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POLLUTANT AND WIND DATA FOR EPISODES IN THE ATMOSPHERE OVER THE SUDETEN, POLAND

In this paper, we present the experimental results of the Emission Abatement Strategies and the Environment Programme to show an evidence of great variability of pollutant levels in the air over the Sudeten mountains. We attempt to define the prototype meteorological conditions that might be conducive to high concentrations of ozone, sulphur and nitrogen oxides observed in the mountains. In order to interpret meteorological conditions during episodes, the hourly measurements taken in the automatic station and the weather maps were used. We conclude that two general categories of meteorological conditions are responsible for pollutant episodes in the region under consideration: the high pressure system and the weather front; however, not all high pressure systems and weather fronts necessarily cause high levels of pollutants.

1. INTRODUCTION

The Black Triangle encompassing highly industrialized areas of Czech Republic, Poland and Germany has become severely polluted. Environmental damages in this area are particularly serious at highly elevated sites. There is evidence that highly elevated sites receive greater amounts of atmospheric pollutants than surrounding low-elevated areas. This phenomenon is primarily due to the additional input of chemicals from cloud water interception [1], [2]. In the upper parts of the mountains, clouds are not only the source of additional wet deposition, but they are also an important medium of SO_2 oxidation [3], [4]. During the investigations carried out over the period of 1988–1993 in upper parts of the Karkonosze Mountains located in the middle of the Black Triangle, this statement was confirmed

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by an efficient conversion of SO_2 to sulphate ions, approaching occasionally 90% [5]. This phenomenon is investigated in detail within the framework of the EASE (Emission Abatement Strategies and the Environment) Project.

This paper describes the first step of the process: the identification of the prototype meteorological conditions that might be favourable to the appearance of high concentrations of sulphur and nitrogen oxides in the Karkonosze and Izerskie Mountains. In order to interpret the meteorological conditions during episodes, the hourly measurements taken in the automatic station in Czerniawa were used.

2. DESIGN OF FIELD INVESTIGATIONS

2.1. SAMPLING SITES AND SAMPLING PROCEDURES

Ambient aerosol samples are taken at three sampling sites located in the western part of the Sudeten Mts. Thus, the site No. 1 is placed at the summit of Szrenica (1362 m asl). The instrumentation is housed in a small caravan inside the meteorological garden. The caravan is equipped with: four experimental setups comprising midget fritted gas absorbers, rotameters, gas-meters, pumps to collect gaseous and particle pollutants, i.e., SO_2, NO_x, ammonia ions, total sulphur and sulphates, and the automatic dichotomous sampler (virtual impactor, Sierra-Andersen SE 245-10-1). The site No. 2 is situated at the altitude of 762 m asl (Rozdroże), 10 km from the summit of Szrenica in the NW direction. It is operated manually. SO_2, NO_x and TSP samples have been collected for two weeks in a month. The site No. 3 is located at the RIVM site (Czerniawa) approximately 15 km from the summit of Szrenica in the NW direction. The automatic station for air monitoring is placed in Czerniawa. It allows the constant measurements of SO_2, NO, NO_2 and O_3 concentrations as well as determination of wind direction and speed. Both stations are under control of the Provincial Inspectorate of Environment Protection in Jelenia Góra.

Measurements were taken during field campaigns at the Szrenica site. Four field campaigns were conducted: in February 1994 (3 weeks) and 1995 (3 weeks), July 1994 (2 weeks) and October (2 weeks). During the campaigns daily aerosol samples were collected.

2.2. ANALYTICAL PROCEDURES

Concentrations of atmospheric aerosols (SO_2, NO_2, NH_4, total sulphur) at the Szrenica site were determined by colorimetric methods following absorption of the gases by suitable reagents. SO_2 was trapped in a sodium tetrachloromercurate bubbler (the pararosaniline methods [6], [7]). Air flow amounted to 2 m^3/day. NO_2 concentration (NO was oxidized by aqueous solution of potassium permanganate) was determined by the Jacobs–Hochheiser method (the arsenite method [7], [8]).

NH_3 was collected by aspiration of air and its passage through 0.01 n H_2SO_4. The concentration of the resultant ammonium ions was determined by the indophenol method [9].

