



# Soil tillage to reduce surface metal contamination – model development and simulations of zinc and copper concentration profiles in a pig slurry-amended soil



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## ABSTRACT

Long-term applications of organic amendments, such as pig slurry (PS), may represent environmental risk of soil and water pollution by trace metals (TM). Our objective was to examine different soil and manure management scenarios that enhance the long-term agricultural use of soils under repetitive PS applications while avoiding environmental risk. Firstly, we developed a new module for simulating the impacts of soil tillage frequencies in Hydrus-1D. Secondly, we used a previously validated modeling approach to predict the surface accumulation and movement of the TM during the next 100-year in the soil under different PS doses (80 and 40 m<sup>3</sup> ha<sup>-1</sup> cultivation<sup>-1</sup>) and tillage frequencies (no-tillage and 20, 10, and 5-year tillage). No-tillage simulations revealed consistent TM surface accumulations, reaching the soil threshold value for Cu in the 0–20 cm layer after 86 years of PS amendments at high doses, but in layers 0–5, 0–10, and 5–10 cm, this concentration was already reached after 17, 38, and 75 years, respectively. While soil tillage reduced TM concentrations over the top 20 cm of the soil profile, it increased their transfer to deeper layers. Periodical soil tillage each 5, 10, and 20 years was found to allow PS applications without reaching the Cu threshold value in soil during 100 years. However, soil solution concentrations of Zn reached the threshold values for groundwater. Therefore, the best manure management practice for the long-term PS disposal with respect to Zn and Cu concentrations in soil is the application of moderate PS rates.

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## 1. Introduction

Long-term applications of organic waste amendments on soils may lead to a notable accumulation of mineral and organic micropollutants in the soil's surface layer. Hence, such practices potentially represent environmental risk of soil and groundwater pollution. Recently, we have carried out validating and prospective modeling work, using the Hydrus software package, to simulate the movements of trace metals (TM) zinc (Zn) and lead (Pb), derived from past industrial metallurgical activities, in agricultural

soils under different long-term land uses in northern France (Rheinheimer et al., 2013; Mallmann et al., 2012a). Then we have successfully applied the same modeling approach to simulate the movement of Zn and copper (Cu) in an Alfisol in Southern Brazil, managed under repetitive applications of pig slurry (PS) (Mallmann et al., 2012b). In addition to major nutrients such as phosphorus (P), nitrogen (N), and potassium (K) (Lourenzi et al., 2013), PS contains consistent amounts of oligo-nutrient elements such as Zn and Cu that at high concentrations become contaminants (Giroto et al., 2010; Berenguer et al., 2008; L'Herroux et al., 1997). Both Zn and Cu are consistently present in pigs' diet, representing mineral supplements or growing elements, aiming to increase the productivity index of these animals (Li et al., 2005). A large fraction of Zn and Cu, however, is not absorbed by the animal

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gastrointestinal tract and is released via pig manure (Nicholson et al., 2003). In the work of Mallmann et al. (2012b), we showed how a continuous application of PS on the Alfisol for 50 years would lead to large Zn and Cu contents in the 0–5 cm soil layer. Particularly for Cu, the simulated soil concentrations in this layer exceeded the Brazilian guideline intervention value for agricultural areas of 200 mg kg<sup>-1</sup>, established by the Conselho Nacional do Meio Ambiente (CONAMA, 2009).

Such strong accumulation of TMs at the soil's surface is directly related to agricultural land management practices of no-tillage, currently widely used in Brazil. More than 60% of cultivated land (23.6 million hectares) is farmed as a no-till system, maintaining the soil covered and unplowed, sowing is performed directly on the soil surface without its previous preparation and crop rotation is used (Mello and van Raij, 2006). Therefore, no-tillage practices in agricultural lands under long-term PS amendments will favor the accumulation of Zn and Cu within the first few centimeters of the soil profile, and reduce the movement of these TMs to layers below the depth of 20 cm. In this view, the no-tillage management represent a disadvantage in comparison to the conventional tillage management, where the surface layer is annually mixed by plowing and, consequently, the concentrations of added micro-pollutants are mechanically homogenized over the entire plow-depth (about 20 cm). Therefore, introducing periodical soil tillage, as a sustainable land management practice, represents an original solution for reducing the strong surface accumulation of the TMs by mixing them over a greater depth. It also represents a solution to comply with Brazilian environmental guidelines on maximum TM values (CONAMA, 2009), which may be restrictive for long-term organic amendments on no-tillage farming systems.

Soil tillage practices, however, by homogenizing the plow layer and thus reducing the soil solution metal concentrations in the uppermost soil layer and increasing them at the bottom of the tilled layer, favor the downward movement of the TMs via the soil solution to depths below the plow layer. They may then propagate down to the bottom of the soil profile and require a more detailed examination, especially at depths close to the groundwater table. Therefore, the threshold values for groundwater established by the Brazilian legislation (CONAMA, 2009), i.e. 1.05 and 2.0 mg L<sup>-1</sup> for Zn and Cu, respectively, should not be reached at deeper soil depths to avoid risks of groundwater pollution.

