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Interactive comment on "Modelling of long term Zn, Cu, Cd, Pb dynamics from soils fertilized with organic amendments" *by* Claudia Cagnarini et al.

Anonymous Referee #2

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Manuscript title: Modelling of long term Zn, Cu, Cd, Pb dynamics from soils fertilized with organic amendments

General comments

This MS addressed the issue of the modelling of the contamination of agricultural soils by trace elements added by the long term application of organic residues and the toxicological and ecotoxicological consequences of such a contamination. This issue is relevant and of a timely interest. In particular, most of previous study were focused on the modelling of the change in total trace element concentration in soil, but the present MS is rather dedicated to the modelling of trace element availability (also called lability in the MS) in soil. This is clearly original and of an environmental interest to make the link with the potential toxicological and ecotoxicological consequences.

I have however a range of comments that to my point of view should be addressed before addressing this MS for publication in SOIL.

Major comments My first major comment is related to the general approach followed and to what is to my point of view the main conclusion of the paper. The author concluded (lines 509-511) that "the IDDM-ag model provided an adequate description of the measured EDTA-extractable concentration trends…". Looking at the figure 5, this is not so obvious. There are indeed several situations for which there is a clear discrepancy between experimental data and modelling (e.g. Zn-SS, Pb-SS, Cd-COM) and also other situations where the Swiss (e.g. Zn-FYM, Pb-FYM) or the ZOFE (e.g. Cu-SS) was alternatively the hypothesis which enables the model to have the best fit. In addition, these fits are based on the modelling approach considering lateral mixing. If the principle of lateral mixing is explained and if the uncertainty related to such a computation is discussed in the MS, there is not any validation of such a computation based on experimental data. In addition to that, there are a lot of uncertainty on some major flux of trace elements for a significant part of the field experiment history, particularly on trace elements added by organic residues. To consider whether the fits obtained were adequate or not, uncertainty in model parameters and input data should be considered and compared to the uncertainty in experimental data. Moreover, considering the question about the adequacy of the fits of EDTA-extractable concentration, it should be to my point of view necessary to show as the first step the fits obtained for total trace element concentration trends in soil. The idea is that if the total trace element trends are not adequately simulated, how the EDTA-extractable trend could be? The simulation of total trace element concentration trends seem to me even more necessary as the accumulation trend expected is not visible for most trace element and organic fertilisation modalities. In particular, total Cu concentration shows a strong decreasing trend that was attributed to the past application of Cu fungicide, then followed by a sharp removal of Cu from the top-soil layer. No convincing explanation is given for this as the authors said that they do not have information about Cu-fungicide applications and assumed that soil ploughing and bioturbation explained the dilution of Cu concentration in soil without any simulation to support these strong assumptions. Without any other explanation, it seems that the Cu dataset is strongly biased and should, to my point of view, be removed from the MS.

My second major comment is related to the second major conclusion of the paper suggesting that Cu and Zn contamination in soil can be harmful to soil organisms. To my understanding, this conclusion is based on the methodology described lines 309-313. It is however really unclear how the related computation of critical limits was effectively achieved. I looked at the cited paper of Lofts et al. 2005, from which I supposed that the free ion approach was based on EDTA-extractable concentration, pH and SOM data. If I am right, I notably wonder how the natural background concentration of trace elements in the soil was considered as regard to the fact that this specific issue is addressed by Lofts et al. (2005). Also, this methodology was tested on two dataset from UK and North America. It is thus not obvious that the methodology is relevant for the specific case and consequently the specific application of the IDDM-ag model studied here.

Additional comments Lines 67-68 and 77-82: Sewage sludge is introduced differently from other organic residues (FYM and COM), notably because of the higher trace element concentration found in SS compared to FYM and COM. However, this is because the FYM and COM had relatively low trace element concentrations. For instance, I suppose that FYM is a cow manure. If a pig or poultry manure had been chosen, the concentration of several trace elements (Cu and Zn more particularly) would have been much higher and likely comparable to the concentrations observed in SS.

Line 199: To what refer the metal input? To the pool of total or available trace element?

Lines 199-211: It is not clear what is considered behind “geogenic input”. It is also very surprising to use data on the weathering rate of deppe layers of peat bogs to estimate the weathering rate in the present field experiment where the soil is clearly not a peat bog.

Doc concentration was fixed at 7 mg C/L. The authors further argued that Doc variation between 7 and 12 mgC/L does not impact the leaching of TE. However, considering the large variation in SOM and pH in the different fertilization modalities, I am surprised that a larger range of Doc concentration was not expected. Several authors (e.g. Araujo et al. 2019, https://doi.org/10.1016/j.envpol.2018.12.070; Cambier et al. 2014, https://doi.org/10.1016/j.scitotenv.2014.06.105; Laurent et al. (2020, https://doi.org/10.1016/j.scitotenv.2019.135927) showed drastic change in Doc concentration in soil amended with organic residues. A way to estimate the initial Doc concentration and its likely evolution over time could have been to use the empirical multi-linear regression suggested by Romkens et al. 2004 (Derivation of Partition Relationships to Calculate Cd, Cu, Ni, Pb, Zn Solubility and Activity in Soil Solutions; Alterra: Wageningen, 2004; p 75).

Line 303: The choice to fix pH and SOM in soil at the value found in 2014 for predictive modelling is really disputable, when considering how these two parameters are strongly impacted by the long-term applications of organic residues and particularly in the context the field experiment studied as showed in figure S4. This point should at least be discussed.

Lines 324-338 and 348-364: These two paragraphs are really too descriptive and speculative. As related to my first main comment, the simulation of total trace element concentration trends seem a prerequisite to assess the adequacy of the model used. But, as far trace element availability in soil is concerned, some (usually found in the literature, but nevertheless strong) hypotheses on the soil parameters explaining the change over time in the EDTA-extractable concentration of trace elements. These hypotheses should be checked, for instance by looking for multi-linear regression between trace element EDTA-extractable concentration or lability and some important parameters such as the input of trace metal in soils, total trace metal concentration in soil, SOM and pH in soil.

Lines 425-445: Basically, the consideration of lateral mixing is interesting. However, the comparison of simulations with and without lateral mixing should be showed clearly (at least in supporting information) to support the conclusion that accounting for lateral mixing is important.

Section 3.4: It is really unclear to me what is the added value of the FTIR and XRD datasets. To my point of view, these datasets should be removed.

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