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# Evaluation of approaches to calculate critical metal loads for forest ecosystems

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*Critical load methods for metals can be used to assess future risks due to metal inputs.*

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ABSTRACT

This paper evaluates approaches to calculate acceptable loads for metal deposition to forest ecosystems, distinguishing between critical loads, stand-still loads and target loads. We also evaluated the influence of including the biochemical metal cycle on the calculated loads. Differences are illustrated by examples of Cd, Cu, Pb and Zn for a deciduous forest on five major soil types in the Netherlands. Stand-still loads are generally lower than critical loads, which in turn are lower than the target loads indicating that present levels are below critical levels. Uncertainties in the calculated critical loads are mainly determined by the uncertainty in the critical limits and the chemical speciation model. Including the metal cycle has a small effect on the calculated critical loads. Results are discussed in view of the applicability of the critical load concept for metals in future protocols on the reduction in metal emissions.

## 1. Introduction

In Europe, there are large areas of forest soils that have been polluted by various metals through atmospheric deposition (Bergkvist et al., 1989) both from industrial emissions (Steinnes and Friedland, 2006) and traffic emissions (Spellerberg, 1998). In situations of extreme metal pollution, such as occurring in the Kola Peninsula in Russia, various impacts have been observed on forest ecosystems, including a decrease in forest productivity, vitality, biodiversity and stand structure (Chernenkova and Kuperman, 1999; Koptsik et al., 2004; Pukacki and Kaminska-Rozek, 2002). Elevated concentrations of metals have been found to damage root growth and functions both in pot experiments or hydroponic cultures (e.g. Arduini et al., 1992; Karolewski and Giertych, 1994; Rautio et al., 2005) and in the field situation (e.g. Helmisaari et al., 1999; Menon et al., 2007), impairing the water relations of affected plants (Menon et al., 2007; Poschenrieder and Barceló, 1999).

While the above mentioned effects are limited to severe metal pollution, impacts on soil organisms related to anthropogenic enhanced accumulation in the organic layer occur at much lower concentrations of metals (specifically cadmium and lead) in forests (Bringmark and Bringmark, 1995; Bringmark et al., 1998; Palmborg et al., 1998). This has lead to the derivation of critical limits for total metal contents, either in the humus layer or the mineral soil, based on No observed effect concentrations (NOECs) from laboratory studies with plants and soil organisms, such as soil microbiota and soil invertebrates (e.g. Tyler, 1992).The risks from metal pollution, however, depends on their availability which in turn is influenced by soil properties such as pH, CEC, clay and organic matter content, etc. (e.g. Boekhold et al., 1993; Groenenberg et al., in press; Sauvé et al., 2000). Recently, critical dissolved free metal ion (FMI) concentrations were thus derived in view of ecotoxicological effects on soil organisms and plants, based on NOEC soil data and soil properties (pH, organic matter content), affecting its availability (De Vries et al., 2007; Lofts et al., 2004).

Common practice in risk assessment is to compare present concentrations with critical concentrations at which adverse effects are not to be expected. With this approach it impossible to assess future risks due to metal inputs which may cause accumulation and possibly leads to future exceedance of critical limits. A method to assess future risks is the critical load approach. This approach determines the maximum level of constant atmospheric deposition that causes no or tolerable damage, (“long-term acceptable load” or “critical load”). It has been applied successfully in international negotiations on the reduction of atmospheric emissions of nitrogen and sulphur. Its potential use in international negotiations on the reduction in metal emissions is under debate. According to its definition, a critical load (CL) for metals equals the load resulting at steady-state in a concentration in a compartment (soil solid phase, soil solution, groundwater, plant, sediment, fish, etc.), that equals the critical limit set for metals, thus preventing ‘significant harmful effects on specified sensitive elements of the environment’ (Nilsson and Grennfelt, 1988).

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In defining a CL one aims at long-term sustainability of the ecosystem. In this context, sustainability can be defined as the situation where (i) no further net accumulation of metals occurs (stand-still load) or (ii) accumulation of metals is below critical limits in defined ecosystem compartments (e.g. soil solid phase, soil solution; CL). The method to calculate CLs of metals is based on the balance of all relevant metal fluxes in and out of a considered ecosystem in a future steady-state situation. First approaches were described in a "manual" for calculation of critical loads of metals in terrestrial ecosystems (De Vries and Bakker, 1998), including various approaches to calculate critical loads. In 2004, The UN-ECE ICP Modelling & Mapping published a formal manual (UBA, 2004), focusing on Cd, Pb and Hg, with related background report describing the various details (De Vries et al., 2005). Because of the time it takes to reach steady-state concentrations, the use of dynamic model approaches should be considered (Lofts et al., 2007). A particular example of a dynamic model approach is the calculation of a target load, which is defined as the load resulting in a concentration in a compartment that equals the critical limit set for metals within a defined time period.

