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Long-term temporal changes of precipitation quality in the mountainous region of Chopok (Low Tatras, Slovakia)

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Abstract

The paper has analysed the data about chemical composition of precipitation at Chopok EMEP station. The database comprised the information about the concentrations of sulphates, nitrates, ammonia ions, base cations and heavy metals (Cd, As, Al, Zn). The results are presented in a graphical form showing long-term temporal trends in the chemical composition of precipitation. The statistical characteristics of temporal trends were tested with Student method analysing the significance of the sampling coefficient of correlation. We revealed that the majority of concentrations of selected elements in precipitation significantly decreased in time.

Key words: precipitation chemistry, atmospheric deposition, temporal trends, acidification

Introduction

Continuous development of industrial and agricultural activities, transport, and the change in natural landscape and its individual components to anthropogenic components contribute to systematic distortion of balance in environment. The atmosphere, that does not recognise borders among countries, is most affected by human activities. Pollutants are transported over long distances of hundreds of kilometres from the pollution sources depending on the time they persist in the atmosphere. Pollutant emissions from stationary and mobile sources undergo chemical changes in the atmosphere, and have direct and indirect effects on human health and environment (SAŽP, 1997). Monitoring of chemical elements and compounds that cause acidification, eutrophication and other chemical processes in environment is still up to date. Acidification and eutrophication can contribute to climate change by influencing the gas exchange between the soil and the atmosphere. The impact of increased emissions of NOx is most discussed (BARTOŇOVÁ, 2009).

The area of the Slovak Republic is situated at the south-eastern edge of the area with the highest regional air pollution and acidity of precipitation in Europe (SHMÚ, 2009). The problems with acid rains occurred several times in the last century. The first significant decrease of pH in precipitation was recorded in the 60s of the last century due to continually increasing concentration of SO_2, and NO_x in the atmosphere (BARANČOK & VARŠAVOVÁ, 1998). The residence time of SO_2 in the air is 1-3 days, while the residence time of nitrogen oxides is 1-10 days.

Several papers have dealt with the changes in precipitation chemistry in relationship to elevation and season (e.g. BUBLINEC & DUBOVÁ 1994). The results from the observations point out at significant acidification of through-fall in mountainous spruce forests, with the gradient of pH decrease equal to 0.32 units per 100 elevation metres. The authors found that the acidity was the highest in spring and winter, while pH slightly increased as autumn was approaching. Similar results were also presented by ŠKVARENINA (1994).

The summary report on the environment in Europe (EEA, 1998) published by the European Environment Agency (EEA) with the headquarters in Copenhagen presents that the emissions of acidifying pollutants have significantly decreased since 1990, particularly in Central and Eastern Europe as a result of economic restructuring and gradual introduction of modern technologies. The reduction in Western Europe is primarily connected with the changes in the use of fuels, desulphurisation and denitrification of combustion gases, and the introduction of three-way catalysts in cars. Due to significant reduction of emissions, no further acidification occurs in the majority of European ecosystems, although several risk areas still exist situated mainly in Central Europe (HŮNOVÁ et al., 2009).

In connection with the significant reduction of SO_2 emissions in Slovakia as well as in the whole Europe and with far more moderate decrease of NO_x emissions, in the last decade we have observed a change in the ratio between sulphates and nitrates, and hence, also a change in their impact on precipitation acidification. While before the sulphates dominated, nowadays nitrates have become more important. This change can also modify the impact of acidic atmospheric deposition on vegetation. Some studies point out that the precipitation containing more sulphates is more toxic than the precipitation comprising more nitrates although their pH is equal (ASHENDEN, 2002).

Materials and methods

Study area

The meteorological observatory of the Slovak Hydrometeorological Institute (SHMÚ) on Chopok is situated at the ridge of the Low Tatras at an elevation of 2,008 m a.s.l., longitude 19°35'32", and latitude 48°56'38". The measurements started in 1977. Since 1978, this observatory has been a member of the EMEP (Environment Monitoring and Evaluation Programme) and GAW/BAPMoN/WMO networks.

Chopok station belongs to a cold climatic region. Long-term mean of precipitation totals (1951-1980) is 1,142 mm, from which 667 mm falls in the summer half-year. A more detailed climatic description is shown in Figure 1.

Emission situation

According to NEIS (National Emission Inventory System), the main polluters in the district of Liptovský Mikuláš are 4 big and 100 medium sources of air pollution concentrated into two industrial centres of two towns – Liptovský Mikuláš and Liptovský Hrádok. In 2003, these sources produced in total 45.562 tons of particulate matter,756.205 t SO_2, 211.959 t NOx, 85.372 t CO and 24.181 t TOC to the atmosphere. In the study area, no significant sources of pollution exist, only small sources from the category of fuel-energy industry. The local air pollution comes from line sources, particularly road II/584 between Liptovský Mikuláš and Demänovská Dolina. CO, NOx, volatile non-methane hydrocarbons are mainly emitted to the atmosphere from these line sources. To a lesser extent, the combustion gases of cars also comprise the compounds of SO_2, CH_4, N_2O, Pb, HN_3, CO_2.

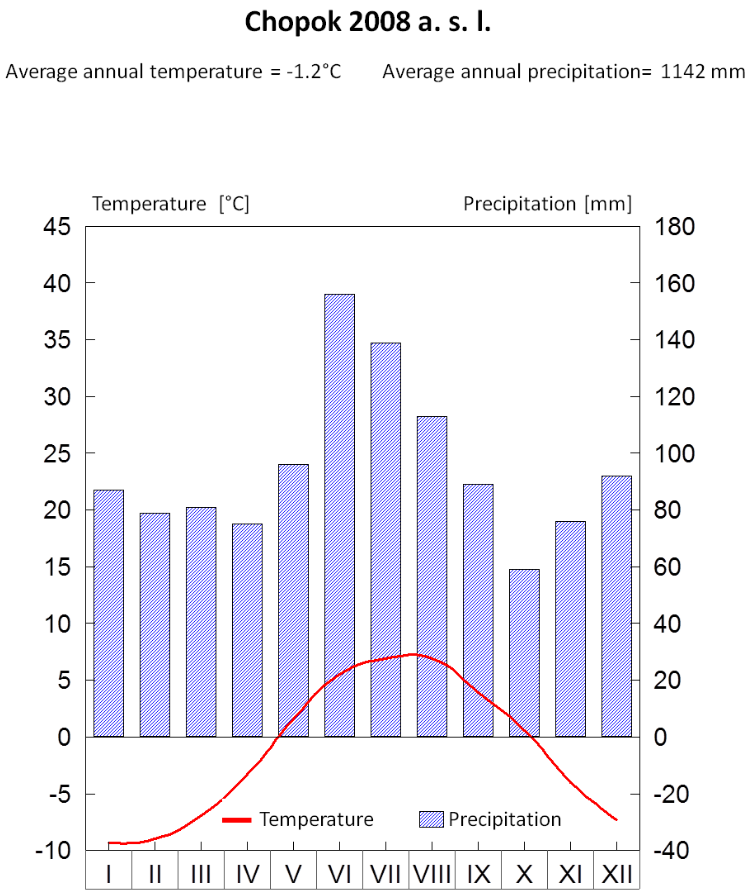

Figure 1 Climate data for Chopok

Pollution situation

The immission load of the area can be characterised only at places where the state of the atmosphere is monitored. For this reason, an automatic monitoring station is installed on Chopok. It is a regional station that belongs to a network of EMEP (European monitoring evaluation programme – programme for monitoring and evaluation of long-range transmission of air pollutants in Europe).

