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Critical Loads of Lead and Cadmium for Different Type of Forest and Aquatic Ecosystems at the Petrohan LTER Site, Bulgaria

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Abstract

The current study was designed to determine the critical loads of lead (Pb) and cadmium (Cd) for two types of vegetation (broadleaves and coniferous) and freshwaters (stream water and reservoir), in order to evaluate the effect of Cd and Pb deposition on investigated ecosystems by comparative simultaneous study at a mountainous LTER site in Bulgaria. Bulk precipitation and throughfall chemistry were monitored during the period 2005-2010 at two forest stands (*Fagus sylvatica* and *Picea abies*). Monitoring of water, sediment and bulk precipitation chemistry at two water bodies (stream and reservoir) were carried out too. The removal of Pb and Cd by the biomass and their vertical migration with drainage water was determined. It has been found that the tolerance of surface water to the pollution of Pb has been comparable with forests but aquatic ecosystems are more sensitive to the deposition of Cd due to the low values of critical loads meaning that critical loads for forest can not protect the surface water from the same catchment.

Steady state critical loads of Pb and Cd for forests based on human health approach have remained stable and have not been exceeded during the study period. Real risk of harmful effect in the future has been discovered for surface waters because of higher input of Cd than critical loads obtained by ecotoxicological approach.

Key words: beech, cadmium, critical loads, lead, reservoir, spruce, stream water

Introduction

Since forest ecosystems are associated with many ecosystem functions related to biodiversity, provision of forest products, water protection and carbon sequestration, it is crucial to know the amount of pollutant deposition above which these ecosystems would be damaged. Critical loads have been defined as quantitative estimates of an exposure to one or more pollutants below which significant harmful effects on specified sensitive elements of the environment do not occur according to present knowledge (Nilsson and Grennfelt 1988).

Critical loads can be used to determine the sensitivity of a given receptor. When the value of the critical load is high, the receptor is more tolerant and less sensitive to the pollutant of concern. In this case the receptor can withstand large amounts of pollutant deposition without showing harmful effects. The risk of damage can be assessed by means of critical loads exceedances by current deposition rates of pollutants. This approach is very effective because it can inform regional emission control policies. This study proposes modeling methodologies that have the capability of providing effect-based support to policies that are aimed at mitigating air pollution and the change of biodiversity and climate in an integrated manner. A lot of results for calculating and mapping critical loads of heavy metals and their exceedances have been published during the last 10 years but for terrestrial ecosystems only. Except monitoring data on concentration of heavy metals in surface waters and sediments (Wilkens 1995; Skjelkvale and Ulstein 2002) and methodological manuals (Posch *et al*. 2003; Modeling and mapping manual 2004) there are not any comparative investigations devoted to the critical loads of Pb and Cd for aquatic ecosystems in the references.

The aim of this study was to carry out comparative investigations on the sensitivity and risk of damage by heavy metal pollution in two types of forest (beech and spruce) as well as two types of surface water (stream and reservoir) at the LTER Petrohan site in the Western Balkan Mountain in Bulgaria by means of critical load calculations and their exceedances based only on simultaneous measured data. From this point of view the following tasks have been taken into account:

1 Determination of Pb and Cd deposition rates in the precipitation at the LTER Petrohan site;
2 Collection of measured data needed for the calculation of local critical loads of Pb and Cd for forests (Fagus silvatica and Picea abies L. Karst) and surface water (stream and reservoir) in order to assess their sensitivity to heavy metal pollution;
3 Assessing the risk of harmful effects and damages to forests and surface waters by computing the exceedances of critical loads of Pb and Cd.

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Materials and methods

Collectors for bulk precipitation and throughfall under beech and spruce crowns have been installed at two plots at the Petrohan site in beech (Fagus silvatica) and spruce (Picea abies L. Karst.) forest ecosystems (Fig. 1).

The bulk precipitation and throughfall has been collected with 6 (for bulk precipitation) and 12 (for throughfall) continuously open plastic funnels (1 m above the ground) per plot with an individual collecting surface of 314 cm^2 in polyethylene bottles stored in the upper soil layer. Individual water samples of bulk precipitation, throughfall and surface water have been collected fortnightly after measuring the pH and water volume in the field, and analyzing the Pd and Cd concentration in each individual sample by inductive coupled plasma atomic emission spectrometry (ICP-OES Vista MPX, Varian Inc, Australia). All samples have been stored in the refrigerator at 1-4°C until analyses, preserved with HNO_3 and pre-concentrated 50 times initially and diluted with concentrated HNO_3 acid to a volume of 10 ml. The values of pH are determined on separate aliquots of the unfiltered sample the same day of sampling in the field. The remaining samples are filtered through a 0.45 μm membrane filter in order to remove any solid materials and to stabilize them for the subsequent analyses. Pb and Cd uptake by the biomass has been derived by multiplying the content of these metals in the beech and spruce stem, measured by inductive coupled plasma atomic emission spectrometry, after cutting 10 representative trees in the buffering area, with the annual growth Yveg (kg.$ha^{-1}yr^{-1}$) determined as follow:

$$Yveg = Va\ \rho / a,$$

where: Va – the mean volume of all trees at the catchment area in $m^3\ ha^{-1}\ yr^{-1}$;
ρ – the wood density in kg m^{-3}; measured by means of the Tsonmis method (1991)
a – the age of trees in years.

Stem samples were digested with a concentrated acid mixture of HNO_3- $HClO_4$ (3:1) and heated at 160°C for 5 hours. After cooling, the extract was diluted, filtered, and made up to 25 ml with 5 % HNO_3. The concentrations of Pb and Cd in the extract were determined by inductive coupled plasma atomic emission spectrometry (ICP-OES Vista MPX, Varian Inc, Australia).

The runoff of water has been measured as mean annual values for a period of 20 years in grid cells of 10 x 10 km for the entire country (Kehayov 1986). The method is based on a splitting of river hydrographs, hydrogeological parameters of the underground water bodies, measurements of the mineral runoff between neighboring hydrometric sites, infiltration of the water source etc. Sediments in surface water were wet digested using mixture of concentrated HCl and HNO_3 acids (3:1) before analyzing the concentration of Pb and Cd by inductive coupled plasma atomic emission spectrometry (ICP-OES Vista MPX, Varian Inc, Australia).

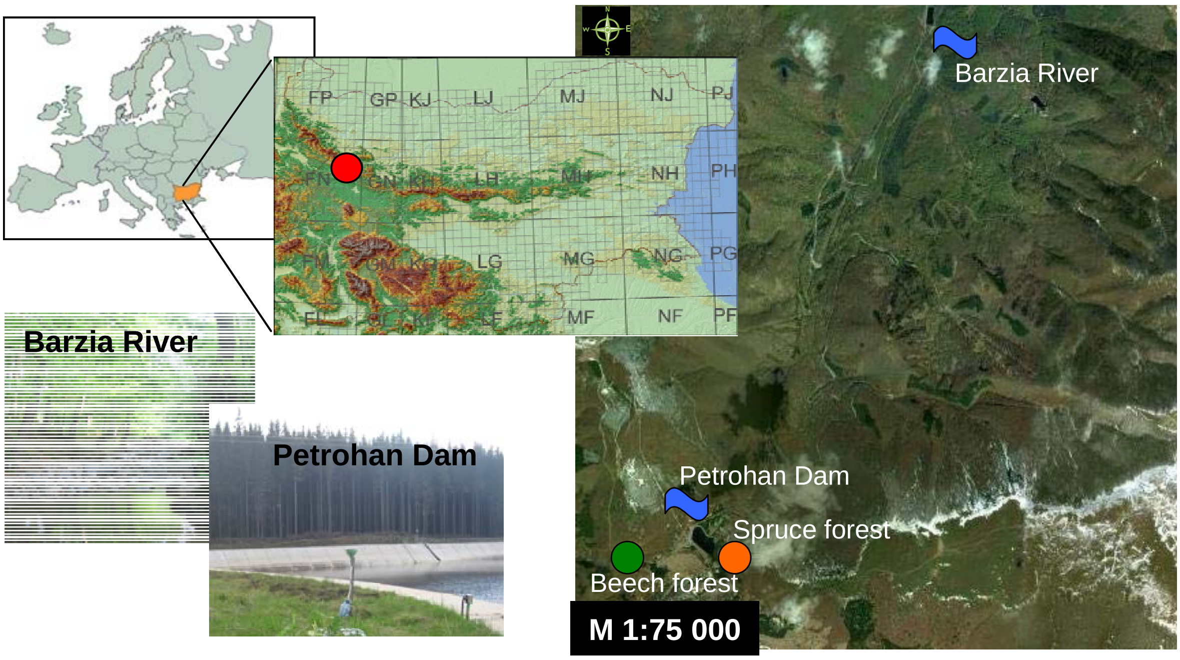

Fig. 1. Position of the Petrohan site and experimental plots.

