

COMPARATIVE RISK ASSESSMENT STUDIES OF HEAVY METAL POLLUTIONS IN BEECH FORESTS

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Abstract

Beech forests in the Western part of Bulgaria have been monitored in order to assess the risk of harmful effects of lead (Pb) and cadmium (Cd) pollution by means of Critical Loads calculations and their exceedances by real deposition. Critical loads of Pb and Cd for two sites (Vitinya and Petrohan) have been determined using the "Steady State Mass Balance" method based on the heavy metal uptake by the biomass and the leaching of the metals in the root zone. Real deposition of Pb and Cd was measured every two weeks during a one-year period by collecting the throughfall in plastic collectors (6 for each site). All samples have been analysed for their Pb and Cd content using atomic emission spectroscopy. The same method has been applied for measuring the content of Pb and Cd in the wood of beech trees. Fluxes of leaching water were measured in grid cells of 10 x 10 km for the entire country. The results obtained show that the critical loads of both Pb and Cd are lower for the Vitinya site demonstrating the higher sensitivity of beech to the pollution of heavy metals in comparison with the Petrohan site. In addition the real deposition of Pb and Cd has been higher at the Vitinya site. Although there were no exceedances of critical loads of Pb for both sites, additional deposition in the future will lead to a sooner exceedance of the critical load at the Vitinya site as compared to the Petrohan site. We conclude that the beech forest at the Vitinya site is at risk of damages by Cd pollution whereas the beech forest at the Petrohan site is more tolerant to heavy metal pollution due to its higher critical loads.

Key words: atmospheric deposition, cadmium, critical loads, exceedances, lead, throughfall.

Introduction

There is ample awareness of interactions at the starting point of the source-receptor chain with respect to sources and emissions of heavy metals and their adverse effects as air pollutants on ecosystems and various services these ecosystems provide, such as a sustainable

biodiversity (Metzger et al. 2005). 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 quantita-

tive 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). The Bulgarian focal center has contributed to the calculation and mapping of critical loads of acidifying pollutants and heavy metals mostly for forest and aquatic ecosystems (Ignatova 2007; Ignatova et al. 1998, 2002, 2005; Ignatova and Damyanova 2006; Slootweg et al. 2007).

Critical loads for different receptors 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 exceedances of critical loads of current deposition rates of the pollutant of interest. This approach is very effective because it can inform regional emission control policies. This study proposes modelling 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.

The aim of this study was to carry out comparative investigations on the sensitivity and risk of damage of heavy metal pollution in beech forests in the Western Balkan Mountain by means of critical load calculations and their exceedances. From this point of view the following tasks have been taken into account:

1. Determination of Pb and Cd deposition rates in the precipitation of beech

forests in two regions of the Western Balkan Mountain differing in air pollution levels.

2. Collection of measured data needed for the calculation of local critical loads of Pb and Cd for beech forest in order to assess their sensitivity to heavy metal pollution.

3. Assessing the risk of harmful effects and damages to beech forests by computing the exceedances of critical loads of Pb and Cd.

Material and Methods

Collectors for throughfall under beech crowns have been installed at two sites of the Western Balkan Mountain: Petrohan and Vitinya (Fig. 1). The collectors were located at 3 positions (Fig. 2) following the forest: 1 – close to the beech stem; 2 – In the middle of the crown projection; 3 – between two trees.

The throughfall has been collected with 18 plastic funnels (1 m above the ground) per site with a collecting surface of 314 cm² in polyethylene bottles (Fig. 3) stored in the upper soil layer to minimize biological activity in the collected solution due to darkness and lower temperatures. Individual water samples have been collected fortnightly after measuring the water volume and analysing the samples for their Pb and Cd content by atomic emission spectrometry.

Pb and Cd uptake by the biomass has been derived by multiplying the content of these metals in the beech stem and branches, measured by atomic emission spectrometry, with the annual growth determined after cutting the representative trees.

The runoff of water under the root zone has been measured and mapped 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 neighbouring hydrometric sites, infiltration of the water source etc.

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). From this point of view the leaching of heavy metals has been obtained by multiplying the runoff of water under the root zone with the critical concentration of the heavy metals in the soil solution (UBA, 2004).

