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Measurements of proton load (H^+) in natural environment of the West Carpathians Mountains (passive sampling method)

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Abstract

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We studied air pollution in selected forest (beech, spruce) ecosystems in Slovakia, the West Carpathians Mts. To monitor forest exposure to air pollutants, we used the method of passive samplers. The comparison between pairs of open plots and forest stands revealed that the load was in general higher on the corresponding open plot. The only exception was the beech experimental site whose former open plot has already been restocked by natural regeneration, and the difference between the load on the regenerated plot and the original stand was not remarkable. The results of the proton load pointed out the dependence on altitude. In comparison with other regions in Slovakia and some other in central Europe, the air pollution with airborne pollutants at the Beech Ecological Experimental Station Kremnické vrchy Mts (470–510 m asl) can be assessed as moderate (9.3 mmol H^+ day^{-1} m^{-2}). The values monitored in beech stands of the Žiarska kotlina basin (470 m asl) in 1992 were 1.3 times higher. The Biosphere Reserve Poľana Mts (1,370–1,380 m asl), with dominant spruce, had pollution levels four times higher than the BEES. In the National Park Nízke Tatry Mts (1,440 m asl), was the mean annual value 3.5 higher compared to the corresponding value at the BEES Kremnické vrchy Mts. Assessment of soil resistance against human-induced acidification revealed that the region of the Kremnické vrchy Mts was the most resistant in this case, too. As for the contamination, the most loaded ones are stands at altitudes above 600–700 m asl. On the other hand, the soil acidity in these areas is not extreme, which manifest a strong buffering and regeneration capacity of the local soils. The method of passive samplers is a simple method for mapping pollutants effects in natural environment of forest ecosystems.

Key words

air pollution, hydrogen ion, passive sampler, forest ecosystem, the West Carpathians Mts

Introduction

Extensive damage to forest stands, extinction of organisms sensistive to alien substances, lowering rate of plant debris decomposition and other similar phenomena can be observed in our conditions since the beginning of the last century. One of the most important factors deteriorating the environment is air pollution with emmitted materials, airborne pollutants and depositions. The levels of airborne pollutants in urban and industrial agglomerations are monitored with automatic monitoring networks. The monitoring possibilities in mountain and rural conditions are limited, and manual supplementing is necessary. It is also possible to use the cummulative method of passive samplers for identification of polluting materials in amounts representing an ecological risk. This method enables us to observe the quantity of proton load (H^+) indicating the acid components of the atmosphere. In model forest ecosystems in the West Carpathians Mts, we studied differences between amounts of air pollutants in forest stands and open plots depending on the altitude. We also considered the distance from pollution source, landscape topology and air masses transport. For selected localities we also evalusted soil resistance to effects of human-induced pollution.

Material and methods

The research in the area has been focussed on assembling sufficiently large data sets for more precise identification of load with airborne pollutants. Former analytic measuring devices are progressively replaced with passive samplers. The second do not require any energy supply in the field, and their demands are low on cost and personnel. They provide a tool for study of field, non-forest and forest vegetation in areas with extreme or toxic episodes in airborne pollutants occurrence, with heavy car traffic, in national parks, etc. Well recognised are results of several authors Krochmal and Kalina (1997), Cox (2003), Bytnerowicz et al. (2004). In Slovakia, these results have been used in the recent years by Šablatúrová and Bičárová (1995), Varšavová and Barančok (1999), Molnárová (2000). The methods are gettting more and more mastered and simple. The results can be compared with the results obtained using continual analysers (Hangartner et al., 1996; Werner et al., 1999; Buffoni, 2002). The precision obtained with passive samplers and continual monitoring was judged as favourable after testing by several authors: Krupa and Nosal (2001).