According to the results of EMEP measurements, the Slovak Republic (SR) is situated at the south-eastern edge of the area with the highest regional air pollution and precipitation acidity in Europe. The acidity of precipitation at this station represented by pH was 4.5 in 2000, which indicates the most acid precipitation from all regional stations in Slovakia (SHMÚ, 2010).

Sampling and analytical methods

Sampling and analytical methods used for the precipitation sampling and chemical analytical methods are presented in Tab.1

Tab 1 Method of sampling and analytical methods, Chopok (EMEP, 2010)

	Sampling	Sampling frequency	Analysis method
Precipitation			
Precipitation amount	SK02: Bulk,	Daily	
Sulphate	SK02: Bulk	Daily	Ion chromatography
Nitrate	SK02: Bulk	Daily	Ion chromatography
Ammonium	SK02: Bulk	Daily	Ion chromatography
Magnesium	SK02: Bulk	Daily	Ion chromatography
Sodium	SK02: Bulk	Daily	Ion chromatography
Chloride	SK02: Bulk	Daily	Ion chromatography
Calcium	SK02: Bulk	Daily	Ion chromatography
Potassium	SK02: Bulk	Daily	Ion chromatography
pH	SK02: Bulk	Daily	pH meter

Statistical characteristics were calculated in Microsoft Excel 2007. From the basic tabular values of weighted concentrations of pollutants in precipitation we first calculated deposition using the following formula (1)

$$\text{Deposition (N)} = \text{RR} \times \text{CON (N)} \qquad (1)$$

Deposition of a pollutant N [$kg.ha^{-1}.year^{-1}$]

RR – precipitation amount (mm);

CON – concentration of a pollutant: $mg.l^{-1}$, of heavy metals in $\mu g.l^{-1}$;

N – pollutant;

The analysed characteristics were visualised in Microsoft Excel 2007 as a time series described by a regression line. Statistical characteristics of temporal trends were tested with Student test of significance of sampling coefficient of correlation with $f=n-2$ degrees of freedom.

Results

The graphs presented below are based on the tabular values from EMEP, which include the values of pH, activity of H+, concentrations of sulphates converted to sulphur, and nitrates and ammonia ions converted to nitrogen. Afterwards, deposition of individual chemical elements was calculated from these values, and time series were evaluated as presented in tables.

From the graphical representation of the values and the trend regression line (Fig.2, Fig.3) it is obvious that from the long-term point of view, precipitation totals slightly increased by 6.4 mm of atmospheric precipitation per year. The correlation in the analysed time series is significant at 95% significance level, which means that the temporal trend is significant.

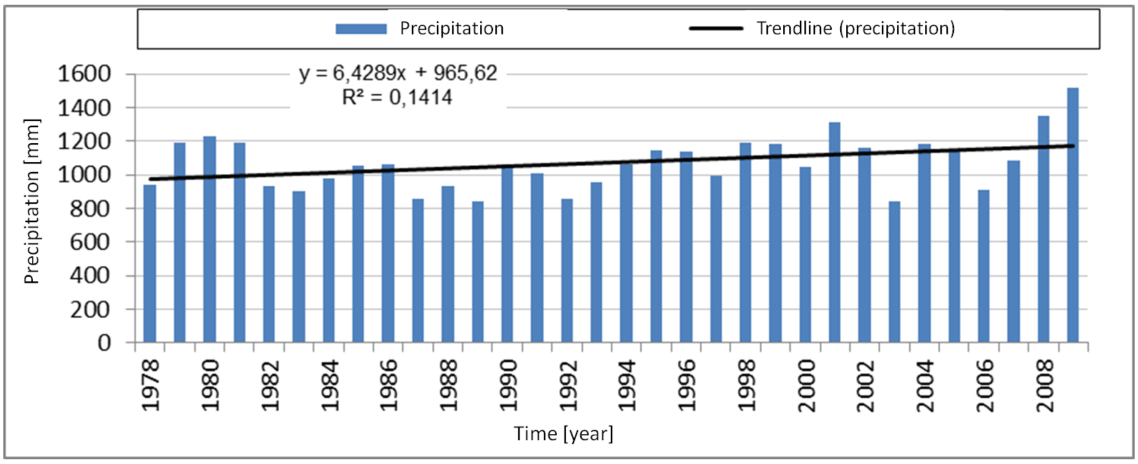

Figure 2 Annual precipitation totals during 1978-2009

The acidity of atmospheric precipitation significantly decreased during the analysed period. This trend is mainly caused by the reduction of primary acidifying ions, such as sulphate and nitrate ions. The reduction in the concentration of these ions is evident from the following graphs (Fig.4, 5) and Tab 2.

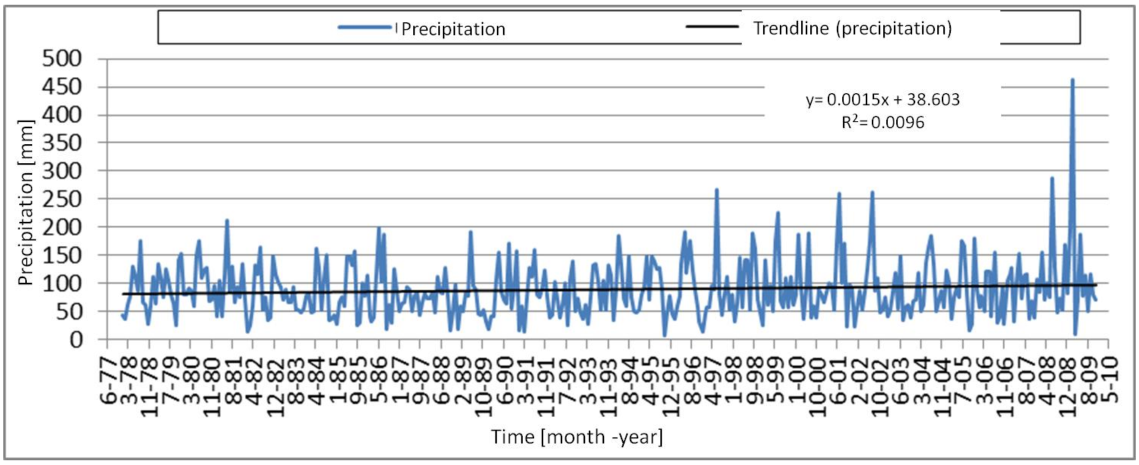

Figure 3 Monthly precipitation totals during 1978-2009

Table 2 Characteristics of the acidifying substances, annual values

Chopok	acidifying substances								
Statistic value	Precipitation			$N\text{-}NH_4^+$		$S\text{-}SO_4^{2+}$		$N\text{-}NO_3^-$	
				concentration	deposition	concentration	deposition	concentration	deposition
	mm	pH	H^+	$mg.l^{-1}$	$kgN.ha^{-1}.year^{-1}$	$mg.l^{-1}$	$kgS.ha^{-1}.year^{-1}$	$mg.l^{-1}$	$kgN.ha^{-1}.year^{-1}$
Rate of change mm; $mg.rok^{-1}$; $kg.ha^{-1}.rok^{-1}$	6.4289	0.0224	-2.00E-06	-0.0341	-0.3443	-0.0669	-0.6557	-0.117	-0.0756
tendency	increasing	increasing	decreasing	decreasing	decreasing	decreasing	decreasing	decreasing	decreasing
P=1-α%.level of significance	95.00%	99.90%	99.90%	99.90%	99.90%	99.90%	99.90%	99.90%	99.90%
max	1519.8	4.95	0.0001	1.37	15.7405	2.59	30.9202	0.61	6.4341
min	840.27	3.93	0.00001122	0.37	4.234169	0.41	4.3584	0.25	2.860925

