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Evaluation of the Risk of Pb and Cd Deposition on Bulgarian Forests Using a Critical Load Approach

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Abstract. The current study was designed to calculate the critical loads of lead (Pb) and cadmium (Cd) for forests in different mountainous sites in Bulgaria and to assess a risk of damage caused by these metals for 2017 year. Steady-state mass balance model for input and output metal fluxes from an ecosystem was applied, based on the criterion for ecotoxicological protection. These mass fluxes were calculated based on measured data. The value of critical load for Cd was considerably lower than those for Pb. That means all forest ecosystems were more sensitive to the Cd deposition compared to the Pb one. It was found that the critical load for Pb and Cd for coniferous stands was higher than those for the deciduous one. Exceedances of critical loads of Pb and Cd for four study sites during the 2017 year were calculated and used as a criterion for risk assessment against heavy metal pollution. Exceedances of critical loads for both metals were found, e.g. a real risk of the harmful effect of Pb and Cd for all study sites.

Key words: critical loads, heavy metals, risk assessment, forest ecosystems, lead and cadmium.

Introduction

Some metals such as lead, cadmium and mercury are extremely toxic in very low concentrations (Breckle & Kahile, 1992; Iqbal & Mehmood, 1991; Kahle & Breckle, 1898). In contrast to other metals (Cu, Zn) that are essential at small amount because play role of cofactors of enzymes, Pb, Cd and Hg only cause damage effect. Organisms have not natural biochemical mechanisms for their elimination. That way metals have remained in ecosystems for a long period damaging the most sensitive organisms initially and causing harmful effect of more organisms in the future.

By definition critical loads are 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 & 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 any damages to occur. The risk of harmful effect can be assessed using critical load exceedance (Agren, 1997). That is a deposition rate of pollutant, which is more than the acceptable level, e.g. the critical load. This effect-based approach is very effective because it takes into account all regional characteristics of a specific receptor and that way reveals the present state of the receptor or the whole ecosystem. Assessing the real state of a site of interest, policymakers can plan and predict measures for prevention and emission control.

The aim of this study was to carry out comparative investigations on the sensitivity and risk of damage of heavy metal pollutants on two types of forests (deciduous and coniferous) in four Bulgarian mountains by means of calculation of critical load and their exceedances based only on simultaneous measured data. From this point of view the following tasks were taken into account:

1. Determination of Pb and Cd deposition rates in the throughfall at the study sites;
2. Collection of measured data needed for the calculation of local critical loads for Pb and Cd for forests in order to assess their sensitivity to heavy metal pollution;
3. Assessing the risk of harmful effects and damages of forests by computing the exceedances of critical loads for Pb and Cd.

Materal and Methods

Sites' location and characteristics

The study sites are located in different mountains regions of Bulgaria and represent typical coniferous and deciduous forests for the country. Site Petrohan is located at Western Balkan and is a part of an international network for long-range ecosystem research (LTER). Site Plana is a part of the same network and is situated at Plana Mountain not far from the capital city Sofia. Site Beklemeto is located at Central Balkan. Sites Pismenovo is located at Strandja Mountain. Three of the study sites are located in a range between 1245 and 1430 m a.s.l. but the site Pismenovo only is at 65 m a.s.l and situated at Strandja Mountain. All study sites were not under anthropogenic influence. At every study site two plots with one coniferous and one deciduous stand were chosen.

Plots and stands characteristics such as altitude, ages, and species are given in Table 1.

Sampling, analysis and database collection

Permanently opened polyethylene plastic collectors were used for collection of throughfall deposition. Each collector had a collection area of 314 cm^2 and stood approximately 1.5 m above ground level. Three throughfall precipitation collectors were used at each plot. Sample collections were done every month. Water quantity and activity (pH) were measured at each sampling time for each collector at the plots. Mix samples were formed for each plot for deposition and stored at 4 °C. For analyzing dissolved element forms, all samples were filtered through 0.45 μm cellulose filter using membrane filtering system. Lead and cadmium were measured by atomic absorbance spectrophotometer.

For an increment estimation a model tree was determined for every sample plot and tree samples were taken. For that purpose heights and diameters of at least 50 trees in every sample plot were measured.

Calculation of an increment, critical loads (CL) of Pb and Cd and their exceedances

The effect-based steady-state mass balance model was used to calculate the critical loads of Pb and Cd (Posch & Hettelingh, 2004; Posh *et al.*, 2003). 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:

$$CL(M) = Mu + Mle(crit) \quad (1),$$

where:

CL(M) = critical load of a heavy metal ($g.ha^{-1}.yr^{-1}$);

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 ($g.ha^{-1}.yr^{-1}$).

