Original Research

Effect of Transregional Transport of Pollutants on Atmospheric Air Quality in the Tricity Area of (Poland)

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Received: 2 April, 2002
Accepted: 29 April, 2002

Abstract

Two-dimensional variance analysis of the results of determination of NO_3^-, SO_4^{2-}, F^-, Al^{3+}, Cl^-, NH_4^+, PO_4^{3-}, Fe^{3+}, Ca^{2+}, K^+ and Mg^{2+}, and pH and electrolytic conductivity of precipitation water samples collected in the Tricity area showed that transregional transport of pollutants affects air quality in this region. Over the large area of the Tricity agglomeration fluctuations in analyte concentrations in precipitation water samples are not affected on a statistically significant level by localizing sampling sites, but the area affected by transport direction of air mass movement.

Keywords: atmospheric precipitation, inorganic pollutants, transregional transport of pollutants, analysis of variance, cluster analysis

Introduction

As a result of human activity and natural processes different substances enter the atmosphere and are transported over long distances due to the movement of air masses. Transregional transport of pollutants present in the atmosphere has been studied since 1997 within the frames of the European Monitoring and Evaluation Programme (EMEP) [1]. The transport of pollutants into other parts of the environment including human and animal bodies is very significant, too. Therefore, it is important to observe the changes in emission rate and concentration of particular pollutants in the atmosphere.

Taking into account the basic areas of human activity, the main sources of selected pollutants emitted to the atmosphere can be specified as given in Table 1 [2].

Pollutant emission rates depend strongly on urbanization of a given area. The Tricity agglomeration lies in a zone of temperate climate. The Tricity climate has transitional character between the maritime and continental climates; it is modified by the Baltic Sea. Tricity lies in the northern part of Poland, close to the Baltic (18°56' E, 54°40' N). Climate differences in the region result from terrain, height, shoreline character and distance from the water body. Atmospheric circulation is generally meridional and frequent cold air advection from the north brings cold in spring and summer. Due to the flatness of the Vistula delta the Baltic effect is stronger here and therefore autumns are warmer

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Table 1. The main sources of selected pollutants emitted to the atmosphere air.

Pollutant	Sources
SO_2	combustion of sulphur containing fuels, metallurgy
NO_x	fuel combustion stationary sources (burners, power plants) and vehicle engines; industrial processes (manufacturing of nitric acid and explosives); dominant domestic sources as smoking, gas and oil heaters and cookers
CH_4	coal mining and farming (domestic animals, mainly ruminants)
CO	noneffective combustioncoal mining and farming (domestic animals, mainly ruminants)oal mining and farming (domestic animals, mainly ruminants)oal mining and farming (domestic animals, mainly ruminants)oal mining and farming (domestic animals, mainly ruminants)oal mining and farming (domestic animals, mainly ruminants)of fuels in low efficiency boilers and house ovens, and also in vehicle engines - this is so-called "low emission"
NH_3	agriculture, mainly animal farming
PO_4^{3-}	manufacturing fertilizers, soldering and welding processes, manufacturing road paints
Mg^{2+}	production of masonry mortar and cement
Cl^-	chemical and plating processes
Ca^{2+}	building and ceramics industries, manufacturing of bearing metals and soda

and longer. The nature of the shoreline modifies the Baltic influence – higher temperatures dominate on the seaside of the Gulf of Gdańsk. The northern rand of the region with the seaside to the open Baltic is characterized by dry and windy maritime climate [3].

The cities of the Tricity agglomeration and the surrounding areas are characterized by the occurrence of pollutants specific for the technological processes applied. Transregional transport of pollutants is relatively easy because of the lack of natural barriers and the fact that maritime and continental climates meet here, which brings about frequent and strong winds.

Apart from commonly known classifications of pollutants, the additional categorization of emission sources was proposed for the needs of this work:

- local sources (those within the agglomeration and its close neighbourhood);
- distant sources.

