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ATMOSPHERIC SULPHATE FORMATION AND AIR POLLUTION EPISODES IN THE UPPER PARTS OF THE KARKONOSZE MOUNTAINS, POLAND

A field programme for the measurement of the chemical composition and potential sources of atmospheric aerosols was conducted in the upper parts of the Karkonosze Mountains in the period from April, 1987 to September, 1988.

The highest concentrations of pollutants in the air over the Karkonosze Mountains (SO₂ up to 47 μ g/m³, NO_x up to 14 μ g/m³, fine sulphate aerosol up to 180 μ g/m³, NH₃ up to 45 μ g/m³) were recorded during the inflow of cold and humid maritime-polar air masses from north-west Europe.

The results of this study suggest that reactions of SO_2 in aqueous phase with H_2O_2 tend to dominate during aerosol formation in the region of interest. There is evidence that SO_2 from the mixing layer undergoes a burst of sulphate formation as the air ascends up the mountains.

1. INTRODUCTION

In the past few years, acidic aerosols in the atmosphere have become a problem of serious concern. Their formation by gas to particle conversion has been confirmed by numerous researchers [1]–[6]. It is a well-established fact that acidic aerosols have both direct and indirect effects on terrestrial and aquatic ecosystems [7]–[10]. Evidence of such an impact is also impressive for the Sudeten Mountains (south-western Poland). Atmospheric pollution not only inhibits the growth of coniferous forest, but also accounts for their ever increasing damage. The degradation comes from the NW part (ridges of the Izerskie Mts.) and moves towards higher summits of Alpine type (the Karkonosze Mts.).

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The problem was first formulated 12 years ago, but until 1986 relatively little has been known about the chemical composition of the aerosol and precipitation in the region of interest.

In this study, we present the field data for the period from April, 1987 to September, 1988. Particular consideration was given to the episodic nature of air pollution and the sulphate formation mechanisms in the area of interest. The investigations included a total of ninety cases which were related to air mass category, wind direction and velocity, air temperature, air humidity, and stability classes.

2. METHODS

Samples of SO₂, NO_x (NO₂ and NO), NH₃ and sulphate aerosol were collected simultaneously at three sampling sites. All of them are located in the western part of the Sudeten Mts., where forest damage has become increasingly frequent. Thus, site No. 1 is placed at the Szrenica summit (1362 m a.s.l.), the first high elevation exposed to the air masses coming from the central and western Europe. Site No. 2 is situated at the meteorological station on the front facetof the Szrenica mountain, about 40 m below the summit. Site No. 3 (which has been operated since December, 1987) is located at an altitude of 762 m a.s.l. (Rozdroże), in the vicinity of a meteorological station, 10 km from the Szrenica summit in the NW direction. Samples were taken in 24 h intervals continuously for five days every month from April, 1987 to September, 1988.

 SO_2 was trapped in a sodium tetrachloromerculate containing bubbler (the pararosaniline method). Air flow amounted to 2 m³/day. Nitrogen dioxide concentration (NO was oxidized to NO₂ by aqueous potassium permanganate solution) was determined using the modified Saltzman technique. NH₃ was collected by aspiration of air and its passage through 0.01 n H₂SO₄. The concentration of the resultant ammonium ions was determined by the indophenol method. Particulate sulphates were collected on a Whatman 41 filter which had been mounted in front of an acid H₂O₂ containing bubbler for the absorption of sulphur compounds. Air flow rate was 2.2 m³/day. The concentrations of sulphates (from filter extracts and H₂O₂ solution) were determined by the barium perchlorate-thorin method.