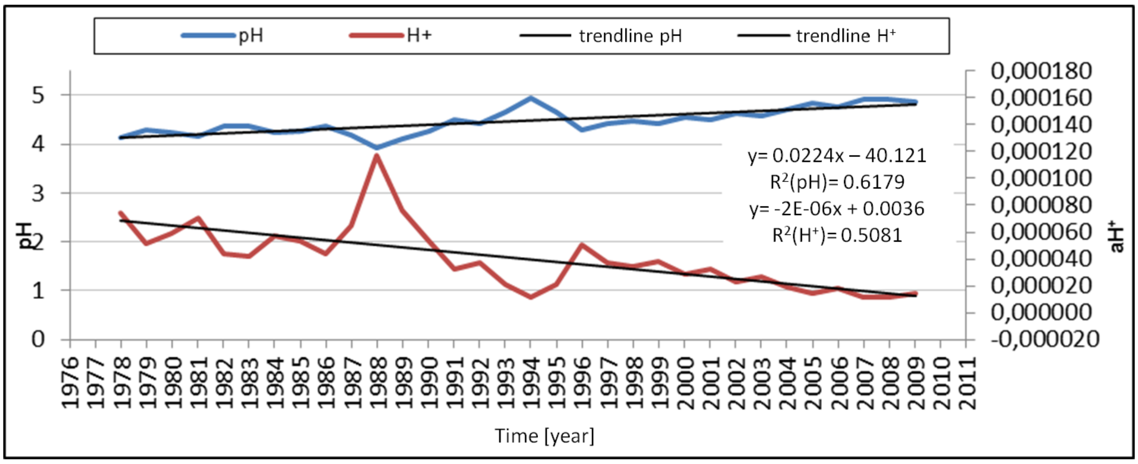

Figure 4 Annual weighted means of pH and aH^+ in precipitation during 1978-2009

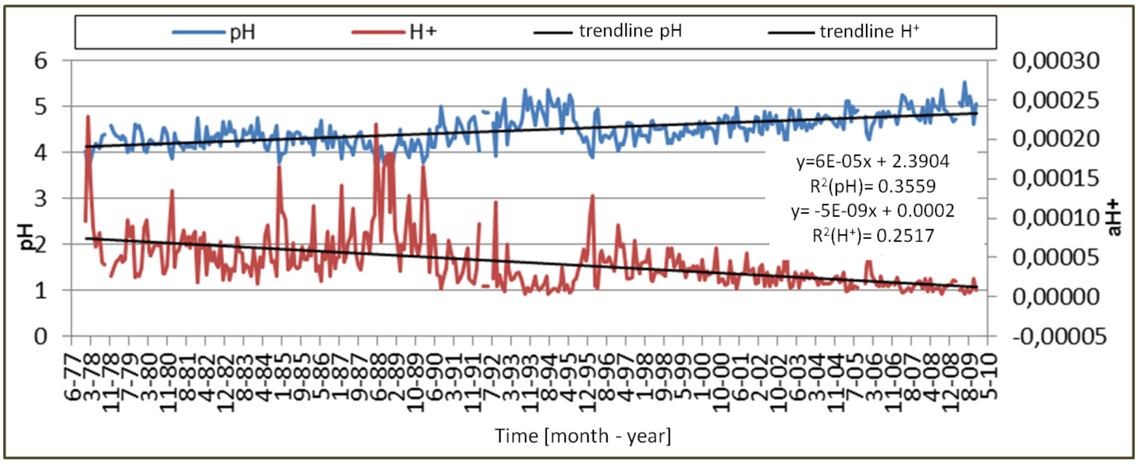

Figure 5 Monthly weighted means of pH values and activity of H^+ in precipitation during 1978-2009

Sulphate ions were dominant in precipitation, and contributed to the acidity of precipitation most. Their concentration was decreasing at a rate of -0.0669 $mg.l^{-1}$ per year and -0.0002 $mg.l^{-1}$ per month over the whole period from 1978 to 2009 as shown in Fig 6. This decreasing trend correlates with the time series at 99.90% significance level. The overall reduction in the concentration of sulphates coincided with the long-term reduction of SO_2 emissions since 1980 (SHMÚ, 2008). The monthly weighted means of sulphate concentrations converted to sulphur fluctuated from 0.23 to 7.07 $mg.l^{-1}$, which indicates that the values of sulphate concentrations are very variable, although their variance gradually decreased (Fig.7).

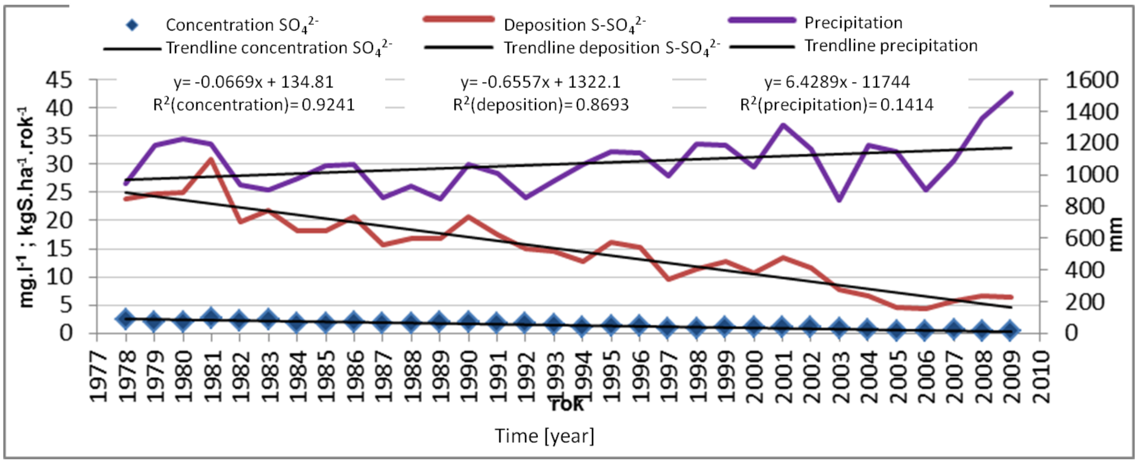

Figure 6 Deposition of S-SO_4^{2-}, precipitation totals, concentration of S-SO_4^{2-} from annual weighted means during 1978-2009

