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# Changes of cloud water chemical composition in the Western Sudety Mountains, Poland

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## Abstract

The changing chemical composition of cloud water and precipitation in the Western Sudety Mountains are discussed against the background of air-pollution changes in the Black Triangle since the 1980s until September 2004. A marked reduction of sulphur dioxide emissions between the early 1990's and the present (from almost 2 million tons to around 0.2 million tons) has been observed, with a substantial decline of sulphate and hydrogen concentration in cloud water ($SO_4^{2-}$ from more than 200 to around 70 μmol $l^{-1}$; $H^+$ from 150 to 50 μmol $l^{-1}$) and precipitation ($SO_4^{2-}$ from around 80 to 20–30 μmol $l^{-1}$; $H^+$ from around 60 to 10–15 μmol $l^{-1}$) samples. At some sites, where fog/cloud becomes the major source of pollutants, deposition hot spots are still observed where, for example, nitrogen deposition can exceed 20 times the relevant critical load. The results show that monitoring of cloud water chemistry can be a sensitive indicator of pollutant emissions.

*Keywords:* Total ionic content; Fog precipitation; Fog deposition; Seeder–feeder effect; Acid rain; Sudety Mountains

## 1. Introduction

The "Black Triangle" (BT), where Poland, Germany and the Czech Republic meet is one of Europe's most industrialized regions (Fig. 1). As the name implies, it is also one of the most polluted areas on the continent — plagued for decades by choking coal dust emitted by electric power plants and district heating plants which can be found within a 150 km (93 miles) radius the heart of the BT. In this heavily industrialized region, electric power plants currently produce a total output of between 14,860 MW and 4000 MW of energy, with specific contributions from Turów Power Station (2000 MW) in Poland, three major plants in Germany (6860 MW) and six locations in the Czech Republic (6000 MW). The highest pollution emission, and as consequence the highest acid deposition in Europe (exceeding 1500 mol $ha^{-1}$ $year^{-1}$ in the BT), was observed at the turn of the 1980s (Fig. 2). Critical loads have been estimated for most natural and semi-natural areas within Europe (Posch et al., 1997).

In 1989 the power plants in the BT burnt 80 million tons of lignite coal, and as a result, 1.9 million ton of $SO_2$ (about 30% of Europe's total $SO_2$ emissions) and approximately 1 million ton of $NO_2$ was emitted (Mańczyk, 1998, 1999).

Chemical composition of rain and cloud water is a particularly sensitive indicator of pollution emissions. According to results of measurements carried out in the

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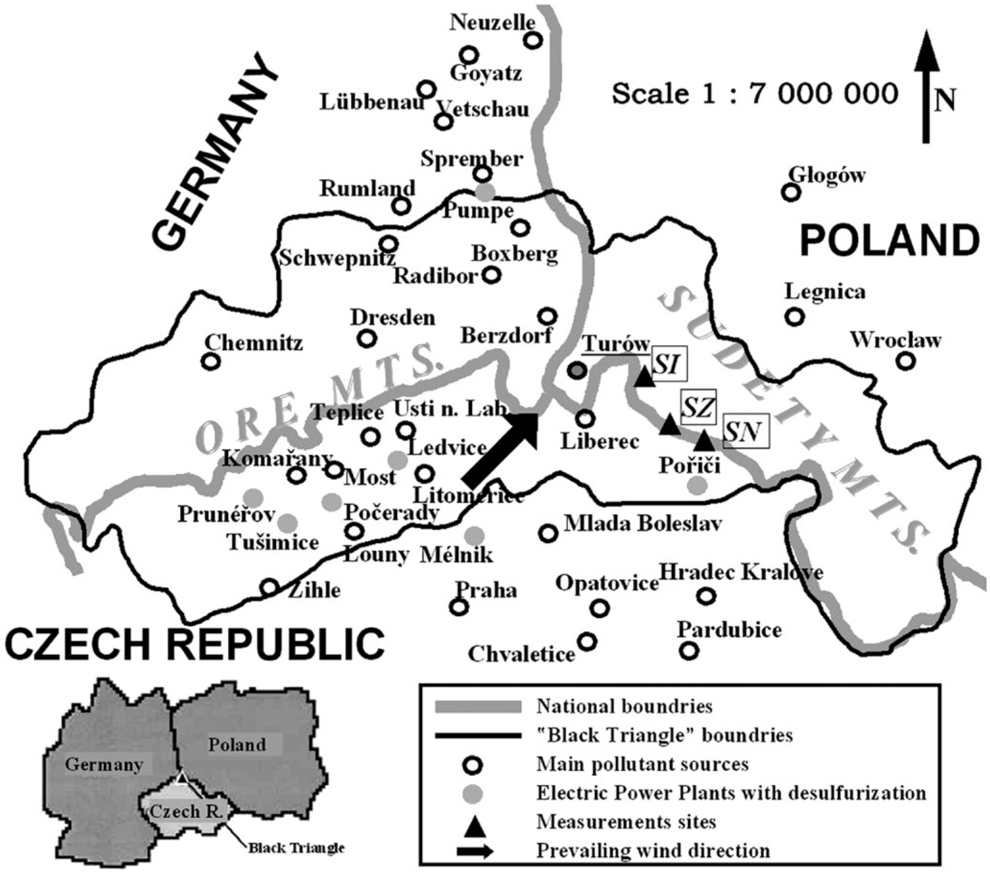