The concentrations of sulphur compounds determined in the H_2O_2 solution were usually far in excess, as compared to the sulphur concentrations established by the pararosaniline method. As each of the two methods involves a different form of sulphuric pollutants, it is not surprising that the calculated concentrations fail to correlate well with one another. But a paired *t*-test has revealed that the two data sets are not statistically similar. Hence, the bubbler that contains H_2O_2 solution is likely to include not only sulphates obtained via oxidation of SO₂, but some other compounds containing sulphate ions. Some of the meteorological data sets (temperature, humidity, wind direction and velocity, pressure, cloudiness, radiation) were established from on-site observations. Others (mass category, height of inversion layer, synoptic situation in central Europe, direction of air mass inflow at 850 mb) were submitted by the National Weather Service of Wrocław.

3. RESULTS AND DISCUSSION

3.1. METEOROLOGICAL BACKGROUND

During the sampling periods, air masses from all wind sectors were monitored. Sudeten were influenced primarily by maritime-polar (MP) air masses from NW, W, SW and S directions (43 out of 60 events in the summer season, and 23 out of 30 events in the winter season). This influence was associated either with low pressure systems (with prevailing warm air masses from central Europe or from central and western Czechoslovakia) or with high pressure systems (with prevailing cool air masses from the northern and central parts of the Germany). The inflow of continental-polar (CP) and arctic (A) air masses from E-S and NW-N-NE directions, respectively, should be attributed to anticyclonic weather conditions. Air mass inflow from the Mediterranean was rarely recorded in the measuring period (6 events).

The data sets obtained were divided into two major groups according to the seasonal pattern - those collected in the summer season (April–September) and those collected in the winter season (October–March). Mean concentrations of all the species studied were calculated for the two seasons and for the air masses to which the Sudeten were exposed within those time spans.

3.2. CONCENTRATIONS OF AIR POLLUTANTS

The concentrations of all species analysed varied widely throughout the measuring periods. Several episodes were characterized by increased concentrations, especially those of the sulphur compounds. Tables 1--3 summarize the results for the Szrenica summit (site No. 1). The concentration patterns were similar in site No. 2, but the difference in the measured values of concentrations between the two sites amounted to $\pm 20\%$.

Tables 1 and 2 give mean concentrations (with standard deviation) of SO_2 and sulphate aerosol and table 3 gives mean concentrations (with standard deviation) of NO_x and ammonium ions for air masses reaching the Szrenica summit during the summer and winter periods, respectively. Table 4 provides 24-hour concentrations of SO_2 , NO_x , sulphates and ammonium ions for selected case studies. Table 5 shows relevant meteorological conditions. The table includes solely data sets for air mass inflow from the westerly wind section.

Table 1

Mean 24 hour concentrations of SO₂ and sulphate aerosol (\bar{s} , $\mu g/m^3$) and standard deviation (σ , $\mu g/m^3$) for air masses reaching the Szrenica summit during the summer period (n – number of observations)

Air mass cat.	Wind direction	s _{so2}	σ_{SO_2}	5 _{SO4(f)}	$\sigma_{\mathrm{SO}_{4(f)}}$	55504(c)	$\sigma_{\mathrm{SO}_{4(\mathrm{c})}}$
MP	S-SW-W n = 14	25.7	8.0	52.5	18.4	30.2	15.6
MP	n = 14 W-NW-N n = 29	33.5	26.1	94.1	64.4	14.4	6.1
СР	E-SE-S n=6	10.8	9.0	< 1.0	-	31.3	11.9
MA	$\frac{1}{n} = 8$	11.0	1.5	27.6	23.1	26.4	13.6

Table 2

Mean 24 hour concentrations of SO₂ and sulphate aerosol (\bar{s} , $\mu g/m^3$) and standard deviation (σ , $\mu g/m^3$) for air masses reaching the Szrenica summit during the winter period (n – number of observations)

Air mass cat.	Wind direction	\bar{s}_{SO_2}	σ_{so_2}	^{\$} so _{4(f)}	$\sigma_{\mathrm{SO}_{4(\mathbf{f})}}$	$\bar{s}_{SO_{4(c)}}$	$\sigma_{{\rm SO}_{4(c)}}$
MP	S-SW-W	3.4	1.4	54.0	30.1	7.9	6.1
MP	n = 10 W-NW-N n = 13	30.9	11.4	64.9	25.5	5.9	1.2
MA	n = 15 NW-N-NE n = 5	8.5	5.2	13.1	12.1	18.0	8.1

