ELSEVIER

Journal of Hazardous Materials 61 (1998) 99–106

JOURNAL OF HAZARDOUS MATERIALS

Calculation and mapping of critical loads for heavy metals and persistent organic pollutants for Dutch forest soils

W. de Vries [a,*], D.J. Bakker [b], J.E. Groenenberg [a], G.J. Reinds [a], J. Bril [c], J.A. van Jaarsveld [d]

[a] *DLO Winand Staring Centre for Integrated Land, Soil and Water Research, P.O. Box 125, 6700 AC Wageningen, Netherlands*

[b] *TNO Institute of Environmental Sciences, Energy Research and Process Innovation, P.O. Box 57, 1780 AB Den Helder, Netherlands*

[c] *DLO Institute for Agrobiological and Soil Fertility Research, P.O. Box 30003, 9750 RA Haren, Netherlands*

[d] *National Institute of Public health and the Environment, P.O. Box 1, 3720 BA Bilthoven, Netherlands*

Abstract

In order to enable application of the critical load approach in the international heavy metal and POP abatement policies, critical loads of lead, cadmium, copper, zinc, lindane and benzo(*a*)pyrene have been calculated for Dutch forest soils. The environmental risk of atmospheric deposition of these substances on Dutch forest soils has been described in terms of the difference between the calculated present atmospheric loads and critical loads. Results indicate that the calculated critical loads strongly depend on the type of environmental quality criterium which serves as a basis for the calculation. More information on the quality criteria to be used is thus essential to gain insight in the risk of the loads of heavy metals and POPs on the terrestrial ecosystem.

Keywords: Heavy metal; Organic pollutant; Dutch forest soil

1. Introduction

Concern on the dispersion and impacts of heavy metals and persistent organic pollutants (POPs) is large. For example, more than twenty working groups and task forces in Europe are presently working on policies to prevent air pollution by these substances. In the past years, several studies have therefore been carried out to assess

* Corresponding author.

0304-3894/98/$19.00
PII S0304-3894(98)00113-7

critical loads of heavy metals and persistent organic pollutants (POPs) for forest soils and surface waters both on a national scale (ATMODEP study [1]) and a European scale (ESQUAD study [2–4]). Based on these studies, DLO Winand Staring Centre and TNO have developed draft manuals for the calculation of critical loads of heavy metals [5] and POPs [6] on soils and surface waters.

Here, we report the results of a study in which critical loads of the heavy metals lead (Pb), cadmium (Cd), copper (Cu) and zinc (Zn) and of the POPs lindane and benzo(*a*)pyrene (B(*a*)P) have been calculated for Dutch forest soils. An indication of the environmental risk of the pollutants is presented by the difference between present loads and critical loads.

2. Methods and data

Critical loads for the heavy metals and POPs considered were calculated for approximately 18 000 forest soil combinations using detailed soil and vegetation maps. Calculations were made for the organic layer and the mineral topsoil (top 10 cm) of major forest soil combinations in each 5 km × 5 km grid cell over the Netherlands. Information on the atmospheric deposition for each grid was derived by using the emission/deposition model TREND. The atmospheric deposition on forests within a grid was then derived by multiplying this deposition with a filtering factor, thus, correcting for forest filtering of dry deposition (see Ref. [5]). The critical load of pollutants has been defined as the deposition level that will ultimately lead to pollutant concentrations in various ecosystem compartments (e.g. soil, groundwater, and vegetation) that equal given environmental quality criteria. Information on the environmental quality criteria, models and input data is summarized below.

2.1. Environmental quality criteria

2.1.1. Heavy metals

With respect to heavy metals, a distinction can be made in effects on soil micro organisms/soil fauna, vascular plants, terrestrial fauna and humans. In this context,

Table 1
Environmental quality objectives for total metal concentrations in soil, soil solution and groundwater (after Ref. [5])

Heavy metal	Soil[a] (mg kg^{-1})	Soil solution[b] (mg m^{-3})	Groundwater[c] (mg m^{-3})
Pb	50 + L + H	100	15
Cd	0.4 + 0.007(L + 3H)	20	1.5
Cu	15 + 0.6(L + H)	20	15
Zn	50 + 1.5(2L + H)	200	150

[a] Based on background concentrations in relatively unpolluted areas. Values depend on the clay (lutum) content (L in %) and the organic matter (Humus) content (H in %).
[b] Based on ecotoxicological data from laboratory experiments, with culture solutions.
[c] Based on background concentrations in relatively unpolluted areas.

criteria can be chosen for the total heavy metal concentrations in (i) the organic layer or the mineral top soil, to protect soil organisms such as earthworms that consume soil solid material, (ii) the soil solution, to avoid effects on vascular plants, such as elevated uptake and reduced growth, (iii) groundwater, to avoid drinking water contamination and (iv) crops/foliage, to avoid toxic effects on humans consuming the crops [5]. In this study, use was made of critical metal concentrations in the soil solid phase, the soil solution and groundwater in order to calculate critical loads (Table 1).