From the viewpoint of the airborne pollutants threatening the environment, there is crucial role of acid components, primarily compounds of sulphur, nitrogen and hydrogen ions H^+. The long-term impact of airborne pollutants load to forest ecosystems in Slovakia is studied using the method of measuring proton load (H^+) according to Obr (1989). The method has been designed at the Forest Research Institute in Zvolen. It is a summary method, and for making comparisons it is necessary to use the same time period. There are provided records of interception of gaseous (SO_2, NO_x, HF), liquid (HNO_3, H_2SO_4) and solid particles (NH_4HSO_4) on filtering paper surface. The proton load is deduced from acid deposition neutralising the alkaline solution exposed in the field. The non-neutralised residue is determined as the difference between the non-exposed and exposed absorption solution by titration with HCl on a Tashiro indicator. The device for exposition of extracted filtration papers consists of a holder with a shelter and a silicone cylinder (passive sampler). The cylinder is protected with a removable case, allowing free access of air. At the same time, it provides protection against precipitation and direct sun radiation. The sampling equipment consists of two samplers fixed to a holder and placed on the open plot and in the stand. More details can be found in Kellerová et al. (1997).

The method of measuring proton load provides us with a pattern of dry and, to some extent also on wet deposition, primarily horizontal. The result is depending on the locality. The territory of Slovakia is situated in the Carpathian bow with highly diversified terrain, and the differences at the plot altitude may be considerable. The plots are situated in areas with frequent fogs occurrence, low cloudiness and inversions. These meteorological phenomena are associated with high concentrations of polluting materials. The areas in concern are exposed to prevailing transport of air masses and, consequently to the transboundary transport of air pollutants. The transboundary transport and larger local sources cause considerable soil acidification, primarily at higher altitudes. From this viewpoint, it is necessary to study the soil resistance against human-induced acidification.

Characteristics of research plots

The testing material is exposed at the Beech Ecological Experimental Site (BEES) and in a network of permanent research plots (PRP). The research plots are situated in the main forest areas of Slovakia, crucially important for forest production, with beech and spruce as the dominant woody plants (Fig. 1).

1. PRP Žiarska kotlina basin, 48° 35´ N, 18° 51´ E, is situated at 470 m above the sea level. The 90-year-old forest stand with dominant beech (*Fagus sylvatica*) and the corresponding open plot are situated in the southern part of the Štiavnické vrchy Mts. In recent years, the basin was strongly polluted with materials emitted by regional sources as an aluminium plant and a power plant. The area is also next to a busy motorway Bratislava – Banská Bystrica.
2. BEES Kremnické vrchy Mts, 48° 38´ N and 19° 04´ E, is situated at 470–510 m, in the beech forest vegetation tier. The stand age is 105 years. The airborne pollutants on plots are transported from several sources in the Zvolenská kotlina basin: three stationary power plants, a network of motorways and a dense railroad network. Under certain weather conditions, these sources may strongly influence the plots with pollutants.
3. Permanent research plots 3. I. Štefanová (690 m asl) and 3. II. Pod Veľkým Rozsutcom (1200 m asl) are situated in the National Park Malá Fatra, 49° 13´ N, 19° 04´ E. The dominant woody plant is spruce (*Picea abies*), the stands are differentiated in terms of age and diameter. The domestic pollution sources are energy plants, chemical plants and car traffic.
4. PRP Zadná Poľana (1360–1380 m asl), 48° 39´ N and 19° 29´ E, is situated at upper forest line in the Biosphere Reserve Poľana. The spruce stand with admixture of beech and rowan (*Sorbus aucuparia*) is 110–125 year old. The pollution sources are power plants, engineering industry and mobile sources.
5. PRP Čertova svadba 48° 54´ N and 19° 45´ E is situated in the eastern part of the National Park Nízke Tatry at an altitude of 1440 m asl. The age of the spruce (*Picea abies* Karst.) stand is 140–190 years.