Fig. 1.

early 1990s in the Western Sudety Mountains, the sulphate aerosol $SO_4^{-2}$ was the most significant component of air pollution, with 80% of the initial $SO_2$ converted into sulphate on average. The major ions present in rain and cloud water were $SO_4^{2-}$ and $NO_3^-$. Ion concentrations in cloud water were typically 3 to 5 times higher than those in precipitation water (Kmieć et al., 1994; Twarowski, 1996).

A number of factors contributed to a high rate of observed pollutant deposition via fog (Błaś and Sobik, 2003; Sobik and Migała, 1993). These include high wind speed, significant liquid water content and the number of days with fog exceeding 250 days per year, combined with high concentrations of pollutants in fog droplets and a large amount of precipitation (up to 2000 mm $year^{-1}$). Besides precipitation, cloud water deposition was also found to be a major contributor to acidification of the natural environment in the Western Sudety Mountains. These processes led to extensive damage, such as edaphic changes and forest decline. The highest pollutant deposition rate was observed at convex landforms on the windward side of a larger massif where fog deposition tends to be the main component of the total flux of pollutants to the ground.

Fog precipitation and pollutant deposition rate varied noticeable over short distances (microscale) in mountainous terrain because of the variability in surface roughness (Błaś and Sobik, 2003).

Since June 1991 the BT countries have initiated a programme to reduce the emissions from power stations, district heating systems and other large industrial plants. To lower $SO_2$ emissions from both electric power stations and district heating plants, low-sulphur fuel and new desulphurization equipment were used. Other methods were taken to reduce $NO_2$ emissions, such as the use of fuels with low nitrogen content, denitrifying equipment, low $NO_2$ emission burners and the introduction of exhaust-gas recirculation methods. The reduction of particulate $SO_2$ emission was achieved by the use of electrostatic precipitators and circulating fluidised bed boilers which, in the Turów Power Plant, absorb 90% of the $SO_2$ that would otherwise have gone into the atmosphere. Thanks to various improvements, particulate emission from the Turów Power Plant was reduced