The default method is based on the Dutch target values for total soil concentrations that are used in the environmental policy (based on background concentrations).

2.1.2. Persistent organic pollutants

The critical load calculations for POPs were based on the Dutch target values for the total soil concentrations of B(*a*)P (0.025 mg kg^{-1}) and lindane (0.00005 mg kg^{-1}). These target values are used in the environmental policy and based on ecotoxicological effects.

2.2. Calculation methods

The calculation methods used are all based on several assumptions including equilibrium partitioning, a homogeneously mixed soil system and the occurrence of oxidized circumstances. This implies that the models can only be applied to homogeneous humus layers and upper mineral soil layers of well-drained soils. More information on the assumptions and inherent limitations is given in Ref. [5].

2.2.1. Heavy metals

The 'default' model used for heavy metals is a steady-state model containing (i) a mass balance equation describing the input–output fluxes of heavy metals, combined with (ii) rate-limited process descriptions for metal cycling due to litterfall and plant uptake, weathering, surface run-off, bypass flow, leaching and (iii) equilibrium descriptions for adsorption and complexation processes, that determine the partitioning of heavy metals between the different soil phases. In this study, the effects of surface run-off and bypass flow were neglected. This implies that the critical load equalled the net metal loss by above- and below-ground forest uptake corrected for litterfall, minus the metal weathering rate (negligible in the humus layer) plus a critical metal leaching rate.

In order to calculate a critical leaching rate, the critical total metal concentration in the soil solution must be known. This may be derived directly (Section 2.1) or indirectly from critical total metal concentrations in soil. The latter approach (default model) requires the inclusion of adsorption and complexation processes. The adsorption/complexation calculations applied in this study were [5] as follows.

(i) A simple linear equilibrium partition equation that relates the total metal content in the soil to the total metal concentration in the soil solution. In this approach, adsorption and complexation are lumped into one partition coefficient.

(ii) A nonlinear (Freundlich) equilibrium adsorption equation partitioning the heavy metals over the solid (adsorbed) phase and the soil solution (dissolved free concentra-

tion), combined with a linear complexation description partitioning the heavy metals over dissolved organic carbon (DOC) and the soil solution. In this approach, complexation of heavy metal with inorganic anions is considered negligible compared to complexation with DOC.

The time-period to reach steady-state in the mineral soil can be very long for heavy metals in the mineral soil. Therefore, an alternative (semi-)dynamic method was also applied. In this approach, a critical metal accumulation rate (the accumulation of heavy metals in soils until a given critical content in a 100-year time period) was added to the calculated critical load. Using this approach, the present metal status is affecting the critical load, or target load. To avoid confusion with the critical loads calculated with a steady-state model, the term target load is preferred since a definite time target (100 years) is included in the calculation. When environmental quality criteria for the soil solution were used, the associated critical metal concentration in the solid phase was calculated with the adsorption and complexation reactions described above.

2.2.2. Persistent organic pollutants

The 'default' model used for persistent organic pollutants is a steady-state mass balance model containing process descriptions for (bio)degradation, volatilization, plant uptake, surface run-off, bypass flow, leaching and equilibrium partitioning between the different soil phases (specifically soil organic carbon and dissolved organic carbon). As with heavy metals, in this study, the effects of surface run-off and bypass flow were neglected. Furthermore, plant uptake was assumed to be negligible. Volatilization of lindane was also disregarded because the effect of this process was already taken into account in the calculation of the present net atmospheric deposition of lindane, which was used to determine the possible excess load. This implies that the critical load was determined by the biodegradation rate.

2.3. Input data

Input data were derived as a function of the substance considered, location (receptor area) and receptor (the combination of land use and soil type). Regarding the type of forest, a distinction was made between pine forests, spruce forests and deciduous forests. Seven major soil groups were distinguished (calcareous and non-calcareous sandy soils, the latter being divided in mineral-poor and mineral-rich soils, loess soils, calcareous and non-calcareous clay soils and peat soils) on the basis of the soil map of the Netherlands 1:50 000. A summarizing overview of the data acquisition approach is given in Table 2. A more detailed overview of the various input data is given in Refs. [5,6].