This classification is similar to that proposed by Ruijgrok [4].

For the Tricity agglomeration the following examples of distant sources can be specified:

- for hydrogen chloride, ammonia, methanol – the "Polpharma" Pharmaceutical Plant in Starogard Gdański [2];
- for compounds of magnesium, potassium and calcium – the Pomeranian Yeast Plant "Polmos S.A." in Tczew;
- for compounds of iron and phosphorous – the Mineral Waters Production Plant in Borkowo, near Żukowo;
- for compounds of calcium, magnesium and aluminium – the Crushed Stone Mining and Production Works in Delovo, Stężyca;
- for compunds of calcium and lead – the Chinaware Plant "Lubiana S.A." in Lubiana.

The main local sources of the Tricity agglomeration are:

- for aromatic and aliphatic hydrocarbons and sulfur dioxide – the Oil Rafinery in Gdańsk;
- for organic solvent vapours: xylenes and toluene - repair docks;
- for fluorine compounds, sulfuric acid, sulfur dioxide, dusts of fertilizers, apatities and phosphorites – the Phosphatic Fertiliser Plant "Fosfory S.A." in Gdańsk.

Within this work an attempt was made to determine the sources of mineral pollutants in precipitation water in the Gdańsk agglomeration. It was assumed that the deposition of the pollutants from the local sources depends mainly on sampling site location and wind direction. Deposition originating from distant sources (transregional pollutants) should depend on predominant wind direction in a given period only.

Materials and Methods

The subjects of chemometric analysis were the results of concentration measurement of: NO_3^-, SO_4^{2-}, F^-, Al^{3+}, Cl^-, NH_4^+, PO_4^{3-}, Fe^{3+}, Ca^{2+}, K^+ i Mg^{2+} in precipitation water samples collected for a period of 24 months (from January 1998 to December 1999) in 10 sampling sites distributed over the Tricity area. Ions concentration determinations were accompanied by pH and electrolytic conductivity measurements [5,6,7,8].

Figure 1 shows the exact positions of sampling sites. Which were distributed in the area in such a way so that some were near the emitters and some far away from the emitters. This would make it possible to find out if the particular areas differ from each other in pollution impact. More specific criteria of sampling site selection were not applied.

In the case of all sampling sites, precipitation was collected in a regular way. Precipitation was collected for a month and transported as a whole to a laboratory for analysis, them a new empty container was placed to collect precipitation. In the case of each sample the results are averages of three measurements. In the course of the project samplers were mechanically damaged several times and samples lost. In the gross, over 1300 determinations of NO_3^-, SO_4^{2-}, F^-, Al^{3+}, Cl^-, NH_4^+, PO_4^{3-}, Fe^{3+}, Ca^{2+}, K^+ and Mg^{2+} were made, and missing a few data did not hinder chemometric analysis. In a few rare cases when analyte concentration was below the detection limit of the analytical method applied the content was assumed to be equal to half the detection limit.

Samples of precipitation water (rainfall, melted snow) were collected with the use of a set composed of a polyethylene container of 2.5 dm^3 volume and a funnel 200 mm in diameter, both made of the same material. They were placed on a stand 1.5 m above ground level to protect