Table 3

Mean 24 hour concentrations of NO_x and ammonium ions (\bar{s} , µg/m³) and standard deviation (σ , µg/m³) for air masses reaching the Szrenica summit during the summer and winter periods (n – number of observations)

Air mass cat.	Wind direction	$\bar{s}_{\mathrm{NO}_{\mathbf{X}}}$	σ _{NO_x} summer	^s _{NH4} period	$\sigma_{{ m NH}_4}$	Š	NOx	σ_{NO_x} winter	^{\$} _{NH4} period	$\sigma_{{ m NH}_4}$
MP	S-SW-W	4.1	2.1	31.7	14.9		4.6	2.1	36.5	18.1
	n = 14					n = 10				
MP	W-NW-N	6.0	3.3	36.6	18.9		8.1	4.7	29.0	19.5
	n = 29					<i>n</i> = 13				
CP	E-SE-S	4.1	1.2	10.2	4.4		_		_	—
	n = 6									
MA	NW-N-NE	3.3	0.6	5.8	2.0		8.2	5.2	5.9	0.8
	n = 8					<i>n</i> = 4				

Г	a	b	1	e	4

D	Concentration, µg/m ³						
Date	SO ₂	NO _x	SO _{4(f)}	SO _{4(c)}	$\rm NH_4$		
11-12 August 1987	47.0	3.6	152.0	17.0	22.0		
13-14 August 1987	19.1	5.5	147.4	19.4	14.0		
15-16 September 1987	13.2	4.8	180.0	21.0	44.8		
18-19 September 1987	23.1	13.8	159.2	16.8	11.0		
14-15 October 1987	2.1	3.5	120.0	7.9	12.5		
9-10 February 1988	24.9	4.0	110.1	54.9	23.2		
10-11 February 1988	20.0	3.9	33.9	30.0	13.5		
8-9 March 1988	16.2	3.2	88.0	5.2	13.2		
9-10 March 1988	23.7	10.1	38.4	5.5	2.5		
11-12 April 1988	14.2	7.8	61.4	16.0	29.1		
12–13 April 1988	12.6	3.8	69.4	16.2	22.0		

24 hour concentrations of SO_2 , NO_x , sulphate and ammonium ions for selected case studies at the Szrenica summit

Table 5

Meteorological parameters for selected case studies

Date	Air mass cat.	Inflow direction [deg]	Temperature [°C]	Wind velocity [m/s]	Relative humidity [%]
1-12 August 1987	MP	270-310	2.7–3.9	10–17	95–100
3-14 August 1987	MP	270-330	3.6-6.3	4-8	94-100
5-16 September 198	87 MP	270-320	2.8-7.2	10-20	86-100
8-19 September 198	87 MP	270-320	4.0-10.8	11-18	96-100
4-15 October 1987	MP	210-250	0.9-3.2	12-40	90-100
9-10 February 1988	MP	280-290	-8.4 - (-6.0)	12-24	99–100
0-11 February 1988	8 MP	270-280	-8.4 - (-7.5)	7-21	97-99
89 March 1988	MP	280-290	-8.4-(-7.0)	5-24	98
9-10 March 1988	MP	270-010	-13.0-(-11.7)	4-19	99-100
1-12 April 1988	MP	220-270	-4.7-0.8	8-14	73–98
2-13 April 1988	MP	220-270	0.8-3.2	7-12	53-72