Sulphates were collected on a Whatman 41 filter which had been mounted in the front of an acid hydrogen peroxide solution containing bubbler for absorption of sulphur compounds [10]. Air flow rate was 2.2 m^3/day. The concentrations of sulphates (from filter extracts and H_2O_2 solution) were determined by the barium perchlorate–thorin method [11].

The following meteorological data were measured at the site: temperature, pressure, relative humidity, wind direction and speed. 48-h backward trajectories (arriving at 0.00 and 12.00 LTM at the Szrenica site) for the field campaign periods were delivered from the Department of Meteorology and Environment Protection, Charles University, Prague.

3. RESULTS AND DISCUSSION

3.1. AIR POLLUTANT DATA

The results of investigations carried out previously in the Karkonosze and Izerskie Mountains over the period of 1988–1993 allowed us to arrive at firm conclusions [5], [12]. The weather conditions in the mountains not only change easily but they also depend on the complex topography. Therefore the interpretation of the pollutant concentrations proved to be more complex than it had been expected. The samples were taken during 24 h, and at the same time the weather conditions changed as frequently as three times and defining the average conditions was often an impossible task.

In order to insight into the concentration variations as well as to detect the main frequency of pollutants and their linear interdependences within time series, the classical methods were applied to the present data (for 1994). The daily and hourly profiles of the pollutant series were investigated to define the basic properties of the pollutant behaviour. In the figures 1 and 2, the time series of sulphur dioxide, nitrogen dioxide, total sulphur and ammonium aerosol concentrations measured during field campaigns in 1994 at the Szrenica site are presented. The time series for Czerniawa and Rozdroże are shown in figure 3 and 4, respectively. It is evident that in the case of Czerniawa we deal with a large number of missing values, particularly for the concentrations of ozone and nitrogen oxides.

The time series represented by figures allow us to draw the general conclusions only. The nitrogen dioxide concentrations are characterized by similar variation over the consecutive measuring periods at all sampling sites, while the sulphur dioxide concentrations exhibit a wider variation during cold months than during warm months. The variations of sulphur dioxide and total sulphur concentrations follow

