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ANNUAL AND SEASONAL COURSE OF PRECIPITATION ACIDITY AND ITS RELATION TO THE DIRECTION OF ADVECTING AIRMASSES IN THE CARPATHIAN FOOTHILLS NEAR BOCHNIA

Abstract: Precipitation acidity for the area situated close to the highly polluted industrial centres of southern Poland was analysed for the period 1991-1996. It was found that in 1991-1996 there was a substantial decline in precipitation acidity with the minimum in 1994, which coincided with minimum emissions of SO_2 and solid particles (ashes) by the Nowa Huta industrial complex (30 km WNW from the research area). Seasonal fluctuations of pH were well expressed with the lower values in winter half-years and the higher ones in the the summer half-years. The direction of advecting airmasses did not significantly influence the pH of daily totals of precipitation. The reason for this might be the general large-scale atmospheric pollution of southern Poland which causes acidification of precipitation independent of the direction of advecting air-masses. It is possible that the direction does influence pH of separate rainfall events but is not easily detected in daily totals which are the result of a mixture of separate rainfall events.

1. Introduction

Poland is among those European countries that suffer from rain acidity most seriously. There are many reasons for this. The first and the most important is extensive burning for industrial (energy production) and domestic purposes. Having vast resources of hard coal, mainly in the Upper Silesia coal basin, and brown coal dispersed in several areas of central and south-western part of the country, the Polish economy has been dependent on coal to a large extent. In 1994 about 60% of the energy production in Poland was based on hard coal and 13% on brown coal. Normally the sulphur content of coal varies between 0.5-1.0% but in some places coal supplies contain more with the maximum values reaching almost 4-5%. Sulphur dioxide (SO_2) emitted by coal-based industries is the main reagent leading to the formation of the acidic precipitation in Poland.

Fig. 1. Sulphur dioxide and dust emisson by steel industry of Nowa Huta in 1952-1995 (Source: Environmental Protection Office of the Sendzimir Steel Works in Nowa Huta).

Ryc. 1. Emisja dwutlenku siarki i pyłów przez kombinat metalurgiczny w Nowej Hucie w latach 1952-1995 (Źródło: Biuro Głównego Inżyniera d.s. Ochrony Środowiska Huty im. Sendzimira).

The late seventies and early eighties were the period of the highest levels of coal exploitation and use. Since that period there has been a substantial decline in production of heavy industries which was followed by a decline in sulphur dioxide (SO_2) emissions. A spectacular example of the long-term changes in SO_2 and solid particles input into the atmosphere is documen-ted for Nowa Huta, one of the largest steel complexes in Europe (Fig. 1, see also Fig. 3). The peak emissions took place in the late 1970s and dropped dramatically in the 80s and 90s. The reason for the decline was the economic crisis of the 1980s followed by industrial restructuring in the 1990s together with change and improvement in production technologies, with more restictive environmental policies and an increasing level of eco-investments.

Being situated in the central part of Europe, Poland is also the recipient of gaseous air pollution advecting mostly from the western part of the continent. Thus two factors: local industry and imported pollution result in the south of Poland having one of the highest levels of rain water acidity (Fig. 2) and atmospheric SO_2 concentrations in Europe (Fig. 3).

The aim of this study is to show and analyse the seasonal and long-term patterns of atmospheric precipitation acidity in the Carpathian Foothills, close to one of the largest pollution emitters in Poland - the steel works of Nowa Huta. This paper will also discuss the extent to which the direction of air-mass advection influences the rain-water pH of daily totals of precipitation in the study area. The question is, should we expect lower pH values (more acidic) in precipitation coming from the direction of the large industrial emitters and higher pH values (less acidic) - from those directions which do not bring high levels of pollution.

2. Site characteristics

The site for the collection of precipitation is located at the meteorological station in Łazy, a village situated at the northern edge of the Carpathian Foothills near the town of

Bochnia (see Fig. 3). The station is a part of the Research Field Centre in Łazy run by the Institute of Geography, Jagiellonian University, Cracow. The meteoro-logical station occupies a local hill-top at an elevation 245 m a.s.l. Due to its geographical position (100 km SE of the industrial region of Upper Silesia, 30 km ESE of the Nowa Huta steel works) and the dominat-ing wind directions (from W and NW), the site seems to be an ideal place for studying relationships between precipitation acidity and the direction of airmass advection. There are no major industrial emitters comparable to Nowa Huta and Silesia situated north, east and south of the area, that could affect air quality and rain-water acidity to a comparable extent. Local emitters are dispersed domestic furnaces in the village of Łazy.