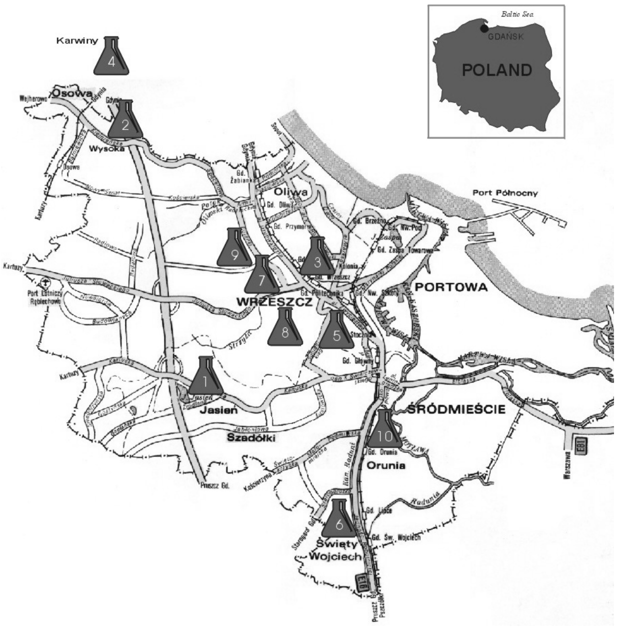

Fig. 1. Distribution of sites for sampling precipitation waters in the Tricity area.

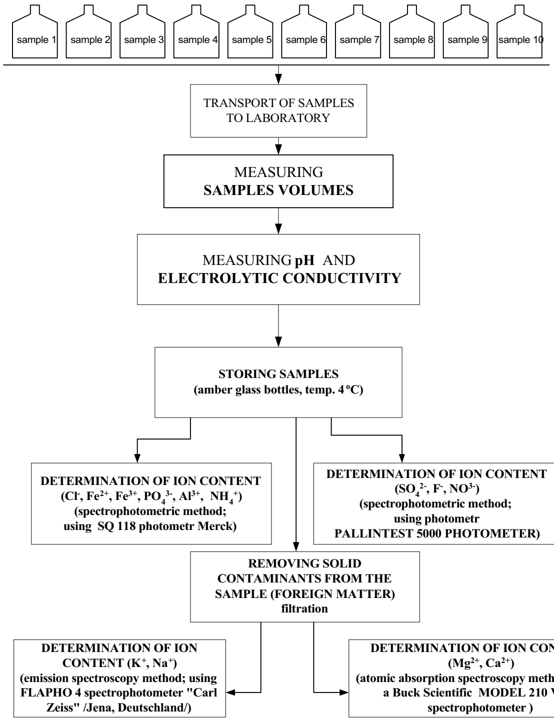

Fig. 2. Schematic presentation of analytical procedure for precipitation water samples.

precipitation against solid contaminants. Before use all parts of the set were subjected to special cleaning procedures [9]. Additionally, the funnel and container were rinsed with demineralised water just before placing them on the stand. The sampling sets were situated in open areas, not too close to building structures. In the period from June to August the sampling containers were wrapped with aluminium foil to minimize the effect of sunlight on analytes in the precipitation water sampled.

After delivery to a laboratory the precipitation samples collected were subjected to analysis according to the procedure depicted in Fig. 2.

Results and Discussion

As a result of analysis of 205 samples a set of 1300 measurement data was obtained which represent the content of different analytes in samples of precipitation water. Table 2 gives statistical descriptions of 14 variables characterizing 205 samples of precipitation waters. Apart from analyte concentrations, Table 2 contains pH, electrolytic conductivity (L) and volume (V) of samples.

The results obtained are close to those presented elsewhere [10, 11] for seaside areas of Washington State, Hong Kong and Turkey affected by different climates. It is quite difficult to evaluate the quality of precipitation water because regulatory values were not given. If permissible concentrations from inland surface waters are taken as a reference then the precipitation water studied can be classified into purity class I. In rare cases the parameters were beyond the range of permissible levels.

Skewness values for variable distribution (except for volume and pH) evidence high abnormality of distribution of the variables analysed. The basic requirement for correct chemometric analysis of a set of variables using variances is that variable distribution is brought to a form close to normal [8]. This was done by using a function of

$$x' = \log(x)$$

This transformation function was selected since each variable has a natural origin at a point of 0, the ratio of the smallest to the highest value is lower than one, a skewness factor is positive (the transformation was not applied for pH and volume). On the basis of logarithms of analytes concentrations it is difficult to accurately evaluate and compare the pollution of given areas. Due to differences in precipitation per area unit, it was necessary to present data as deposition or the mass of pollutants falling on area unit, D [kg/ha]. Therefore, in calculations the data for the analytes determined were used in the form of logD [kg/ha].