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:

$$Mu = Yveg\ [M]veg \qquad (2),$$

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:

$$Mle(crit) = cle\ Qle\ [M]ss(crit) \qquad (3),$$

where:

Qle = Flux of drainage water ($m.yr^{-1}$);

[M]ss(crit) = Critical limit for the total concentration of heavy metal.

The exceedances of critical loads of Pb and Cd for forest ecosystems (CL(M)ex) (SCHUTZE & HETTELINGH 2004; HETTELINGH *et al.*, 2008) were calculated by the following equation:

$$CL(M)ex = Dep(M) - CL(M) \qquad (4),$$

where:

Dep(M) = Trhoughfoll deposition rate ($g.ha^{-1}.yr^{-1}$).

In the present study, the growth rate has been evaluated for the growing stock as a periodic annual increment over the past 10 years.

It was calculated by the equation:

$$Z_v^{mek} = \frac{V_a - V_{a-n}}{n} \qquad (5),$$

where:

Z_v^{mek} = a periodic annual increment, $m^3.ha^{-1}.yr^{-1}$

V_a = volume per 1ha at the ending of the growing period, $m^3.ha^{-1}$

V_{a-n} = volume per 1 ha at the beginning of the growing period, $m^3.ha^{-1}$

n = years of the growing period (10).

The volume at the end of the period V_a is calculated by a method developed by Mihov (MIHOV, 2000) as a function of three variables: an average diameter ($d_{cp.}$),a mean height ($h_{cp.}$) and an average tree spacing ($a_{cp.}$):

$$V_a = \left(\frac{d_{cp.}}{a_{cp.}}\right)^2 \left(b_1 + b_2 h_{cp.} + b_3 h_{cp.}^2\right) \quad (6),$$

where regression coefficients b_1, b_2, b_3 are different for each tree species, and an average tree spacing is calculated by the equation (7):

$$a_{cp.} = \sqrt{\frac{10000}{N}} \qquad (7),$$

where N is the number of trees per 1 ha.

The volume at the beginning of the period was calculated using the above-mentioned method. For establishing the d_{cp} values increment cores were taken with increment borer and h_{cp}. To reconstruct height growth a partial stem analysis was performed. In none of the sample plots there are no fellings and dead trees, so the number of trees 10 years ago we assume to be the same as at the present.

Results and Discussion

The results of the periodic annual increment for the all study sample plots are presented in Table 2. The data showed the highest increment for both coniferous and deciduous stands at site Pismenovo. That results were logical because of the fact that both stands were the youngest from all study stands. Very similar values were calculated for the forests at site Plana. The biggest difference in the increment (almost double) was found between stands of Norway spruce and Common beech at site Petrohan with 30 years of age difference.