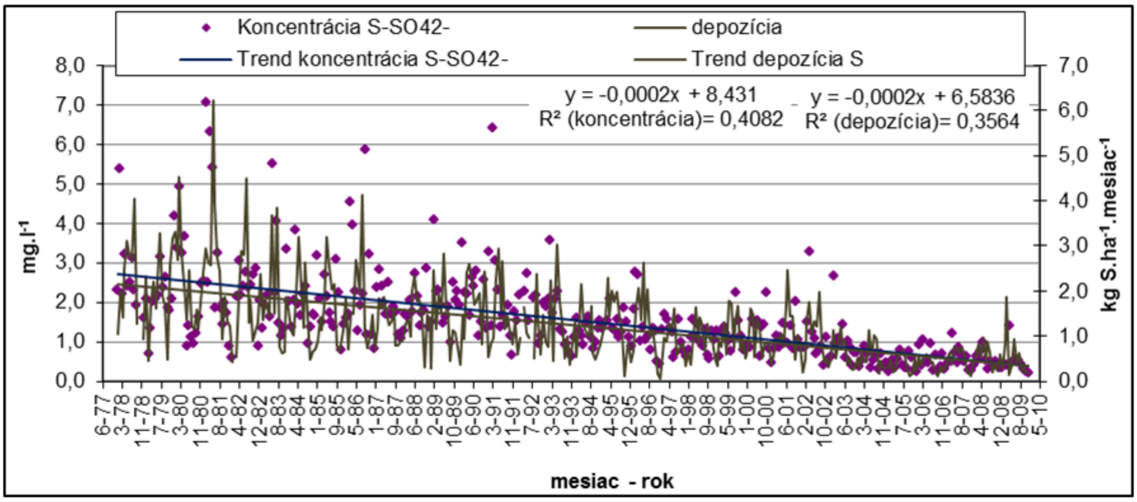

Figure 7 Monthly weighted means of concentration of S -SO_4^{2-}, deposition of S-SO_4^{2-}, and precipitation during 1987-2009

At Chopok EMEP station, the concentration of nitrates started to be measured and assessed in October 1985. The results of the analyses showed a decreasing trend in the concentration of nitrates in precipitation presented in Fig. 8 and 9, which was also statistically confirmed at 99.90% level. The concentration was decreasing at a rateof -0.117 mg.l^{-1} per year. The amount of nitrates in precipitation did not change as fast as the amount of sulphates. Considering the factors and the sources of elements affecting chemical composition of precipitation, the emissions from fossil fuels and car traffic are the main source of nitrates in precipitation.

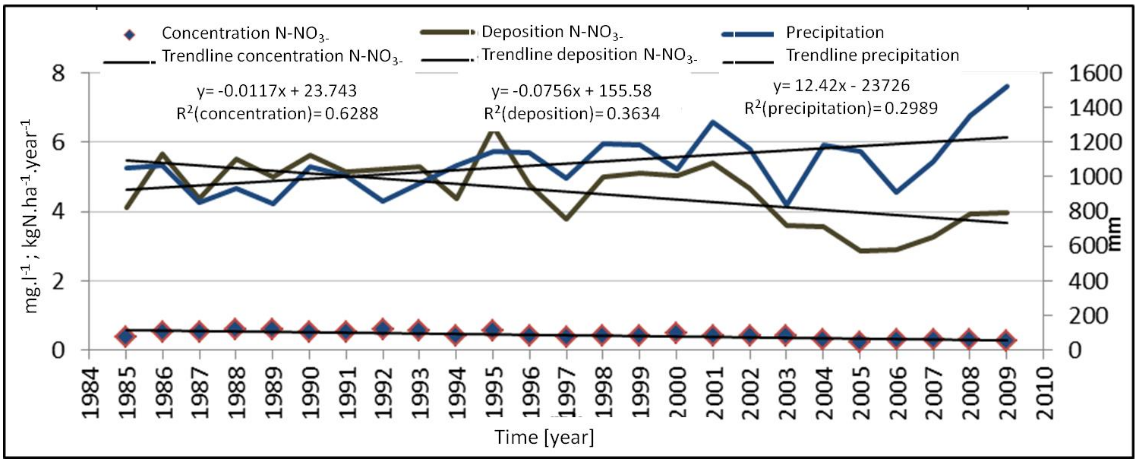

Figure 8 Deposition of N-NO_3^- , precipitation totals, concentration of N-NO_3^- from annual weighted means during 1985-2009

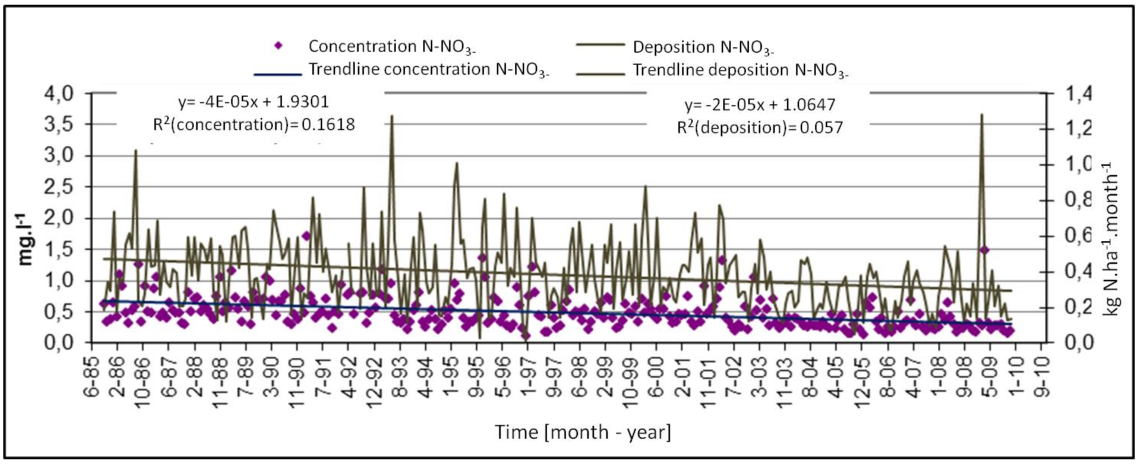

Figure 9 Monthly weighted means of N-NO_3^- concentrations and N depositions in precipitation during 1985-2009

The measurements of ammonia ions started at Chopok EMEP station in 1994. The values of the annual weighted means of NH_4^+ cations converted to nitrogen were almost two-fold of the values of nitrate nitrogen in precipitation. The analysis of the temporal changes in concentrations of ammonia nitrogen revealed a decreasing trend in the time series at a rate of -0.0341 mg.l^{-1} of nitrogen per year. The decreasing trend is evident from Fig. 10 and 11 and was confirmed at 99.90% significance level. In spite of the decreasing tendency of the time series, high concentrations were also recorded, e.g. in April 2009, which was, however, caused by low precipitation total. The results indicate that the higher concentration was not coupled with the higher deposition of ammonia nitrogen.