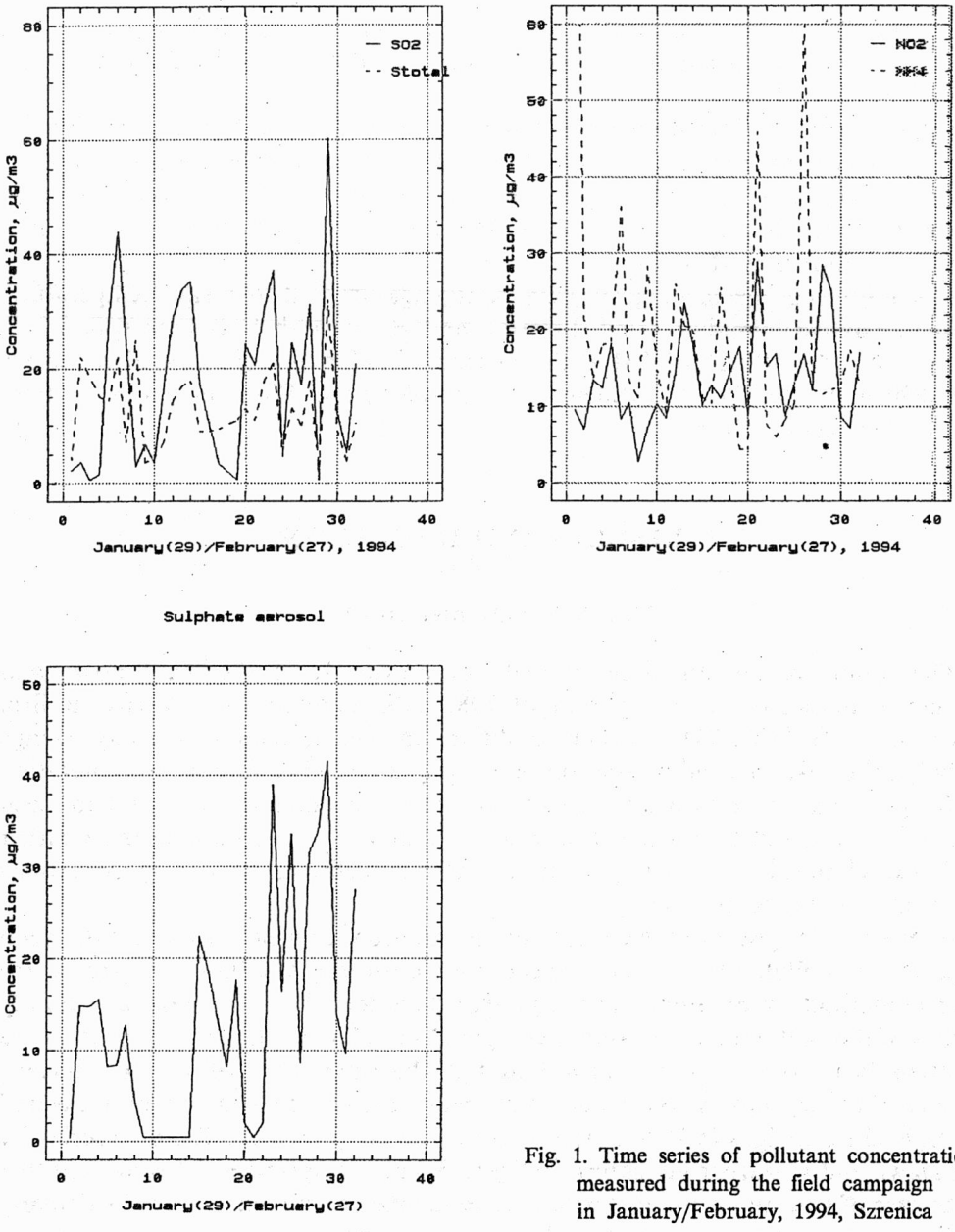

Fig. 1. Time series of pollutant concentrations measured during the field campaign in January/February, 1994, Szrenica

different pattern at the Szrenica site. In contrast to SO_2 concentrations, total sulphur exhibits a wider variation during warm months (figures 1, 2). The variation in ammonium aerosol concentrations is similar to that of sulphate aerosol (a wider variation in winter). On the other hand, the sulphate aerosol reveals a uniform variation at the Rozdroże site (figure 4). It is difficult to establish the variation of ozone concen-