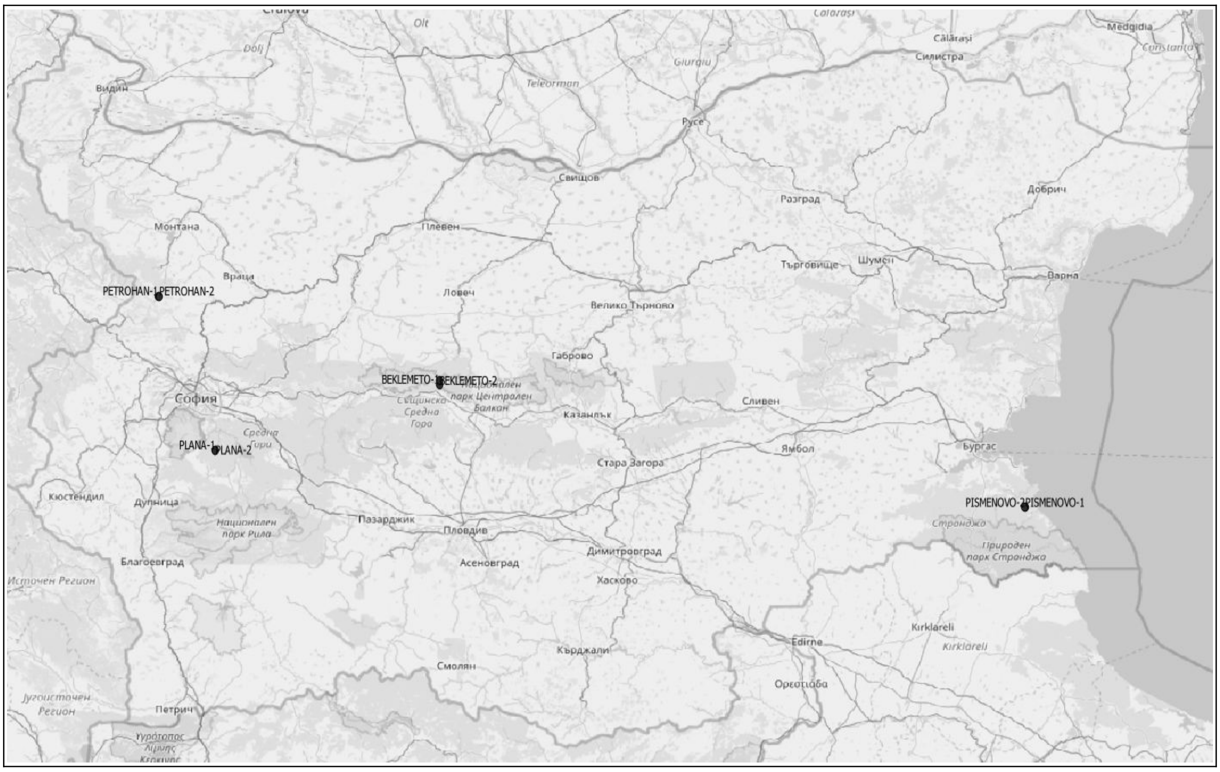

Fig. 1. Indicative map of the sampling plots (OpenStreetMap®, OpenStreetMap Foundation (OSMF)).

Table 1. Plots' characteristics.

Plots	Altitude, m a.s.l.	Age, year	Species
Petrohan 1	1420	84	Norway spruce
Petrohan 2	1430	114	Common beech
Plana 1	1245	37	Scots pine
Plana 2	1250	48	Common beech
Beklemeto 1	1300	108	Austrian pine
Beklemeto 2	1410	124	Common beech
Pismenovo 1	65	26	Austrian pine
Pismenovo 2	65	26	Pedunculate oak

Table 2. Net increment for 2017 year in g.ha^{-1}.yr^{-1}.

Site	Petrohan 1	Petrohan 2	Plana 1	Plana 2	Beklemeto 1	Beklemeto 2	Pismenovo 1	Pismenovo 2
Increment	8366	4320	12948	10440	3100	2592	13206	10665

Table 3. Pb net uptake in vegetation (M(Pb)), critical leaching of Pb (Le(Pb)) and critical loads of Pb (CL(Pb)) for 2017 in $g.ha^{-1}.yr^{-1}$.

Sites	Mu(Pb), $g.ha^{-1}.yr^{-1}$	Le(Pb), $g.ha^{-1}.yr^{-1}$	CL(Pb), $g.ha^{-1}.yr^{-1}$
Petrohan 1	58.56	10.64	69.20
Petrohan 2	25.92	10.64	36.56
Plana 1	90.64	2.84	93.48
Plana 2	62.64	2.84	65.48
Beklemeto 1	21.7	1.26	22.96
Beklemeto 2	15.5	1.26	16.81
Pismenovo 1	92.44	5.96	98.40
Pismenovo 2	63.99	5.96	69.95

Using the data obtained for the annual increment and drainage water from the catchment, the mass fluxes for both toxicant leaving the ecosystems as biomass uptake and leaching were calculated. Their values and those for calculated critical loads are presented in Tables 3 and 4.

It can be seen that net uptake by vegetation fluxes were considerably higher than those by leaching for both metals lead and cadmium (Tables 3, 4). The highest biomass uptake corresponded to the highest annual increment. Taking into account both factors influencing on vegetation uptake: increment and metal content in biomass, it could be figure out the higher biomass metal fluxes for coniferous than deciduous stands at every site. Critical leaching fluxes are depended mainly on drainage water fluxes from an every site that depended on catchment characteristics. Hereby site Petrohan characterised by highest critical metal leaching fluxes for both metals: lead and cadmium.

Calculated critical loads for Pb (Table 3) for all study plots varied between 22.96 and 98.40 $g.ha^{-1}.yr^{-1}$. The comparison between the different receptors indicates that higher values were typical for the coniferous forests, while deciduous species from the same site had lower levels of critical loads. The difference was about 40% but for Petrohan site only it reached double value. According to the main principle of the theory, the lower the critical load, the more sensitive the receptor is. Therefore, the broadleaves forests were more sensitive than the coniferous for all study sites. The most tolerant to the Pb pollution appeared stands of Austrian pine at site Pismenovo and Scotch pine at site Plana, which were the younger one, despite the difference of about 1200 m above sea level between the two sites. The most vulnerable were both forest species at the site Beklemeto, which were the oldest of all the studied. Moreover, comparison between two beeches stands at Petrohan and Beklemeto with the similar ages, revealed double sensitivity of the stand of Beklemeto. That means the assessment of the sensitivity is very specific and regional distinct factor, which have to be taken into account when an environmental protection measures are planned.