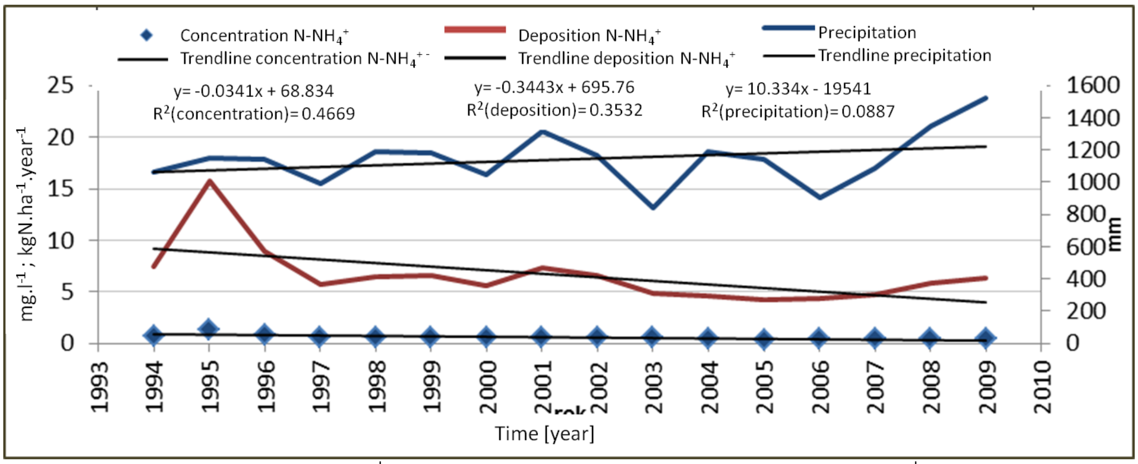

Figure 10 Deposition of N-NH_4^+, precipitation totals, concentration of N-NH_4^+ from annual weighted means during 1994-2009

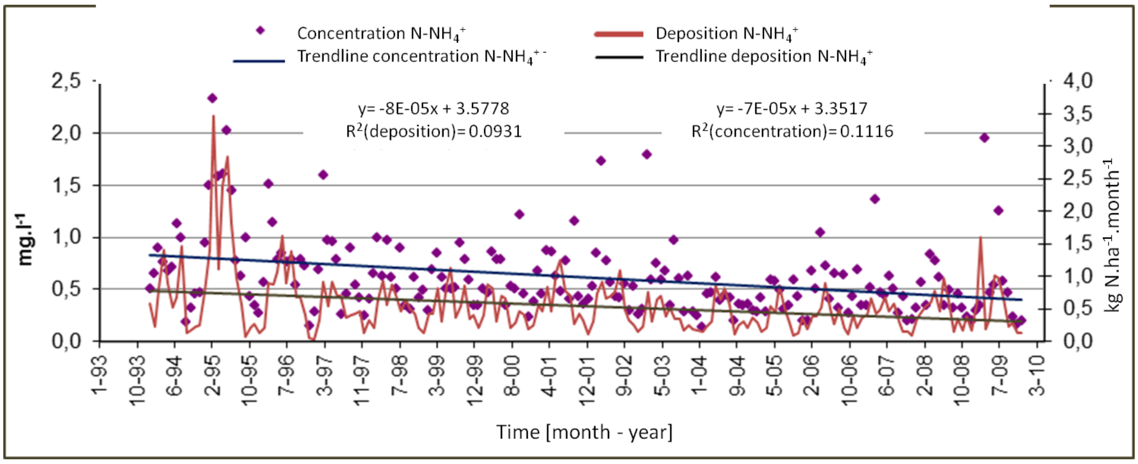

Figure 11 Monthly means of N-NH_4^+ concentrations and N-NH_4^+ depositions in precipitation during 1994-2009

Tab.3 presents the concentrations and depositions of alkaline cations from the tabular values obtained from EMEP and their statistical characteristics.

Table 3 Characteristics of alkaline cations, annual values

Chopok	basic cations								
Statistic value	**Precipitation**	**Calcium**		**Magnesium**		**Potassium**		**Sodium**	
		concentration	deposition	concentration	deposition	concentration	deposition	concentration	deposition
	mm	$mg.l^{-1}$	$kgCa.ha^{-1}.year^{-1}$	$mg.l^{-1}$	$kgMg.ha^{-1}.year^{-1}$	$mg.l^{-1}$	$kgK.ha^{-1}.year^{-1}$	$mg.l^{-1}$	$kgNa.ha^{-1}.year^{-1}$
Rate of change mm; $mg.rok^{-1}$; $kg.ha^{-1}.rok^{-1}$	15.112	-0.0583	-0.5845	-0.009	-0.0891	-0.017	-0.1707	-0.0154	-0.1411
tendency	increasing	decreasing	decreasing	decreasing	decreasing	decreasing	decreasing	decreasing	decreasing
P=1-α%. level of significance	90.00%	99.90%	99.00%	99.00%	95.00%	99.00%	95.00%	99.00%	95.00%
max	1519.8	1.85	21.2554	0.34	3.9064	0.7	8.0426	0.65	7.4681
min	840.27	0.1	0.908	0.02	0.1816	0.06	0.5448	0.068	0.7264

The concentrations of alkaline cations in precipitation were relatively stable. Annual weighted means of concentrations of individual base cations fluctuated as follows: Na^+: 0.068 – 0.65 $mg.l^{-1}$, Mg^{2+}: 0.02 – 0.34 $mg.l^{-1}$, Ca^{2+}: 0.1 – 1.85 $mg.l^{-1}$, K^+: 0.06-0.7 $mg.l^{-1}$. Fig.12 presents the decreasing temporal development in the concentration of base cations and also we can see an extreme value in 1995 and the variation of Ca^{2+} and Na^+ concentrations. In the last period, the concentrations of K^+ and Mg^{2+} were balanced. Fig.13 presents the relative portion of individual base cations in precipitation where calcium is the dominat base cation with relative portion higher than 50%.

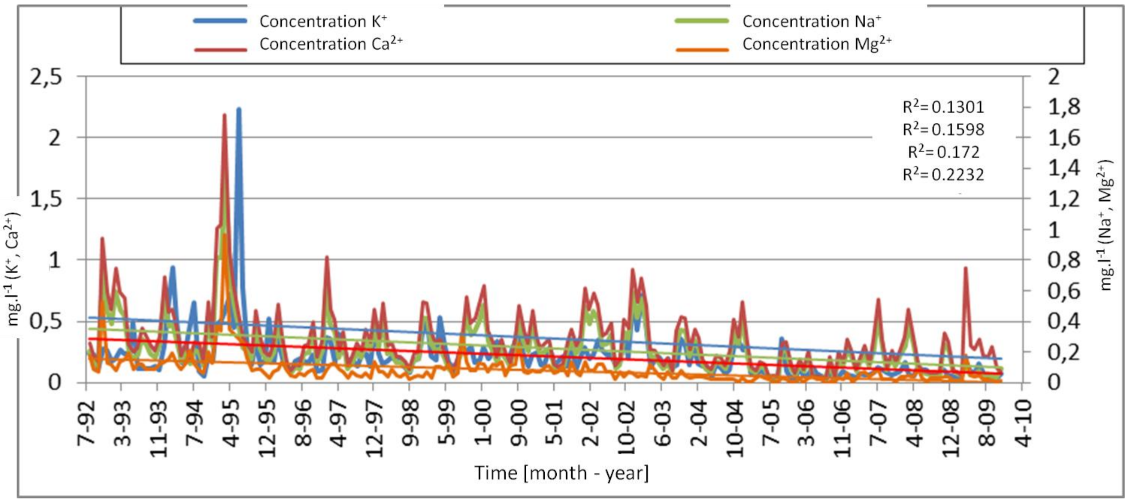

Figure 12 Monthly weighted means of alkaline cations in precipitation during 1992-2009