The effect-based steady-state mass balance model was used to calculate the critical loads of Pb and Cd. 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), $g \cdot ha^{-1} \cdot yr^{-1}$;

Mu = Metal net uptake in the harvestable parts of plants under critical load conditions, $g \cdot ha^{-1} \cdot yr^{-1}$;

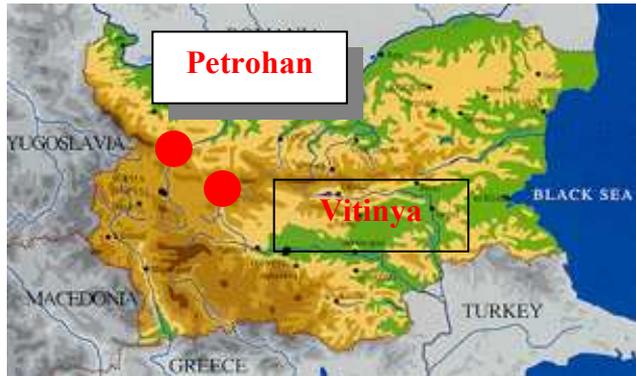


Fig. 1. Two monitoring sites in a beech forest of the Western Balkan Mountain in Bulgaria (Petrohan and Vitinya).

$Mle(crit)$ = Critical leaching of a metal M from the considered soil layer, $g \cdot ha^{-1} \cdot yr^{-1}$.

The **metal net uptake** of harvestable parts of plants was calculated by multiplying the annual yield by the fraction of metal net uptake within the considered soil depth and the metal content of the harvestable parts of the plants as follow:

$$Mu = fMu Yha [M]ha,$$

where: Mu = Metal uptake in harvestable parts of plants, $g \cdot ha^{-1} \cdot yr^{-1}$;

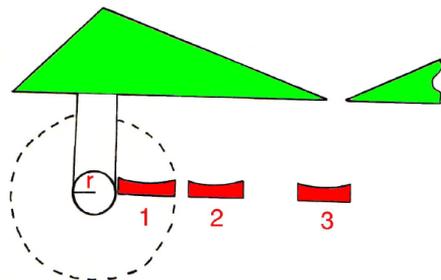


Fig. 2. Distribution of collectors for the deposition of heavy metals under beech crowns: 1 – close to the stem; 2 – under the half of the crown projection; 3 – between two trees.



Fig. 3. Permanently open collectors for the deposition of heavy metals.

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, $g \cdot ha^{-1} \cdot yr^{-1}$;
 CL(M) = Critical load for Pb and Cd, $g \cdot ha^{-1} \cdot yr^{-1}$.

Results and Discussion

fMu = Fraction of metal net uptake within the considered soil depth, accounting also for the metal uptake due to deposition on vegetated surfaces (the fraction of metal net uptake within the considered soil depth has been set to 1);

Yha = Yield of harvestable biomass (dry weight), $kg \cdot ha^{-1} \cdot yr^{-1}$;

[M]ha = Metal content of the harvestable parts of the plants, $g \cdot kg^{-1} \cdot dw$.

The **critical leaching flux** of heavy metals from the topsoil was calculated according to the follow equation:

$$Mle(crit) = cle \cdot Qle \cdot [M]_{ss}(crit),$$

where: Mle(crit) = Critical leaching flux of heavy metal from the topsoil, $g \cdot ha^{-1} \cdot yr^{-1}$;

Qle = Flux of drainage water leached from the regarded soil layer, $m \cdot yr^{-1}$;

[M]_{ss}(crit) = Critical limit for the total concentration of heavy metal in the soil solution ($10 \text{ mg} \cdot m^{-3}$ for Pb and $3 \text{ mg} \cdot m^{-3}$ for Cd) (UBA 2004);

cle = 10; this is a factor for appropriate conversion of flux units from $mg \cdot m^{-2} \cdot yr^{-1}$ to $g \cdot ha^{-1} \cdot yr^{-1}$.

The differences between the monitored and the critical possible loads of

Given that the amount of the through-fall in the beech forest at the Petrohan site was higher (898 mm) than at the Vitinya site (807 mm), it was necessary to determine the acidity and heavy metal concentrations in order to calculate the real HM deposition at these sites. The mean annual acidity of the throughfall under the crowns of the beech forest in Petrohan was 5.88 pH compared to 5.69 in Vitinya, showing that the precipitation entering the soil was in general not acidic and that the difference between the pH values was not significant. Under these conditions both the heavy metal concentration levels and the deposition rates have been higher in Vitinya as compared to Petrohan. The mean concentration of Pb for the study period was $0.8 \mu g \cdot dm^{-3}$ in Vitinya whereas in Petrohan it reached $0.6 \mu g \cdot dm^{-3}$. The concentration of Cd was two times higher in Vitinya ($0.2 \mu g \cdot dm^{-3}$) than in Petrohan ($0.1 \mu g \cdot dm^{-3}$) (Fig. 4). Although the amount of throughfall was higher in Petrohan, the deposition of both Pb and Cd was lower in comparison with the Vitinya site. The Pb deposited in Vitinya was $5.76 \text{ g} \cdot ha^{-1} \cdot yr^{-1}$ and $4.98 \text{ g} \cdot ha^{-1} \cdot yr^{-1}$ in Petrohan. The respective val-

ues for Cd deposition were $1.47 \text{ g}\cdot\text{ha}^{-1}\text{yr}^{-1}$ (Vitinya) and $0.90 \text{ g}\cdot\text{ha}^{-1}\text{yr}^{-1}$ (Petrohan) (Fig. 5).