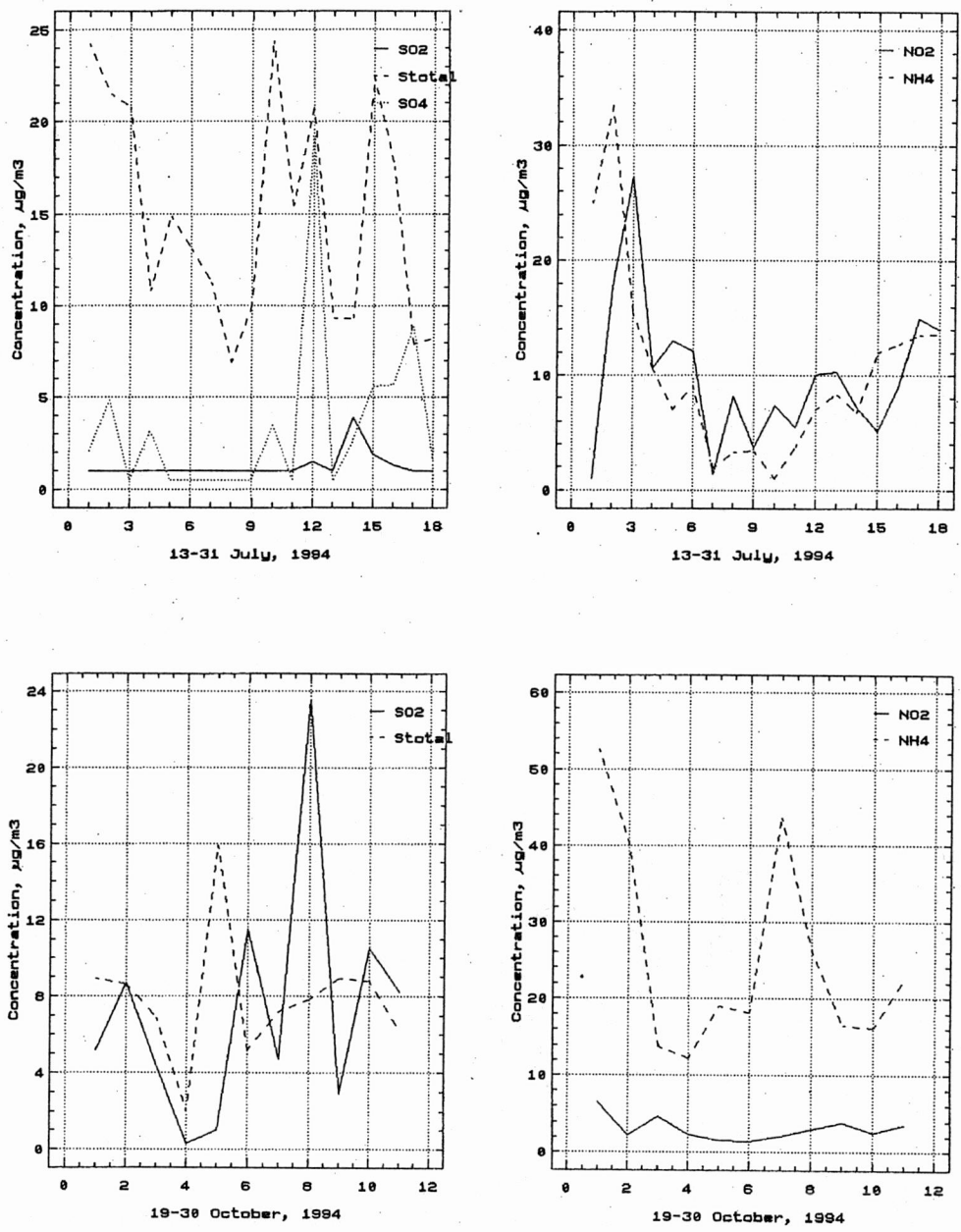

Fig. 2. Time series of pollutant concentrations measured during field campaigns in July and October, 1994, Szrenica

trations during cold months, except for December, because of too small number of the measurements. It is widely known, however, that the ozone daily concentrations are