Fig. 2. Rain water acidity in Europe (after Hanssen, 1990).

Ryc. 2. Zakwaszenie opadów w Europie (wg Hanssena, 1990).

The precipitation water-sampler comprising four teflon-covered aluminium cones each with a reception area of 1000 cm^2 (Phot. 1). The water was stored in PVC bottles and collected for pH measurements on a daily basis (6.00 GMT). The reason for the installation of four separate collectors was to obtain water volumes large enough for pH measurements of precipitation events amounting to less than 0.1 mm.

3. Study period

The study covers a time span of six years (November 1, 1990 to April 30, 1997). Annual precipitation for the period averaged 620 mm (Fig. 4), with the summer-half year (May-October) having a larger share of the total than the winter one (November-April). Due to technical problems, sampling was not possible between August 31 and November 30, 1992. The total number of samples collected between November 1, 1992 and April 30, 1997 was 752.

4. Method of data elaboration

Monthly, half-year and annual means of pH were calculated from mean values of hydrogen ion concentration (H^+). The similar procedure was applied for means representing each of the directions of air-masses advection.

Fig. 3. Sulphur dioxide mean concentration ($g\ m^{-3}$) in air in Poland in 1990 (after Juda et al., 1995). The arrow shows the dominanting direction of rain-bearing winds in southern Poland.

Ryc. 3. Koncentracja dwutlenku siarki ($g\ m^{-3}$) w powietrzu w Polsce w 1990 r. (wg Juda et l.,1995). Strzałka wskazuje przeważający kierunek napływu deszczonośnych wiatrów w południowej Polsce.

In order to analyse the relationship between the direction of air-mass advection and water acidity, the calendar of synoptic situations for South-East Poland (The upper Vistula drainage basin), elaborated by T. Niedźwiedź (1988), was applied. The calendar was partly published (Niedźwiedź, 1988, 1992) and partly made accessible for the needs of this study by its author.

The synoptic situations for the area can be classified according to two factors: the spatial pattern of atmospheric pressure (cyclonic and anticyclonic) and the direction of air-mass advection. The following anticyclonic and cyclonic situations were distinguished ('c' and 'a' are for cyclonic and anticyclonic situations respectively):

- Na, Nc - situations with air-masses advection from the north,
- NEa, NEc - situations with air-masses advection from the north-east,
- Ea, Ec - situations with air-masses advection from the east,
- SEa, SEc - situations with air masses advection from south-east,
- Sa, Sc - situations with air-masses advection from the south,
- SWa, SWc - situations with air-masses advection from the south-west,
- Wa,Wc - situations with air-masses advection from the west,
- NWa, NWc - situations with air-masses advection from the north-west.

Above are listed 16 situations representing distinct directions of advection. The next four situations represent no advection or unstable directions. These are:

- Ca - central anticyclonic situation, no advection, high pressure centre over south Poland or Slovakia,
- Ka - high-pressure wedge, sometimes several undistinct centres or unclearly defined area of high pressure, axis of the ridge of high pressure,
- Ca - central cyclonic situation, low pressure centre over south Poland or Slovakia,
- Bc - low-pressure trough, unclearly defined area of low pres-sure or axis of the low-pressure trough with different directions of advection and frontal systems separating different air-masses.

Finally, pressure cols and undefined situations were distinguished using the symbol X.

For the needs of this study the distinction between cyclonic and anticyclonic situations was omitted and neighbouring directions were joined to obtain the following groups:

- N+NE - situations with an advection of air masses from the north and north-east,
- E+SE - situations with an advection of air masses from the east and south-east,
- S+SW - situations with an advection of air masses from the south and south-west,
- W+NW - situations with an advection of air-masses from the north and north-west,
- C+K+B - central cyclonic and anticyclonic situations, anticyclonic wedges or low pressure troughs,
- X - undefined situations and pressure cols.