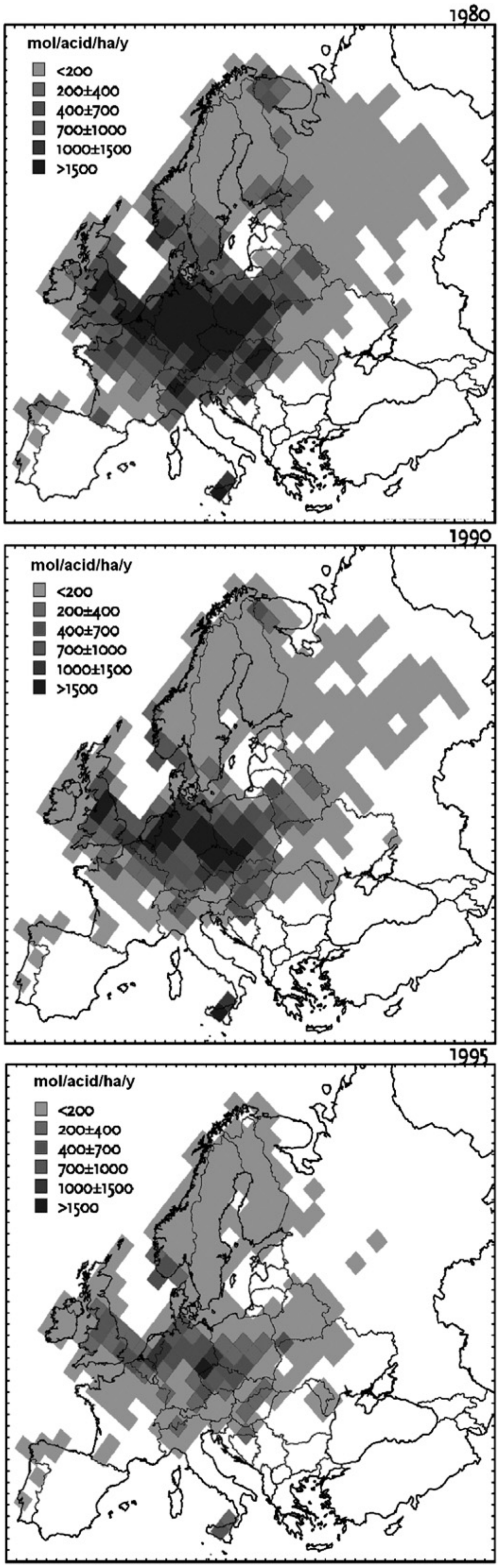

Fig. 2.

by 88% and $SO_2$ emissions by 83% from the 1989 levels (Libicki, 1998; Marszalik, 1995).

As the result of a national pollutant abatement policy since the beginning of the nineties, a substantial reduction in gaseous emissions have been observed, with $SO_2$ being reduced most significantly (Fig. 3; Abraham et al., 2003). The emission reduction achieved by Poland so far meets the targets of the Gothenburg protocol (Mitosek et al., 2004). Apart from the overall decrease of pollutant concentration and deposition rate, significant changes in the distribution of both concentration and deposition are observed. However, one should stress that deposition is still high, and frequently exceeds critical load limit values in the Western Sudety Mountains. where serious environmental problems remains (Fig. 2). The area of the BT remained the most polluted region in Europe, with critical loads of acid deposition exceeding standards over 1000–1500 mol $ha^{-1}$ in 1995.

The aim of this study is to describe these changes in relation to atmospheric processes with the emphasis on cloud water composition and pollutant deposition by fog.

## 2. Site and data description

The Western Sudety Mountains are located along the Polish/Czech border, which is east of the central part of the BT. The region experiences major downwind pollution for most of the year. Data from two sites were taken into account, namely SI (Stóg Izerski, 1060 m above mean sea level (AMSL)) that is located on the westernmost orographic threshold, and SZ (Szrenica, 1332 m AMSL) at the next orographic threshold and about 20 km eastward from SI in the central part of the Western Sudety (Fig. 1, Table 1). Apart from continuous long-term pH and conductivity data in cloud water samples collected daily at SZ since 1988, there is a lack of long consistent data sets on cloud water chemistry. Two separate cloud water data series from SZ, namely 2 winter months in 1993 (Kmieć et al., 1994) and the summer of 1995 (Dore et al., 1999), which both represent the late stage of intense although decreasing emissions, were compared with the results of current measurements at SZ during the period June 2003 to September 2004. In all cases cloud water samples were taken daily with the use of simple passive collectors set 2 m above the ground in grassy areas and wind-exposed sites. The individual samples after instant pH and conductivity measurements, were integrated into monthly samples and was subsequently analysed to measure concentrations of major ions in the Laboratory of Water

