

## TRENDS IN ATMOSPHERIC POLLUTION, DEPENDENT ON DISTANCE FROM A POLLUTION SOURCE

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### Abstract

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A research on the polluted atmosphere was conducted in three different localities over a ten-year period. On all the plots, there was pursued parallel monitoring of trends, with focus on intensity of the immission load impact in dependence on altitude, season, meteorological and climatic processes in the boundary layer of the atmosphere and on the situation in the forest stand or in open area. The highest values were measured on the research monitoring plots (RMP) Poľana Mts – both on the open plot  $21.8 \text{ mmol H}^+ \text{ day}^{-1} \cdot \text{m}^2$ , and in the forest stand  $17.5 \text{ mmol H}^+ \text{ day}^{-1} \cdot \text{m}^2$ ; the lowest at the EES Kremnické vrchy Mts – on the open plot  $12.1 \text{ mmol H}^+ \text{ day}^{-1} \cdot \text{m}^2$ , in the stand  $11.5 \text{ mmol H}^+ \text{ day}^{-1} \cdot \text{m}^2$ . In both cases, the phenomenon is local. The linear trend at the RMP Žiar and EES Kremnické vrchy Mts was moderately increasing; opposite it was the situation at the RMP Poľana Mts where it the local trend was influenced by the transboundary transport of polluting substances. The supposition that the deposition of acid components is higher in the out-of-vegetation period than in the vegetation period was found true for the RMP Poľana Mts. Over the whole 10-year period the load to open plot higher was compared to the stand interior on the RMP Žiar and Poľana Mts; this was also confirmed exactly, with statistical characteristics. The trends on plots at the EES were different, owing to the changes in density.

*Key words:* air pollution, proton load, forest ecosystems, deposition

### Introduction

Unfavourable influence of atmospheric deposition in the last twenty years has had negative consequences on health condition both of coniferous and broadleaved woody plants. The actual state of the air quality is monitored using an automatic system with measuring equipments installed in industrial regions. Long-term measurements of pollutants concentrations in forest ecosystems can be provided using a manual sorption-accumulation method according to Obr (1989) that provides data about the immission load and allows extrapolation

tion in the case of localities not covered by the mentioned industrial immission monitoring network. We pursued the research at three different plots in three main vegetation zones in Slovakia. The examined localities had different immission load intensity, altitude and tree species composition.

## Material and methods

In terms of threat to the environment by airborne pollutants (immissions), acid components are highly dangerous, primarily the compounds of sulphur, nitrogen and the hydrogen ions  $H^+$ . Observations on long-term influence of immission load to forest ecosystems and the differences between the seasonal concentrations can be performed using the method of proton load ( $H^+$ ). This method is based on the interception of gaseous ( $SO_2$ ,  $NO_x$ , HF), liquid ( $HNO_3$ ,  $H_2SO_4$ ) and solid particles ( $NH_4HSO_4$ ) on the filtration paper surface. Proton load is connected with acid deposition neutralizing the alkalinity of the testing solution exposed in the field. The non-neutralised remainder is obtained as the difference between the non-exposed and exposed absorption solutions by titration with HCl on the Tashiro indicator (Kellerová, 1999). The indication is realised at Research Monitoring Plots (RMP) Žiar, Poľana Mts and at the Ecological Experimental Site (EES) Kremnické vrchy Mts. The pairs of parallel collectors (samplers) were situated on non-forested plots (stocking 0.0) and in forest stands. The region of Žiarska kotlina basin belonged for long years to the most polluted areas in the Slovak Republic. However, since the beginning of the 90s years of the last century, there has been evident decrease in the emitted industrial pollutants.

The research monitoring plot (RMP) Žiar is situated in the southern part of the Štiavnické vrchy Mts at the altitude of 470 m a.s.l. The mean annual temperature at the site is 8.0-8.5 °C and in the vegetation period 14.5 -15.5 °C, mean annual precipitation total is 700–750 mm. The mean wind speed is low, with prevailing east and northwest directions. The proportion of calm events with practically zero dispersion of air pollutants is 40.9%. These facts are to a considerable extent determining the distribution of airborne pollutants over the considered area. The research-monitoring plot is situated in a beech ecosystem, the parallel pairs of collectors have been placed in the open space and in the forest stand with an original stocking of 0.9. The principal contaminants at the locality were found to be oxides of sulphur and fluorine.