5. Results

Arithmetic average of daily sums of precipitation-water pH in Łazy for the whole study period was 4.35. Thus the annual load of hydrogen ion introduced into the local environment may vary from 24.6 to 33.5 kg m^{-2} a^{-1} depending on annual total of precipitation (Tab. 1).

The overall tendency of annual averages showed substantial rises from 1991 to 1994 (Fig. 5) falling to a lower value in 1995 and 1996 (4.42 and 4.43 respectively).

Phot. 1. Precipitation-water samplers in Łazy.

Fot.1. Stanowisko poboru wód opadowych w Łazach.

Fig. 4. Annual totals of atmospheric precipitation in Łazy in 1991-1996.

Ryc. 4. Sumy roczne opadów atmosferycznych w Łazach w latach 1991-1996.

Tab. 1. Annual load of H^+ ions with precipitation water at Łazy.

Tab. 1. Roczny ładunek jonów wodorowych (H^+) dostarczanych z opadami w Łazach.

pH	Precipitation, mm Opad, mm	Load, kg km^{-2} Ładunek, kg km^{-2}
4.35	550	24.57
	600	26.80
	650	29.03
	700	31.27
	750	33.50

Fig. 5. Annual averages of precipitation-water pH in Łazy in 1991-1996.

Ryc. 5. Średnie roczne wartości pH wody opadowej w Łazach w latach 1991-1996.

Seasonal fluctuations of precipitation acidity were well expressed (Fig. 6 and 7) with lower values for the winter half-year (4.24) and higher ones for the summer half-year (4.55). The only exception to this was 1995 when the winter pH was slightly higher than the summer one. Generally over the last three years the seasonal differences in precipitation acidity have been getting smaller.

The average values of pH for particular directions of air mass advection (Fig. 8 and Tab. 2) show some differentiation, however the differences are not large enough to be statistically significant. The only significant difference is between the WNW and WSW-ESE sectors for the summer half year. The lowest average values are for the direction supplying airmasses that have passed through the industrial areas of Upper Silesia and Cracow-Nowa Huta. In case of annual and winter half-year averages the differences between the directions are poorly expressed with slightly lower values for the ENE sector than for the others. The general conclusion for the studied area is that the direction of air-mass advection does not influence the precipitation acidity, and the documented differences do not reach the significance level of 0.05.

6. Discussion and conclusions

The substantial drop of precipitation acidity reaching a minimum in 1994 is undoubtedly the effect of falling industrial emissions. Proving the relationship between the two is however difficult or even impossible. The average pH values for particular years or seasons depend not

Fig. 6. Seasonal (winter half-year/summer half-year) fluctuations of precipitation-water pH in Łazy in 1991-1997.

Ryc. 6. Sezonowe (półrocze zimowe/ półrocze letnie) fluktuacje pH wody opadowej w Łazach w latach 1991-1997.

Fig. 7. Monthly averages of precipitation-water acidity at Łazy in 1991-1997.

Ryc. 7. Średnie miesięczne wartości pH wody opadowej w Łazach w latach 1991-1997.

Tab. 2. Mean values of pH of precipitation water at Łazy for different synoptic situations in November 1, 1990 to April 30, 1997.

Tab. 2. Średnie wartości pH wody opadowej w Łazach przy poszczególnych sytacjach synoptycznych w okresie od 1 listopada 1990 r. do 30 kwietnia 1997 r.

Period Okres	Type of synoptic situation Typ sytuacji synoptycznej					
	N+NE	E+SE	S+SW	W+NW	C+K+B	X
Winter half-year Półrocze zimowe	44 4.14	60 4.37	46 4.43	164 4.27	94 4.15	9 3.96
Summer half-year Półrocze letnie	61 4.61	21 4.97	37 4.74	90 4.46	122 4.49	4 5.24
Year Rok	105 4.35	81 4.46	83 4.54	254 4.43	216 4.31	13 4.11

Fig. 8. Precipitation-water pH for different direction of advecting airmasses in Łazy.

