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Modeling the migration of fallout radionuclides to quantify the contemporary transfer of fine particles in Luvisol profiles under different land uses and farming practices

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ABSTRACT

Soil mixing and the downward movement of solid matter in soils are dynamic pedological processes that strongly affect the vertical distribution of all soil properties across the soil profile. These processes are affected by land use and the implementation of various farming practices, but their kinetics have rarely been quantified. Our objective was to investigate the vertical transfer of matter in Luvisols at long-term experimental sites under different land uses (cropland, grassland and forest) and different farming practices (conventional tillage, reduced tillage and no tillage). To investigate these processes, the vertical radionuclide distributions of ^{137}Cs and ^{210}Pb (xs) were analyzed in 9 soil profiles. The mass balance calculations showed that as much as 91 ± 9% of the ^{137}Cs was linked to the fine particles (≤2 μm). To assess the kinetics of radionuclide redistribution in soil, we modeled their depth profiles using a convection–diffusion equation. The diffusion coefficient represented the rate of bioturbation, and the convection velocity provided a proxy for fine particle leaching. Both parameters were modeled as either constant or variable with depth. The tillage was simulated using an empirical formula that considered the tillage depth and a variable mixing ratio depending on the type of tillage used. A loss of isotopes due to soil erosion was introduced into the model to account for the total radionuclide inventory. All of these parameters were optimized based on the ^{137}Cs data and were then subsequently applied to the ^{210}Pb (xs) data. Our results show that the ^{137}Cs isotopes migrate deeper under grasslands than under forests or croplands. Additionally, our results suggest that the diffusion coefficient decreased with depth and that it remained negligible below the tillage depth at the cropland sites, below 20 cm in the forest sites, and below 80 cm in the grassland sites.

1. Introduction

Pedogenesis is a non-linear process in time and space (Montagne et al., 2013). Although the occurrence of major pedogenic processes can be easily identified in soil, their chronological succession and their rates are difficult to quantify due to the difficulty of finding relevant tracers to investigate the processes of interest in the appropriate temporal scale. Among these processes, lessivage, which is defined as the migration of fine particles (≤2 μm or lutum fraction) from subsurface horizons (eluviation) and their accumulation in the B horizon (illuviation), has not been quantified, and its kinetics remain poorly understood (Quénard et al., 2011).

Lessivage was traditionally described as occurring at a timescale ranging from tens to thousands of years (Jamagne, 1973; Finke and Hutson, 2008). Although some authors considered that lessivage is an ancient pedogenic process that occurred exclusively under warm and humid climates (Jamagne, 1978; Jamagne and Pédro, 1981), Fedoroff (1997) showed that it is still currently active in regions with a Mediterranean climate. Therefore, we decided to study (i) the occurrence of current lessivage in a non-Mediterranean temperate climate and (ii) the impact of land use and agricultural practices on this lessivage.

Lessivage is considered to be the major pedogenic process occurring in several soil types (Bockheim and Gennadiyev, 2000), mainly in European Luvisol and Albeluvisol soils. These soil types are also particularly widespread in the USA and in China as they

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develop in all loess deposits. In Europe, these soils cover 20% of the surface (Jones et al., 2005). Several authors hypothesized that agricultural activities could have a crucial impact on their evolution (Khan et al., 2005; Kühn, 2003; Fedoroff, 1997). Therefore, they represent an ideal case study.

Lessivage is not the only pedogenic process that affects vertical solid matter transfers in soils. Other processes that may act in synergy or in opposition to lessivage must be considered. Pedoturbation processes, such as clay shrinking–swelling cycles, soil freezing and bioturbation, tend to mix or, conversely, to segregate soil matter. Bioturbation is the movement of matter in the soil due to the presence of living organisms (Gobat et al., 2004), soil fauna and/or root systems, as well as tree falls (Wilkinson et al., 2009). This has several important consequences on soil structure, such as the mechanical loosening of the soil, the oxygenation of the deepest layers, the redistribution of organic matter, the transport of buried elements to the surface and the neutralization of the pH. Overall, it is clear that bioturbation plays an essential role in clay distribution in soils. Although there are many agents that act in bioturbation, in a temperate climate, earthworms are considered to be the main actor (Müller-Lemans and van Dorp, 1996; Gobat et al., 2004; Persson et al., 2007), particularly anecic worms when vertical movement is considered (Persson et al., 2007).