There is agreement that the effect of heavy metals on forests is in better correlation with the metal concentration in the soil solution than in the soil itself (Crommentuijn *et al.* 1997, Lamersdorf *et al.* 1991, Tyler 1992, Wilkens 1995). Critical loads for terrestrial ecosystems addressing human health effects could be calculated, either in view of not violating food quality in crops or in view of ground water protection regarding its potential use as drinking water. (Modeling and Mapping manual 2004).

The effect-based steady-state mass balance model was used to calculate the critical loads of Pb and Cd. Human health effects of deteriorating forest ecosystem services through heavy metal deposition that represented here as a change in the quality of drinking water has been taken into account in our study. The model implies that the critical load equals the net uptake by the forest growth plus an acceptable metal leaching rate, according to the follow equation:

CL(M)=Mu+Mle(crit)

where: CL(M) = critical load of a heavy metal M (Pb or Cd) (g ha^{-1} yr^{-1});
Mu = Net uptake of metal M in the vegetation under critical load conditions (g ha^{-1} yr^{-1});
Mle(crit) = Critical leaching of a metal M (g ha^{-1} yr^{-1}).
For a more detailed description of methods see Modelling and Mapping Manual (2004) (www.icpmapping.org)

The metal net uptake in the vegetation was calculated by multiplying the annual yield by the metal content in the stem of trees as follow:

Mu =Yveg [M]veg,

where: Yveg = Net increment of stem (dry weight) (kg $ha^{-1}yr^{-1}$);
[M]veg = Metal content in the stem of trees (g kg^{-1}dw).

The critical leaching flux of heavy metals Mle(crit) (g ha^{-1} yr^{-1}) was calculated according to the follow equation:

Mle(crit) = cle Qle [M]ss(crit),

where: Qle = Flux of drainage water (m yr^{-1}); [M]ss(crit) = Critical limit for the total concentration of heavy metal. In agreement with the joint task force on health aspects of air pollution it was decided to use internationally accepted critical limits to protect ground water quality (Recommendations for maximum metal concentration in drinking water of World Health Organization: for Cd 3 mg m^{-3} and for Pb 10 mg m^{-3}) (WHO 2004); cle = 10; this is a factor for appropriate conversion of flux units from mg m^{-2} yr^{-1} to g ha^{-1} yr^{-1}.

Critical loads of Pb and Cd for ecotoxicological effects on terrestrial ecosystems were calculated for Bulgarian Forests by the Coordination Centre and were compared to the human health- drinking water values (Slootweg *et al.* 2005). It was found that both values for the entire forested area of Bulgaria were between 4 and 6 g $ha^{-1}yr^{-1}$. Later critical loads of Pb and Cd for Bulgarian forests based on eco-toxicological effects on soil organisms were published in the CCE Status Report 2008 (Hettelingh *et al.* 2008).

The differences between the monitored and the critical possible loads of Pb and Cd by present atmospheric depositions as exceedances of critical loads were calculated by the following equation:

CL(M)ex = PL(M) – CL(M),

where: PL(M) = Present deposition of Pb or Cd; CL(M) = Critical load for Pb and Cd, g ha^{-1} yr^{-1}. As with terrestrial ecosystems, the critical loads of Pb and Cd for surface fresh waters is the acceptable total load of heavy metals inputs corresponding to the sum of tolerable outputs by harvest within, and outflow from a catchment according to the follow equations:

CL(Pb, Cd) = Mu + Mlo (crit),

The uptake into harvestable parts of plants (Mu) is calculated in analogy to terrestrial ecosystems, while harvest in this context means in general the harvest of wood in forested catchments (Modeling and Mapping manual 2004; Schutze and Hettelingh 2006):

Mu =Yveg [M]veg,

In the calculation of the critical outflow of metals the lateral water flux off the catchment is multiplied with the critical concentration in the surface water:

In order to calculate critical loads of metals for fresh water ecosystems it is necessary to know the total aqueous concentration, i.e. the concentration of dissolved metal and of metal bound to suspended particle matter (SPM). The critical lateral outflow Mlo(crit) (g ha^{-1} yr^{-1}) is described as the product of the flux of water and the critical limit for the total concentration of the heavy metal in the surface water according:

Mlo(crit) = 10 Qlo[M]tot sw(crit)

where: Qlo = lateral outflow flux of water from the aquatic system (m yr^{-1});
[M]tot sw(crit)= total aqueous metal at the critical limit (mg m^{-3})
[M]tot sw(crit) = [M] sw(crit) + [M] spм(crit) [SPM]sw
Where: [M] sw(crit)= critical dissolved concentration; [M]spм(crit)= metal concentration bound to suspended particle materials (SPM) in the surface water measured by atomic emission spectrometry; [SPM]sw= concentration of SPM in the surface water determined by gravimetric analysis (kg m^{-3}).