In general, critical loads of pollutants can be used for the assessment of the sensitivity of receptors to a given pollutant. Higher values of critical loads indicate higher tolerance and lower sensitivity of receptors to pollutants. From a practical point of view it is common to compare the critical load with the real deposition of pollutants.

Using this comparison, the real risk of damage and disturbance of the sustainable development of a beech forest can be assessed. As mentioned above, the deposition of both Pb and Cd was higher in Vitinya than in Petrohan, but these deposition rates are not suitable for assessing the risk of harmful effects of these pollutants to forest ecosystems, because their critical loads have been lower in Vitinya as compared to Petrohan.

When comparing the variables used for the determination of critical loads of Pb at the two experimental sites, it can be seen that both Pb uptake by the harvestable part of the biomass and its leaching by the water runoff under the root zone were much higher in Petrohan than in Vitinya. On one hand this can be related to the higher concentration of Pb in the beech stems and branches in

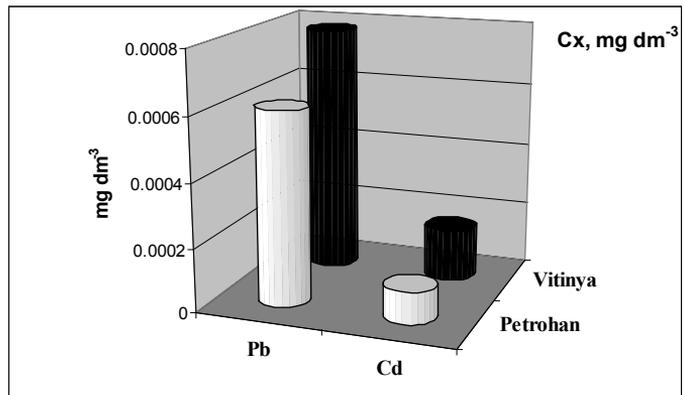


Fig. 4. Mean annual concentration of Pb and Cd in the throughfall of beech forests in Petrohan and Vitinya.

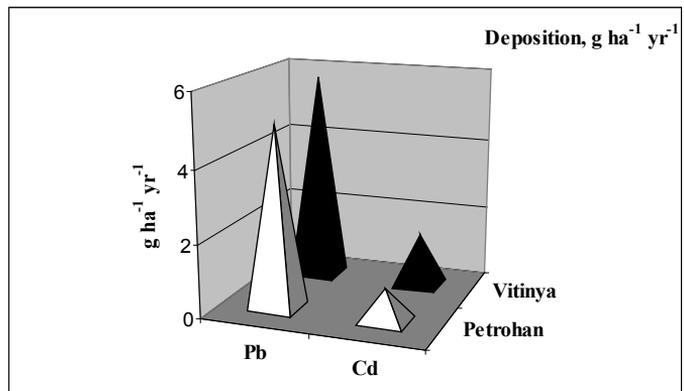


Fig. 5. Annual deposition of Pb and Cd in the throughfall of beech forests in Petrohan and Vitinya.

Petrohan ($0.0014 \text{ g}\cdot\text{kg}^{-1}$) than in Vitinya ($0.0004 \text{ g}\cdot\text{kg}^{-1}$), leading to a biomass uptake of Pb of $10.24 \text{ g}\cdot\text{ha}^{-1}\text{yr}^{-1}$ in Petrohan as compared to $2.79 \text{ g}\cdot\text{ha}^{-1}\text{yr}^{-1}$ in Vitinya. On the other hand the leaching of Pb by the water runoff in Petrohan was higher ($10.63 \text{ g}\cdot\text{ha}^{-1}\text{yr}^{-1}$) than in Vitinya ($3.63 \text{ g}\cdot\text{ha}^{-1}\text{yr}^{-1}$). In this case the value of the critical load of Pb at the Petrohan site was 3 times higher ($20.87 \text{ g}\cdot\text{ha}^{-1}\text{yr}^{-1}$) than in Vitinya ($6.42 \text{ g}\cdot\text{ha}^{-1}\text{yr}^{-1}$). The obtained

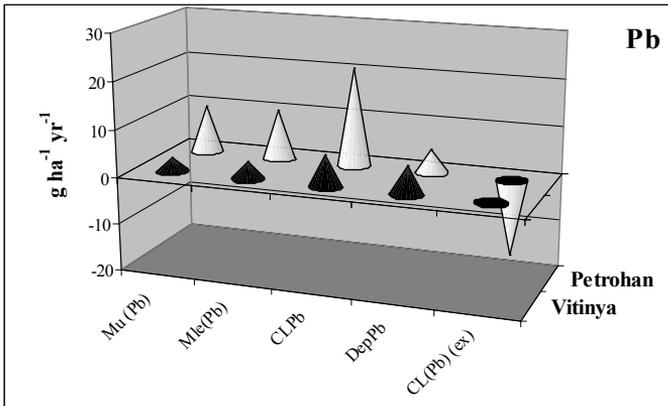


Fig. 6. Annual biomass uptake (MuPb), leaching (MlePb), critical load (CLPb), deposition (DepPb) and exceedance of critical load (CLPbex) of Pb in beech forests in Petrohan and Vitinya.