The ten sampling sites (Fig. 1) were divided into four categories depending on urbanisation characteristics and local sources of pollutants:

- **category RO** – sampling sites 2, 3, 7, 8 and 10 localized close to heavy traffic roads (Gdańsk);
- **category LA** – sampling sites 1, 6 and 9 localized in areas of low dispersed residential housing (Gdańsk);
- **category KA** – sampling site 4 localized in the north-west fringe of the agglomeration close to a large residential area with high buildings (Gdynia);
- **category TUG** – sampling site 5 on the roof of the Chemical Faculty of GUT; this place was treated in a

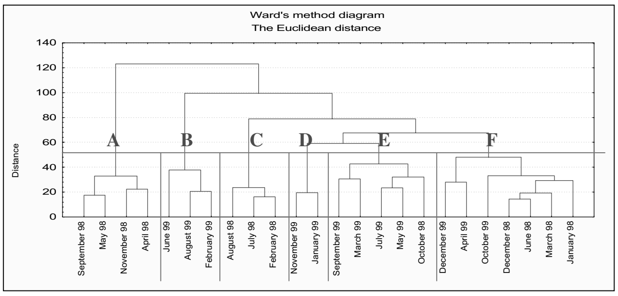

Fig. 3. Cluster diagram showing similarity of months with respect to wind direction.

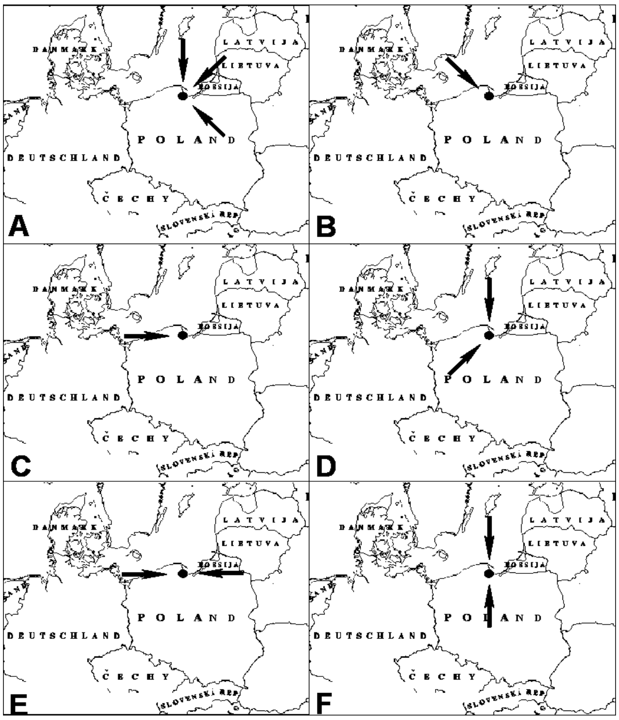

Fig. 4. The main directions of winds blowing in the Tricity area.

special way due to expected specificity of the pollutants emitted here (result of didactic and research activities of the faculty).

For each of 24 months, when data on the content of pollutants in precipitation water were collected the number of days with winds from 8 selected direction (N, NE, E, SE, S, SW, W and NW) was also noted down. The data were presented as percentages of winds blowing from given directions. For determination of similarity of particular months with respect to distribution of wind directions cluster analysis was made according to Ward's method [12] (the same method was applied by researchers from National Central University, Taiwan in 1977 [13]). The cluster diagram (Fig. 3) shows the similarity of particular months with respect to wind directions.

As results from this analysis monthly distributions of wind direction frequency in the Tricity agglomeration can be classified into six main types:

- **type A** – predominance of N, NE and SE winds;
- **type B** – high frequency of NW winds;
- **type C** – western winds;
- **type D** – high contribution of N and SW winds;
- **type E** – high percentage of winds blowing evenly with a parallel latitude;
- **type F** – high influence of meridional winds.