Critical limits referring to total dissolved metal concentration have been adopted. Recommended critical limits for dissolved Cd and Pb concentrations in surface waters {[Cd]sw(crit) = 0.38 mg m^{-3}; [Pb]sw(crit) = 11 mg m^{-3}} are all related to ecotoxicological effects (Modeling and Mapping manual, chapter 5.5 2004).;

The exceedances of critical loads of Pb and Cd for surface water were calculated by the following equation:

$$CL(M)ex = PL(M) – CL(M)$$

All symbols and abbreviations used in this article are in accordance with the Modelling and Mapping Manual (2004) (chapter 5.5).

Results

The trend of mean annual deposition of Pb and Cd by the bulk precipitation shows a clear decreasing from 89.48 g ha^{-1} yr^{-1} in 2005 to 5.41 g ha^{-1} yr^{-1} in 2010 for Pb and from 13.39 g ha^{-1} yr^{-1} to 1.17 g ha^{-1} yr^{-1} for Cd whereas after 2007 the deposition of both Pb and Cd remains relatively stable (Fig. 2). Values for 3 years period (2006-2008) have been used for calculating procedure in this study.

Although the amount of bulk precipitation was the same for the beech and spruce plots (1020 mm), the throughfall (789 mm) and the deposition of both Pb and Cd were lower under the spruce crowns in comparison with the beech (Table 1).

When comparing the variables used for the determination of critical loads of Pb at two experimental plots, it can be seen that the Pb uptake by the harvestable part of the biomass was higher for spruce than for beech but the leaching of Pb by the water runoff was the same for both beech and spruce plots. In this case the value of the critical load of Pb for spruce forest was higher than for beech one (Table 1). A comparison of the critical loads with the current deposition rates of Pb and Cd revealed that there were no exceedances of critical loads at both plots (beech and spruce) for the entire study period. The negative

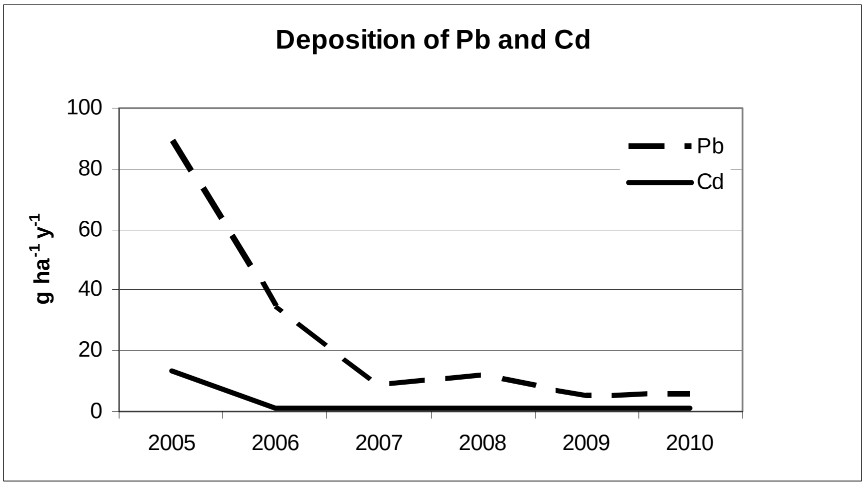

Fig. 2. Mean annual deposition of Pb and Cd during the period 2005-2010 at the Petrohan site, g $ha^{-1}yr^{-1}$.

Table 1. Critical loads of Pb and Cd for beech and spruce forests, g.ha-1yr-1 [Mu- vegetation uptake; Mle-metal leaching flux; CL- critical load; Dep- metal deposition; CL(M)(ex)- exceedance of critical loads]

	Pb		Cd	
g ha^{-1} y^{-1}	Beech	Spruce	Beech	Spruce
Vegetation uptake (Mu)	10.24	11.65	0.37	0.33
Leaching (Mle)	10.63	10.63	3.18	3.18
Critical loads (CL)	20.87	22.25	3.55	3.51
Deposition throughfall (Dep)	14.98	14.40	0.90	0.43
Exceedance CL(M)ex	-5.89	-7.85	-2.65	-3.08

value of the exceedance of critical load of Pb was -7.85 $g.ha^{-1}yr^{-1}$ for spruce forest against -5.89 $g.ha^{-1}yr^{-1}$ for beech one. Similar results have been found for Cd. Despite higher Cd deposition rates in beech forest than in spruce the critical loads were comparable for spruce and beech which leads to the higher negative value of the exceedance of the critical load for spruce than for beech (Table 1).