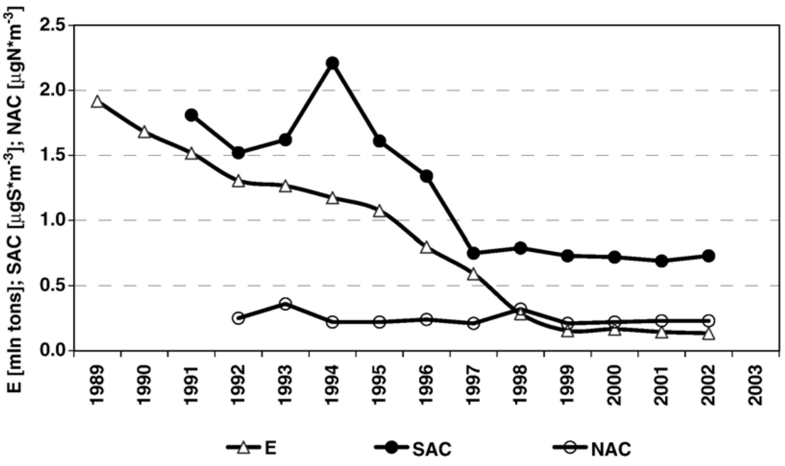

Fig. 3.

Chemistry of the Institute of Meteorology and Water Management. This laboratory performed the analytical measurements of bulk precipitation collected at Polish EMEP stations and the National Network of Precipitation Chemistry. The quality control consists in intercomparison of analytical methods within EMEP (Co-operative Programme for Monitoring and Evaluation of the Long Range Transmission of Air Pollutants in Europe). All characteristics integrated for longer periods were weighted by water volume.

## 3. Discussion

### 3.1. Long-term ionic composition changes

Cloud water composition at the SZ site in the early 1990s was characterised by a very large content of dissolved pollutants expressed by average total ionic content (TIC) close to 1000 µmol $l^{-1}$ (Table 2), with sulphates, ammonia, nitrates and hydrogen as major ions. Before the early 1990s only precipitation chemistry data was available, meaning that cloud water composition could be estimated by the use of later quasi-constant cloud water to precipitation concentration ratio extrapolated in the past. If the ratio is 3.0, the cloud water TIC in the late 1980s would have been around 1200–1300 µmol $l^{-1}$. In the case of average cloud water chemistry at the SZ and SI sites during the period June 2003 to September 2004, only insignificant reduction in concentration was noted when compared to 1993 and 1995 data (Tables 2 and 3). Taking into account the estimated TIC in the late 1980s, the reduction becomes clearer.

Table 1

| Point name | Altitude [m a.s.l.] | Geographical coordinates | Institution |
|---|---|---|---|
| Stóg Izerski (SI) | *1110* | $\lambda = 15°17'$E<br>$\varphi = 50°53'$N | Department of Meteorology and Climatology Wrocław University |
| Szrenica (SZ) | *1330* | $\lambda = 15°44'$E<br>$\varphi = 50°44'$N | Department of Meteorology and Climatology Wrocław University |
| Śnieżka (SN) | *1602* | $\lambda = 15°31'$E<br>$\varphi = 50°48'$N | Institute of Meteorology and Water Management |

Much more profound is the different behaviour of the constituents. Concentrations of sulphates and hydrogen ion dropped significantly, but concentrations of $Ca^{2+}$ and $Na^{+}$ increased slightly. The concentration changes of nitrates and ammonia were weaker and varied from season to season. The largest relative decrease (percentage of a given ion in TIC) occurred for sulphates (from 29% at SZ and SI in the summer of 1995 to 10% in 2003/4), and the $H^{+}$ cation (from 13% at SZ and 10% at SI to 8% and 4%, respectively, in 2003/2004).

Table 2

| | II–III 1993 | | V–X 1995 | | | |
|---|---|---|---|---|---|---|
| | CW SZ | PW SZ | CW SZ | CW SI | PW SZ | PW SI |
| pH | 3.83 | 4.20 | 4.08 | 4.01 | 4.27 | 4.27 |
| Con. | – | – | 109 | 135 | 38 | 39 |
| $SO_4^{2-}$ | 215.5 | 69.4 | 181 | 284 | 77 | 102 |
| $NO_3^-$ | 166.5 | 85.4 | 104 | 242 | 49 | 59 |
| $Cl^-$ | 69.1 | 22.6 | 22 | 41 | 20 | 21 |
| $NH_4^+$ | 196.3 | 77.3 | 139 | 205 | 25 | 48 |
| $Ca^{2+}$ | 17.3 | 18.8 | 12 | 19 | – | – |
| $Na^+$ | 54.3 | 16.1 | 18 | 34 | – | – |
| $K^+$ | 11.4 | 5.6 | 12 | 16 | – | – |
| $Mg^{2+}$ | 8.3 | 3.5 | 9 | 12 | – | – |
| $H^+$ | 147.9 | 63.1 | 83 | 98 | 54 | 54 |
| Σ TIC | 888.3 | 361.8 | 529 | 826 | 269 | 284 |