Fig. 4 presents maps showing the main directions of winds in the Tricity area.

It should be stressed that classification of a month into a given type is independent of time of the year.

The two-dimensional analysis of ANOVA variances with respect to wind direction type month (six types) and sampling site localization (four categories) shows that, on a significance level (α) of 0.05, statistically significant differences were observed in concentration of a given analyte with respect to wind direction distribution and site localization. The effect of site localization was found out

Table 2. Statistical characteristics of variables used to describe the precipitation water samples collected at sampling sites in Tricity in the period 01.1998-12.1999.

	Average value	Minimum value	Maximum value	Standard deviation	Skewness factor
V [dm^3]	1.447	0.155	3.277	0.738	0.445
pH	6.89	3.90	9.87	0.93	0.131
L [μS]	42.693	7.000	420.000	52.472	4.339
NO_3- [mg/dm^3]	2.971	0.004	19.400	3.660	2.076
PO_4- [mg/dm^3]	0.171	0.010	4.000	0.422	6.805
SO_4- [mg/dm^3]	10.466	1.088	75.550	12.686	2.181
NH^{4+} [mg/dm^3]	0.587	0.005	5.960	0.696	3.067
F^- [mg/dm^3]	0.274	0.020	2.040	0.230	3.139
Fe^{3+} [mg/dm^3]	0.078	0.010	0.820	0.124	3.795
Al^{3+} [mg/dm^3]	0.017	0.001	0.130	0.021	2.232
Cl^- [mg/dm^3]	3.817	0.100	24.570	4.844	2.468
K^+ [mg/dm^3]	1.554	0.010	29.340	4.087	4.423
Mg^{2+} [mg/dm^3]	0.694	0.010	13.890	1.761	4.794
Ca^{2+} [mg/dm^3]	2.708	0.010	87.500	6.967	8.834

to be significant for sample electrolytic conductivity and for deposition of five of the studied analytes (NO_3^-, SO_4^{2-}, Cl^-, Mg^{2+}, Ca^{2+}). A more detailed analysis shows that the samples collected at GUT (sampling site 5) differ from samples from all the other sites. The samples from site 5 have much higher electrolytic conductivity ($\overline{\log L}$=1.87 ± 0.20 µS) than those from all nine other sites ($\overline{\log L}$=1.42 ± 0,32 µS). GUT samples are also characterised by increased deposition of such ions as NO_3^-, SO_4^{2-}, Cl^-, Mg^{2+}. These depositions are statistically higher than the corresponding depositions in all nine other sites (Fig. 5).

It should be emphasized that the increased analytes depositions observed for site 5 are independent of wind directions. Fig. 6. shows electrolytic conductivity of samples ($\overline{\log L}$) collected at different localization categories with respect to wind direction types; plots for concentration of NO_3^-, SO_4^{2-}, Cl^- and Mg^{2+} are similar.

As results from this study, the samples collected at site 5 are heavily polluted with local pollutants, which in this case most probably originate from the Chemical Faculty. During laboratory exercises large quantities of gaseous pollutants and particulates are emitted to the atmosphere; among them vapours and mists of inorganic acids such as HNO_3, H_2SO_4 and HCl and their volatile derivatives. Due to different characteristics site 5 was excluded from further analysis. When this site was not taken into account, variance analysis shows that, at a significance level of 0.05, localization does not have a statistically significant effect on the deposition of any of the analytes studied. The same concerns sample volumes and electrolytic conductivity. With this respect the Tricity area can be considered integral; the fact that distances between some stations are quite large does not have a statistically significant effect on deposition of the analytes studied. This also means that, excluding site 5 (GUT), local emission sources do not influence deposition to a statistically significant degree.

However, the significant effect on deposition of pollutants, and sample volumes and electrolytic conductivity of precipitation is observed when considering wind directions bringing air masses. This is illustrated in Table 3.