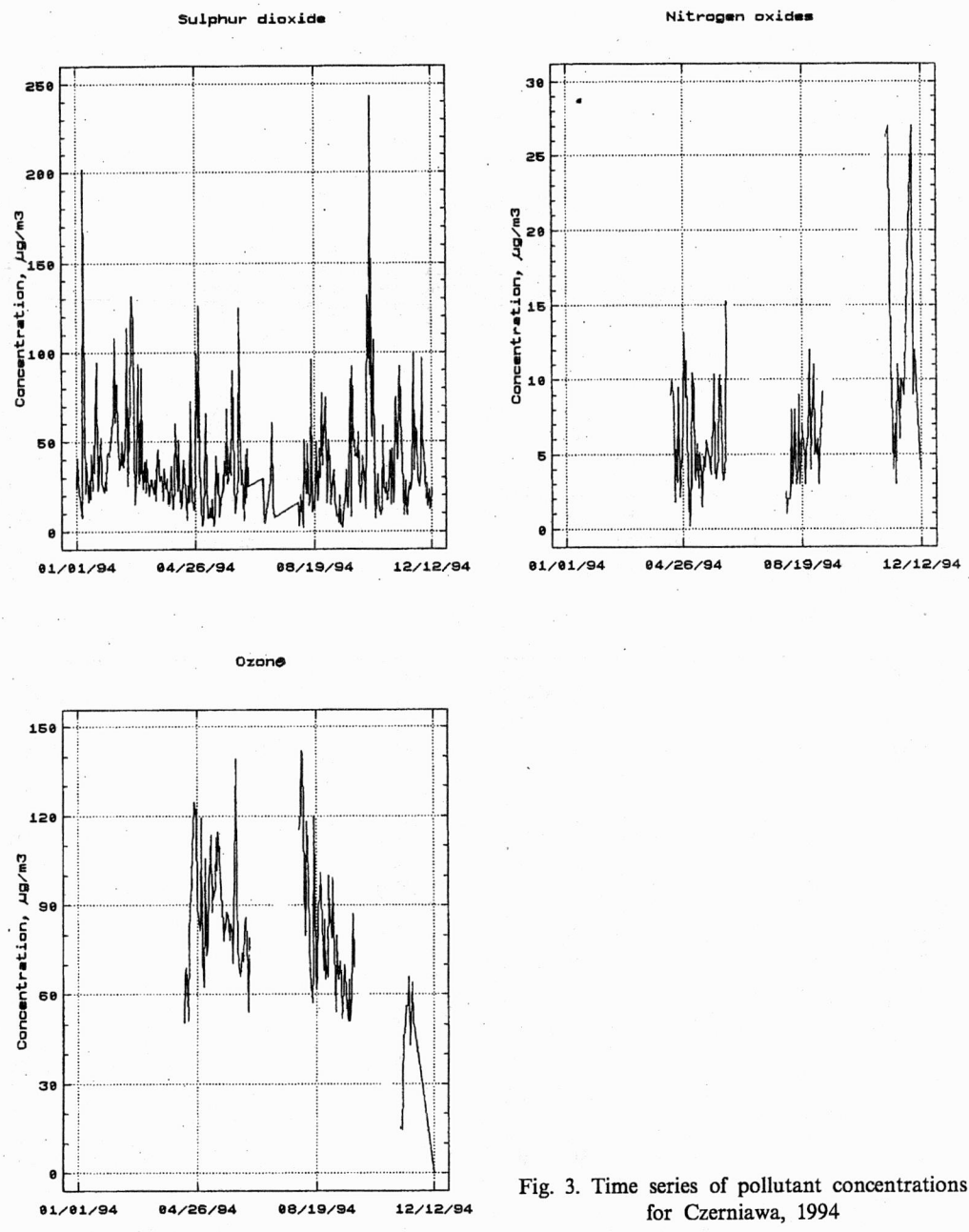

Fig. 3. Time series of pollutant concentrations for Czerniawa, 1994

higher over the warm months, and the variation appears attenuated during the cold months. This phenomenon may be explained by attenuation of photochemical activity in the atmosphere during winter.