The area of the research-monitoring plot Poľana Mts is less loaded with local airborne pollutants, however, it is much more threatened by transboundary transport of noxious materials, primarily oxides of sulphur and nitrogen. The values of sulphur oxides determined in the atmosphere at the regular monitoring by the Slovak Hydro-meteorological Institute, Centre Banská Bystrica, have not exceeded the allowable limit (Šipikalová, Kamenský, 2001). The plot is situated in a spruce ecosystem belonging to the territory of the PLA Poľana, Zadná Poľana Mts, at 1360–1380 m a.s.l. The site exposure is SE, and it is covered with a 110–125 year old beech stand with an original stocking of 0.6–0.7. The mean annual air temperature is about 3.7 °C, mean annual precipitation total ranges between 700–1300 mm.

The experimental site EES Kremnické vrchy Mts is localised at 480–510 m a.s.l., in SE part of the Kremnické vrchy Mts, in the upper part of the catchment area of Kováčovský potok stream (Bublinec, Dubová, 1989). The annual precipitation total ranges from 510–1040 mm, in the vegetation period from 160–530 mm (Dubová, 2001). The mean annual temperature at the site according to Janík (2005) is 6.8 °C. The EES mainly consists of a beech stand, the collectors are situated on an open plot and in the stand with an original stocking of 0.7. In the years following a cutting intervention responded the local vegetation to the actual changes; and Barna (2000, 2001), considering the actual facts, précised in 1996 the stand density value as 0.78. The EES is classified as a so – called. “Clear locality“, that means a locality outside of direct influence of important industrial zones.

The individual partial plots of the BEES Kremnické vrchy Mts have been provided with precipitation samplers and there have also been supplied data from the SHMI meteorological stations in the vicinity. In the beginning with the plot establishment, there have been performed regular monthly calculations of the precipitation amounts collected in the samplers. These values are further processed to obtain mean montly and annual precipitation totals, maximum and minimum values and mean annual values over long periods. The precipitation is considerably variable with time and space, depending on the altitude and on the wind strength and direction.

## Results and discussion

The actual status and trends in air pollution were examined in three territories, over a ten-year period. We studied the trends in changes, focussing to the intensity of the immission load impact in dependence on the altitude, season, meteorological and climatic events in the boundary layer of the atmosphere and the situation in open area or in forest stand. For the period 1993–2002 we calculated and compared mean daily values for the proton load on the individual plots. The highest values were measured on the RMP Poľana Mts, both on the open plot 21.8 mmol H<sup>+</sup> day<sup>-1</sup> · m<sup>-2</sup>, and in the stand 17.5 mmol H<sup>+</sup> day<sup>-1</sup> · m<sup>-2</sup>, the lowest at the EES Kremnické vrchy Mts: on the open plot 12.1 mmol H<sup>+</sup> day<sup>-1</sup> · m<sup>-2</sup>, in the stand 11.5 mmol H<sup>+</sup> day<sup>-1</sup> · m<sup>-2</sup>.

In 1994–1997 the values of load with airborne pollutants to the RMP Žiar had in general decreasing trend. This trend was connected with the decrease in industrial activities and with updated technology and improved burning processes (Závodský, 1995). A moderate increase in the recent years followed from a failure in the separator of one of the polluters and also from the increase in electric energy production. Similar situation was at the EES where was evident influence of more remote pollution sources (Zvolen, Banská Bystrica, Žiar nad Hronom). In both cases we deal with a local phenomenon, apart from emission conditions, also dependent on the meteorological differences between individual years. The situation is different at the RMP Poľana (Fig. 1), with a decreasing trend – influenced, the confidence value is, however, lower than the threshold of statistic significance ( $P = 0.05$ ). In the recent past, the main source of pollutants was considered to be the Cement-mill in Banská Bystrica (Válka et al., 2001). At present a stagnation in the emitted SO<sub>2</sub> amounts can be supposed – entailed by the overall decrease in the number of the polluting sources.

