Geophysical Research Abstracts
Vol. 15, EGU2013-12652, 2013
EGU General Assembly 2013

Fate and transport of mercury in soil systems : a numerical model in HP1 and sensitivity analysis

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Mercury (Hg) poses threats for human health and the environment, notably due to its persistence and its ability to bioaccumulate in ecosystems. Anthropogenic activities are major contributors of mercury release to soils. Main sources of contamination include manufacturing (chlor-alkali plants, manometer spill), mine tailings from mercury, gold and silver mining industries, wood preservation. The objective of this study was to develop a reactive transport model for simulating mercury fate and transport in the unsaturated zone, and to gain insight in the fate and transport of Hg following anthropogenic soil contamination. The present work is done in the framework of the IMaHg project, which aims at providing recommendations to improve management of sites contaminated by mercury within the SNOWMAN funding framework.

A model of mercury fate and transport in soil systems was developed using the reactive transport code HP1 (Jacques and Šimůnek, 2010). The geochemical database THERMODDEM (Blanc et al., 2012) is used, augmented with some speciation data from (Skyllberg, 2012).

The main processes accounted for in the model are : Hg aqueous speciation (including complexation with dissolved organic matter (DOM) – humic and fulvic acids, and thiol groups), Hg sorption to solid organic matter (SOM), dissolution of solid phase Hg (e.g. cinnabar HgS(s)), dissolution of Hg non-aqueous liquid phase (NAPL), sunlight-driven Hg(II) reduction to Hg(0), Hg(0) diffusion in the gas phase and volatilization, DOM sorption to soil minerals. Colloid facilitated transport is implicitly accounted for by solute transport of Hg-DOM complexes.

Because we focused on soil systems having a high Hg contamination, some processes showing relatively smaller Hg fluxes could be neglected such as vegetation uptake and atmospheric wet and dry deposition. NAPL migration and entrapment is not modelled, as pollution is assumed to be historical and only residual NAPL to be present. Mercury methylation and demethylation was not implemented, because it could be neglected in an oxidising environment. However, if the model is to be tested in more reducing conditions (e.g. shallow groundwater table), methyl- and dimethylmercury formation can be non negligible.

Using 50 year time series of daily weather observations in Dessel (Belgium) and a typical sandy soil with deep groundwater (free drainage, oxic conditions), a sensitivity analysis was performed to assess the relative importance of processes and parameters within the model. We used the elementary effects method (Morris, 1991; Campolongo et al., 2007), which draws trajectories across the parameter space to derive information on the global sensitivity of the selected input parameters.

The impact of different initial contamination phases (solid, NAPL, aqueous and combinations of these) was also tested. Simulation results are presented in terms of (i) Hg volatilized to the atmosphere; (ii) Hg leached out of the soil profile; (iii) Hg still present in the soil horizon originally polluted; and (iv) Hg still present in the soil profile but below the original contaminated horizon.

Processes and parameters identified as critical based on the sensitivity analysis differ from one scenario to the other ; depending on pollution type (cinnabar, NAPL, aqueous Hg), on the indicator assessed and on time (after 5, 25 or 50 years). However, in general DOM in soil water was the most critical parameter. Other important parameters were those related to Hg sorption on SOM (thiols, and humic and fulvic acids), and to Hg complexation with DOM. Initial Hg concentration was also often identified as a sensitive parameter. Interactions between factors and non linear effects as measured by the elementary effect method were generally important, but also dependent on the type of contamination and on time.

No model calibration was performed until now. The numerical tool could greatly benefit from partial model calibration and/or validation. Ideally, detailed speciation data on a contaminated sites would be required, together with a good characterization of the pollution source.

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