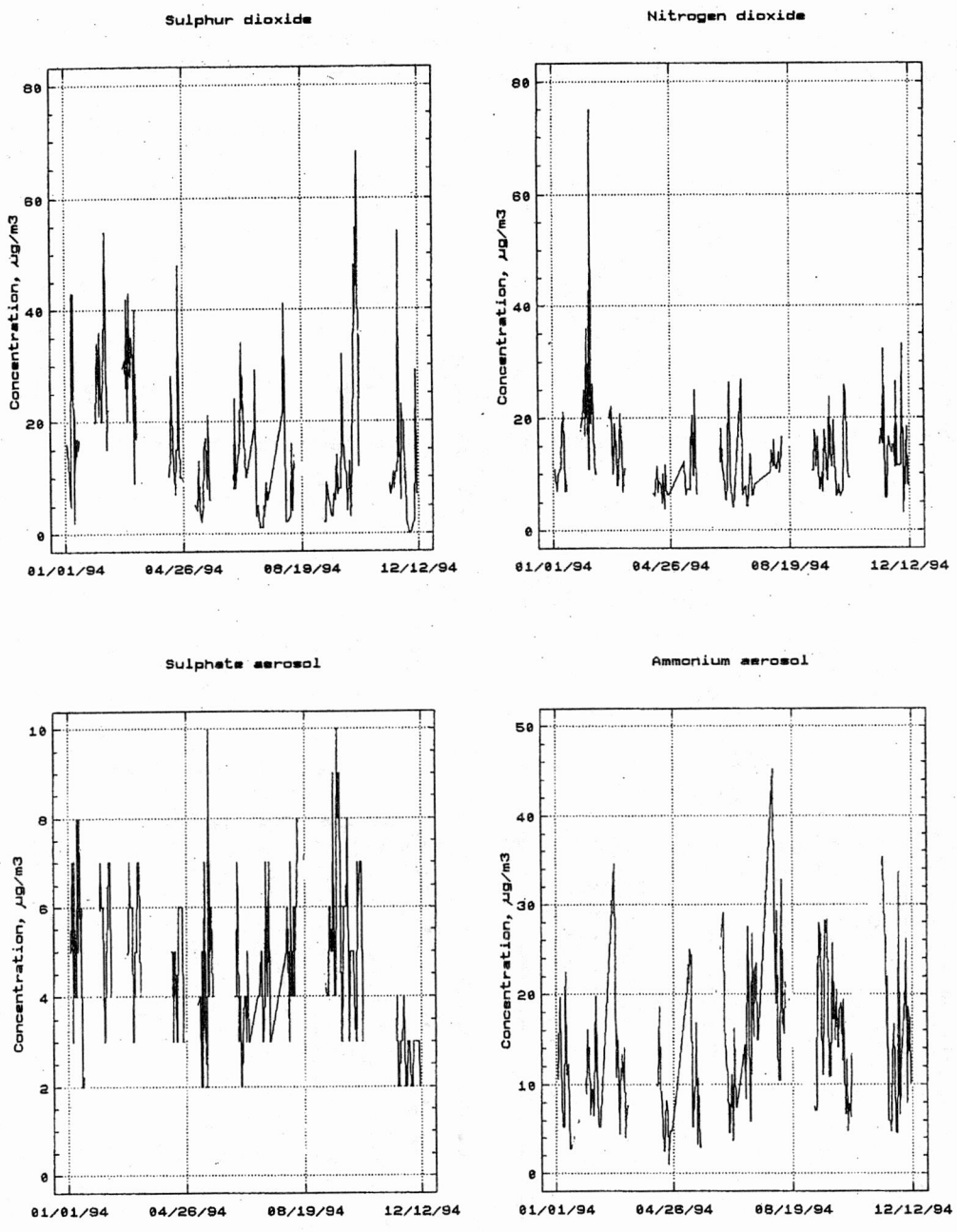

Fig. 4. Time series of pollutant concentrations for Rozdroże, 1994

If we deal with the environmental problems, the examination of the hourly profiles of the time series is informative. Two kinds of such profiles were found. The

hourly profiles represent the average hourly concentration from 12 a.m. to 12 p.m. for each month or the hourly concentration from 12 a.m. to 12 p.m. for some selected