The mean concentration of SPM for the period 2006-2008 in the surface water of the Petrohan reservoir is higher (0.0047±0.0017 kg m^{-3}) than in the Barzya river (0.0032±0.0015 kg m^{-3}) as well as the concentration of Pb in surface water (0.0011±0.0003 mg m^{-3} in Petrohan dam against 0.0006±0.0001 for Barzia river) and the content of Pb in the SPM (25.763±0.467 mg kg^{-1} for Petrohan dam and 20.047±0.567 mg kg^{-1} for Barzya river). Although the total concentration of Pb in both types of water bodies is similar the total leaching of Pb from the catchment area remains higher for Petrohan dam than for Burzya river. As a result the value of critical load for Petrohan dam is higher compared to Burzya river. There were no exceedances of critical loads of Pb for both studied water bodies due mainly to the lower deposition of Pb at their catchment areas. The negative value of the exceedance of the critical load of Pb for Petrohan dam is two times higher than the exceedance for Burzya river (Table 2).

The critical load of Cd was lower than of Pb but similar for both water bodies due to the both similar uptake of Cd by the biomass at the catchment areas and the leaching of Cd with the water outflow. Although the higher content of Cd in the SPM in the surface water of Barzia river than in Petrohan reservoir the total concentration of this metal is rather similar for both water bodies because of the opposite relationship for the concentration of SPM in the waters of concern. All mean annual values of variables used in the study and standard deviations for each year (2006, 2007 and 2008) can be seen in the table 3. All measured concentration of Pb and Cd in water are below the critical limits for the ecotoxicological effect.

The main difference between the risk assessment of damage by the pollution of Pb and Cd is that unlike for Pb the critical loads of Cd were exceeded for both Petrohan reservoir and Barzia river (Table 2).

Discussion

The trend of deposition of Pb and Cd shown in figure 2 demonstrates lower level of pollution of Cd than of Pb at the Petrohan site. The significant decreasing of the deposition of heavy metals in forested catchments during the studied period (2006-2008) has not affected the concentration of Pb and Cd in surface water and their content in suspended particle matter (Table 3). Forest and aquatic ecosystems could be considered to be in a steady state for applying the steady state mass balance approach in critical loads calculating procedure.

In general, a critical load of pollutants indicates only the sensitivity of an ecosystem against the anthropogenic input of the metal of interest. Higher values of critical loads indicate higher tolerance and lower sensitivity of receptors to pollutants. As mentioned above, the deposition of Pb and Cd was higher in beech forest than in spruce one, but these deposition rates are not suitable for assessing the risk of harmful effects of these pollutants to forest ecosystems, because the critical load of Pb has been lower for beech as compared to spruce. The obtained values of critical loads have demonstrated that the spruce forest in Petrohan was more tolerant and less sensitive to the Pb deposition than the beech one at the same site.

A potential risk of damage for the studied ecosystems is possible only when the critical load is exceeded. A comparison of the critical loads with the real deposition

Table 2. Critical loads of Pd and Cd for surface water of Petrohan reservoir and Burzia river. Mean values for the period 2006-2008

Parameter	Pb		Cd	
	Petrohan reservoir	Barzia river	Petrohan reservoir	Barzia river
SPM (kg m^{-3})	0.0047	0.0032	0.0047	0.0032
Concentration in water (g m-3)	0.0011	0.0006	0.0001	0.0001
Content in SPM (mg kg^{-1})	25.763	20.047	0.2839	0.4533
Total concentration (mg m-3)	11.1130	11.0637	0.3811	0.3813
Leaching (g ha^{-1} y^{-1})	11.78	9.57	0.40	0.33
Plant uptake (g ha^{-1} y^{-1})	11.97	11.35	0.61	0.58
Critical load (g ha^{-1} y^{-1})	23.72	20.93	1.02	0.91
Deposition (g ha^{-1} y^{-1})	17.82	17.82	1.42	1.42
Exceedance (g ha^{-1} y^{-1})	-5.90	-3.11	+0.40	+0.51

Table 3. Mean annual values of metal concentration in surface water (Msw) and in deposition [M]dep, concentration of suspended particle material in water ([SPM]sw) and content of metal in suspended particle material (Mspm) for 2006-2008

