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# The response of metal leaching from soils to climate change and land management in a temperate lowland catchment



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#### ABSTRACT

Changes in soil hydrology as a result of climate change or changes in land management may affect metal release and leaching from soils. The aim of this study is to assess the cascading response of SOM and DOC levels and metal leaching to climate change in the medium-sized lowland Dommel catchment in the southern part of the Netherlands. We implemented the CENTURY model in a spatial setting to simulate SOM, DOC, and water dynamics in topsoils of the Dutch portion of the Dommel catchment under various climate and land management scenarios. These CENTURY model outputs were subsequently used to calculate changes in the topsoil concentrations, solubility, and leaching of cadmium (Cd) and zinc (Zn) for current (1991-2010) and future (2081-2100) conditions using empirical partition-relations. Since the metal leaching model could not be evaluated quantitatively against measured values, we focus mainly on the trends in the projected metal concentrations and leaching rates for the different scenarios. Our results show that under all climate and land management scenarios, the SOM contents in the topsoil of the Dommel catchment are projected to increase by about 10% and the DOC concentrations to decrease by about 20% in the period from present to 2100. These changes in SOM and DOC only have a minor influence on metal concentrations and leaching rates under the climate change scenarios. Our scenario calculations show a considerable decrease in topsoil Cd concentrations in the next century as a result of increased percolation rates. Zinc, however, shows an increase due to agricultural inputs to soil via manure application. These trends are primarily controlled by the balance between atmospheric and agricultural inputs and output via leaching. While SOM and DOC are important controls on the spatial variation in metal mobility and leaching rates, climate-induced changes in SOM and DOC only have a minor influence on metal concentrations and leaching rates. The climate-induced changes in metal concentrations in both the topsoil and the soil leachate are primarily driven by changes in precipitation and associated water percolation rates.

#### 1. Introduction

In many parts of the world, anthropogenic activities have led to soil contamination by heavy metals (Xue et al., 2003; Azimi et al., 2004; Peng et al., 2009). Apart from local soil contamination by waste dumps and mining activities, soils have become enriched with metals by diffuse inputs via atmospheric deposition or application of manure, fertilisers, sewage sludge or other agrochemicals. Leaching of heavy metals from diffusively contaminated soils can be a major source of metals in surface waters (Bonten et al., 2008b; Wijngaard et al., 2017).

Changes in soil hydrology as a result of climate change or changes in

land management may accelerate or reduce metal release and leaching. For example, Visser et al. (2012) projected reduced future metal leaching rates in response to reduced precipitation in a small metal-contaminated catchment in the southern part of the Netherlands. In contrast, Joris et al. (2014) predicted a climate-induced increase in metal fluxes in northern Belgium.

Furthermore, lowering of the soil pH due to, for example, acid deposition or a raise in the soil redox potential due to, for example, soil drainage may cause an accelerated release of bound metals into the soil solution of metal-contaminated soils. During the early 1990s this phenomenon was referred to as 'chemical time bombs' (Stigliani, 1991).

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Climate change or changes in land management may contribute to such changes in environmental soil conditions. Initially, it was argued that climate-change induced losses of SOM in European soils could cause a reduction in the cation exchange capacity, thereby promoting the mobilisation of metals (Ter Meulen-Smidt, 1995). The response of SOM content to climate change is however not unambiguous as besides negative responses (Cramer et al., 2001; Smith et al., 2005; Stergiadi et al., 2016), positive responses have also been reported (Liski et al., 2002; Álvaro-Fuentes et al., 2012; Gottschalk et al., 2012; Smith, 2012). Moreover, climate change affects the DOC concentrations in soil and these changes are mainly associated with changes in precipitation and evapotranspiration patterns (Harrison et al., 2008; Stergiadi et al., 2016). An increase in precipitation has been suggested to result in enhanced DOC concentrations and leaching rates (Whitehead et al., 2009), whereas a decrease in net precipitation is predicted to result in reduced DOC concentrations and leaching rates (Stergiadi et al., 2016).

The above-mentioned studies indicate that the response to climate change of the solid-solution partitioning, mobility and leaching of metals is equivocal and involves many feedback processes. Quantification of these responses is essential for a proper assessment of leaching to groundwater and surface water and related risks to ecosystems and humans. The most common approaches for such quantifications that consider changing soil properties involve the use of process-based multi-surface models describing solution chemistry and binding to reactive surfaces (Weng et al., 2002; Bonten et al., 2008a; Dijkstra et al., 2009; Groenenberg et al., 2012) or empirical partition-relations that relate the distribution of metals to soil properties (McBride et al., 1997; Sauvé et al., 2000; Tipping et al., 2003; Rodrigues et al., 2010; Groenenberg et al., 2012).

The aim of this study is to assess the cascading response of SOM and DOC levels and metal leaching to climate change in a medium-sized lowland catchment in the southern part of the Netherlands (Dommel). For this purpose, we implemented the CENTURY model (Parton et al., 1987, 1988, 1993) in a spatial setting to model SOM, DOC, and water dynamics in topsoils of the Dommel catchment under various climate and land management scenarios. These CENTURY model outputs were subsequently used to predict changes in the topsoil concentrations, solubility, and leaching of cadmium (Cd) and zinc (Zn) for current (1991-2010) and future (2081-2100) conditions using the empirical metal partition relations developed by Römkens et al. (2004) and Groenenberg et al. (2012). As the metal leaching model could not be evaluated quantitatively against measured values, the model results are subject to an unknown degree of uncertainty. Therefore, we focus the results description and discussion on the trends in the projected metal concentrations and leaching rates and their differences between the climate and land management scenarios.