The soil parameters used for the heavy metal model (pH, organic carbon concentration in the soil and soil solution, clay content, CEC and the dissolved Ca concentration of both the organic layer and mineral layer) and the additional soil parameters used for the POP model (bulk density of both the organic layer and mineral layer and the thickness of the organic layer) were based on available data for 200 forested stands for the year 1995. Values for each forest type/soil type in each 5 km $\times$ 5 km grid cell were derived by regression relationships with available data on tree species, soil type,

Table 2
Data acquisition approaches for input data

Input data	Data acquisition approach
Precipitation	Estimate per grid, interpolating data of weather stations
Interception	Relationship with precipitation amount and forest type
Transpiration	Calculated as a function of precipitation, forest type and soil type
Litterfall/foliar uptake	Relationship with deposition and forest type
Root uptake	Relationship with deposition and forest type
Adsorption	Relationship with soil characteristics such as pH, organic matter content, clay content and CEC based on literature data
Complexation	Relationship with pH based on literature data
Biodegradation	Literature data

modelled atmospheric deposition level, etc. A similar approach was used to derive the present heavy metal contents, used in the (semi-)dynamic approach [7].

3. Results and discussion

3.1. Heavy metals

Calculated critical loads based on a steady-state model strongly depended on the environmental quality criterium used and the soil type considered as illustrated in Table 3.

Use of a critical metal concentration in the soil (the 'default' steady-state method) resulted in higher critical loads for (acid) sandy soils and lower critical loads for calcareous soils (Table 3). This is because the critical load increases with a decrease in adsorption constant (occurs in the direction from calcareous clay soils to acid sandy soils), due to a decrease in metal accumulation. The adverse effects of elevated dissolved metal concentrations on vegetation, soil fauna and groundwater (the latter through metal leaching) are not accounted for in this approach.

Table 3
Median critical loads of heavy metals for the mineral topsoil as a function of the environmental quality criterium

Soil	Critical load ($mg\ m^{-2}\ yr^{-1}$)							
	Soil criterium				Soil solution criterium			
	Pb	Cd	Cu	Zn	Pb	Cd	Cu	Zn
Sand	7.1	4.1	208	930	44	9.0	12	106
Loess	8.2	3.6	357	602	54	13	16	131
Clay	2.6	0.70	121	86	46	12	12	112
Peat	15	1.4	548	930	50	12	14	121
Calcareous	0.03	0.07	15	18	48	13	13	119
All	7.0	4.0	205	907	45	9.3	12	108

Table 4
A comparison of median values of modelled present loads (year 1995) and critical loads of heavy metals for the mineral topsoil related to various environmental quality criteria

Heavy metal	Present load (mg m^{-2} yr^{-1})		Critical load (mg m^{-2} yr^{-1})		
	Grid	Forests[a]	Soil	Solution	Groundwater
Pb	4.5	9.1	7.0	45	6.1
Cd	0.10	0.26	4.0	9.3	0.79
Cu	0.93	2.1	205	12	9.2
Zn	4.0	8.4	907	108	79

[a]The modelled deposition on a grid multiplied by a forest filtering factor (see Section 2).

Use of critical metal concentrations in the soil solution caused higher critical loads for Pb and Cd and lower critical loads for Cu and Zn as compared to the soil criterion. Furthermore, use of the soil solution criterium resulted in only small differences between the different soil groups (Table 3). This can be expected since adsorption and complexation descriptions are not needed when using critical dissolved metal concentrations, which implies that the critical load mainly depends on the precipitation excess.

Comparison of present loads on forests and critical loads (Table 4) showed that exceedances hardly ever occurred for Cd, Cu and Zn in relation to both the soil and soil solution criterium. Use of a groundwater criterium sometimes caused exceedances for Cd but not for Cu and Zn. For those metals, calculated critical loads were quite comparable to those calculated with the soil solution criterium (compare the results in Table 4 and the quality criteria in Table 1). Unlike the other metals, present loads of Pb often exceeded the critical loads when using the soil criterion (Table 4), especially in calcareous soils, clay soils and peat soils. Use of the groundwater criterion also caused exceedances at more than 50% of the forested area. Note, however, that the results for the groundwater criterium are only indicative, since the assumption of a homogeneous soil system is not valid for a soil profile with a depth until groundwater level.

The alternative (semi-)dynamic approach showed that the highest target loads were calculated for calcareous clay soils and lowest for acid sandy soils (Table 5). The reason for this is that for a given critical metal concentration in the soil solution, the associated critical metal content in the solid phase increases when the adsorption constant is higher.