For the purpose of this study, we have made a distinction between the fine sulphate aerosol, $SO_{4(f)}$, equivalent to the difference in the concentration between $S_{H_2O_2}$ (concentrations of sulphur compounds measured in the H_2O_2 solution) and S_{TCM} (concentrations of sulphur compounds determined by the pararosaniline method), and the coarse sulphate aerosol, $SO_{4(c)}$, determined from filter analysis. Thus, for the majority of the species investigated, the highest concentrations were associated with the inflow of MP air masses from western and north-western Europe. During CP air mass inflow from eastern Poland and A air mass inflow from northern Poland and northern Germany only $SO_{4(c)}$ (which is the longest-lived portion

of atmospheric aerosol) displayed increased concentrations. In the case of CP air mass inflow, only minute quantities of $SO_{4(f)}$ were monitored. It is interesting to note that concentrations of the species investigated (particularly those of the sulphur compounds) were highly variable even within the same air mass. And this means that the influent air masses, to which the Sudeten were exposed, were characterized by variable pollution levels. As far as the calculated mean concentrations are concerned. we note the following: the concentrations of the pollutants which influenced the Sudeten in the summer season of 1987 were especially high. This holds for the extremely high concentrations measured in August, 1987 (from 10th to 16th) and in September, 1987 (from 14th to 19th). At that time the meteorological situation was influenced alternately by cyclonic and anticyclonic weather conditions. There was inflow of cool and humid MP air masses with prevailing W-NW directions. Air temperature in the mountains fell by 5 degrees Centigrade (from 10°C to 5°C). The inversion layer persisted on the slopes till 10 or 11 h (local time). Near stable or stable conditions prevailed during daytime; stable conditions were dominant in the night hours. The summits remained above the stratus cloud base.

Air pollutants emitted from high stacks in that span of time were slowly undergoing dilution while they were moving downwind in stable air with low vertical exchange. It is a fact that air ascends to the mountain, and so do the pollutants in the mixing layer. Thus, sulphur dioxide may be rapidly converted to H_2SO_4 during entrainment into the upper atmospheric layers of enhanced air humidity and lower temperature. Further support for such a burst of sulphate formation comes from the work reported previously, where aerosol samples were collected on electron microscopic screens and then examined under a transmission electron microscope [11], [12]. Numerous fine particles were found in the smallest size fraction collected; most of them occurred within the size range below 0.1 μ m. A burst of sulphate formation during the upward transport is a more likely explanation for very high concentrations of sulphates measured in the mountains far away from the emitter. It also may explain the forest damage which comes from the front face of the Sudeten.

Increased concentrations of sulphur compounds were also measured in February and March, 1988. The Sudeten were influenced by cyclonic weather conditions with humid and cold air masses from W and NW directions. In that time span stable conditions prevailed. Unstable conditions (observed on February 10–11, as well as on March 7–8 and 9–10) brought about a decrease in the concentrations of sulphates. No decrease was found to occur in the concentrations of SO₂ and NO_x. This finding indicates that local emission is a potential contributor to the enhanced concentrations of primary pollutants at that time.

Equally high concentrations of sulphur compounds were recorded with the inflow of MP air masses from S-W sector. It is interesting to note that, at the air mass inflow from S-SW-W directions, SO_2 and NO_x concentrations were distinctly lower in the winter season than in the summer season. Air mass inflow from the S-W wind sector was monitored in October-November, 1987. At that time, there was observed a strong inversion just below the summit (about 1000 m a.s.l.), therefore vertical mixing was significantly limited. The concentrations of sulphur compounds measured above the inversion layer should be attributed primarily to long-range transport. In October–November, 1987, air temperature ranged from 3 to 7°C, relative humidity was higher than 85% and clear sky prevailed. The high extent of conversion (above 70%), low air temperature and high humidity suggest that liquid-phase oxidation of SO₂ by H_2O_2 rather than gas-phase oxidation of SO₂ might have been responsible for the high sulphate concentrations recorded in that span of time [4], [5]. This suggestion has additional support in the MIDDLETON et al. [1] and SAXENA and SEIGNEUR [6] models which allow predicting the oxidation of SO₂ to sulphate in atmospheric aerosols. According to the SAXENA and SEIGNEUR model [6], the reaction of dissolved SO₂ with H_2O_2 is very sensitive to temperature, thus contributing appreciably to the total sulphate formed at 5°C (and not at temperatures higher than 5°C). The solubility of both SO₂ and H_2O_2 increases at lower temperatures.