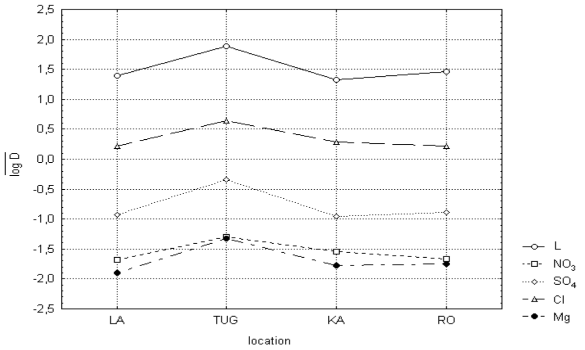

Fig. 5. Calculated parameters of deposition ($\overline{\log D}$ [kg/ha]) of NO_3^-, SO_4^{2-}, Cl^-, Mg^{2+} and electrolytic conductivity ($\overline{\log L}$ [µS]) for different localization categories.

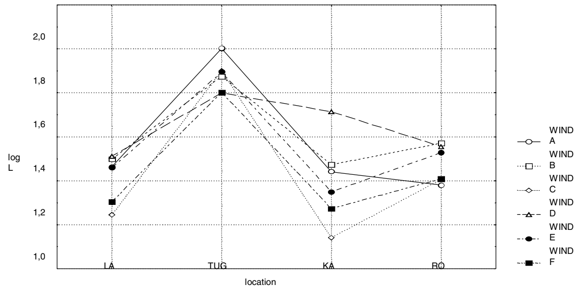

Fig. 6. Electrolytic conductivity parameters $\overline{\log L}$ [µS] for different sampling site categories and wind direction types.

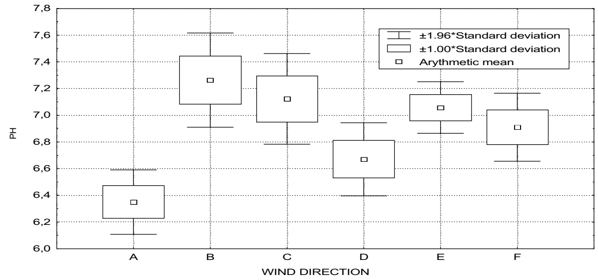

Fig. 7. Effect of wind direction (six main types) on pH of precipitation water.

Table 3. Dependence of analytes concentration in precipitation water on direction of advection of air masses to the Tricity area.

	WIND DIRECTION							WIND DIRECTION					
Variable	**A**	**B**	**C**	**D**	**E**	**F**	**Variable**	**A**	**B**	**C**	**D**	**E**	**F**
V	▼						**Fe^{3+}**	**no dependence**					
PH	▼						**Al^{3+}**		▼				
L	**no dependence**						**Cl^-**		▲				
No^{3-}		▼		▲	▲	▼	**K^+**		▼	▲			
Po_4^{3-}		▼		▲			**Mg^{2+}**		▼	▲	▲	▼	▲
SO_4^{2-}	▼						**F^-**				▼		
NH^{4+}	**no dependence**						**Ca^{2+}**		▼				

▲ - level significantly higher than average level
▼ - level significantly higher than average level

In the case of samples belonging to group A, less precipitation is observed; the effect of continental climate brings about advection of dry air masses from eastern Europe and Scandinavia. Measurement data indicate that air masses coming from the northern and eastern directions bring small amounts of precipitation. The lack of natural barriers to the free movement of air masses causes increased advection of air with high content of acidic components from Russia, Lithuania and Latvia. In the case of winds of type A, pH of atmospheric precipitation ranges from 6.1 to 6.6. This can be related to the presence of reduced nitrogen and sulphur compounds. Precipitation pH, when winds blow from other directions (types B to F), is always higher than 6.4. The described relationships are given in Fig. 7.

