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## Cadmium, copper and zinc leaching and surface run-off losses at the Öjebyn farm in Northern Sweden—Temporal and spatial variation

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## Abstract

Quantitative estimates of cadmium (Cd), copper (Cu) and zinc (Zn) flows in percolating soil water and in surface run-off on arable land are rather scarce. Such information is essential for establishing reliable mass balances of trace metals, e.g. on field level, due to the fact that the trace metal outputs in water are quantitatively significant in field balance calculations. The aim of this study was to quantify trace metal leaching and surface run-off for a 5-year period at sites representing different soil types (Eutric Regosol, Thionic Gleysol, Dystric Cambisol) at the Öjebyn experimental farm in Northern Sweden. The metal outputs in water were estimated by combining chemical analysis of elements in soil water with simulations of water flow using the hydrological SOIL model. Average Cd, Cu and Zn concentrations (for three soils and 5 years) in soil water at 50 cm depth were 0.10, 21 and 11  $\mu$ g L<sup>-1</sup>, respectively. However, there was a large variation between years and soil types. The relative variation (coefficient of variation) between soil types for average Cd, Cu and Zn concentrations was in the range 20–60% depending on year. The average Cd, Cu and Zn output by leaching were  $0.22 \pm 0.20, 50 \pm 33$  and  $24 \pm 20$  g ha<sup>-1</sup> year<sup>-1</sup>, respectively, while the average outputs by surface run-off were  $0.13 \pm 0.09$ ,  $43 \pm 18$  and  $16 \pm 9$  g ha<sup>-1</sup> year<sup>-1</sup>, respectively. Both the simulated amount of water run-off and metal concentrations in the soil water had a significant impact on the variability of the metal flows. In this study the differences in metal concentrations in soil water due to differences in farming practice were much less important than the inter- and intra-field differences and temporal variability. The definition of the lowermost soil layer boundary of the system was found to have a great influence on the resulting metal mass balance.

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## 1. Introduction

Long-term accumulation of trace metals can affect soil fertility and product quality and promote trace metal leaching. Data on trace metal leaching and surface run-off within and from arable land are scarce ([Moolenaar and](#page--1-0) [Lexmond, 1998; McGrath, 2001](#page--1-0)). Such information is

essential both for establishing reliable mass balances of trace metals and for quantifying inputs to surface waters. Earlier studies have shown that trace metal outputs in water are quantitatively significant in field balances [\(Moolenaar and](#page--1-0) [Lexmond, 1998; Bengtsson et al., 2003](#page--1-0)). Römkens et al. [\(2002\)](#page--1-0) reported that the contribution of arable soils to the total load of cadmium (Cd), copper (Cu) and zinc (Zn) to Dutch surface waters could be as high as 60%.

Copper and Zn are essential micronutrients for plants and animals but may be harmful at excessively high concentra-

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There is a concern about excess inputs of trace metals such as Cd, Cu and Zn to Swedish agroecosystems [\(Andersson,](#page--1-0) [1992](#page--1-0)). Whether these inputs eventually lead to a net loss from soil is largely determined by the chemical characteristics of the elements in question. Management practices (e.g., crop rotation, tillage/no tillage), soil type and climate also affect the mobility [\(Watson and Atkinson, 1999](#page--1-0)). Soil pH is one of the most important variables controlling Cd and Zn solubility in soils. Metal sorption by soil decreases, and solution concentrations of the metal cations generally increase, with decreasing pH ([McLaren et al., 2004](#page--1-0)). [Gray et al. \(2003\)](#page--1-0) examined the relationship between the amount of Cd leached from soils and various soil properties to see whether they could predict the leaching potential. They found no statistically significant relationships between the amount of Cd leached and the soil properties investigated, including the Cd sorption and desorption characteristics. However, they concluded that the volume and pH of drainage water might be important factors controlling the amount of Cd leaching from these soils. Copper is known to form stable complexes with dissolved organic matter (measured as dissolved organic carbon; DOC). Movement through the soil of  $Cu^{2+}$  complexed by organic ligands could be expected in soils with high concentrations of DOC ([McLaren et al., 2004\)](#page--1-0).  $Cu^{2+}$  ions usually form inner-sphere complexes with organic matter even at low pH values (pH 4 according to [Kang et al., 1997\)](#page--1-0). Therefore, the leaching of organically complexed  $Cu^{2+}$  is important within a wide range of pH values.

