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Lead and Zinc status evolution during pedogenesis in a 100 years old Technosol

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1. Introduction

In the frame of the transposition of the European directive on waste in French law, a national technical committee has been implemented in France since 2009 to set up a methodology of management of excavated lands. The reuse of excavated lands in a lot of city planning and landscaping configurations is designed for long-term uses. Their reuse and deposition on site should lead to the formation of new soils, generally called Technosols (WRB 2006). Although contaminated, these materials present a pedologic interest since they fulfill several functions such as biomass production, storage-filtration and transformation of nutriment, trace elements and water, or formation of ecological niches (Morel et al. 2015). However, the long-term behavior of these materials is difficult to study since the pedological processes exceed the human timescale. Nevertheless, early pedological transformations could sometimes be observed in Technosols (*e.g.*, Séréet al. 2006). The study of these processes needs to focus on “old” Technosols, *i.e.* on materials that have been settled for hundreds of years. The first industrial activities started in Europe in the early 19th century. At this period, the earthworks carried out for factory building led to the backfill of wide areas. These materials are still in place in some old brownfields. Their investigation could highlight several early pedological processes, as well as assessing the status of some pollutants, especially by studying soil profiles where the deeper horizons are considered less evolved than the upper horizons. For this study, a ~2m depth profile was characterized in a 100 years-old Technosol developed on backfills that contain significant concentrations of lead (Pb) and zinc (Zn).

2. Material and Methods

The study was conducted on a soil profile of the “Union” former industrial site (Tourcoing, France). Historical investigations carried out at the archives of the French railways evidenced that the area of study was not reworked for a century (Coussy et al. 2017). The whole soil profile was sampled as a function of depth (Figure 1). Two types of samples were collected: composite samples for bulk analyses and samples representative of a given depth in the profile for punctual investigations (hereafter denominated “slices”). Composite samples were taken within the following range of depth: 0-10 cm, 30-40 cm, 110-130 cm and 175-185 cm. At each depth range, three replicates were taken on a same horizon. Soil slices were taken at 5 cm, 90 cm and 180 cm deep. The soil structure was preserved (non-destructive sampling using cylinders) and 3 thin sections (30 µm) were elaborated. In laboratory, the soil pH, total organic carbon content, grain size distribution and total elemental concentrations were

determined according to standardized methods (Coussy et al. 2017). The release of trace elements was assessed by leaching of the composite samples. The leaching tests were conducted in closed PTFE reactors with continuous stirring during 24h, with a liquid/solid ratio of 10. Pb and Zn status was assessed by bulk analyses such as selective dissolution, and by punctual analyses (scanning electron microscope, electronic microprobe). Selective dissolutions were carried out using both Dithionite-Citrate-Bicarbonate (DCB) and pyrophosphate methods, done in parallel on aliquots. DCB selectively solubilizes pedogenic oxides and hydroxides (Mehra, Jackson 1960). The pyrophosphate method was used to dissolve amorphous complexes and/or very poorly crystalline compounds. Finally, Zn status investigations were refined by X-ray absorption spectroscopy (XAS) and nuclear microprobe (μ -PIXE and μ -RBS).

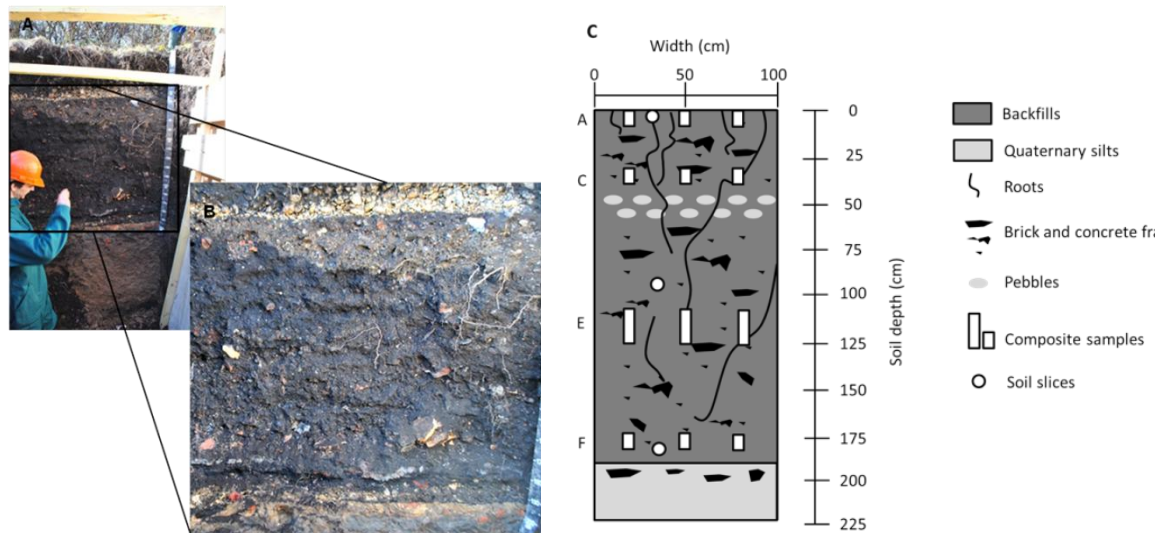


Figure 1. Soil profile. A General overview. B. Details of the central zone of the profile. C. Schematic representation of the soil profile.

