h^{-1}) on three decimetric undisturbed luvisol E-horizon cores to systematically study the impact of the rainfall interruption duration (RID) on retention. Successive rains were doped with 0.5 μm fluorescent microspheres of different colors. Microsphere release at the base of the soil column was monitored by flow cytometry. When preferential macropore flow was preponderant, increasing the RID from 5 to 1200 h led to the retention of up to an additional 27% of the microspheres rained onto the soil. This increased retention was ascribed to a higher absorption of the colloidal suspension into the macropore walls for longer RIDs, when water redistribution far from the macropores was more important. When matrix flow was preponderant, RID effect on retention was overwhelmed and massive microspheres retention was observed. These results are useful to provide guidelines for the safe use of reclaimed water in agriculture, and to estimate the importance of clay translocation (a colloid transfer based soil formation mechanism).

1. Introduction

Groundwater quality can be affected by the arrival of both dissolved and suspended contaminants. Harmful colloids (e.g. viruses, bacteria, man-made nanoparticles) conveyed by infiltrating water can make their way through the soil and reach the water table (Gerba and Smith, 2005). Harmless colloidal-sized soil constituents can carry adsorbed contaminants that would otherwise have a low mobility (De Jonge et al., 2004; DeNovio et al., 2004; McCarthy and Zachara, 1989). Also, these autochthonous soil colloids are actors of soil formation, as their mobilization from an upper soil layer, their transport to — and their retention in — a lower layer is a widespread soil formation process known as clay translocation or lessivage (Mackeague and Arnaud, 1969; Bockheim and Gennadiyev, 2000).

The mechanisms through which colloidal particles are mobilized, transported and retained in the soil have been the focus of a great amount of research. Colloid retention has been extensively studied in homogeneous porous media of e.g. glass beads and sand. Mechanisms (and factors) that affect colloid retention in such porous media have been identified and included into colloid transport models (Bradford et al., 2003, 2014; Wan and Tokunaga, 1997). Retention was comparatively poorly studied in undisturbed soils, where attention mostly focused on autochthonous soil colloid mobilization (Cornu et al., 2014; Mohanty et al., 2014, 2015b; Ryan et al., 1998). Although the retention mechanisms identified in model porous media may also be at work in undisturbed soils, their relative importance is probably different (Jacobs, 2007; Jacobsen et al., 1997; Mishurov et al., 2008).

In undisturbed soils, depending on initial and boundary conditions, gravity-driven water transport can occur through a small number of preferential flow paths (e.g. earthworm burrows, space left by decayed roots, cracks). In these macropores (that have diameters above about 300 to 500 μm) the water bypasses most of the soil porosity (Gerke, 2006; Jarvis, 2007). A few phenomenological studies highlighted the role of macropores on colloid retention (Burkhardt et al., 2008; Cey and Rudolph, 2009; Cumbie and McKay, 1999; Driese and McKay, 2004; Nielsen et al., 2011; Passmore et al., 2010). They showed that colloids dispersed in input water were mostly retained (i) in the first few centimeters below the soil surface, (ii) onto the walls of the macropores through which water flow occurred and (iii) in the soil matrix next to these macropores. However, the factors controlling this pattern as well as the associated mechanisms have not been thoroughly investigated yet.

Abbreviations: RID, rain interruption duration; SC, autochthonous soil colloids; FCM, flow cytometry; AWI, air water interface.

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Most of the studies on colloid retention and mobilization have focused on single events, although in natural conditions, wetting and drying cycles follow one another. Nonetheless, some studies showed that mobilization of autochthonous soil colloids (SC) and of previously deposited allochthonous colloids were highly affected by these cycles (El-Farhan et al., 2000; Majdalani et al., 2008; Michel et al., 2010; Mohanty et al., 2015a, 2015b; Schelde et al., 2002; Zhuang et al., 2007). It was shown that the duration of the dry period before a rainfall – because it controlled the initial water content of the active macropores – affected significantly the leaching of SC. We propose that during successive rainfall events, the rain interruption duration (RID) may also control (i) the amount of particles retained in the soil and (ii) their retention pattern in the soil porosity.

Based on this premises, the aims of this paper was to assess and quantify the effect of successive rainfall events separated by rain interruptions of increasing duration on colloid retention in undisturbed soil columns. Rainfall experiments spiked with fluorescent microspheres were conducted. A flow cytometry-based protocol allowed us to (i) determine the state of the microspheres (freely suspended or adsorbed onto SC), and (ii) to quantify the remobilization –during a rainfall event – of microspheres retained during previous rainfall events. We proposed a conceptual model to explain the variation of microsphere retention observed when the duration of the rain interruption increased from 5 to 1200 h. The importance of the proposed mechanism — compared to other mechanisms that affect particle retention — was discussed, together with its environmental consequences.