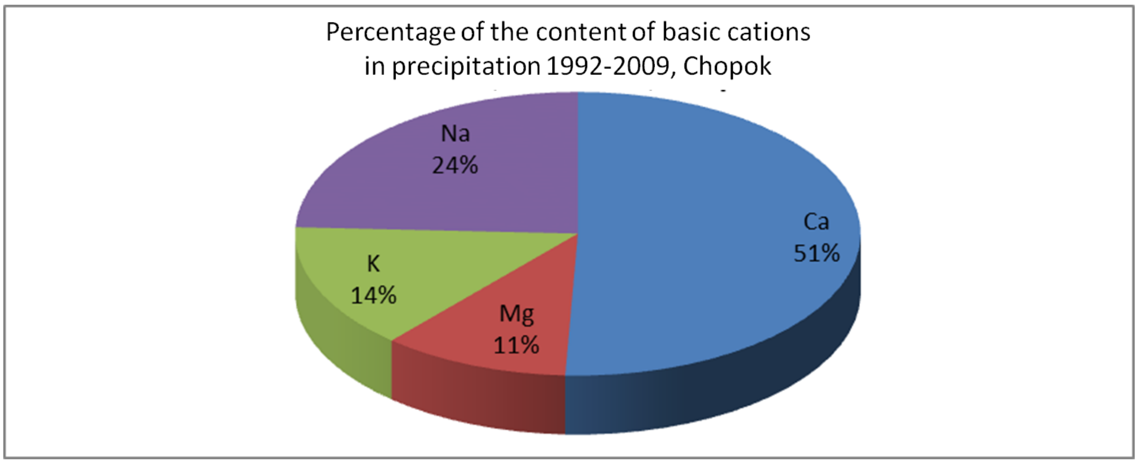

Figure 13 Percentual proportion of alkaline cations in precipitation

Graphical visualisation of long-term temporal trends in concentration of heavy metals in precipitation and their wet atmospheric deposition are presented in Tab.4

We can see great variation in monthly values of aluminium concentrations (see Fig.14) as well as the deposition values (Fig.15). Overall, the concentration decreased, which was statistically confirmed at 99.00%.

Table 4 Characteristics of heavy metals, annual values

Chopok	Heavy metals								
Statistic value	**Precipitation**	**Zinc**		**Aluminum**		**Cadmium**		**Arsenic**	
		concentration	deposition	concentration	deposition	concentration	deposition	concentration	deposition
	mm	$\mu g.l^{-1}$	$gZn.ha^{-1}.year^{-1}$	$\mu g.l^{-1}$	$gAl.ha^{-1}.year^{-1}$	$\mu g.l^{-1}$	$gCd.ha^{-1}.year^{-1}$	$\mu g.l^{-1}$	$gAs.ha^{-1}.year^{-1}$
Rate of chase mm; $mg.rok^{-1}$; $kg.ha^{-1}.rok^{-1}$	15.502	-2.3898	-20.788	-5.3076	-41.57	-0.0788	-0.8117	-0.0181	-0.1002
tendency	decreasing	decreasing	decreasing	decreasing	decreasing	decreasing	decreasing	decreasing	decreasing
P=1-α%. level of significance	99.00%	99.90%	99.00%	99.00%	95.00%	99.00%	99.00%	80.00%	**
max	1519.8	99.416	1004.5788	107.136	1031.5687	0.946	9.8936	0.603	5.4752
min	840.27	16.118	221.3044	22.138	237.9472	0.062	0.6736	0.146	1.5863

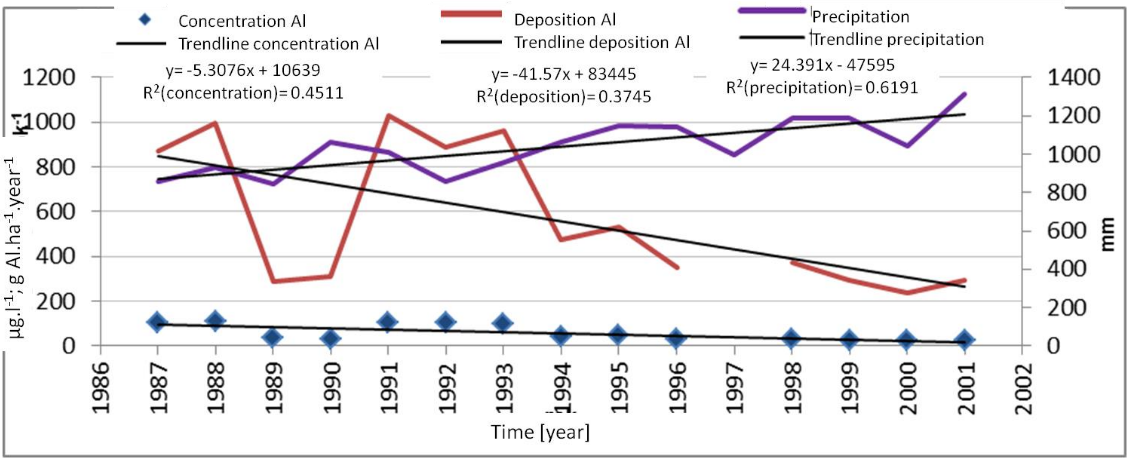

Figure 14 Deposition of Al, precipitation totals, concentration of Al from annual weighted means during 1987-2009

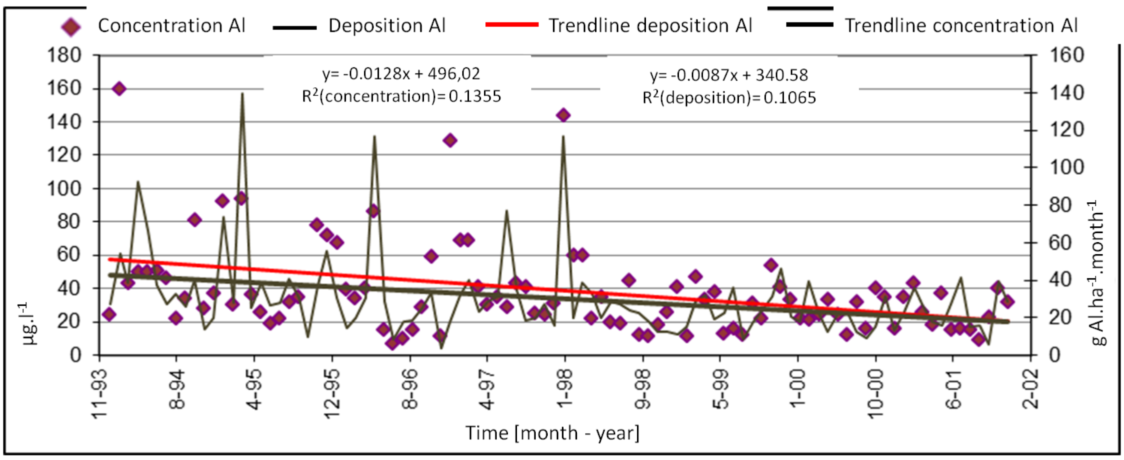

Figure 15 Monthly weighted means of Al concentration and deposition in precipitation during 1994-2001

The concentration of zinc in precipitation has been observed in Chopok EMEP station since 1987. The analysis of temporal change in the zinc concentration in precipitation revealed a significant decreasing trend (Fig.16, 17) confirmed by a statistical test at 99.00%. Monthly values of weighted means of zinc concentration in precipitation were very variable as shown in Fig.17. From this figure we can also see increased concentrations in 2009 due to low precipitation totals in some months.