Other mechanisms of sulphate formation (e.g., reactions of dissolved SO_2 with O_3 or its oxidation in the gas phase) do not seem to have contributed significantly to sulphate formation in the region of interest. The aqueous-phase oxidation of SO_2 with O_3 becomes significant at comparatively high pH [6], [13], [14], [15]. The atmospheric aerosols in the Sudeten are highly acidic (pH 2.8-4.4) [12]. The gas-phase reaction of SO_2 with OH radicals is temperature-sensitive [6], [16]. At decreased temperatures (which prevailed in the area investigated) the concentrations of OH radicals are very low.

But we can also adopt the mechanism of trace-metal-catalyzed oxidation of SO_2 by O_2 [13]. Such reactions occur on the available airborne particulates, and they are likely to contribute to the formation of $SO_{4(c)}$ which is efficiently retained by the filter. Water extracts of atmospheric aerosol particulates contained iron and manganese ions (from minute quantities to 640 µg Fe²⁺/g particulate and from minute quantities to 120 µg Mn²⁺/g particulate). Concentrations of zinc ions measured in water extracts were exceptionally high (1700 to 2500 µg/g). No trace metals were found in the H₂O₂ solution.

Table 6 summarizes the results for the sampling site Rozdroże, which had not been operated before December 1987. It should be noted that the Rozdroże site has been located in a valley. Forests grow along the northern and southern sides of the valley which is open to the East and to the West. Thus, polluted air masses from easterly or westerly directions have a free pathway and circulation through the valley.

Taking into account the data sets for the period of January to September 1988 (table 6), we can see that concentrations of the species investigated in the valley (and specificaally those of sulphur compounds) are noticeably higher than these at the Szrenica summit in the winter season. The difference in the concentrations is likely to be due to the non-uniform distribution of concentrations which vary with altitude.

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standard deviation (σ , µg/m ³) for air masses reaching Rozdroże ($n - n_{\rm c}$	
standard deviation (0, μ g/m) for an masses reaching Kozdroze ($n - m$	uniber of observations)

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Air	Wind		Concentration, $\bar{s} \pm \sigma$						
cat.	direction	SO ₂	NO _x	SO _{4(f)}	SO _{4(c)}	NH4			
			Winter peri	od					
MP	W-NW-N $n = 11$	28.8 ± 16.8	7.9 ± 3.3	138.3±49.6	16.4 ± 12.0	28.8 ± 10.7			
			Summer per	iod					
MP	S-SW-W n = 9	20.2 ± 13.8	7.0 ± 3.1	23.0 ± 12.9	20.6 ± 13.3	19.4± 2.7			
MP	W-NW-N n = 5	17.0±10.2	6.5 ± 3.4	19.0 ± 7.6	36.8±17.6	36.0±22.3			

A likely explanation may also be the influence of local emission sources (this sampling point often stays below the inversion layer). As a result of wind velocity (which amounted to one third of the value measured at the Szrenica summit) the residence time of air pollutants in the valley was longer and might have been responsible for the higher degree of conversion during the winter season.

3.3. POLLUTION SOURCES

The inflow of highly polluted air masses from westerly directions results in forest damage. It seems therefore reasonable to describe some major pollution sources which can be regarded as potential contributors to the concentrations of many species in the Sudeten aerosol.

One of these sources is an industrial complex which consists of four large lignite-fired power plants. They are situated at the border of Poland with Germany, 40 to 50 km from the Szrenica summit in the W-NW direction. Another source is the "golden triangle" of Leipzig, Karl-Marx-Stadt and Dresden located 150–200 km in the W-NW direction. The "black belt" of northern Bohemia (Czechoslovakia), 100 to 150 km from the Karkonosze Mts. in the W-SW direction, is the last important contributing source.