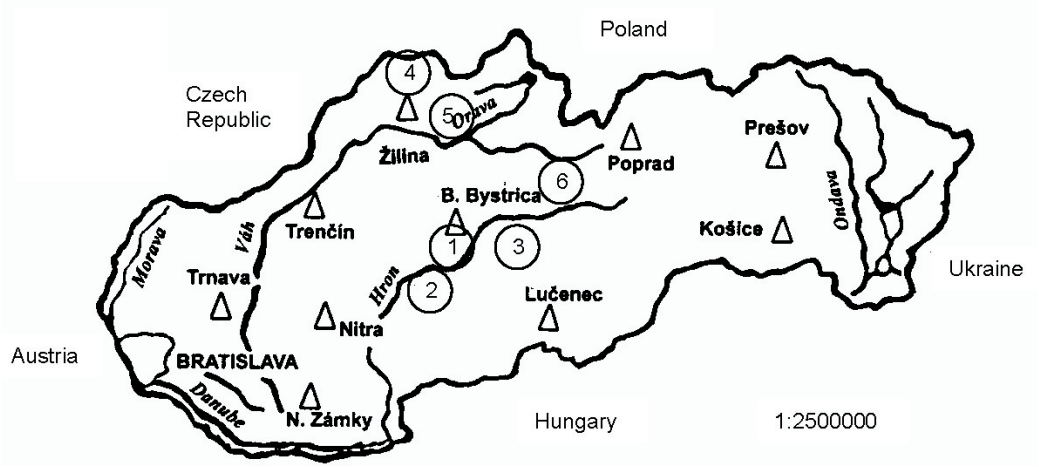

Fig. 1. Network of permanent research plots in Slovakia – 1 – PRP Žiarska kotlina basin, 2 – BEES Kremnické vrchy Mts, 3 – I. PRP Štefanová and II. PRP Pod Veľkým Rozsutcom in NP Malá Fatra Mts, 4 – PRP Zadná Poľana Mts, 5 – PRP NP Nízke Tatry Mts

The air is polluted by heating and transport. The close mountain saddle Čertovica is crossed with an important busy international motorway.

Apart from the pollutants produced by domestic sources, the Slovak territory is exposed to the transboundary pollution. The second reaches up to 60% of the total amount. The stands at 700–800 m asl in the Malá Fatra, Poľana, Nízke Tatry Mts are mainly polluted with long-range transported pollutants.

Results and discussion

This work provides the pollution patterns in selected forest areas in Slovakia over the time period 1992–2002. We evaluated trends and dynamics of the proton load input. The comparison between proton load amounts in forest stands and corresponding open plots revealed that the load on the open plot was in general higher than the load to the stand (Fig. 2).

This trend was kept for long in the beech stand in the Žiarska kotlina basin, with the mean value on the open plot being 17.0 and in the stand 11.2 mmol H^+ day^{-1} m^{-2}. In 1997–1999 was similar situation also on permanent research plots with dominant spruce in Malá Fatra Mts, with mean values on the open plot 3.I. being 21, on 3.II. being 20.6, in the stand 3.I. being 16.6 and in the stand 3.II. being 13.7 mmol H^+ day^{-1} m^{-2}. In the Biosphere Reserve in Zadná Poľana Mts we obtained 23.0 on the open plot and 17.7 mmol H^+ day^{-1} m^{-2} in the stand. In the Nízke Tatry Mts (1992–1995), it was on open plot 30.5 and in the stand 25.5 mmol H^+ day^{-1} m^{-2}. The situation is also illustrated with the results of statistical analysis presented in Table 1.

Beginning with the year 1995, the trend was not kept unambiguous any longer at the Beech Experimental Site Kremnické vrchy Mts. The mean value on the open plot 12.2 was many times found higher than the value in stand 11.0 mmol H^+ day^{-1} m^{-2}, they were, however, years when the reverse was true (1995, 1996, 1999, 2002). It is the result of natural regeneration of the former open plot. The crowns of the regenerating stand become with filtering corresponding to their progressively closed canopy.

The research plots have been established in terrain with diversified topography and different altitudes. The results obtained with linear regression analysis show dependence of proton load on altitude for separate plots (Fig. 3). Each value has been obtained from mean annual values.

In the Žiarska kotlina basin (1), the mean value 14.1 was somewhat higher than in the BEES Kremnické vrchy Mts (2) 11.6 mmol H^+ day^{-1} m^{-2}, in spite of lacking difference in altitude. The values in Žiarska kotlina basin were continually higher over the last ten years of the twentieth century, as the result of pollution sources in close neighbourhood (aluminium plant, power plants, dense traffic). Higher values on plots situated in the mountain spruce tier 18.8 (3.I.), 17.2 (3.II.), 20.3 (4.) and 28 mmol H^+ day^{-1} m^{-2} (5.) are also caused by long-transported and transboundary air pollution.