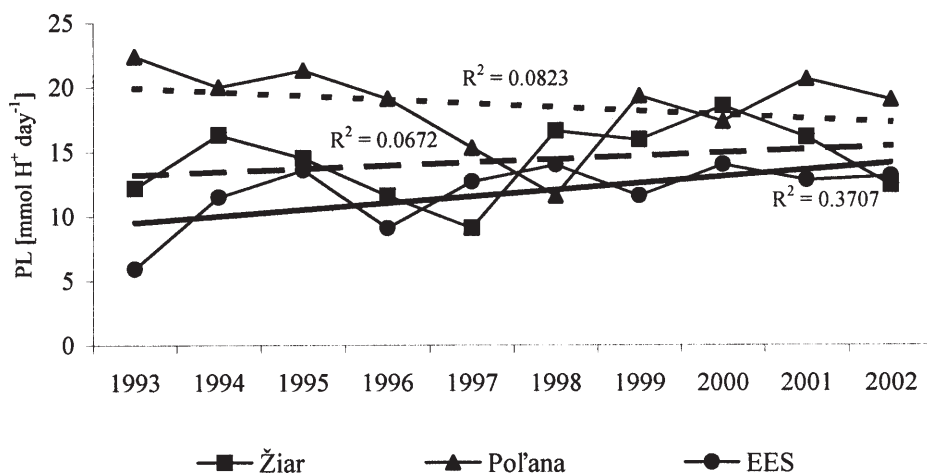


Fig. 1. Linear trends in proton load on plots with different degrees of air pollution (PL – proton load).

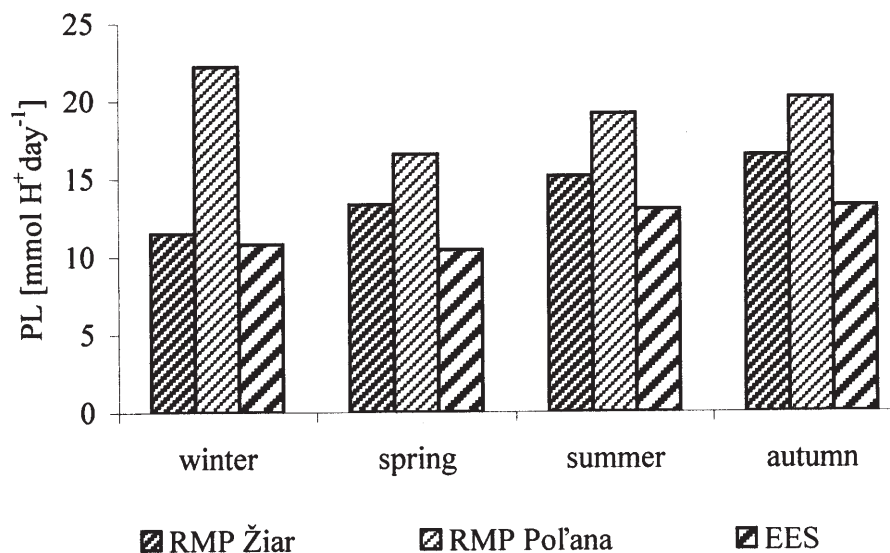


Fig. 2. Deposition load and the differences in seasonal concentrations (PL – proton load).

As protons are characteristic for the acid component of airborne pollutants, we can compare the mean annual values to the emitted amounts of sulphur dioxide that show, since the 90s of the last century, a decreasing trend (Závodský, Pukančíková, 1995).

From the literature we know that the deposition of acid components in out-of-vegetation period is lower than in vegetation period. Consequently, we supposed that the highest values should be observed in the winter months. Nevertheless, the daily mean values calculated over the examined 10-year period show that the situation was different in dependence on plots. In winter we observed the highest value of 22.2 mmol H<sup>+</sup> day<sup>-1</sup>·m<sup>-2</sup> only at the RMP Poľana Mts (Fig. 2).

One of the reasons why the values of concentrations of proton load found in Žiar and at the EES were found surprisingly higher in the period when it was not expected is temperature, primarily temperature extremes. Soták (1998) means that in 1987–1996 increased the mean annual temperature in the Žiarska kotlina basin by 0.7 °C, also as a consequence of milder and drier winters. Relatively higher daily temperatures during mild winters decrease the rate of airflow determining the distribution of polluting materials and creating favourable conditions for their dispersal.