M	Water body	Year	Msw, g m^{-3}	[M]dep, g m^{-3}	[SPM]sw, g kg^{-1}	Mspm mg kg^{-1}
Pb	**Petrohan**	2006	0.0011±0.0006	0.0028±0.0019	0.0076±0.0035	24.7500±2.9564
		2007	0.0013±0.0006	0.0009±0.0005	0.0041±0.0018	23.4940±2.3492
		2008	0.0009±0.0003	0.0013±0.0009	0.0025±0.0017	29.0350±0.5676
		Average	**0.0011** Vr=0.45	**0.0016** Vr=0.68	**0.0047** Vr=0.48	**25.7630** Vr=0.07
	Barzia	2006	0.0009±0.0001	0.0028±0.0019	0.0016±0.0007	18.6180±1.6131
		2007	0.0006±0.0003	0.0009±0.0005	0.0028±0.0018	23.4080±2.1876
		2008	0.0004±0.0001	0.0013±0.0009	0.0053±0.0015	18.1150±0.7675
		Average	**0.0006** Vr=0.33	**0.0017** Vr=0.64	**0.0032** Vr=0.40	**20.0470** Vr=0.07
	Critical	limit	0.011	0.011		
Cd	**Petrohan**	2006	0.00011±2.71E-6	0.0002±0.0001	0.0076±0.0035	0.2200±0.0431
		2007	0.00010±7.11E-6	0.0001±2.61E-5	0.0041±0.0018	0.3990±0.0301
		2008	0.00010±3.76E-6	0.0001±4.22E-5	0.0025±0.0017	0.2327±0.0177
		Average	**0.0001** Vr=0.04	**0.0001** Vr=0.56	**0.0047** Vr=0.48	**0.2839** Vr=0.16
	Barzia	2006	0.00020±0.00001	0.0002±0.0001	0.0016±0.0007	0.5990±0.2639
		2007	0.00010±3.57E-5	0.0001±2.61E-5	0.0028±0.0018	0.4468±0.0207
		2008	0.00010±2.63E-6	0.0001±4.22E-5	0.0053±0.0015	0.3141±0.1054
		Average	**0.00013** Vr=0.15	**0.00013** Vr=0.43	**0.0032** Vr=0.40	**0.4533** Vr=0.29
	Critical	limit	0.00038	0.00038		

rates revealed that there were no exceedances of critical loads for both forest and surface water ecosystems for the entire study period, hence the studied forest ecosystems were not at risk of damages due to Pb and Cd pollution. The negative values of the exceedances at the Petrohan site suggest that the spruce forest there could withstand more additional deposition of Pb (-7.85 g ha^{-1} yr^{-1}) before reaching the critical load value as compared to the beech, where the critical load value would be reached after an additional Pb deposition of 5.89 g ha^{-1} yr^{-1} (Table 1).

Despite higher Cd deposition rates in beech forest than in spruce the tolerance and the sensitivity of both beech and spruce forests to the pollution of Cd are similar because of their close values of critical loads of this pollutant. The forests ecosystems are not at risk of damage because the values of the deposition of Cd are lower as compared to the critical loads which leads to the high negative values of the exceedance of the critical load for spruce (Table 1).

Lead and cadmium have been widely recognized as highly toxic to plants decreasing seed germination and seedling, root and plant growth, and affecting many plant metabolic processes (Iqbal & Mehmood 1991; Breckle & Kahile 1992; Iqbal & Siddiqui 1992; Shafiq *et al.* 2008; Farooqi *et al.* 2009). At combined treatments of Pb and Cd all the measured variable were greatly reduced showing that Pb, when combined with Cd has got adverse effects on plants as compared to individual treatments (Kahile & Breckle 1989; Iqbal & Shafiq 1998; Iqbal *et al.* 2001; Shafiq *et al.* 2008). As a first step the individual critical loads of Pb and Cd for forest ecosystems have been determined in this study in accordance with the Modeling and Mapping manual (2004). Further research is needed to develop the methodological approach for calculation of critical loads based on combined effect of Pb and Cd for different forest species.

The surface water of Barzya river is more sensitive and less tolerant to the pollution of both Pb and Cd due to the lower values of their critical loads as compared to Petrohan dam. The tolerance of both water bodies is much more higher to the pollution of Pb than to Cd.

When comparing the values of critical loads of Pb and Cd for forest and water ecosystems, it can be seen that the sensitivity of forests is comparable with waters only for Pb because of their similar critical loads. Critical loads of Cd for water ecosystems are very low (0.91- 1.02 g ha^{-1} yr^{-1}) demonstrating higher sensitivity of surface water to the pollution of Cd in comparison with forests (3.51- 3.55 g ha^{-1} yr^{-1}).