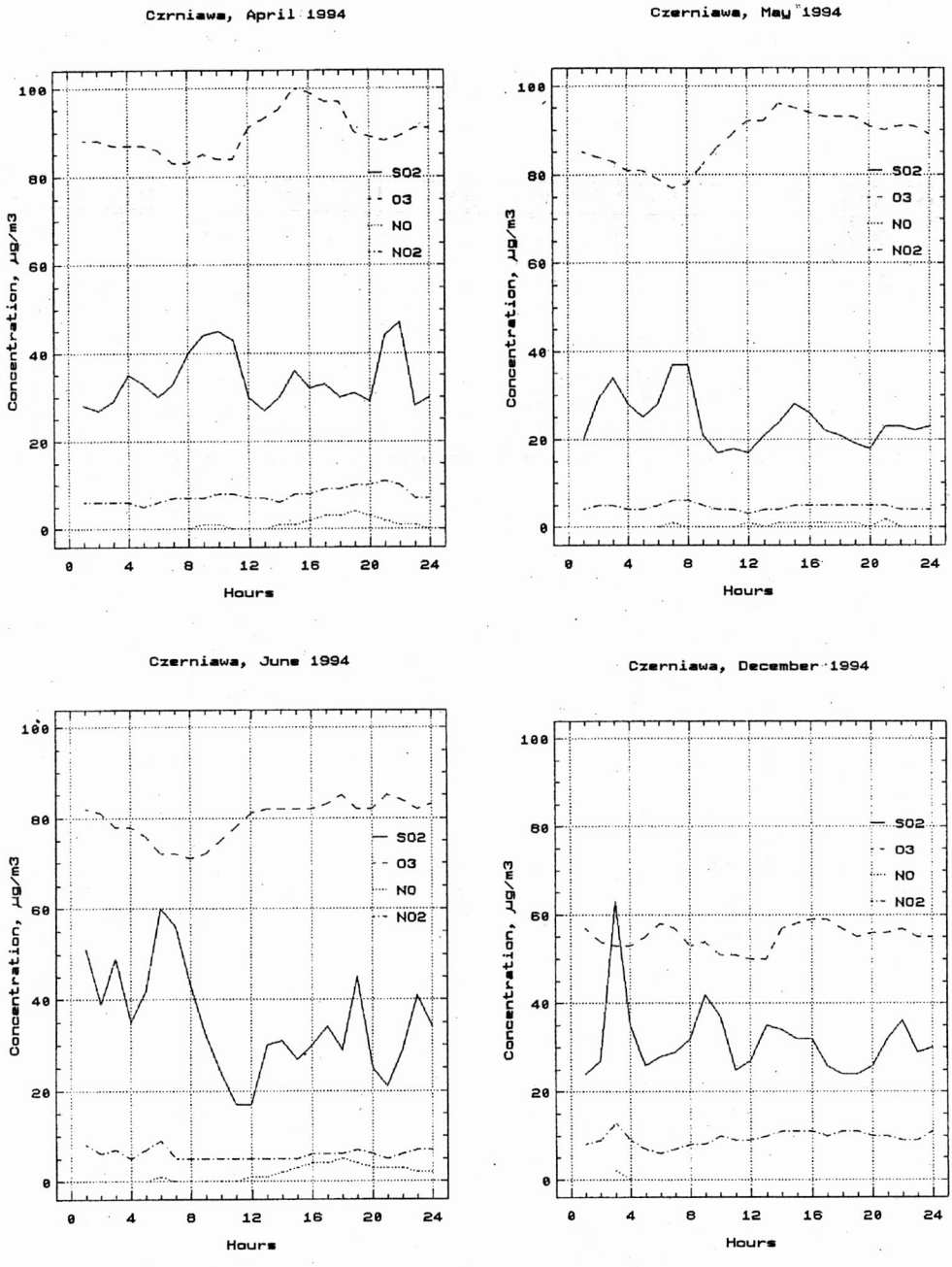

Fig. 5. Average hourly concentrations of SO_2, NO_x and O_3 for April, May, June and December, 1994, Czerniawa

days. We were able to prepare the hourly profiles for Czerniawa only because it was the only station equipped with automatic analysers. Because of the large number of missing values, the hourly profiles were prepared for three months only, i.e. for April, May and June. For a sake of comparison, the results for December were also compiled, although the samples of particular pollutants varied in number. The results are shown in figure 5. The hourly profiles for the days selected, i.e. for the episodes chosen, are described in a further part.

In the case of ozone during spring, there is observed a noticeable peak at about 4 p.m. It probably indicates an intensive ozone formation that begins at about 10 a.m. and ends after the sunset. It may also be a result of the downward flux of ozone due to turbulent mixing [13]. It should be stressed that although the ozone concentration decreases past 4 in the afternoon, it is still high during the night, and decreases slightly in the morning. In the mountains, the ozone decline is considerably slower than that in the built-up area due to the lower concentrations of NO and thus the ozone is accumulated during the night. The decrease in ozone concentration in the morning may be attributed to the intensified convection below inversion layer (if it occurs above Czerniawa) or to the rise of inversion layer which is evidenced by the increase in SO_2 and NO_2 concentrations. The lowest levels of ozone coincide with the highest levels of SO_2 and NO_x. It is difficult to discern the second peak in SO_2 concentration in the evening or late afternoon (the first was in the morning) but it is likely that the increase in SO_2 concentrations is the result of a lowering of the boundary layer at that time.

In the case of N_2O, the first peak, not very well depicted, occurs in the morning simultaneously with the increase in SO_2 and NO_2 concentrations and the decrease in ozone concentration. The second peak during warm months is between 4 p.m. and 9 p.m. Its appearance may be explained by accumulation of the products of NO_2 photolysis. The presence of NO_2 in the atmosphere causes the decrease in the ozone concentrations. Though the ozone concentrations decrease only slightly, but it results from the low concentrations of NO which approach some μg per m^3.