In terms of long-range transport, these distances are not too long. At the average air mass flow of 50 to 70 km/h (measured on September 14 to 16, 1987) the air pollutants emitted from the coal-fired power plants reach the mountains only after 1 hour or so, whereas those released by the "golden triangle" within 2–3 hours.

The three contributing sources are likely to be responsible for the environmental degradation of the Sudeten. This degradation comes from the ridges of the Izerskie Mts. and moves towards the higher parts of the area investigated, the Karkonosze Mts.

4. CONCLUSIONS

There is evidence that a number of episodes with a duration from 2 to 5 days took place in the periods studied. During the episodes the daily concentrations of SO_2 , NO_x , $SO_{4(f)}$ and NH_3 amounted to 47, 14, 180, and 45 µg/m³, respectively.

The highest concentrations of pollutants in the atmosphere over the Karkonosze Mts. were recorded during the inflow of cold and humid maritime-polar air masses coming from south-western to north-western Europe. High concentrations of pollutants were observed simultaneously at the summits and the valleys.

During the winter periods higher concentrations of $SO_{4(f)}$ were recorded in the valleys. This phenomenon is likely to be associated with the frequent occurrence of inversion layers. The lack of ventilation led to the accumulation of SO_2 and sulphate. Local emissions should also be considered as a potential contributor to the enhanced concentrations of primary pollutants at that time.

Under realistic conditions in the mountains, oxidation of SO₂ appears to occur in the aqueous phase. The high extent of conversion (above 70%), low air temperature (below 10°C) and high humidity (90 to 100%), suggest that liquid-phase oxidation of SO₂ by H_2O_2 might have been responsible for the high sulphate concentrations in the region of interest.

A burst of sulphate formation during the upward transport is a more likely explanation for the very high concentrations of $SO_{4(f)}$ measured in the mountains far away from the emitter. It also may explain the forest damage which comes from the front face of the Sudeten and moves towards the higher parts of the mountains.

The major pollution sources which can be regarded as the potential contributors to the Sudeten aerosol are three industrial areas located mainly outside Poland, from 50 to 200 km in the SW-NW directions.

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TWORZENIE SIĘ SIARCZANÓW W ATMOSFERZE ORAZ EPIZODY ZANIECZYSZCZEŃ POWIETRZA W WYŻSZYCH PARTIACH KARKONOSZY

Pomiary chemicznego składu aerozolu atmosferycznego w wyższych partiach Karkonoszy wykona w okresie od kwietnia 1988 do września 1989. Najwyższe stężenia zanieczyszczeń rejestrowano podczas napływu chłodnych i wilgotnych mas powietrza polarno-morskiego z kierunków zachodnich. Wyniki pomiarów sugerują, że na badanym obszarze dominuje mechanizm utleniania SO_2 w fazie ciekłej z udziałem H_2O_2 . Stwierdzono, że reakcje te zachodzą najintensywniej podczas wznoszenia się mas powietrza wzdłuż zboczy górskich.

ОБРАЗОВАНИЕ СУЛЬФАТОВ В АТМОСФЕРЕ И ЭПИЗОДЫ ЗАГРЯЗНЕНИЙ ВОЗДУХА В ВЫСШИХ ПАРТИЯХ КАРКОНОШИ

Измерения химического состава атмосферного аэрозоля в высших партиях Карконоши проводились с апреля 1988 по сентябрь 1989. Самые боьшие концентрации загрязнений были зарегистрированы во время адвекции прохладных и влажных масс полярно-морского воздуха с запада. Результаты исследований показывают, что на исследуемой территории преобладает механизм окисления SO₂ в жидкой фазе при участии H₂O₂. Было установлено, что эти реакции происходят найболее интенсивно во время подъема масс воздуха вдоль горных скатов.