Table 3

| A) | CW SZ | | | | CW SI | | | |
|---|---|---|---|---|---|---|---|---|
| | Total | Warm VI 2003–X 2003 | Cold XI 2003–IV 2004 | Warm V 2004–IX 2004 | Total | Warm VI 2003–X 2003 | Cold XI 2003–IV 2004 | Warm V 2004–IX 2004 |
| pH | 4.25 | 4.19 | 4.33 | 4.20 | 4.44 | 4.51 | 4.43 | 4.36 |
| Con. | 66 | 86 | 51 | 74 | 69 | 73 | 62 | 79 |
| $SO_4^{2-}$ | 66.7 | 80.8 | 50.1 | 84.1 | 68.3 | 61.2 | 65.8 | 84.4 |
| $N-NO_3^-$ | 172.6 | 234.4 | 109.3 | 231.8 | 177.4 | 170.6 | 140.1 | 269.7 |
| $Cl^-$ | 65.6 | 87.5 | 43.4 | 86.2 | 96.7 | 118.0 | 74.9 | 112.4 |
| $N-NH_4^+$ | 166.6 | 193.1 | 118.0 | 231.0 | 189.5 | 178.0 | 168.5 | 253.2 |
| $Ca^{2+}$ | 26.2 | 37.0 | 16.0 | 34.9 | 32.4 | 30.2 | 26.5 | 48.7 |
| $Na^+$ | 67.3 | 97.5 | 41.2 | 87.5 | 100.9 | 127.7 | 70.4 | 127.1 |
| $K^+$ | 5.7 | 7.1 | 4.1 | 7.5 | 20.7 | 45 | 8.5 | 10.7 |
| $Mg^{2+}$ | 10.3 | 16.6 | 5.4 | 13.6 | 13.6 | 15.3 | 9.4 | 20.0 |
| $H^+$ | 55.7 | 65.2 | 47.0 | 62.8 | 36.2 | 30.8 | 36.7 | 43.4 |
| Σ TIC | 636.7 | 819.2 | 434.5 | 839.4 | 735.7 | 776.8 | 600.8 | 969.6 |
| B) | PW SZ | | | | PW SI | | | |
| pH | 4.85 | 4.98 | 4.82 | 4.81 | 4.87 | 4.80 | 4.74 | 5.08 |
| Con. | 20 | 19 | 21 | 18 | 25 | 31 | 29 | 19 |
| $SO_4^{2-}$ | 25.3 | 27.5 | 27.4 | 21.8 | 31.5 | 38.8 | 35.0 | 24.2 |
| $N-NO_3^-$ | 37.6 | 34.1 | 43.2 | 34.5 | 53.4 | 65.1 | 63.1 | 38.5 |
| $Cl^-$ | 15.7 | 18.0 | 19.8 | 10.2 | 23.7 | 22.3 | 26.9 | 22.1 |
| $N-NH_4^+$ | 62.0 | 68.2 | 57.1 | 62.7 | 74.2 | 94.4 | 80.8 | 56.3 |
| $Ca^{2+}$ | 10.5 | 10.4 | 14.0 | 7.2 | 18.7 | 17.6 | 15.0 | 22.2 |
| $Na^+$ | 12.8 | 11.5 | 15.9 | 10.6 | 22.7 | 21.5 | 30.2 | 17.6 |
| $K^+$ | 3.5 | 4.8 | 2.7 | 3.6 | 7.0 | 13.0 | 4.0 | 5.7 |
| $Mg^{2+}$ | 2.4 | 2.9 | 2.6 | 1.9 | 4.2 | 5.5 | 4.3 | 3.3 |
| $H^+$ | 14.2 | 10.5 | 15.2 | 15.6 | 13.5 | 16.0 | 18.1 | 8.3 |
| Σ TIC | 184.0 | 187.9 | 197.9 | 168.1 | 248.9 | 294.2 | 277.4 | 198.2 |