Acid sulphate soils are common in arable land in for instance the coastal area of the Bay of Bothnia in northern Finland and Sweden. Metals bound in minerals in these soils are mobilised under oxic conditions when iron sulphides (FeS or FeS<sub>2</sub>) are oxidised and sulphuric acid ( $H_2SO_4$ ) and trivalent iron oxides/hydroxides are formed (Ö[born, 1994\)](#page--1-0). Enhanced metal leaching has been observed in drainage water from these soils and in surface water in areas where acid sulphate soils are frequent ([Andersson et al., 1988;](#page--1-0) Áström, 2001).

Metal input–output balances can be used to indicate whether metal accumulation or depletion occurs in soils, and to quantify potential metal inputs to surface waters. A soil system mass balance (or field mass balance) should include elements entering the soil and leaving it at a predefined soil depth, within the horizontal boundaries of a field. These balances may also take into account the internal element flows that occur within a farm [\(Oenema et al., 2003](#page--1-0)). In farming systems based on livestock (dairy or meat production), the main sources of trace metal inputs to soil are atmospheric deposition, mineral fertilisers, farmyard manure and urine (in which feed additives of metals constitute an important source) as well as liming substances (e.g. [Bengtsson et al., 2003](#page--1-0)). The main trace metal outputs are represented by water run-off, crop removal and

(particularly in a global perspective) soil erosion. A general soil system balance could be written according to Eq. (1)

$$
\Delta \text{ soil} = \sum \text{inputs} - \sum \text{outputs} \tag{1}
$$

If there is a positive element balance, i.e.  $\sum$  inputs  $>$   $\sum$  outputs, the element(s) must accumulate in the soil provided that all quantitatively significant inputs and outputs have been accounted for.

Leaching losses have sometimes been calculated indirectly, using a mass balance approach where the surplus, i.e. the amounts of elements not accounted for, have been assumed to be available for water run-off ([Moolenaar and](#page--1-0) [Lexmond, 1999](#page--1-0)). [Moolenaar and Lexmond \(1998\)](#page--1-0) calculated trace metal balances for the Minderhoudhoeve arable system in the Netherlands and estimated average leaching rates based on published values that ranged between 0.5 and 2.8, 5 and 87 and 2.6 and 88 g ha<sup>-1</sup> year<sup>-1</sup> of Cd, Cu and Zn, respectively. [Andersson \(1992\)](#page--1-0) calculated trace metal balances of arable soils for the whole of Sweden and used data from [Andersson et al. \(1988\)](#page--1-0) to estimate the leaching losses. In the latter study, the mean metal leaching in drainage water was measured during 2 years at five sites, the soils of which represented the whole texture range from sand to clay. Water run-off was monitored and samples were taken at least every 2 weeks during the maximum run-off periods. The average leaching from the drainage depth (approximately 1 m) was 0.06, 4.3 and 7.5 g ha<sup>-1</sup> year<sup>-1</sup> of Cd, Cu and Zn, respectively. [Van Driel and Smilde \(1990\)](#page--1-0) calculated heavy metal balances for Dutch agricultural soils. They estimated the leaching output by collecting and analysing drainage water, which had passed through a 50 cm column of loamy soil. The estimated mean leaching losses were 1, 75 and 120 g ha<sup>-1</sup> year<sup>-1</sup> for Cd, Cu and Zn, respectively. In a Swiss study of regional metal balances, [Keller and Schulin \(2003\)](#page--1-0) calculated trace metal leaching by applying a Freundlich adsorption isotherm model (via a pedotransfer function) to estimate concentrations of soluble Cd and Zn. They then used the precipitation surplus of the investigated region to obtain the metal leaching. According to their calculations the leaching of Cd was  $0.4$  g ha<sup>-1</sup> year<sup>-1</sup> with a standard deviation (S.D.) of 1.2 g ha<sup>-1</sup> year<sup>-1</sup> while the leaching of Zn was  $116 \text{ g ha}^{-1} \text{ year}^{-1}$  (S.D. 96), regardless of farm or crop type. [Eriksson \(2001\)](#page--1-0) reported Cd leaching from the plough layer in different Swedish soils within the range 0.12–0.46 g Cd ha<sup>-1</sup> year<sup>-1</sup> (based on Cd concentrations in saturation extracts; [Jansson, 2002\)](#page--1-0).

Several of the studies described above have more or less serious drawbacks. [Moolenaar and Lexmond \(1998\)](#page--1-0) pointed out that water samples from drains do not reflect the situation just below the plough layer or below the main rooting depth, which are two commonly used soil boundaries in balance calculations. Information from adsorption experiments is very difficult to extrapolate to a field situation and is not related to the actual water fluxes and conditions of equilibrium/ disequilibrium between the soil solution and sorption sites

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