The fallout radionuclides ^{137}Cs and ^{210}Pb (xs) have different sources in the environment. ^{137}Cs was released and dispersed during the nuclear weapon tests that occurred between 1953 and the 1970s, as well as by nuclear power plant accidents (Chernobyl in 1986 and Fukushima Dai-ichi in 2011). In contrast, ^{210}Pb is naturally present because it is a product of ^{238}U decay and is a daughter product of gaseous ^{222}Rn. Some ^{222}Rn may escape from the soil and lithosphere and continue to decay in the atmosphere, resulting in ^{210}Pb, which then returns to the Earth's surface through wet fallout. Therefore, this is referred to as unsupported or excess (xs) ^{210}Pb, in contrast to supported ^{210}Pb that is naturally present in the soil. The fallout radionuclides ^{137}Cs and ^{210}Pb (xs) have been used as tracers in soil studies because they are exclusively of atmospheric origin. Furthermore, they are strongly sorbed to the soil solid phases: ^{137}Cs to clay particles (Tamura and Jacobs, 1960; Sawhney, 1972; Cremers et al., 1988) and ^{210}Pb to clay particles and organic matter (Dörr and Münnich, 1989). Thus, their transfer occurs only in association with the solid matter in the soils that are not dominated by soluble organic matter transfers (Jarvis et al., 2010; Matisoff et al., 2011). Finally, the choice of these radionuclides was also motivated because their half-lives (30.2 years for ^{137}Cs and 22.3 years for ^{210}Pb) are favorable for investigating the impact on the transfer processes due to modification in agricultural practices, occurring at a decadal timescale.

These reasons justify the use of ^{137}Cs and ^{210}Pb (xs) as suitable tracers for estimating the kinetics of solid matter transfer in soils, including lessivage and bioturbation. Diffusion–convection modeling of the vertical distributions of these radionuclides (e.g., He and Walling, 1997; Schuller et al., 1997) could provide a way to differentiate between these two processes because the diffusion equation, which corresponds to the random mixing (or movement) of matter upwards and downwards across short distances, is classically associated with bioturbation (Boudreau, 1986; Elzein and Balesdent, 1995; Schiffers et al., 2011), whereas convection as a long distance transport may be associated with lessivage. However, part of the long distance transport may also be associated with bioturbation, such as that resulting from the activity of anecic or epianecic earthworms. This bioturbation may result in more complex processes, such as litter incorporation into deeper layers or, reciprocally, deposits of deep horizon matter at the surface in the form of casts. In the diffusion–convection modeling, long-distance transport by bioturbation was assessed using a supplementary term (Jarvis et al., 2010). Other modeling approaches of worm mixing have been developed based on worm mixing experiments (Barnett et al., 2009; Schiffers et al., 2011). However, the experimental data provided by these studies show that they may be reasonably represented by a diffusion equation.

The aims of this study are (1) to quantify the current particle transfer by ^{137}Cs and ^{210}Pb (xs) in a soil sequence, as defined by Jenny (1941), considering human activity as a pedogenic factor (anthroposequences); (2) to determine the kinetics of these transfers by modeling the vertical distributions of ^{137}Cs and ^{210}Pb (xs) with a diffusion–convection equation using both constant and depth-dependent coefficients; and (3) to identify the impact of both land use and agricultural practices on the intensity and kinetics of particle transfer in soils.

2. Materials and methods

2.1. Choice and description of the sampled sites

We selected three Luvisol sequences from Northern France that experienced different land uses (cultivation, grassland and forest), as land use impacts both the soil pH and the soil organic matter content, which are known to influence soil particle stability and dispersion (Le Bissonnais and Arrouays, 1997; Chenu et al., 2000). Additionally, two agricultural practices, manure input and tillage reduction were investigated because tillage reduction modifies the

Table 1
General description of the studied sites.

Site	Mons	Feucherolles	Boigneville
Experiment name	Essay system – ORE-ACBB[a] Mons	ORE-PRO[b]-Qualiagro	Wheat monoculture
Managing institution	INRA	INRA	Arvalis (Plant institute)
Geographic coordinates	49°52′01″N – 3°01′53″ E	48°53′49″ N – 1°58′19″ E	48°19′30″ N – 2°22′58″ E
Elevation	88 m	120 m	116 m
Mean annual rainfall	680 mm	660 mm	630 mm
Mean annual temperature	11 °C	11.2 °C	10.4 °C
Considered landuses and farming practices	- Conventional tillage - Reduced tillage (since 2000) - Grassland (since 1939)	- Conventional tillage without manure or fertilizers (since 1998) - Conventional tillage with manure (since 1998) - Oak forest (since 1815 at least according to Cassini maps)	- Conventional tillage - Reduced tillage (since 1970) - No tillage (since 1970)
Crop rotation	Wheat – corn – sugarbeet	Corn – wheat	Wheat
Liming	Not since 1986 under cultivation and 1939 for pasture	Not since 1998 under cultivation	Not since 1970
Maximal distance among plots	2 km	2 km	A few tens of meters
Sampling date	March 2011	April 2011	March 2012

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