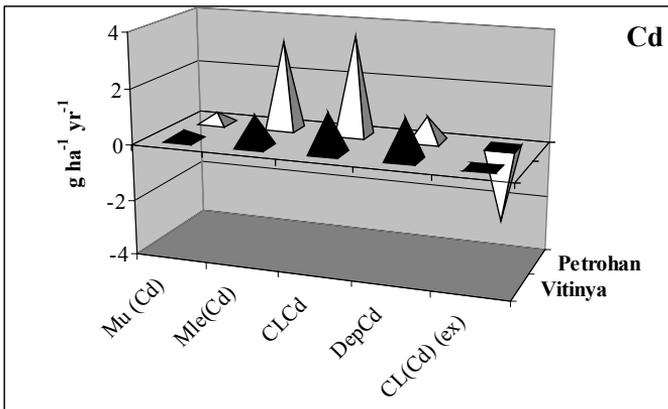


Fig. 7. Annual biomass uptake (MuCd), leaching (MleCd), critical load (CLCd), deposition (DepCd) and exceedance of critical load (CLCdex) of Cd in beech forests in Petrohan and Vitinya.

values of critical loads have demonstrated that the beech forest in Petrohan was more tolerant to the Pb deposition than the beech forest at the Vitinya site. A comparison of the critical loads with the real deposition rates revealed that there were no exceedances of critical loads at both sites for the entire study period,

hence the beech forests were not at risk of damages due to Pb deposition. The negative values of the exceedances at the Petrohan site suggest that the beech forest there could withstand much more additional deposition of Pb ($15.89 \text{ g} \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$) before reaching the critical load value as compared to the Vitinya site, where the critical load value would be already exceeded after an additional Pb deposition of only $0.66 \text{ g} \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$ (Fig. 6). Therefore, the risk of harmful effects and damages after a low increase in Pb deposition is higher in Vitinya than in Petrohan.

Similar results have been found for Cd (Fig. 7). Despite higher Cd deposition rates in Vitinya, the critical load was higher in Petrohan ($3.55 \text{ g} \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$) than in Vitinya ($1.44 \text{ g} \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$). However, the most significant difference found is that the critical load of Cd was not exceeded in Petrohan ($-2.65 \text{ g} \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$), whereas it was exceeded in Vitinya ($0.03 \text{ g} \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$) which increases the risk of damage at the latter site.

Human health effects of deteriorating forest ecosystem services through HM deposition – represented here as a change in the quality of drinking water – has been taken into account in our study. Critical

loads of Pb and Cd for ecotoxicological effects on terrestrial ecosystems were calculated for Bulgarian Forests by the Co-ordination 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⁻¹yr⁻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 values were between 1 and 4 g.ha⁻¹yr⁻¹ and between 10 and 30 g.ha⁻¹yr⁻¹ for Cd and Pb, respectively. In this study, the use of the human health effects- drinking water approach led to Pb critical load values of 20.87 g.ha⁻¹yr⁻¹ and 6.42 g.ha⁻¹yr⁻¹ for the Petrohan and Vitinya site, respectively. Similar differences were found for Cd: The critical load of Cd was higher in Petrohan (3.55 g.ha⁻¹yr⁻¹) than in Vitinya (1.44 g.ha⁻¹yr⁻¹). This means that the values of critical loads for both Pb and Cd were similar despite the existence of different critical limits and the use of different approaches.

Conclusions

The risk of damage to Bulgarian beech forests 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 beech forests to heavy metal pollution. Of the two sites examined, the beech forest in Petrohan can accept higher deposition rates of Pb and Cd before any damages to the forest would be expected as compared to the forest in Vitinya.

The real risk of damage can be assessed through the calculation of the exceedances of critical loads of Lead and Cadmium using measured deposition data. The results obtained in this study have shown that critical loads of Pb have not been exceeded for the beech forests at two Bulgarian sites. However, the possibility to accept additional deposition of Pb before reaching the critical load was lower for the Vitinya site in comparison with the Petrohan site. Special attention has to be paid to Cd whose critical load was exceeded in Vitinya, which increases the potential risk of forest damage at that site.

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