The results of research on the quantity of proton load point out the different intensity of the airborne pollutant impact upon the stand interior and the open plot. Higher load to the open plot in comparison with the stand interior was found on the plots RMP Žiar and Poľana Mts over all the ten years, on the EES over four years only. This fact has also been confirmed by the statistical characteristics: mean, median, minimum and maximum values (Table 1, Fig. 3).

T a b l e 1. Basic statistical characteristics of proton load for RMP Žiar, RMP Poľana and EES Kremnické vrchy Mts in 1993–2002

Site	RMP Poľana		RMP Žiar		EES	
	OP	S	OP	S	OP	S
Mean	22	18	17	12	12	12
Mean error	0.8	0.7	0.9	1.1	0.9	1
Median	22	18	17	11	13	12
Standard deviation	2.6	2.2	2.8	3.5	2.7	3.2
Variation	6.6	5	7.9	12.6	7.3	10.4
Variation range	8.1	8	9.7	9.7	7.6	11.4
Minimum	17.5	13	10.3	7.4	8	3.8
Maximum	25.6	21	20	17.1	15.6	15.2
Sample size	10	10	10	10	10	10
Variation coefficient	11	12	16	29	23	27

OP – open plot, S – stand

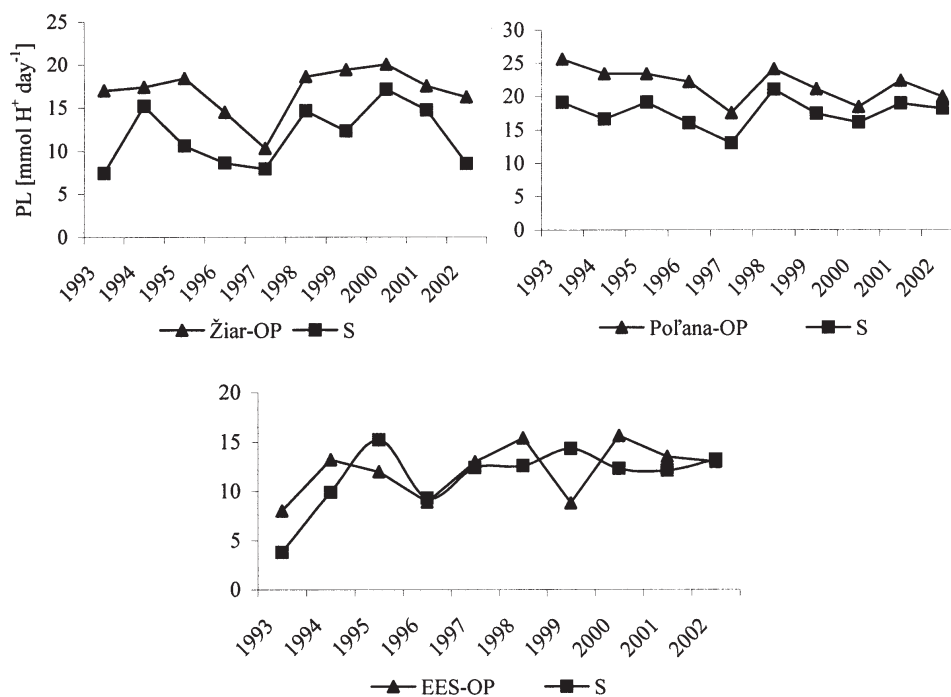


Fig. 3. Influence of forest stand and open area on proton deposition.

Similar facts are known from the literature according to which the amounts of pollutants (e.g.  $\text{SO}_2$ ,  $\text{NO}_x$ ) decrease in direction inwards the stand (Grék, 1991; Škvarenina, 1998; Škvarenina, Mindáš, 1998). The case is interception of acid atmospheric components by tree crowns in fully closed stands (Dubová, Bublínec, 1993).

It was unlike the situation at the EES Kremnické vrchy Mts in other years, primarily in 1995 and 1999, when the values in the stand were higher than in the open area. Negligible differences between the forested and non-forested plots were also found in 1996, 1997, 2001 and 2002. The cutting intervention in 1989 caused gradual changes in the stand density. The dynamics in changes was also reflected on the open plot where we observed increased precipitation totals in 1989–2000 with average values higher than the long-term mean. Practically no difference was found in the stand (Dubová, 2002).