A statistically significant decrease in the deposition of SO_4^{2-} ions is observed in precipitation related to air masses coming from direction A. The main source of oxidized sulphur compounds emitted to the atmosphere is fuel combustion for energy production to supply municipal and domestic sectors of towns and villages. It was shown that transregional transport of sulphur compounds from countries belonging to the former commonwealth of independent states constitutes only a small fraction of pollutants coming with air masses to Poland (Fig. 8). The fact that the content of oxidized sulfur compounds is decreased in precipitation from the air coming from northern, northeastern and southeastern directions is consistent with the profile of the flux of these species from and to Poland.

The Tricity area is under the strong influence of the Baltic Sea. The closeness of sea makes the climate milder and gives high humidity to the air. The advection of air masses from the northwest (type B) leads to precipitation with statistically significant decrease in deposition of ions: Ca^{2+}, K^+, PO_4^{3-}, NO_3^-, Mg^{2+} and Al^{3+}. This precipitation is characterized by high content of Cl^- and hence comparatively high electrolytic conductivity of samples. This fact seems quite reasonable since the air from this direction is characterized by negligibly small content of anthropogenic pollutants but relatively high content of ions characteristic for sea water [14, 15, 16]. Due to the possibility of air advection from areas of Sweden, Denmark and Finland, which are characterized by low emission of chemical compounds, an increase in concentration of inorganic pollutants was not observed. As an example diagrams (Fig. 9) are presented which show dependencies of $\overline{\log D}$ (Al^{3+}) [kg/ha] and $\overline{\log D}$ (K^+) [kg/ha] on wind direction type for precipitation caused by advection of air masses.

It is difficult to draw conclusions concerning the effects of the four other profiles of wind directions (C, D, E, F) on pollutant deposition in the Tricity area due to the occurrence of opposing wind directions (winds blowing with a parallel to latitude – types C and E; meridional winds – types D and F).

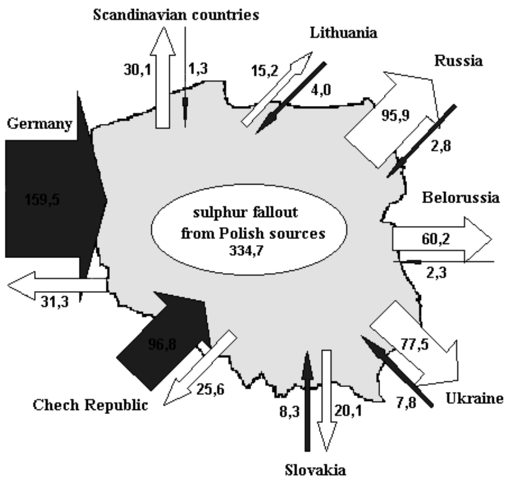

Fig. 8. Flux of oxidised sulphur compounds (thousands of tons of S/year) to and from Poland in 1995.

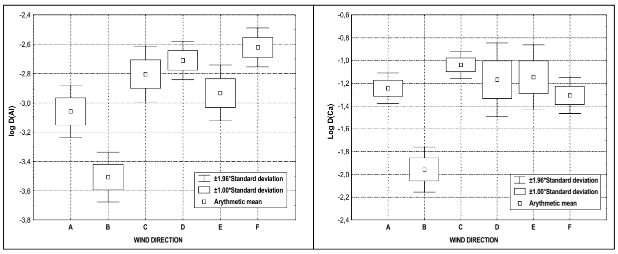

Fig. 9. Dependence of parameters $\overline{\log D}$(Al^{3+}) and $\overline{\log D}$(Ca^{2+}) on the direction of air advaction (precipitation related to all six types of wind direction).

Nevertheless, the statistically significant increase in deposition of Mg^{2+} and NO_3^- occurs when air masses come from western regions. In the areas of western Poland and northern Germany heavy industry is generally not present (mainly due to lack of raw materials); however, the land there is used for farming quite intensively and therefore fertilizers are widely applied.