Because the lack of compatibility between the plots and time periods, we decided for the year 1992. In this year, the research was running parallel on the research plots in the Žiarska kotlina basin, at the BEES Kremnické vrchy Mts, in the Biosphere Reserve Poľana Mts, and in the Nízke Tatry Mts. The mean annual vales were the lowest at the BEES Kremnické vrchy Mts:

9.3 mmol H^+ day^{-1} m^{-2}, in Žiarska kotlina basin, they were 1.3 times higher (11.8 mmol H^+ day^{-1} m^{-2}). The mean value in the Biosphere Reserve Poľana Mts (27.1 mmol H^+ day^{-1} m^{-2}) was three times higher than the value at the BEES Kremnické vrchy Mts. The strongest air pollution was found in Nízke Tatry Mts (33 mmol H^+ day^{-1} m^{-2}), which was three and half times higher compared to the BEES.

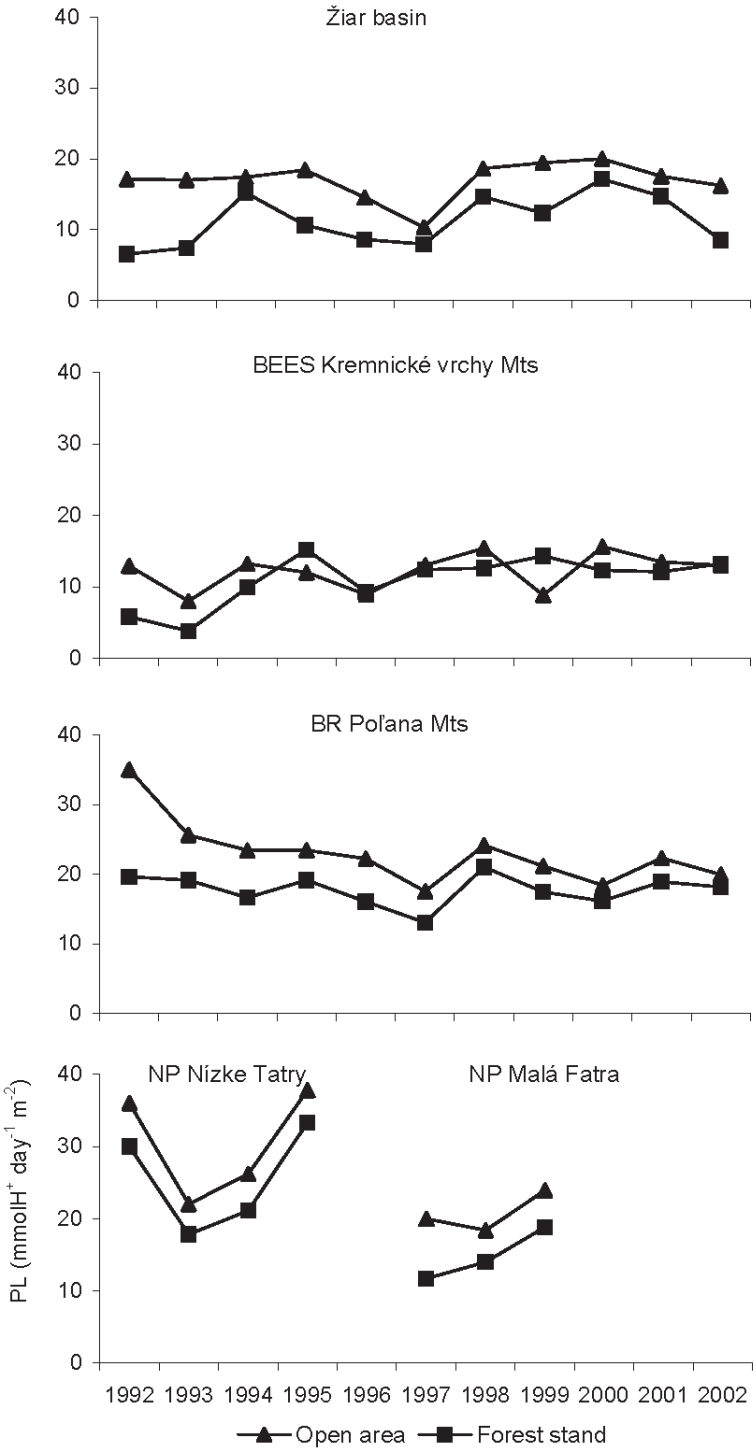

Fig. 2. Comparison of mean annual values of proton load (PL) on open plots and in forest stands in selected beech and spruce ecosystems in Slovakia