The risk of harmful effects and damages is real for aquatic ecosystems due to the exceedances of their critical loads of Cd but not for forests where critical loads of Cd are not exceeded (Table 1 and 2). Exceedance of critical loads of heavy metals means that critical limits will be exceeded in the future, but not necessarily at present. On the other hand non-exceedance of critical loads can include a present risk, if critical limits are already exceeded due to historical inputs (Posch *et al.* 2003). Comparing our results to the expected development of metal concentration in environment in comparison to critical limits for different situations summarized in Posch et al.

(2003) it could be seen that our case for Pb corresponds to the situation N 6 (Fig. 3). The present concentration of Pb in surface water of both Petrohan reservoir and Barzia river is below critical limit and inputs of Pb are below critical load meaning that critical load will not be exceeded at a defined time period. In this case keeping the present load more stringent than critical load is needed.

In the reverse case of Cd, the metal input is larger than the critical load for both water bodies not exceeding the critical concentration of Cd in the surface water (situation N 1) (Fig. 3). In order to avoid future damage emissions of Cd must decrease. However, only keeping the critical load of Pb and Cd will not lead to exceedance of their critical limits in the long run.

Conclusion

The risk of damage to forest and water ecosystems due to the deposition of Pb and Cd cannot thoroughly be assessed by the calculation of the deposition rate of these pollutants only. Additional calculations of critical loads are needed to determine the tolerance of these receptors to heavy metal pollution. Of the two forest species examined, the spruce forest can accept higher deposition rates of Pb before any damages to the forest would be expected as compared to the beech one.

Using comparative simultaneous investigation based on the measured variables only it has been found that the tolerance of surface water to the pollution of Pb has been comparable with forests but aquatic ecosystems are more sensitive to the deposition of Cd due to the low values of critical loads.

Steady state critical loads of Pb and Cd for forests based on human health approach have remained stable and have not been exceeded during the study period. Real risk of harmful effect in the future has been discovered for both surface waters because of higher input of Cd on their catchment areas than critical loads obtained by ecotoxicological approach.

The reason for providing different critical loads for different types of ecosystems is because the critical loads for forest ecosystems do not automatically protect aquatic ecosystems receiving much or most of their metal load by drainage from the catchment area.

Although the measured concentrations of Pb and Cd in surface waters and precipitation have been below the critical limits keeping the calculated critical loads and decreasing the Cd deposition are needed for protecting forest and aquatic ecosystems in studied area.

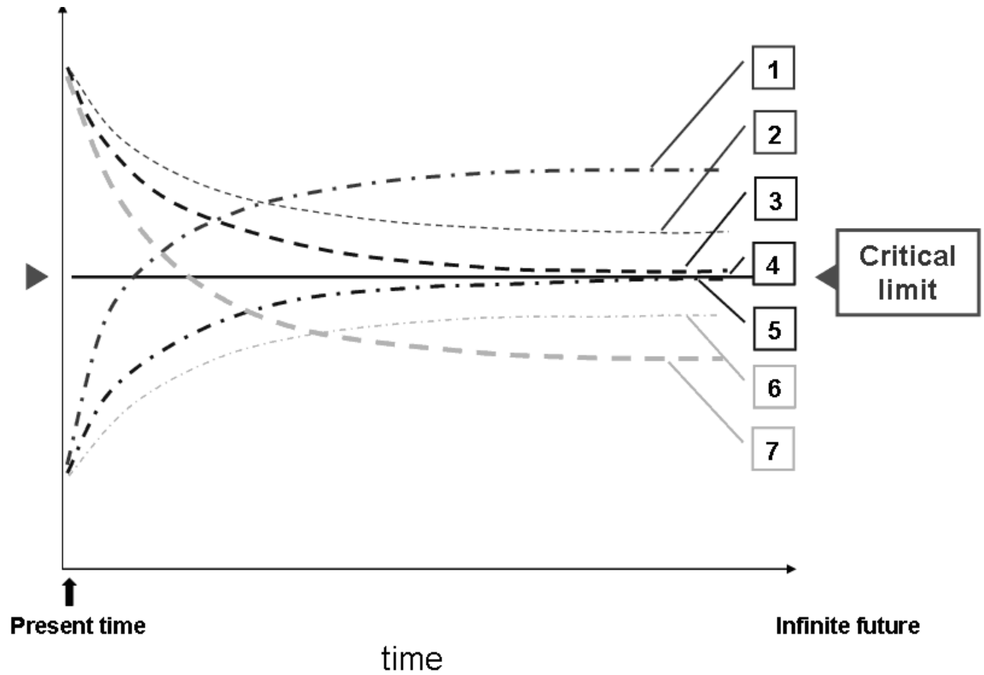

Fig. 3. Expected development of metal concentrations in environmental media in comparison to critical limits for different environmental situations (after Posch *et al.* 2003) Environmental situations:

1 The present concentration is below critical limit, inputs are above critical load;

2 The present concentration is above critical limit, inputs are above critical load;

3 The present concentration is above critical limit, inputs are exactly at critical load;

4 The present concentration is at critical limit, inputs are and remain stable exactly at critical load (the theoretical critical load situation);

5 The present concentration is below critical limit, inputs are exactly at critical load

6 The present concentration is below critical limit, inputs are below critical load;

7 The present concentration is above critical limit, inputs are below critical load.

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References

Breckle, S. W., Kahile, H. (1992) Effects of toxic heavy metals, Cd, Pb on growth and mineral nutrition of beech (Fagus sylvatica L.). Vegetation, 101: 43–53.

Crommentuijn, T., Polder, M. D., Van de Plassche, E. J. (1997) Maximum permissible concentration and negligible concentrations for metals, taking background concentrations into account. RIVM report № 601501001, pp.138.

Farooki, Z. R., Iqbal, M. Z., Kabir, M., Shafiq, M. (2009) Toxic effects of lead and cadmium on germination and seedling growth of Albizia Lebbeck (L.) Benth. Pakistan Journal of Botany, 41: 27–33.

Iqbal, M. Z., Mehmood, T. (1991) Influence of cadmium toxicity on germination and growth of some common trees. Pakistan Journal of Scientific and Industrial Research, 34: 140–142.

Iqbal, M. Z., Shafiq, M. (1998) Effects of lead and cadmium on plants. Abstract pp. 14. Six national conference of plant scientist. Oct. 1998: 20–22.

Iqbal, M. Z., Shafiq, M., Kausar, A. S. (2001) toxic effects of lead and cadmium individually and in combination on germination and growth of Leucaena leucocephala. Pakistan Journal of Botany, 33: 551–557.

Iqbal, M. Z., Siddiqui, D. A. (1992) Effect of lead toxicity on seed germination and seedling growth of some tree species. Pakistan Journal of Scientific and Industrial Research, 35: 139–141.

Kahle, H., Breckle, W. S. (1989) Single and combined effects of lead and cadmium on young beech trees. IUFRO, Biomensdorf: 442–444.

Kehayov, T. (1986) Underground waters in Bulgaria, mapping in 1:2000000, S., BAS, vol.V: 304–307.

Lamersdorf, N. P., Godbold, D. L., Knoche D. (1991) Risk assessment of some heavy metals for the growth of Norway Spruce. Dordrecht. Water, Air, Soil Pollution, 57/58: 535–543.

Modeling and Mapping Manual (Posch M, Hettelingh J-P, Slootweg J eds) (2004) Manual on methodologies and criteria for modeling and mapping critical loads & levels and air pollution effects, risks and trends, Federal Environmental Agency (Umweltbundesamt) Berlin, UBA-Texte 52/04 (download via internet: www.icpmapping.org).

Nilsson, J., Grennfelt, P. (1988) Critical Loads for Sulfur and Nitrogen Miljorapport North 1988:97. Nordic Council of Ministers, Copenhagen, Denmark, pp. 418

Posch, M., Hettelingh, J-P., Slootweg, J. (2003) Manual for the dynamic modeling of soil response to atmospheric deposition. RIVM report, 259101012, 2003 (www.mnp.nl/cce).

Shafiq, M., Iqbal MZ, Athar, M. (2008) Effect of lead and cadmium on germination and seedling growth of Leucaena leucocephala. Journal Appl. Sci. Environ. Manage, 12: 61–66.

Skjelkvale, BL., Ulstein, M. (2002) Proceeding from the workshop on heavy metals in surface waters: Monitoring and biological impact. March 2002, Lillehammer, Norway.

Schutze, G., Hettelingh, J-P. (2006) Results of modeling and mapping of critical loads of Pb, Cd and Hg and critical concentrations in Europe. In: Sufficiency and Effectiveness Review of the HM Protocol, Chapter A3, pp.19.

Tsonmis, G. (1991) Science and Technology of wood. Structure. Propertis. Utilization. Van Nostrand Reinhold, New York.

Tyler, G. (1992) Critical concentrations of heavy metals in the mor horizon of Swedish forests. Solna, Sweden, Swedish Environmental Protection Agency, Report 4078, pp.38.

World Health Organization (2004) Guidelines for drinking water quality. Third edition, vol. 1-Recommendations, Geneva.

Wilkens, B. J. (1995). Evidence for ground water contamination by heavy metals through soil passage under acidifying conditions. Ph D. Thesis, University of Utrecht, pp.146.