3. Results

A comparison between Pb and Zn behavior after leaching and selective dissolution is shown in Figure 2. Pb and Zn follow the same trends after leaching, indicating that they are very little labile throughout the whole profile ($< 0.1\%$ release). These elements are probably under the form of mineral phases or strongly bound to mineral phases.

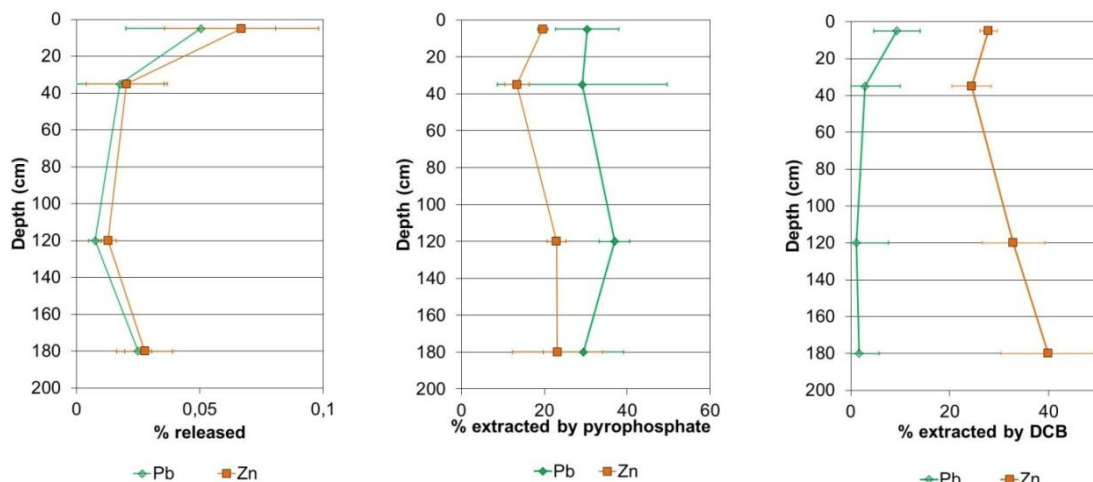


Figure 2. Pb and Zn extracted by leaching, pyrophosphate and DCB, throughout the soil profile (normalized by the total amounts in Pb and Zn of each sample).

Pyrophosphate extractions show that between 20 to 40 % of the total Pb and Zn is bound to organic matter or to slightly crystallized minerals, with no dependence to depth. DCB extractions indicate different trends for Pb and Zn. The Pb associated to Fe (hydr)oxides does not exceed 10% of the total Pb in surface, and this amount decreases along depth. On the contrary, Zn extraction yield (35% on average) is, within uncertainties, constant over the whole profile. However, as Zn concentration in the deepest sampling point is 10 times lower than in the surface horizon, a small quantity of Zn-bearing Fe (hydr)oxides that would have migrated from the upper horizons may have led to such apparent change. Electronic and nuclear microprobe investigations have confirmed that various proportions of Pb and Zn are trapped in Fe (hydr)oxides – mainly identified as ferrihydrite - at the surface of the profile. XAS analyses of the composite samples indicate that Zn coordination is tetrahedral, but in the deeper horizons, Zn is associated to phases having a higher degree of crystallinity than in the surface horizons. The other Pb and Zn minerals identified are sulphides, as well as intermediate phases such as sulphates.

4. Discussion and conclusion

In the studied Technosol, pedogenesis does not induce a dramatic increase of Pb and Zn solubility since efficient scavengers are concomitantly formed in the system. However, the weathering processes during soil evolution influence Pb and Zn speciation. Primary Zn and Pb bearing minerals have been identified as sulphides. However, these phases have evolved at the surface of this Technosol to form sulphates, not stable in conditions of pH observed here. During this evolution, various proportions of Pb and Zn have been trapped in Fe (hydr)oxides (ferrihydrite) neoformed at the surface of the profile since the initial deposit of the backfills. The input of organic matter at the surface is slightly visible after 100 years of evolution. Technosols are highly heterogeneous and widely differ from one place to another because of the variety of their constitutive man-made materials. In this case, 100 years of soil evolution has led to the precipitation of secondary minerals at the surface of the profile. This behavior is similar to pedogenic processes occurring in natural soils, but other specific evolutions due to large thermodynamic disequilibrium can be highlighted, in particular sulphide oxidation and sulphate precipitation.

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