Calculated critical loads for cadmium (Table 4) were considerably lower than those were calculated for the lead. Their values were in the interval 1.16 to 7.07 $g.ha^{-1}.yr^{-1}$. The same tendency was found for cadmium pollutant in respect of the type of receptor: the coniferous three species had the higher critical loads in every study site than the deciduous. That implies the coniferous forests have a lower sensitivity to the Cd pollution. On the contrary, the broadleaves stands would be damaged easily by Cd compared to the coniferous at the same site. The oldest stands at site Beklemeto had the lowest critical loads for Cd and appeared to be the least tolerant to this pollutant from all study sites. The most resistant were the stands at site Pismenovo and site Petrohan.

To assess the present risk for the study forests, it is necessary to compare the value of calculated critical loads to the rate of lead and cadmium deposition.

Data in the figures 2 and 3 showed that for 2017 all critical loads for both heavy metals were exceeded. For some plots such as Plana 2 the exceedance of critical load for Pb was only 1.5%, which is negligible and can be considered as the acceptable value of that toxicant for the beech forest. It have to be mentioned that the exceedance of Pb for Scotch pine at Plana 1 was 15% only, which was the lowest value from four study coniferous species. For the most of the study forest, critical loads were exceeded from two to four times of the lead deposition. Even the most tolerant Austrian pine stand could not withstand to the damage influence of Pb pollution because deposition of the metal was one and the half times more than the acceptable level.

In respect to the cadmium was established the exceedances of critical loads such as for the lead (Fig. 3). The difference was in the levels that ranged from 50% to 90% times above the critical loads. The most vulnerable were both types of forests on the Belkmeto sites, where exceedances of critical loads for Cd were about 95% for deciduous and 90% for the coniferous stands. Even the forests at site Plana with the lowest exceedances for Pb, had exceedances for Cd 80% for beech and 55% for black pine stands.

Table 4. Cd net uptake in vegetation (Mu(Cd)), critical leaching of Cd (Le(Cd)) and critical loads of Pb (Cd(Cd)) for 2017 in g.ha^{-1}.yr^{-1}.

Sites	**Mu(Cd), g.ha^{-1}.yr^{-1}**	**Le(Cd), g.ha^{-1}.yr^{-1}**	**CL(Cd), g.ha^{-1}.yr^{-1}**
Petrohan 1	3.35	3.19	6.54
Petrohan 2	1.3	3.19	4.49
Plana 1	5.18	0.85	6.03
Plana 2	3.13	0.85	3.99
Beklemeto 1	1.24	0.38	1.62
Beklemeto 2	0.78	0.38	1.16
Pismenovo 1	5.28	1.79	7.07
Pismenovo 2	3.2	1.79	4.99

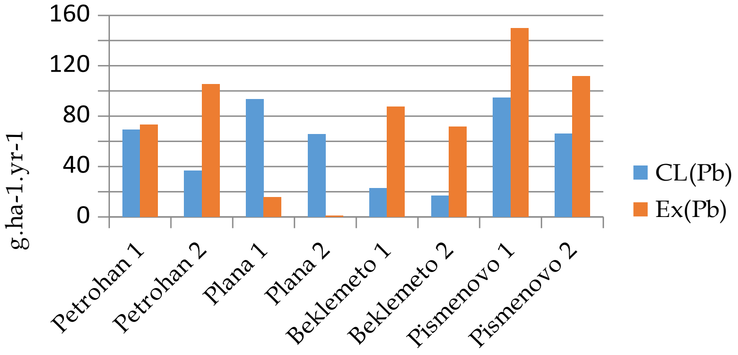

Fig 2. Critical loads (CL) and exceedances of critical loads (CLex) for Pb for 2017 in g.ha^{-1}.yr^{-1}.

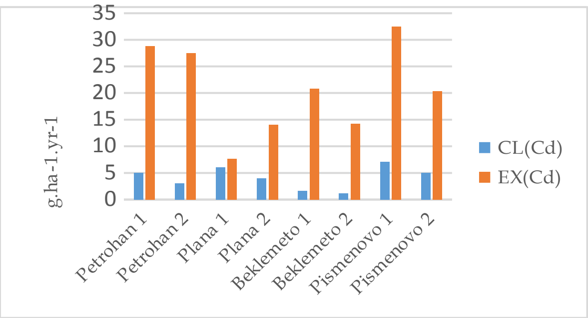

Fig 3. Critical loads (CL) and exceedances of critical loads (CLex) for Cd for 2017 in g.ha^{-1}.yr^{-1}.

Conclusions

The critical loads for Pb were higher than those for Cd. Critical loads for coniferous species for both metals were higher that means the higher tolerance to the metal influence than these for the broadleaves one. The main component formed critical loads was metal uptake flux by vegetation. The younger stands were more tolerant of both Pb and Cd pollutants than the older ones. Exceedances of critical loads for both pollutants were established. The present risk of damage from Pb and Cd for study forests was found. It is urgent measures for reduction of heavy metals emission to be taken.

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