Table 1. Basic statistical characteristics of proton load (H^+) on open plots, and in beech and spruce forest stands (mmol H^+ day^{-1} m^{-2})

Site	1.		2.		3. I.		3. II.		4.		5.	
Area	OA	FS	OA	FS	OA	FS	OA	FS	OA	FS	OA	FS
Sample size	11	11	11	11	3	3	3	3	11	11	4	4
Mean	17.0	11.2	12.2	11.0	21.0	16.6	20.6	13.7	23.0	17.7	30.5	25.5
Median	17.5	10.6	13.0	12.3	21.6	16.5	19.0	14.2	22.3	18.1	31.1	25.5
Mode	17.5	–	13.0	–	–	–			23.4	19.1	–	–
Standard deviation	(±2,7)	(±3,7)	(±2,5)	(±3,5)	(±2,9)	(±2,9)	(±3,2)	(±6,5)	(±4,6)	(±2,2)	(±7,3)	(±7,6)
Standard error	0.8	1.1	0.7	1.0	1.7	1.6	1.9	3.8	1.4	0.6	3.8	3.6

OA – open area, FS – forest stand, 1. – PRP Žiarska kotlina basin, 2. – BEES Kremnické vrchy Mts, 3. I. – PRP Štefanová in NP Malá Fatra Mts, 3. II. – PRP Pod Veľkým Rozsutcom in NP Malá Fatra Mts, 4. – PRP Zadná Poľana Mts, 5. – PRP NP Nízke Tatry Mts

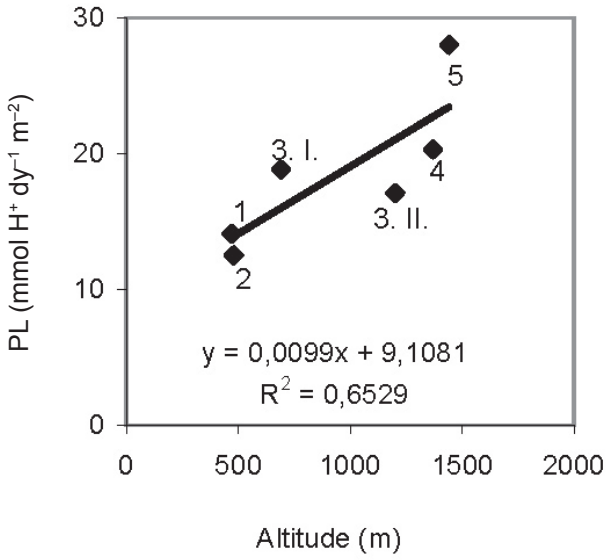

Fig. 3. Dependence of proton load (quantity) on altitude on research plots in Slovakia. 1. – PRP Žiarska kotlina, basin (470 m) 2. – BEES Kremnické vrchy Mts (470–510 m), 3. I. – PRP Štefanová (690 m) and 3. II. – PRP Pod Veľkým Rozsutcom (1200 m) in NP Malá Fatra Mts, 4. – PRP Zadná Poľana Mts (1370–1380 m), 5. – PRP NP Nízke Tatry Mts (1440 m)

The harmful impact of air pollutants on herbs and forest woody plants can be either direct or indirect – through soil and root systems. The soil is subjected to changes in its chemistry, to acidification and subsequent leaching of soil nutrients. The basic physiologic functions are disturbed; forest ecosystems become weaker and die. Soils in selected localities in Slovakia were assessed for their resistance against impact of anthropogenic pollutants.

The main contamining substances in the Žiarska kotlina basin were found oxids of fluorine, sulphur, nitrogen, further arsenic, cadmium, ozone and suspended particulates. Very adverse was impact of fluorine and SO_2 on forest ecosystems. These materials also deteriorated agricultural land, caused contamination of water courses and deteriorated health state of the local people (Mihálik and Bublinec, 1995; Holub and Kontrišová, 1998; Pariláková, 2000; Kulfan et al., 2002). The soils in Žiarska kotlina basin, originally nutritive, have been acidified, with pH ranging from 5.6 to 6.5. Long-term impact of pollutants from local sources lowered the soil quality, which had, together with climate conditions,