Sulphates, which were the most important constituent of cloud water chemistry in the early 1990s, became the fifth most important ion in 2003/4 after nitrates, ammonia, $Cl^-$, and $Na^+$. Nitrates and ammonia became major ions in 2003/4, with an increase of relative concentration at the SZ station from 17% to 28% and from 22 to 26%, respectively. The associated changes at SI were from 25% to 27% and from 21% to 26%. A distinct increase in the relative role is visible for $Na^+$ (from 3% to 10–13%, $Cl^-$(from 4% to 10%) and $Ca^{2+}$ (from 2% to 4–5%). These changes resulted in spectacular decrease of acidity expressed by almost half a unit raise of pH values (from 4.08 in 1995 to 4.20 in the summer of 2004 at SZ and from 4.01 to 4.51 in the summer of 2003 at SI) with a threefold drop in the $H^+$ cation concentration.

Such transformation in cloud water chemistry can be explained by reduced emissions from large sources in the BT region, with $SO_2$ emissions being the most efficiently reduced. The observed relative increase in the role of $Na^+$ and $Cl^-$, with almost unchanged absolute values, resulted from the stable natural source of these components in sea salt aerosols. The relatively stable concentration of $NO_3^-$ and $NH_4^+$ can be clarified by the reduction of industrial emission with parallel high emission from traffic (nitrates) and agriculture (ammonia). A high relative increase of $Ca^{2+}$ concentration can be attributed to the use of $CaCO_3$ in the desulphurisation process in some electric power plants.

### 3.2. Acidity of atmospheric water

To reduce the role of seasonal and inter-seasonal anomalies of circulation patterns (i.e. the drought of 2003), which might influence an existing trend of cloud water chemistry, two 4-years series of pH data from SZ were used to characterise changes in cloud water and bulk precipitation acidity (Fig. 4). The average pH of cloud water, derived from the accumulated $H^+$ content, and weighted by volume increased from 3.61 to 3.96 between the 1990–1993 and 1999–2002 periods.

The relevant pH change in bulk precipitation was from 3.84 to 4.31. From the frequency distribution of individual daily samples, there is evidence of a shift of measured values towards higher pH classes. Alkaline samples of cloud water and bulk precipitation were observed in the later period, which did not occur previously.

Changes in annual volume weighted pH values at the SZ and SN stations are shown in Fig. 5. Since the mid

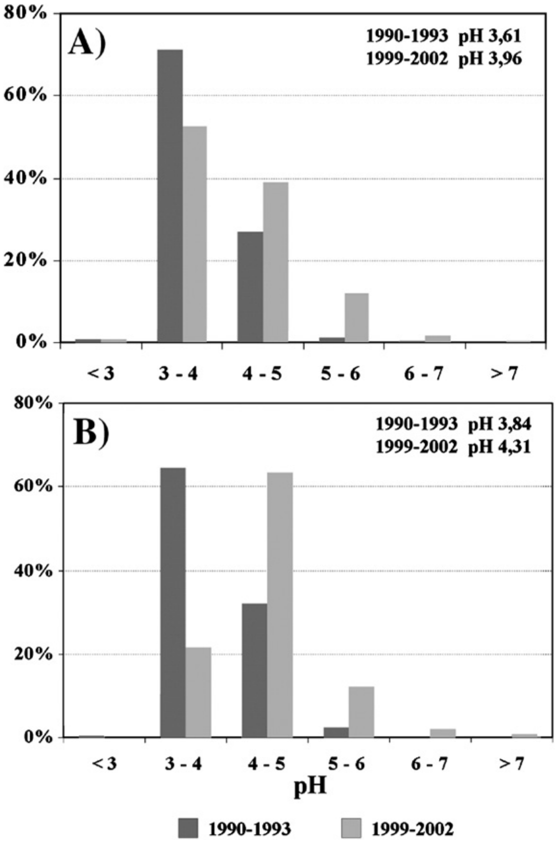

Fig. 4.