The quantity of the proton load in all localities is obviously influenced by horizontal (in fog, dew, low cloudiness) and indirectly also by vertical constituents (in rain, snow) of wet deposition; by dry deposition and in higher situated parts also by hidden deposition. Fogs, with their high concentrations of polluting materials, have considerable impact on the overall immission load. They are formed in the boundary atmospheric layers where the concentration of pollutants is higher than in the higher layers (Škvarenina, 1998). As in certain periods, namely during inversions when dispersion conditions are reduced, the fogs maintain for a long time, even several days, not only several hours (Petrovič, Šoltís, 1981; Kuchaříková, Prošek, 1983), the concentration of contaminating materials considerably increases. Such conditions are common for all examined plots; however, there are great differences in their duration. Another significant factor in the process is the vertical precipitation. Mihálik, Slávik (1991) found that precipitation water fallen through a spruce stand accumulated an appropriate amount of protons. That means that the self-cleaning effect of precipitation can influence the proton amount in atmosphere. In such a way, the proton load values corresponding to lower precipitation totals can be higher compared to those corresponding to more abundant precipitation (Kellerová, Dubová, 2002).

## Conclusion

We studied deposition of proton load to the research monitoring plots Žiar and Poľana Mts and to the Beech Ecological Experimental site Kremnické vrchy Mts over a period of 10 years. The studied localities are different by different intensities of the immission load, altitude, and tree species composition. The research was pursued parallel on all the plots, consequently, the obtained results are comparable. As we expected, the highest values were obtained on the RMP Poľana Mts, the lowest on the EES Kremnické vrchy Mts. However, the differences between the seasonal concentrations were not unambiguous. In the out-of-vegetation periods, the highest values were found on the RMP Poľana Mts. It has been confirmed that the fully closed stands are functioning as a filter for precipitation, gaseous substances and fog drops and that the contents of pollutants ( $\text{SO}_2$ ,  $\text{NO}_x$ ) decrease inwards

the stand. The results of observations on the input of proton load from atmosphere to forest ecosystems complete the overall pattern of the total surface deposition.

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**Kellerová D.: Trendy a závislosť znečistenia atmosféry od vzdialenosti zdroja.**

Výskum znečisteného ovzdušia sme uskutočnili v troch rozličných lokalitách v období desiatich rokov. Paralelne na všetkých plochách sme skúmali trendy zmien s dôrazom na intenzitu pôsobenia imisnej záťaže v závislosti od nadmorskej výšky, sezón, meteorologických a klimatických procesov v prízemnej vrstve atmosféry, voľnej plochy a porastu. Najvyššie hodnoty na voľnej ploche  $21,8 \text{ mmol H}^+ \text{ deň}^{-1} \cdot \text{m}^{-2}$ , v poraste  $17,5 \text{ mmol H}^+ \text{ deň}^{-1} \cdot \text{m}^{-2}$  boli namerané na Výskumnej monitorovacej ploche (VMP) Poľana, najnižšie na Ekologickom experimentálnom stacionári (EES) Kremnické vrchy na voľnej ploche  $12,1 \text{ mmol H}^+ \text{ deň}^{-1} \cdot \text{m}^{-2}$ , v poraste  $11,5 \text{ mmol H}^+ \text{ deň}^{-1} \cdot \text{m}^{-2}$ . V oboch prípadoch ide o lokálny jav. Lineárny trend na VMP Žiar a EES Kremnické vrchy bol mierne stúpajúci, iná situácia bola na VMP Poľana, kde bol trend opačný, ovplyvnený diaľkovým prenosom kontaminantov. Predpoklad, že v mimovegetačnom období je depozícia kyslých zložiek väčšia ako vo vegetačnom období sa potvrdil na VMP Poľana. Počas desiatich rokov bolo zaťaženie voľnej plochy vyššie oproti interiéru porastu na VMP Žiar a Poľana a potvrdzujú to aj štatistické charakteristiky. Na plochách EES Kremnické vrchy dôsledkom zmeny hustoty zakmenenia bol vývoj odlišný.