negative influence on growth conditions and health state of forest stands. Towards the turn of the century, the situation became to improve. The industrial process has been modernised and the changes in legislation forced the reduction in amounts of polluting substances. The amount of fluorine has been reduced to the carrying limit of 1 μg m^{-3} (Urminská et al., 2000). Since 1994, there has not been recorded any important change in air pollution with sulphur dioxide, and the values of this pollutant were mainly kept below the critical limit (20 μg m^{-3} r^{-1}). Somewhat less extensive was reduction of concentrations of nitrogen oxides. There were fluctuations in protone load quantity over the study period. In 1995–1997, the mean annual values in the Žiarska kotlina basin decreased. In 1998, there followed an increase again. In the following years, there was stagnation, and a final drop towards the end of the study period.

The reduction in 1995–1997 was evident also at the BEES and in the Biosphere Reserve Poľana Mts. It could be associated with the drop in industrial production. The increase in the following years was influenced, apart from other factors, with an immense increase in car traffic – amount of motor vehicles.

The soil quality at the BEES Kremnické vrchy Mountains is high; the soils are rich in nutrients, water and humus content. Humus has a high absorption capacity for pollutants interception – these can be accumulated and not allowed to percolate into the ground water. The soil in site is an andosolic cambisol – one of soil substrates most resistant against influence of atmospheric deposition (Bublinec et al., 2002). The soil quality is improved by fully connected broadleaved stands, ensuring filtration of precipitation and gases. The crown structure of beech stands has favourable influence on the air condition in the stand interior. In comparison with other localities in Slovakia and several other in central Europe, the beech stand at the BEES Kremnické vrchy Mts is still a locality with a very low load with airborne pollutants (Kellerová et al., 2001). The results of many research works allow us to asses the ecological stability of the BEES Kremnické vrchy Mts as a resistent one.

In the National Park Malá Fatra Mts, there are dominant west and north-western winds transporting pollutants from the adjacent industrial areas in the region Ostrava-Karviná and pollutants produced by metallurgical plants in the Sliezska basin in Poland. The reseach resulted in assessment of 45% of the forests as forest with disturbed stability and another 6% as having lowered tolerance (Koráň, 1997). In the area of the Malá Fatra, the most endangered are higher situated localities (Chleb Mt, 1647 m asl), exposed to extensive photo-oxidation stress. The precipitation is acid or strong acid, significantly indicating the pollution of the environment (Bublinec et al., 1995). Important pollution sources are local fossil-fuel power stations producing solid contaminants, SO_2 and NO_x. Other pollutants are produced by chemical industry and heavy car traffic (Klinda and Lieskovská, 2001; Ronchetti et al., 2004). The reaction of most soils on the permanent research plots in the Malá Fatra Mts is moderate acid, with pH ranging from 5.7–6.4. The humus quality is medium or very good. The soils have been formed on parent rock material consisting of calcites and dolomites, they are high resistant against airborne pollutants, and their regeneration capacity is very good. Negative factor is the altitude, because the same holds for all mountain localities situated at above 750 m asl. In the research period, this area was under influence of acid and very acid precipitation. A moderate increase in proton load was detected in 1999.

The area of the Biosphere Reserve Poľana Mts is ranked to category with intensive deposition of airborne pollutants, primarily in its exposed mountain parts. The air pollution is produced by local sources and also transported by long-range transport. The research results obtained in 1992–2002 indicate a decrease in mean annual values of the proton load. It seems that after a transition period of dumping, the dominant pollution sources, metallurgical and power plants will increase their production. Also in this area we expect a moderate increase in NO_x produced by small local sources and by mobile sources. The mountain range is volcanic, the soils are humic, deep and sufficiently loose. In connection with human-produced pollutants, the dominant andosols have a good buffering capacity and a good capacity for regeneration.

In the ridge part of the Nízke Tatry Mountains are dominant north, southward blowing winds, but there are also frequent winds blowing from north-west and south-west. The ridge parts are mainly polluted by long-range transported pollutants (Bublinec and Dubová, 1995) from transboundary sources. From the local sources is important heating and car traffic. The close mountain seddle Čertovica is crossed with a rushy international motorway. The traffic has negative inpacts on all the components of the environment (air, water, soil, fauna, flora). The most severe is impact of carbohydrate fuels combustion, resulting in increase in solid contaminants, VOC and NO_x. On the other hand, the trend in pollution by SO_2 is kept decreasing or without change. The soils on plots in the Nizke Tatry Mts are podzoles, but their acididy does not reach extreme values. Ecological conditions for growth of spruce stands are not very favourable – also based on the reason that the stands are situated in the 7-th and 8-th vegetation tier.