1980s till 2004 the precipitation acidity decreased by a factor of approximately 5–6 at the Snieżka station and 7–8 during the past 15 years at the SZ station. This indicates that hydrogen annual load dropped at SN from 125 µmol $l^{-1}$ in 1985 to ca. 25–30 µmol $l^{-1}$ between 2000 and 2004, and from 150 to 30–40 µmol $l^{-1}$ at SZ with some inter-annual changes at both stations. Such strong upward trends in pH values have been observed in Poland only in the mountainous regions of the south-west (Mitosek et al., 2004). Thus the influence of an extensive modernisation of technology in the energy sector became clear.

Moreover, the precipitation acidity at the SZ station is consistently 25% higher than at SN, located 15 km further towards the east–south-east. The Izera Mountains and the westernmost threshold of the Karkonosze Mountains, represented by the SI and SZ stations, respectively, form two subsequent orographic barriers which protect the downwind eastern parts of the Karkonosze Mountains (represented by the SN station) from high pollution concentrations and deposition rates (Błaś et al., 2002).

### 3.3. Cloud water versus bulk precipitation chemistry

The major difference between cloud and precipitation water was TIC and absolute values of chemical constituents, while the proportions between them at a given time were almost identical (Tables 2 and 3). In all cases TIC of cloud water was much higher than that of precipitation water with the TIC ratio between cloud water and precipitation water ranging from 2.2 at SZ and SI in the winter of 2003/2004 to 5.0 and 4.9, respectively, in the summer of 2004. These different concentrations are caused by the fact that fog and low cloud droplets form in the polluted environment of the atmospheric boundary layer where condensation nuclei are abundant while raindrops and snowflakes typically come from much cleaner higher zones of the

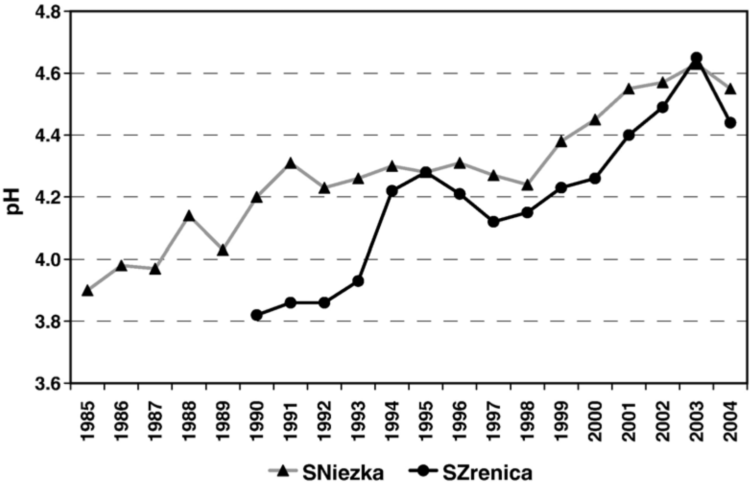

Fig. 5.

troposphere. The overall trend of precipitation chemistry between the early 1990s and 2003/04 was very similar to the transformation of the cloud water ionic structure with the sulphate ion being reduced most.

### 3.4. Spatial differences

Measurements showed differences in TIC and its seasonal variation between the two hill peaks (SI and SZ). The higher annual total of chemical components in cloud water at SI was due to its upwind position in relation to the prevailing wind direction (Tables 1 and 2). Such differences are more evident in 1995 and less evident during 2003/04, with the TIC ratio between CWSI to CWSZ being 1.56 and 1.15, respectively. This is due to the "seeder–feeder" effect which involves scavenging of orographic cloud droplets by precipitation particles — a process that leads to both precipitation and pollutant wet deposition enhancement (Bergeron, 1965; Dore et al., 1999). SI as the upwind hill had received the bulk of pollutant deposition, thus to a certain extent "sheltering" the successive hill peaks downwind. This effect occurred for the whole year, but is more pronounced during the winter because of a higher orographic cloud frequency and the very efficient scavenging of cloud droplets by snow crystals, which have a high surface area to volume ratio (Dore et al., 1992). That is why the TIC is about 38% bigger at SI in the winter. With regard to the anomalous meteorological conditions during the summer of 2003, the TIC was an exceptionally 5.4% higher at SZ. This is mainly due to a significantly higher frequency of fog with a small liquid water content at SZ. Moreover, between June and September 2003 only convective rainfall was observed with no mesoscale cap clouds present, meaning that circumstances for the "seeder–feeder" effect were unfavourable.