#### 2. Methods

#### 2.1. Study area

We modelled metal leaching in the Dutch portion of the Dommel catchment, which covers an area of approximately  $1500\,\mathrm{km}^2$  (Fig. 1). The Dommel River is a 146 km long lowland tributary river to the river Meuse. It rises on the Kempen plateau in the northeastern part of Belgium at an altitude of about 80 m above mean sea level and confluences with the Meuse near the city of 's-Hertogenbosch at about sea level (Bleeker and van Gestel, 2007; De Jonge et al., 2008). The climate is temperate with an average temperature of 10.3 °C and an average annual precipitation of 750 mm y $^{-1}$  over the period 1981–2010 (KNMI, 2011).

The soils consist mainly of Pleistocene fluvial and aeolian sands and loamy sands. (De Mulder et al., 2003) with generally low topsoil pH values between 3.6 and 5. Land use in the Dommel catchment consists of intensively used agricultural land (27% arable land, 23% pastures), natural and semi-natural areas (22% forest, 7% grassland, 3% heather)

and urban areas (14%).

The predominantly sandy soils of the Dommel catchment naturally contain relatively small amounts of heavy metals. The background concentration of Zn and Cd depend on the clay content is generally around 20 mg kg<sup>-1</sup> for Zn and around 0.14 mg kg<sup>-1</sup> for Cd (Bonten et al., 2007). However, the area is considerably polluted by Zn and Cd originating from the historical operation of four zinc-ore smelters in both the Dutch and Belgian part of the catchment and the application of artificial fertilisers and manure on agricultural land (Bonten et al., 2012). Emissions from the zinc ore smelters in the region caused atmospheric deposition of Zn and Cd during about one century, especially in the southern part of the catchment. Since the early 1970s, the atmospheric metal emissions have been greatly reduced as a result of the switchover to the cleaner electrometallurgical ore processing. In addition, the use of ore slags in road constructions and gardens' paving contributed to further dispersal of metals in the area (Copius Peereboom-Stegeman and Copius Peereboom, 1989; Bonten et al., 2012), although the pollution derived from ore slags has a more local character than the pollution due to diffuse atmospheric deposition. Intensive livestock farming in the catchment has led and leads to additional metal loads to soil, including Zn and Cd to agricultural land. However, in the study area, the agricultural load of metals is relatively small compared to the historic atmospheric deposition. For instance, the cumulative Zn load to soil from atmospheric deposition between 1880 and 2000 is estimated to be 435 kg ha<sup>-1</sup> while the cumulative Zn load from agricultural sources equals 95 kg ha<sup>-1</sup> (Bonten et al., 2012). The present-day topsoil concentrations amount to between 20 mg kg<sup>-1</sup> and  $> 200 \text{ mg kg}^{-1}$  for zinc between  $0.1 \text{ mg kg}^{-1}$  and  $> 5 \text{ mg kg}^{-1}$  for cadmium (Mol et al., 2012). These high levels of diffuse metal contamination has led to the degradation of soil ecosystems and potential health risks related to the uptake of these metals by arable crops (Copius Peereboom-Stegeman and Copius Peereboom, 1989). Furthermore, leaching to groundwater and surface water potentially affect drinking water resources (Pedroli et al., 1990; Crommentuijn et al.,

#### 2.2. Model description

#### 2.2.1. Spatial implementation of the CENTURY model

To assess the effects of climate change and land management on SOM and DOC, we implemented the CENTURY 4.6 model (Parton et al., 1987, 1988, 1993) in a spatial setting using the PCRaster-Python framework (Karssenberg et al., 2010). The CENTURY model simulates the dynamics of carbon (C), nitrogen (N), phosphorus (P), and sulphur (S) in the top 20 cm of the soil profile on a monthly time step. The model has different plant production submodels for grassland, agricultural land, and forests, which are linked to a common soil organic matter submodel. It also includes a straightforward water budget submodel that calculates soil moisture content and drainage from the topsoil.

The concepts, variables, and parameters of the various submodels that constitute the CENTURY model have been described in detail by Parton et al. (1987, 1988, 1993) and Metherell et al. (1993). Below, we describe in brief the model setup and for details about the CENTURY model we refer to these references. In the CENTURY model, plant residues and soil organic carbon (SOC) are apportioned to various conceptual pools with different potential decomposition rates. Soil surface and root litter are classified into structural and metabolic material based on their lignin to nitrogen ratio. The first category represents material resistant to decomposition and the second category easily decomposable material. Likewise, the SOC is classified into three pools with different potential decomposition rates (an active pool with a turnover time of several years, a slow pool with a turnover time of 20-50 years, and a passive pool, with a turnover time of 400-2000 years). Decomposition products of each litter or SOC pool flows successively into one or two SOC pools with longer turnover times. The actual turnover rates of the various litter and SOC pools and

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