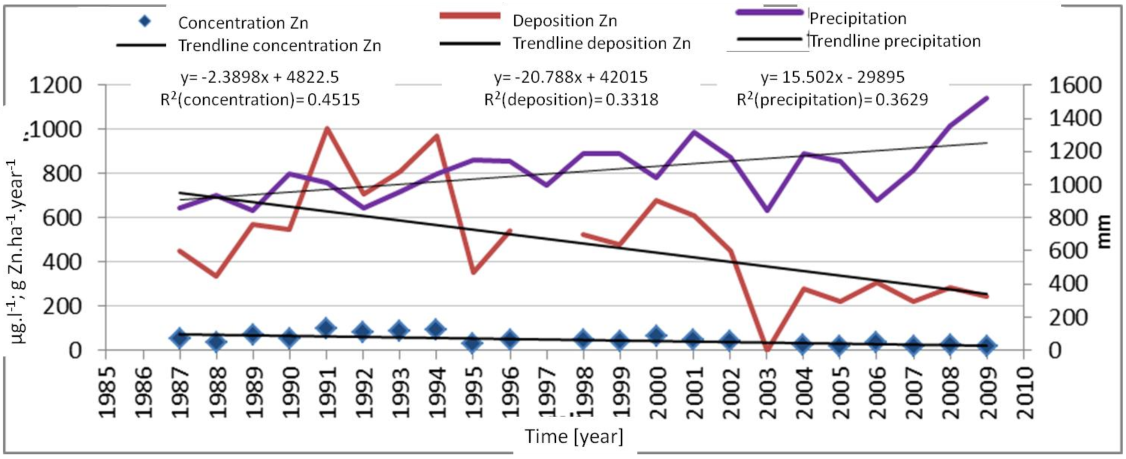

Figure 16 Deposition of Zn, precipitation totals, concentration of Zn from annual weighted means during 1987-2009

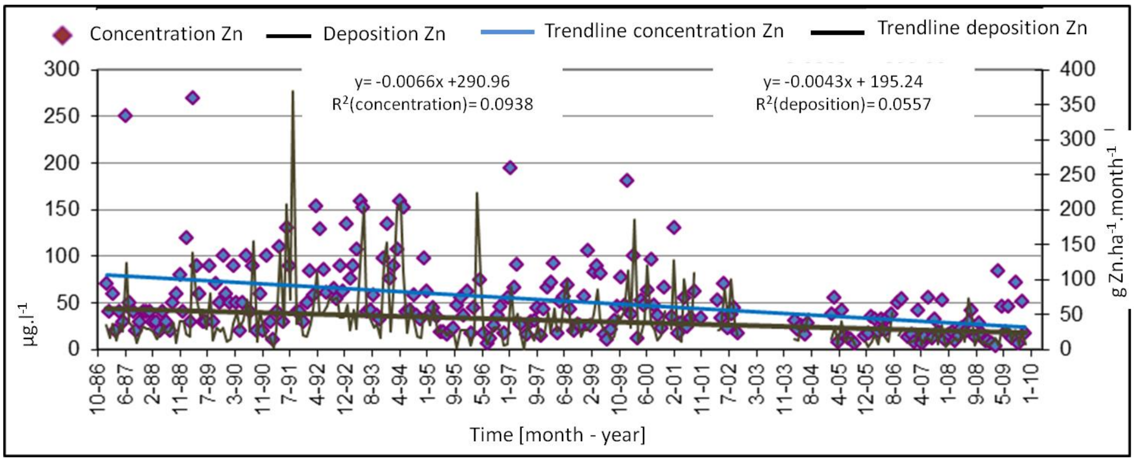

Figure 17 Monthly weighted means of Zn concentration and deposition in precipitation during 1987-2009

The concentrations of Cd did not vary greatly during the ten-year-long measurements. However, the temporal changes in concentrations of Cd had a decreasing trend that was statistically confirmed at 99.00%. Higher concentrations were rare. At the end of the year 2008 and during 2009 we can see an increase in Cd concentrations in precipitation, as well as in its deposition (Fig.18, 19). This is caused by the increased emissions of Cd due to the increased production of copper.

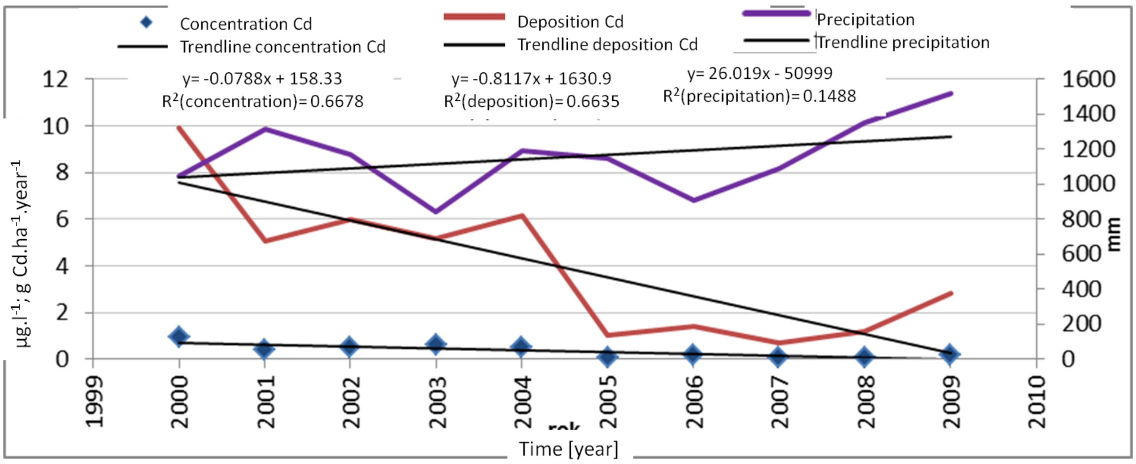

Figure 18 Deposition of Cd, precipitation totals, concentration of Cd from annual weighted means during 2000-2009

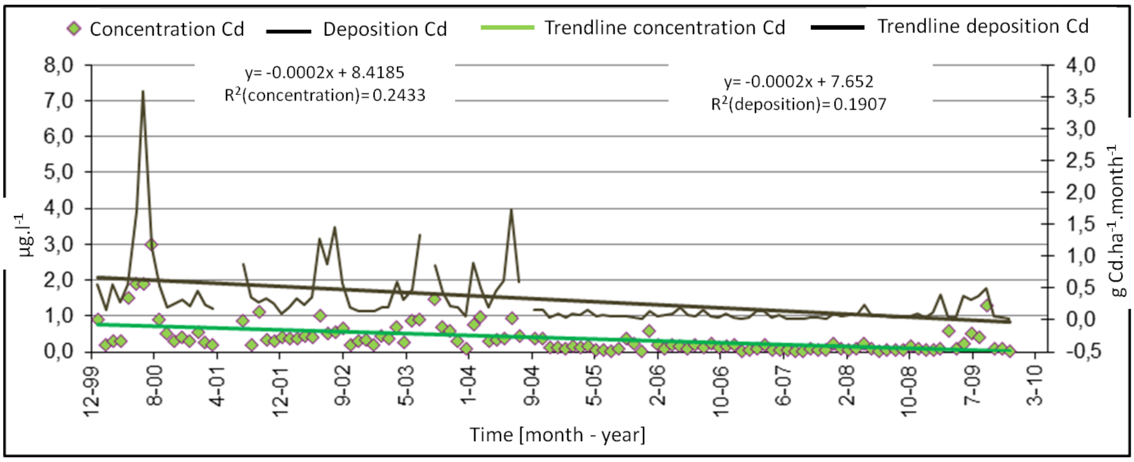

Figure 19 Monthly weighted means of Cd concentration and deposition in precipitation during 2000-2009

Although the trend line of As concentration in precipitation presented in Fig. 20 and 21 has a decreasing character, the analysis confirmed the significance of the sampling coefficient of correlation only at 80.00%. In the years 2008 and 2009 we can see a slight increase in both As concentration and deposition. The maximum annual deposition value was 5.5 gAs.ha^{-1}.year^{-1}.