The aims of the present work were to examine current and adapted soil and manure management practices for a sustainable agricultural land use of soils under repetitive PS applications, while avoiding environmental pollution risks. For that, we used Hydrus-1D to model the future movement of Zn and Cu in an Alfisol contaminated by repetitive applications of PS. First, we developed a new module that simulates the effects of soil tillage at different time intervals and implemented it in Hydrus-1D. Second, we used the validated modeling approach of Mallmann et al. (2012a,b) to predict the surface accumulation, soils solution concentrations, and redistribution of TMs in the Alfisol profile for different PS doses and varying soil tillage frequencies during the next 100 years. Simulated soil and solution concentrations of Zn and Cu were then compared with current Brazilian guideline values.

## 2. Materials and methods

### 2.1. General and site characteristics, soil sampling and analysis

The studied soil profile, a typical Hapludalf (USDA, 2003), is located at the experimental fields of Federal University of Santa Maria, Rio Grande do Sul State, Brazil (29°42'52"S and 53°42'10"W, 90 m altitude). An experiment that was conducted during eight years (01/2000–01/2008) under the no-tillage management and

that received applications of different PS doses (0, 20, 40, and 80 m<sup>3</sup> ha<sup>-1</sup>) serves as the basis of this study. The PS doses were applied at the soil's surface before each cultivation; one time in 2007, two times in 2000, 2004, and 2005, and three times in 2001–2003, and 2006.

Soil samples from a plot with the 80 m<sup>3</sup> ha<sup>-1</sup> PS treatment were collected in January 2008 from soil layers of 0–5, 5–10, 10–25, 25–35, 35–50, and 50–60 cm depths, with three subsamples per layer. We determined the soil's particle density, water retention curve (SWRC), particle size distribution, saturated soil hydraulic conductivity ( $K_s$ ), bulk density, total porosity, pH<sub>H2O</sub>, total content and desorption curves of Zn and Cu, and organic carbon. Analytical methods and procedures used for the determination of these parameters are extensively described in Mallmann et al. (2012b). The results are shown in Table 1.

### 2.2. Model parameterization and solute transport simulations

Hydrus-1D (Šimunek et al., 2008) was used to simulate one-dimensional unsaturated water flow and Zn and Cu transport in a vertical domain for a one century period. The analytical models of van Genuchten and van Genuchten–Mualem were used to describe water retention curves and unsaturated hydraulic conductivity functions (van Genuchten, 1980), respectively. This software also requires additional hydro-physical and chemical parameters to run all desired simulations. For Brazilian climatic conditions with high intensity rainfalls, the most important physical parameter is the saturated hydraulic conductivity ( $K_s$ ), since it determines water infiltration and percolation through the soil profile, the transport of dissolved solutes, and their transfer to greater depths. Additional key parameters include the soil residual ( $\theta_r$ ) and saturated ( $\theta_s$ ) water contents, the shape parameters  $\alpha_{VG}$  and  $n_1$  obtained from SWRC, and the pore connectivity/tortuosity factor ( $L$ ).

Mallmann et al. (2012a,b) and Rheinheimer et al. (2013) used the TM profile concentrations collected in field experiments to show that the two-site sorption model reproduced better the TM movement in the soil compared to the equilibrium sorption model. Similar conclusions were obtained by many other researchers who collected metal breakthrough curves from soil column studies. For example, Chotpantarat et al. (2012) showed that the two-site sorption model fitted both rising and declining branches of breakthrough curves of heavy metals much better than the equilibrium sorption model. Pang et al. (2002) showed that metal interactions with the soil indicate the presence of chemical non-equilibrium in the system, due to the presence of long tailing in breakthrough curves. Moreover, according to Pang and Close (1999), non-equilibrium sorption models describe better the transport of heavy metals and provide better explanations for the spreading and asymmetry of breakthrough curves.

In this way, the chemical parameters required by the two-site model include the initial Zn and Cu solution concentrations, and the Freundlich's  $n$  and  $K_F$  parameters, describing the equilibrium adsorption/desorption process. The most important parameters characterizing the sorption kinetics are  $\beta$  and  $\lambda$ . The  $\beta$  parameter defines the amounts of sorption sites in chemical equilibrium with the soil solution. In our conditions, this parameter decreases with depth. The lower the value of this parameter, the more important are the sorption sites that react slower with the dissolved heavy metals. These sites are sometimes called kinetic sorption sites. As the fraction of kinetic sorption sites increases, the  $\lambda$  parameter, describing the sorption rate, becomes more important. The lower its value, the slower the adsorption to the soil sorption sites, and, consequently, the higher the solute concentration in the soil solution and the faster the transport to deeper layers. However, during the desorption phase, the low value of the  $\lambda$  parameter

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