Seasonal changes of rainfall water concentrations are opposite to cloud water concentrations because of the fact that rainfall chemistry is more prone to emissions rates which are higher during the winter. In the case of cloud water chemistry, the seasonal changes are more dependent on cloud base height. In general it is typical that precipitation is more polluted at SI where cap clouds are efficiently washed out.

### 3.5. Deposition hot spots

At some sites high chemical concentrations of rain and cloud water is accompanied by higher than average rainfall totals and by very efficient horizontal precipitation through fog and clouds. These are areas situated at wind-exposed mountain summits or ridges covered by forest or at least groups of trees or bushes which are efficient receptors of wind-driven fog/cloud droplets. In such places, high enough to be frequently immersed in orographic clouds, the amount of horizontal precipitation can be comparable with bulk precipitation which makes wet deposition (rain/snow + fog) 2 to 3 times higher than forest free sites in the same landform, and even 10 to 15 times higher than in the neighbouring low lands.

A typical example of the latter is Stog Izerski where extreme deposition rates are still observed. As shown at Fig. 6 the total deposition, measured as a chemical input by throughfall at the edge of a spruce forest site during 5 months of the 2004 growing season, equals 1.98 mol $m^{-2}$ with nitrogen (from both nitrate and ammonia) being the main component (0.91 mol $m^{-2}$). This value is equivalent to 305.8 kg of N $ha^{-1}$ $yr^{-1}$, which can be compared to 20.2 kg of N $ha^{-1}$ $yr^{-1}$ under a spruce canopy at a nearby low land site. Thus, the current nitrogen deposition at such hot spots situated in an igneous bedrock environment is around 20 times larger than the relevant critical value of nitrogen deposition stated by Mill et al. (2003).

At the same site (forest edge at Stóg Izerski) the total deposition was composed of around 30% precipitation and 70% direct fog/cloud deposition, while the associated proportion at the nearby lowland site was around 5% and 95%, respectively.

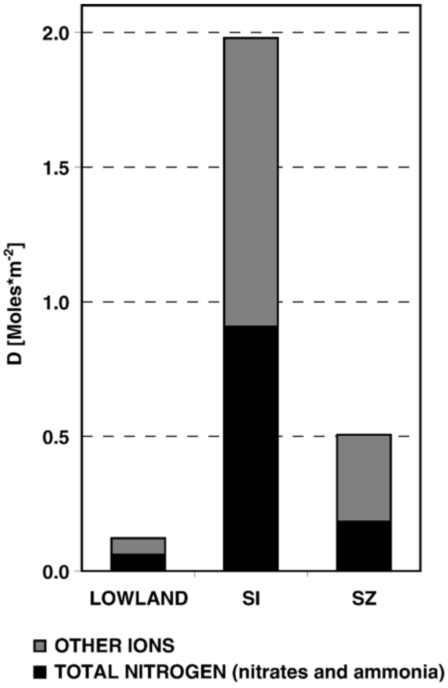

Fig. 6.

## 4. Conclusions

A substantial reduction of $SO_2$ emissions between the early 1990's and 2004 has been observed with a threefold decline of sulphate concentrations in both cloud water and precipitation water samples. Nitrates and ammonia became the dominant ions with an increase of relative concentration. A distinct increase of relative role is evident for $Na^+$, $Cl^-$ and $Ca^{2+}$.

Due to the more polluted nature of the boundary layer than the free atmosphere, cloud water droplets (originating in low-level air) had higher pollutant concentrations than precipitation, implying that cloud droplets may be a more sensitive indicator of emission levels than precipitation itself.

Since the early 1990's, pH values of cloud water samples show a decreasing trend of acidity and an increased frequency of alkaline events. The wider spectrum of pollutant concentration in cloud water, relative to precipitation, points towards the greater utility of cloud chemistry to identify pollution origin at a given site.

Despite of the observed decrease in pollution concentrations and deposition, some "hot spots" still exist where annual deposition of nitrogen can be as high as 300 kg $ha^{-1}$ $y^{-1}$, with fog/cloud water forming around 70% of this value.

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