One can presume that the occurrence of opposing winds brings about advection averaging air masses from highly urbanized areas, (i.e. areas with numerous different sources of gaseous pollutants released to the atmosphere and air masses from nonpolluted areas or from the areas where, in a natural way, sources of substances detrimental to the environment (the open Baltic, northern part of Europe) do not occur).

Conclusion

The aim of these studies was to find out the origin of mineral pollutants present in precipitation water in the area of the Tricity agglomeration.

The subject of chemometric analysis were the results of determination of NO_3^-, SO_4^{2-}, F^-, Al^{3+}, Cl^-, NH_4^+, PO_4^{3-}, Fe^{3+}, Ca^{2+}, K^+ and Mg^{2+} in samples of the precipitation water collected for 24 months (from January 1998 to December 1999) at 10 locations in the Tricity area. The additional data were sample volumes and conductivity. The chemometric analysis of the data obtained reveals that the Tricity agglomeration area is characterised by the occurrence of different sources of gaseous air pollutants. There were few periods when deposition of inorganic ions at some sampling sites were very large. Those rare cases do not cause high increases in average values.

As an example the following cases can be specified:

- Ca^{2+}_{max} = 87.500 mg/dm^3 (Nov. 1998, point 9); $Ca^{2+}_{av.}$=2.708 mg/dm^3;
- K^{+}_{max}= 29.340 mg/dm^3 (Dec.1999, point 7); $K^{+}_{av.}$ = 1.550 mg/dm^3;
- $SO_4^{2-}{}_{max}$ = 75.500 mg/dm^3 (Nov. 1999, point 5); $SO_4^{2-}{}_{av.}$ = 10.466 mg/dm^3;
- L_{max} = 420.00 µS (Dec. 1998, point 10); $L_{av.}$ = 42.69 µS.

These extreme results suggest that the distribution of deposition values for the area studied is log-normal. The pH average value of 6.89 for 205 analysed samples is emphasized to indicate that precipitation water in the Tricity area does not include the nature of acid rain. From the fact that pH value of precipitation water is close to neutral it can be concluded that local sources, as well as distant sources in a long-term period do not emit acid rain precursors to the atmosphere. In the precipitation water collected in the Tricity area ions can be specified which are present at low concentrations as compared to data for the seaside areas of Turkey and Japan published by Vong et al. and Kaya et al. [10, 11]:

- $PO_4^{3-}{}_{av.}$ = 0.171 mg/dm^3;
- $Fe^{3+}_{av.}$ = 0.078 mg/dm^3.

It was expected that deposition related to local sources would depend on sampling site localization or localization and dominant wind directions. In the case of distant sources (transregional pollutants) deposition level should depend only on wind direction in a given period.

The two-dimensional analysis of variances has shown that, excluding sampling site 5 (TUG), there are no differences in average deposition levels for different categories of sampling sites. The results obtained in the case of site 5 indicate that the pollution effects of local sources including strong emitters are rather short-ranged. Emission at site 5 has been found not to influence sites 3 and 9 situated ca 400 m away. If the results for specific site 5 are not included in chemometric analysis, it can be concluded that the Tricity area constitutes an integral part of the environment and the real atmospheric pollution and pollutant deposition with precipitation water is strongly affected by transregional transport of pollutants. Moreover, the effect of local emission sources is very limited, at least in the case of the analytes studied. The cities of Gdańsk, Sopot and Gdynia and their surroundings similar to other areas of northern Poland receive large quantities of pollutants coming from the soutwest and, to a lesser degree, from the east. Air masses from the nort carry a rather small load of pollutants.

It should be realised that variability and fluctuations of climatic conditions make it difficult to analyse pollutants emission sources on a global scale, and studies in this field should be continued. The only way to obtain reliable results and analyze them correctly is the availability of many years' measurement data. This is because only then so occasional fluctuations in analytes concentrations in precipitation waters have negligible effects and only then is it feasible to conduct analysis of possible seasonal changes of mineral pollutants concentrations in precipitation samples.

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