2. Materials and methods

2.1. Undisturbed soil core sampling

The sampled soil horizon was the E horizon (at a depth of 35 cm) of a luvisol (IUSS Working Group WRB., 2006) already extensively studied by Cornu et al. (2014) and sampled in a cultivated field next to La Pilotière (France, 47°49′50.9″ N, 0°36′52.7″ E). After removing the A-horizon, three undisturbed soil cores (A, B and C) were taken by gently pushing polyvinylchloride (PVC) cylinders (internal diameter 12 cm, height 15 cm). The soil principal characteristics determined by Quénard (2011) and Cornu et al. (2014) are summarized in the Supplementary Material, Table S1. To protect their bottom part from mechanical perturbation, the cores were positioned onto a PVC screen (holes of 1.5 mm every 2.5 mm) and inserted into an annular sample holder that was glued onto the pipe (Sammartino et al., 2012).

2.2. Experimental setup

A rainfall simulator made of a tank terminated at the bottom end by 57 hypodermic needles (25 gauge), and connected to an impulse pump was used to control rainfall intensity and duration. During a rainfall, the weight of the core and of the outflowing water were recorded every 10 s. The amount of water that accumulated inside the cores, V_{acc} (mm), and the hydrographs (water flow rate as a function of time) were determined from these measurements. Effluents were collected throughout the experiment, about every three minutes during the transient flow regime, and five minutes when the permanent flow regime was reached. They contained both SC and microspheres. Microsphere concentration was determined by flow cytometry (FCM, see next section). The SC concentration was determined by light extinction (LE) at 400 nm using a Cary 50 UV–Visible spectrophotometer. The microsphere contribution to LE was computed from the microsphere concentration determined by FCM and a calibration curve between microsphere concentration and LE. This contribution was negligible compared to that of SC when the SC concentration was high. When it was not, essentially during the permanent flow regime, the effluent LE was corrected from the microsphere contribution using the additive property of Beer's law, Effluent conductivity was measured using a standard electrode.

2.3. Experimental procedure

To test the effect of rain interruption periods on microsphere retention, the soil cores were submitted to seven successive rainfall events separated by rain interruptions of increasing duration. All rainfalls lasted one hour. The rain interruption durations (RID) were: 5, 23, 48, 118, 335, 644 and 1200 h for core A and B, and 5, 23, 48, 148.5, 306, 670 and 1200 h for core C. To prevent modifications of the macropore network that may result from the formation of drying cracks at the longest RIDs, the columns were loosely wrapped in plastic bags and left at room temperature (22 °C) during rain interruptions. The absence of cracks on the surface, or at the PVC-soil interface was systematically checked visually before every rainfall. The water content remained close to the water saturation all through the experiment. Drainage was free at the bottom of the columns at all time. The rainfall intensity was 21 ± 1 mm h^{-1}. The rainwater contained fluorescent 0.5 μm diameter carboxylated microspheres in a 5×10^{-5} M $CaCl_2$ background solution of pH close to 5.5. At this pH the carboxylate groups were deprotonated, and the microspheres were negatively charged (Rogers et al., 2005). Their concentration was about 2×10^{10} microspheres per liter (about 5×10^9 microspheres were brought to the soil cores during each rain). To quantify the remobilization – during a rainfall event – of microspheres retained during the two previous rainfall events, three sets of microspheres that differed only in the nature of their fluorescent dye were used successively in the following order: blue, purple, red, blue, purple, red and blue. We checked that the different dyes did not affect the surface and transport properties of these internally dyed microspheres: (i) their electrophoretic mobility measured at 20 °C in ultrapure water was similar (Supplementary Material, Table S2), and (ii) when present simultaneously in the rainwater during a preliminary rainfall, microsphere recovery was similar within experimental error for all three microsphere types (Supplementary Material Fig. S1). The experimental setup in contact with the microsphere suspension was thoroughly rinsed between each rainfall and the absence of contamination of rainwater by microspheres from a previous rainfall checked systematically.

2.4. Flow cytometry measurements and analysis

Thanks to a hydrodynamic focalization setup, flow cytometry (FCM) can characterize the colloids of a suspension individually. For each colloid or aggregates detected (also called ‘event’), the flow cytometer records (i) its fluorescence intensity at a number of preset wavelengths and (ii) the intensity of the light scattered at 90° (side scattering, SSC-A) and at small angles (forward scattering FSC-A). Light scattering intensity depends on a complex way on the size and shape of the colloids and on the refractive index(es) of the material(s) they are made of (Mishchenko et al., 2002).

The characteristics of the lasers and filters of the BD LSRFortessa flow cytometer used to excite/detect each microsphere type are shown in the Supplementary Material, Table S2. We checked experimentally that there was no interference between the excitation and emission wavelengths of microsphere with different dyes (Supplementary Material, Flow cytometry section). The microsphere populations were identified and counted independently of one another and of SC using one or two-dimensional representations of their fluorescence and scattering characteristics (Fig. 1a and Flow cytometry section of the Supplementary Material). Once individualized, each microsphere population was further analyzed in a light scattering vs. fluorescence representation. This allowed identifying singlet, doublet and multiplet populations of microspheres and hetero-aggregation of fluorescent microspheres with non-fluorescent SC (Fig. 1b, c) (Li et al., 2004; Mishchenko et al., 2002; Rollié and Sundmacher, 2008, 2010).