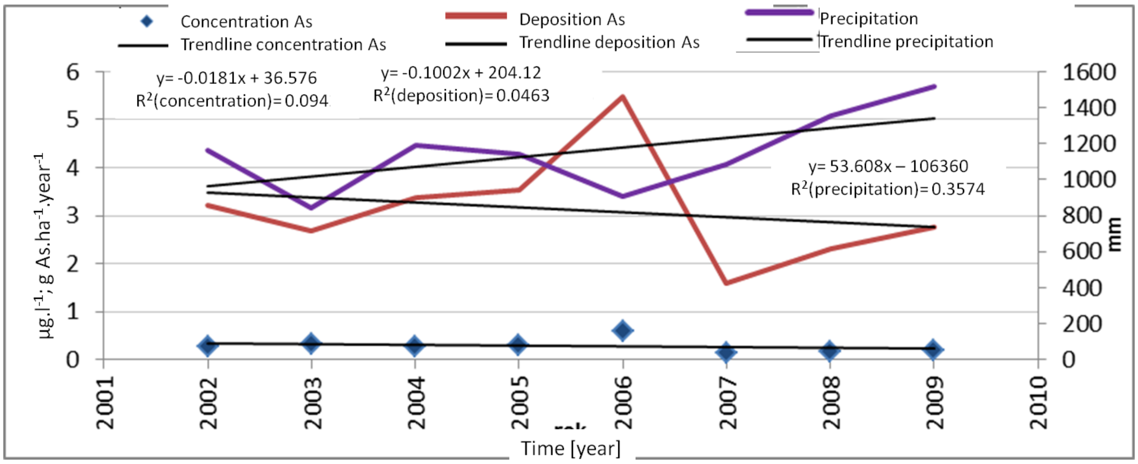

Figure 20 Deposition of As, precipitation totals, concentration of As from annual weighted means during 2002-2009

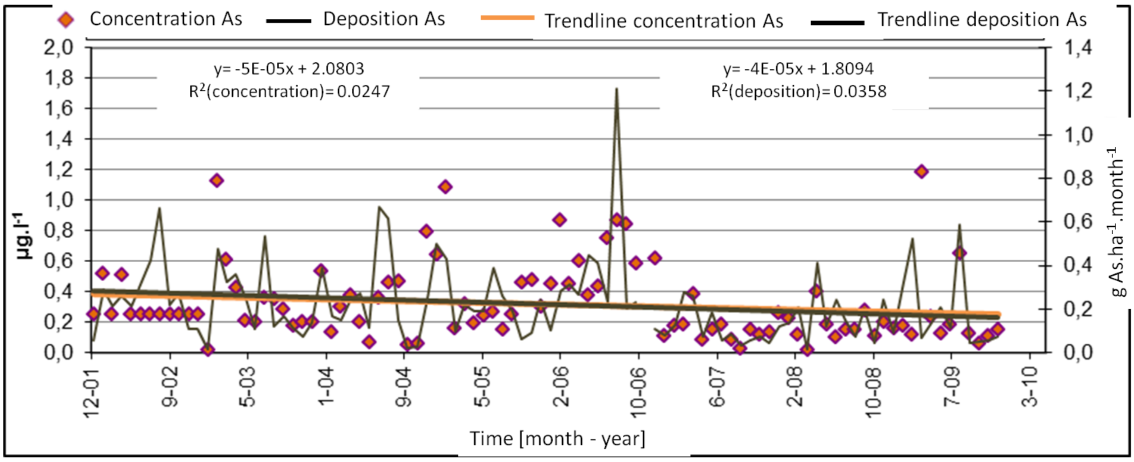

Figure 21 Monthly weighted means of As concentration and deposition in precipitation during 2002-2009

Discussion

The assessment of long-term changes in air quality is very important for evaluating the efficiency of measures taken in order to reduce the emissions of pollutants as well as for evaluating the impacts on e.g. ecosystems (Kunca 2007). A number of authors analysed the trends in the concentration of pollutants in precipitation, e.g. Klemm and Lange (1999) found similar trends of sulphates and nitrates in mountainous regions of Bavaria as on Chopok. Andersson et al. (2007) examined the trends in the concentration of selected parameters of air quality using a complex chemical model and came to similar conclusions as we observed on the selected station of EMEP network.

Interesting changes in the concentrations of sulphates and nitrates in horizontal precipitation were found in Poľana – Hukavský grúň locality by Škvarenina and Minďaš (2001) who also observed a decreasing trend of sulphates in fog water.

Conclusion

The presented work deals with the long-term changes of precipitation quality at Chopok EMEP station situated in the mountainous region of Slovakia. From this station we had a long time series of measured data from monitoring during the period from 1978 to 2009. This period is sufficiently long for the analysis of the development of precipitation quality. The work examined the trends in the concentration of the following elements: S-SO42-, N-NH4+, N-NO3-, Mg2+, K+, Na+, Ca2+, Al, Zn, Cd, As. Processing the precipitation chemistry data and the evaluation of long-term temporal changes of vertical precipitation revealed the following conclusions:

- Sulphur concentration significantly decreased on the monitoring station. In the last years, sulphur deposition on Chopok represented 20% of the deposition at the beginning of the monitored period.
- Nitrogen concentration from ammonia ions significantly decreased. The concentration of nitrogen was reduced by 40% from maximum values.
- The concentrations of nitrate ions converted to nitrogen were decreasing on Chopok – EMEP station. However, during the last years higher concentration of nitrates was recorded, particularly in 2007 and 2008.
- The concentration of all alkaline cations in precipitation was decreasing.
- Depositions and concentrations of heavy metals in precipitation significantly decreased on Chopok monitoring station of EMEP. Slight increase of cadmium concentration was observed in the last years. Cadmium remains one of the priority heavy metals (Cd, Pb, Hg) in the Convention on heavy metals. The overall trend in the concentration and deposition has a decreasing character.

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Summary

Práca sa venuje spracovaniu databázy chemizmu zrážok pre stanicu EMEP – Chopok. V databáze boli spracovávané koncentrácie síranov, dusičnanov, amónneho iónu, bázických katiónov a z ťažkých kovov Cd, As, Al, Zn. Výstupom tejto databázy sú grafické znázornenia. Údaje boli spracované vo forme dlhodobých časových trendov chemizmu zrážok. Štatistické charakteristiky časových trendov boli testované Studentovým t-testom metódou významnosti výberového koeficienta korelácie. Bolo zistené, že väčšina koncentrácií vybraných elementov v zrážkach štatisticky významne klesá.

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