Conclusions

In 1992–2002 we studied air pollution in selected forest areas in Slovakia, the West Carpathians Mountains, by

means of passive samplers. The research has contributed to the knowledge about the relationships of open area and forest stands and proton load quantity. The proton load values were in general higher on the open plot than in the stand. The only exception was the Beech Ecological Experimental Site Kremnické vrchy Mts. The former open plot at the site was in phase of natural regeneration, and since 1995, the difference between the load on the open plot and the load to the forest stand has not been kept unambiguous, any longer. From the results, it is evident that the airborne pollutants are intercepted by tree crowns in stands with closed crown canopy. The important filtration and hygienic function of forest stands is evident.

In 1992 we conducted parallel research on all the plots, with exception of Malá Fatra Mts. The mean annual values were the lowest at the Beech Ecological Experimental Site Kremnické vrchy Mts (9.3 mmol H^+ day^{-1} m^{-2}). In comparison with localities in central Europe we can see that the contamination level at the BEES is low. The values observed in beech stands in the Žiarska kotlina basin were 1.3 times higher, in the Biosphere Reserve Poľana three times higher, and in the National Park Nízke Tatry even 3.5 times higher than at the BEES Kremnické vrchy Mts. The results and analysis of linear regression point out the dependence of proton load quantity on altitude for all the studied plots.

The soils are under permanent influence of airborne pollutants, in the recent years, however, less intensive. The assessment of soil resistance against acidification due to human activities showed that the most resistent are soils in area of the Kremnické vrchy Mts. The contamination of stands situated at 600–700 m asl is the most severe. However, the soil acidity in these areas does not reach extremes, and we can see that the soils buffering and regeneration capacity is good. The passive samplers allow us to quantify the load with airborne pollutants and to identify time and spatial trends in amounts of polluting substances.

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Meranie protónovej záťaže (H^+) v prírodnom prostredí Západných Karpát (metóda pasívnych zberačov)

Súhrn

Vo vybraných lesných (bukových, smrekových) oblastiach Slovenska v Západných Karpatoch sa sledovala úroveň znečisteného ovzdušia prostredníctvom pasívnych zberačov. Vzájomným porovnávaním nezalesnených plôch a porastov sa ukázalo, že zaťaženie voľnej plochy bolo spravidla vyššie ako zaťaženie porastu. S výnimkou bukového stacionára, kde bola pôvodne voľná plocha prirodzene zmladená a preto rozdiel medzi záťažou obnovenej plochy a starého porastu nebol výrazný.

Výsledky protónovej záťaže poukazujú na závislosť od nadmorskej výšky. V porovnaní s inými oblasťami Slovenska, i strednej Európy, ovzdušie v oblasti bukového stacionára Kremnické vrchy (470–510 m n. m.) možno považovať za mierne zaťažené imisiami (9,3 mmol H^+ deň$^{-1}$ m^{-2}). V bučinách Žiarskej kotliny (470 m n. m.) boli v roku 1992 hodnoty 1,3-krát vyššie. V Biosférickej rezervácii Poľana (1370–1380 m n. m.), s prevahou smrečín boli štvornásobne vyššie ako znečistenie v oblasti bukového stacionára. V Národnom parku Nízke Tatry (1440 m n. m.) bola priemerná ročná hodnota 3,5-krát vyššia ako v BEES Kremnické vrchy.

Pri hodnotení odolnosti pôd voči ich acidifikácii antropogénnou činnosťou sa najodolnejšou znova javí oblasť Kremnických vrchov. Porasty v polohách nad 600–700 m n. m. sú z pohľadu kontaminácie zaťažované najviac. Avšak acidita pôd v týchto oblastiach nie je extrémna a možno konštatovať, že pôdy majú dobrú tlmivú i regeneračnú schopnosť.

Použitá metóda pasívnych zberačov umožňuje nenáročným spôsobom mapovať pôsobenie polutantov v prostredí lesných ekosystémov.

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