Fig. 8. pH wody opadowej dla różnych kierunków napływu mas powietrza w Łazach.

only on industrial and domestic input of SO_2 but also on the annual distribution of the precipitation events, the type of rainfall (torrential, shower, long-lasting etc.) and the distribution of rainfall intensity within particular events (Ciszewski, Żelazny, 1995a). In spite of many factors that may affect the water acidity it is worth noting that, for the Nowa Huta complex, the year 1994 was the one with the lowest SO_2 emission since 1956 (see Fig. 1).

Another factor to discuss is the buffering capacity of ashes and other solid particles originating in industrial areas. It is well documented that within and close to industrial centres, in spite of large SO_2 emissions, the precipitation acidity is buffered, so that values are reaching pH 6.0 or more, which is well above the norm (5.6) for rain water in areas recognized as unpolluted (Fiszer, 1990). In the case of study site the buffering role of industry-sourced particles is probably limited or negligible due to the distance between the potential sources (Nowa Huta, Silesia) and the recipient (Łazy). Even if we assume that the suspended particles coming from Nowa Huta could act as a buffer for acidic precipitation in the foothill area of Łazy, the result obtained was opposite to that which could have been expected. In 1994, when the particle emissions from Nowa Huta were the lowest, the precipitation water acidity was lowest too. Thus the industrial emission of solid particles can probably be excluded as a factor influencing precipitation acidity in Łazy.

Seasonal fluctuations of pH is in accordance with seasonally changing SO_2 emissions connected with coal burning for heating purposes, both in power-plants and domestic furnaces. The smaller differences between the seasons documented for 1995 and 1996 compared with earlier years is difficult to interpret.

The last issue to consider is the only slightly expressed and statistically insignificant influence of the direction of air-masses on precipitation-water acidity. The first and most probable reason for this is that southern Poland is part of a larger and generally highly polluted region of central Europe. Thus, independent of the direction of advecting air-masses, the approaching precipitation is polluted, which is manifested in low pH. The probable factor influencing differences between summer and winter periods is that in summer the water pH is not changed by local emissions while in winter it is. Consequently, detailed interpretation becomes more difficult. Another reason for the lack of the expected relationship is that we have analysed the daily totals of precipitation which, in many cases, consisted of separate rainfall events. It is possible that in many cases the different events were connected with different directions of advection. The data obtained by Ciszewski and Żelazny (1995b) show that the chemistry of precipitation water may change dramatically even during one rainfall event and might be strongly influenced by rapidly changing synoptic situations as, for example, a passing frontal system. Even though we have used daily pH values, the method used might give results that are too generalized and do not fully represent the outcomes of chemical atmospheric processes. Finally, variability of the direction of advection on a time-scale shorter than the sampling period might influence the pH of separate rainfall events but not be reflected clearly in the daily totals.

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Wieloletni i sezonowy przebieg zakwaszenia opadów atmosferycznych oraz jego zależność od kierunku napływu mas powietrza na Pogórzu Karpackim koło Bochni

Streszczenie

Analizie poddano przebieg zakwaszenia opadów w Łazach koło Bochni (Pogórze Wielickie). Obszar ten znajduje się w niewielkiej odległości (30-100 km) od dużych ośrodków przemysłowych południowej Polski: Śląska i okręgu krakowskiego.

W przebiegu wieloletnim stwierdzono stopniowy wzrost pH opadów, a zatem spadek zakwaszenia. Związane jest to zapewne ze znacznym ograniczeniem emisji zanieczyszczeń przemysłowych, w tym - kombinatu metalurgicznego w Nowej Hucie.

Przebieg zakwaszenia opadów atmosferycznych odznacza się sezonowymi fluktuacjami. Średnie pH wody opadowej w półroczu zimowym (4,24) jest niższe niż w półroczu letnim (4,55) wobec średniej rocznej wartości wynoszącej 4,35. Różnice pH opadów atmosferycznych w odniesieniu do różnych sytuacji synoptycznych są wyraźniejsze w półroczu letnim niż w półroczu zimowym. Spodziewanne niższe wartości pH wody opadowej przy napływie powietrza

z kierunków W+NW, a więc od strony największych źródeł zanieczyszczenia powietrza, obserwowane są jedynie w półroczu letnim. Generalnie, wielkoobszarowe zanieczyszczenie powietrza w południowej Polsce powoduje, że kierunek napływu mas powietrza nie wpływa istotnie na zakwaszenie opadów atmosferycznych.