Maps of critical loads for metals have been published for Europe (2007) and Canada (Doyle et al., 2003), using various approaches, but an evaluation of the various concepts in terms of their assumptions and effects has not yet been published. This paper aims at such a review. It summarises and evaluates possible approaches to calculate acceptable loads for metal deposition to forest ecosystems, distinguishing between critical loads, stand-still loads and target loads. These approaches differ with respect to the criterion used on which the maximum load is based in either (i) using present metal concentrations for the soil solid phase (stand-still loads) or effect based critical limits (critical loads and target loads) and (ii) with respect to the timescale used, applying a steady-state approach (infinite timescale) for CLs and stand-still loads and a finite timescale with a dynamic approach to calculate target loads. We also evaluate the influence of the complexity of the models with respect to inclusion of the plant metal cycle (metals in litterfall and uptake to replenish litterfall) on the calculated loads. Differences are illustrated by calculation examples of Cd, Cu, Pb and Zn for a generic deciduous forest, where the metal load is due to atmospheric deposition only. Results are discussed in view of the applicability of the CL concept for metals in future protocols on the reduction in metal emissions.

## 2. Calculation methods

### 2.1. Steady-state model approach to assess stand-still loads and critical loads

Stand-still loads and CLs are calculated with a mass balance model on the basis of a present or critical metal leaching rate, respectively, which in turn is defined by a present or critical metal concentration in soil solution.

#### 2.1.1. Steady-state mass balance equation

In deriving CLs use was made of a steady-state mass balance model according to:

$$M_{td} = -M_{lf} + M_{fu} + M_{ru} + M_{le} \tag{1}$$

where $M_{td}$ is the total load of metal M by atmospheric deposition and where $M_{lf}$, $M_{fu}$, $M_{ru}$ and $M_{le}$ are the fluxes of metal M by litterfall, foliar uptake, root uptake and leaching, respectively (all in mg $m^{-2}$ $yr^{-1}$). Apart from the assumption of a steady-state situation, implying that the calculated CL is intended to be valid for an indefinitely long period, various assumptions apply to the model, such as: (i) the soil system is homogeneously mixed which implies that the CL can only be calculated for a distinctive homogeneous layer, (ii) the soil is in an oxidised state and metal partitioning can be described with equilibrium sorption and complexation with DOC, (iii) transport of water and metals only takes places in vertical direction (no seepage flow, surface runoff and bypass flow), (iv) metal weathering and erosion is not considered and (v) the metal deposited is chemically reactive and is 100% available for exchange with the solution phase. The related limitations due to these assumptions are discussed in De Vries et al. (2005).

#### 2.1.2. Calculation of metal fluxes

The various fluxes included in Eq. (1) are litterfall, foliar uptake, root uptake and leaching. The flux of metals in litterfall is calculated as a function of the total metal deposition according to:

$$M_{lf} = M_{lf}(nd) + frM_{lf} \cdot M_{td} \tag{2}$$

Where $M_{lf}(nd)$ is the flux of metal M in litterfall at negligible deposition (mg $m^{-2}$ $yr^{-1}$) and $frM_{lf}$ is the litterfall fraction (–). Foliar uptake of metals is described as a fraction of the total deposition according to:

$$M_{fu} = M_{fu}(nd) + frM_{fu} \cdot M_{td} \tag{3}$$

Where $M_{fu}(nd)$ is the foliar uptake flux of metal M at negligible deposition (mg $m^{-2}$ $yr^{-1}$) and $frM_{fu}$ is a foliar uptake fraction (–). Root uptake in the layer for which the CL is calculated is described as a fraction of the maintenance uptake, to resupply the amount released by litterfall minus foliar uptake, and uptake due to net growth according to:

$$M_{ru} = fr_{ru} \cdot (M_{lf} - M_{fu} + M_{gu}) \tag{4}$$

where $fr_{ru}$ is a depth dependent cumulative root uptake fraction in the root zone (–) and $M_{lf} - M_{fu}$ is the flux of metal M needed to maintain a constant metal concentration in the foliage (mg $m^{-2}$ $yr^{-1}$). At the bottom of the root zone, $fr_{ru}$ equals 1. Growth uptake is derived by the multiplication of the forest yield and the metal content in stem wood, according to:

$$M_{gu} = Y \cdot ctM_{st} \tag{5}$$

Where Y is yield (kg $ha^{-1}$ $yr^{-1}$) and $ctM_{st}$ is the metal content in stem wood (mg $kg^{-1}$). Leaching is described as the product of water flux and soil solution concentration according to:

$$M_{le} = Q_{le} \cdot [M]_{tot,ss} \tag{6}$$

Where $Q_{le}$ is the water flux leaching from the soil (m $yr^{-1}$) and $[M]_{tot,ss}$ is the total metal concentration in soil solution (mg $m^{-3}$). The water flux at steady-state is calculated according to:

$$Q_{le} = (1 - fr_i) \cdot P - E_{se} - fr_{ru} \cdot E_t \tag{7}$$

Where P is precipitation (m $yr^{-1}$), $E_{se}$ is soil evaporation (m $yr^{-1}$) and $fr_i$ is interception fraction (–).

#### 2.1.3. Calculation of critical loads

The steady-state dissolved concentration of a certain metal can be calculated by combining the mass balance equation (Eq. (1)) with Eqs. (2)–(7) according to:

$$[M]_{tot,ss} = (fr_{re} \cdot M_{td} - M_{re})/Q_{re} \tag{8}$$

With:

$$fr_{re} = (1 - frM_{fu}) - fr_{ru} \cdot (frM_{lf} - frM_{fu}) + frM_{lf} \tag{9}$$