Table 5
Median target loads for heavy metals for the mineral topsoil

Soil group	Target load (mg m^{-2} yr^{-1})			
	Pb	Cd	Cu	Zn
Sand	333	10	108	11
Loess	376	17	117	9.3
Clay	1055	26	300	9.2
Peat	75	14	111	12
Calcareous	5424	97	1031	36
All	341	11	11	111

Table 6
A comparison of median values of modelled present loads (year 1995) and critical loads of lindane for the organic layer

Soil group	Present load (g ha^{-1} yr^{-1})	Critical load (g ha^{-1} yr^{-1})	Excess load (g ha^{-1} yr^{-1})
Sand	1.22	0.80	0.40
Loess	1.72	0.36	1.39
Clay	1.30	0.33	0.89
Peat	0.91	0.51	0.38
Calcareous	0.92	0.16	0.75
All	1.26	0.79	0.46

This in turn lead to an increase in the acceptable accumulation rate during the 100-year period, since the difference between the critical and actual metal content increased. For Cd, Cu and Zn, the calculated target loads were comparable to the critical loads related to the soil solution criterion, especially for the sandy soils. For Pb, however, the target load was much higher (compare Tables 4 and 5).

3.2. Persistent organic pollutants

Comparison of present loads and critical loads for lindane and B(*a*)P showed that an exceedance of the critical load only occurred for lindane in the organic layer, as illustrated in Table 6. Results show that the critical loads for lindane (and POPs in general) were lowest in places with thin organic layers, such as calcareous soils and highest in places with thicker organic layers, such as acid sandy soils (see Table 6).

Present atmospheric deposition of lindane (ranging from approximately 0.5 to 2.0 g ha^{-1} yr^{-1}) exceeded the critical load, based on the maximum permissible concentration (MPC), for the organic layer of almost all Dutch forest soils. The calculated excess load ranged in 90% of the cases between from 0.04 and 1.14 g ha^{-1} yr^{-1}. The critical load of lindane in the upper 10 cm of the mineral forest soil ranged from 5 to 20 g ha^{-1} yr^{-1} and were therefore exceeded nowhere.

The calculated critical load of B(*a*)P for the organic layer ranged in 90% of the cases between 7 and 21 g ha^{-1} yr^{-1}. As the present atmospheric deposition of B(*a*)P was calculated to range from 0.5 to 5.0 g ha^{-1} yr^{-1}, it was concluded that the critical load is not exceeded in the organic layer of Dutch forest soils. The critical load for the uppermost 10 cm of the Dutch mineral forest soil was much higher than those for the organic layer, so excess loads did not occur here either.

4. Conclusions

(1) Calculated critical loads based on a steady-state model strongly depend on the type of environmental quality criterium used. More information on the quality criteria to be used is thus essential to gain insight in the risk of the loads of heavy metals and POPs on the terrestrial ecosystem.

(2) Present atmospheric loads on Dutch forests mainly exceed critical loads of Pb for soil and groundwater and of lindane for soil (organic layer). Results are, however, strongly influenced by the uncertainty in descriptions and input data for adsorption/complexation of heavy metals and biodegradation of POPs.

References

[1] D.J. Bakker, The influence of atmospheric deposition on soil and surface water quality in The Netherlands, Delft, The Netherlands, TNO Institute of Environmental Sciences, Report R95/013, 1995, 84 pp.
[2] K.D. van den Hout (Ed.), The impact of atmospheric deposition of non-acidifying pollutants on the quality of European forest soils and the North Sea, Delft, The Netherlands, TNO Institute of Environmental Sciences, Report R93/329, 1994, 143 pp.
[3] D.J. Bakker, K.D. van den Hout, G.J. Reinds, W. de Vries, Critical loads and present loads of lindane and benzo(*a*)pyrene for European forest soils, Delft, The Netherlands, TNO Institute of Environmental Sciences, Report R94/288, 1994, 44 pp.
[4] G.J. Reinds, J. Bril, W. de Vries, J.E. Groenenberg, A. Breeuwsma, Critical and present loads for cadmium, copper and lead for European forest soils, Wageningen, The Netherlands, DLO Winand Staring Centre, Report 96, 1995, 91 pp.
[5] W. de Vries, D.J. Bakker, Manual for calculating critical loads of heavy metals for soils and surface waters, Preliminary guidelines for environmental quality criteria, calculation methods and input data, Wageningen, The Netherlands, DLO Winand Staring Centre, Report 114, 1996, 173 pp.
[6] D.J. Bakker, W. de Vries, Manual for calculating critical loads of persist organic pollutants for soils and surface waters, Preliminary guidelines for environmental quality criteria, calculation methods and input data, Delft, The Netherlands, TNO Institute of Environmental Sciences, Report R96/509, 1996, 114 pp.
[7] E.E.J.M. Leeters, H. Hartholt, W. de Vries, L.J.M. Boumans, Effects of acid deposition on 150 forest stands in the Netherlands. 4. Assessment of the chemical composition of foliage, mineral soil, soil solution and groundwater on a national scale, Wageningen, the Netherlands, DLO Winand Staring Centre for Integrated Land, Soil and Water Research, Report 69.4, 1994, 156 pp.