The nitrogen dioxide profiles are the smoothest and the flattest. The wider variation is observed for the episodes.

3.2. METEOROLOGICAL FACTORS AFFECTING POLLUTANT CONCENTRATIONS

As it has been mentioned before, the pollutant concentrations, especially those of sulphur compounds, are characterized by a large variation. Based on a detailed analysis of several case studies we attempted to define some general categories of meteorological conditions that can be responsible for the occurrence of high pollutant concentrations. The episodes of concentrations were chosen from the recorded hourly data in Czerniawa for 1994. We deal with the episode when the SO_2 concentrations in the air exceed 200 $\mu g/m^3$. Over a period of one year, 34 episodes were recorded. The results are summarised in the paper [14]. In this paper, in figures 6–11 some examples of episodes are presented to show their typical features.

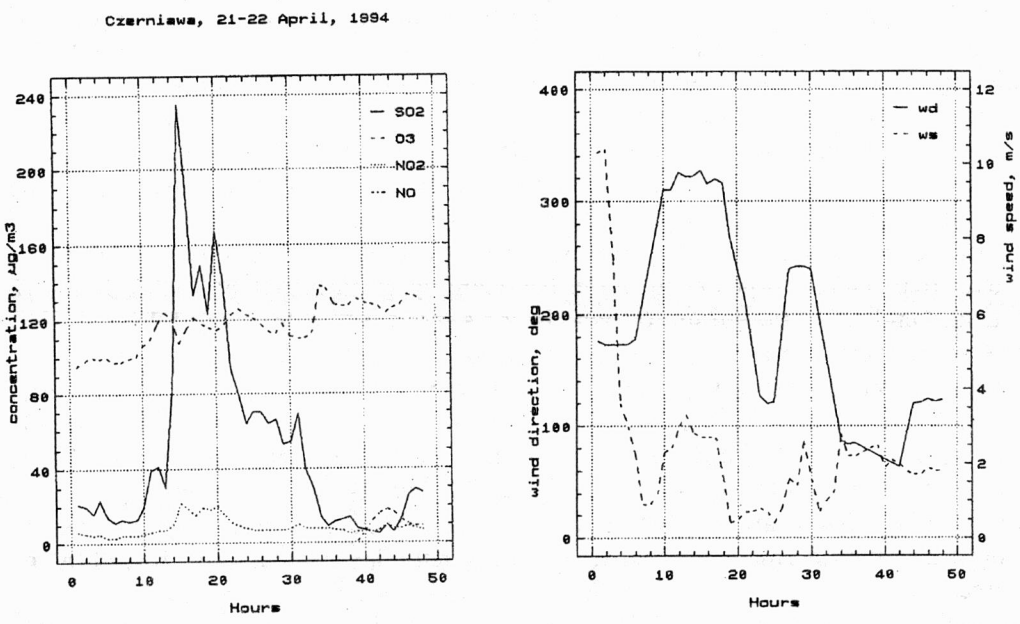

Fig. 6. Hourly variations in pollutant concentrations as well as in wind direction (wd) and wind speed (ws) for April, 21–22, 1994, Czerniawa

On 21–22 April the weather conditions in the Sudeten were affected by a high pressure system with the centre in eastern Europe. At the beginning it was observed the inflow of arctic air from the SE direction, and by day the effect of the high pressure system was reduced. The Sudeten were in the area of weak gradient pressure. The winds dropped at 4 a.m. and changed their direction from SE to NW (fig. 6); the inflow of a polar maritime air masses began. Light speed of wind allowed for accumulation of pollutants analogous to these in the stagnant air of a high pressure system. Polluted air masses reached the Sudeten about 2 p.m. At about 8 p.m. the wind changed its direction from NW to SE–S and the pollutant concentrations decreased. During the inflow of air masses from eastern directions at night, the concentration of NO increased slightly; at the same time the concentrations of SO_2 and NO_2 were very low. Such a phenomenon was typical of the eastern air circulation. The conditions on May 8–9th can also be an example (fig. 7). The weather conditions in the Sudeten were created by a strong high pressure system from Finland. These days the winds were blown from the SE and E directions. The ozone concentrations were high, while those of SO_2 and NO_2 low. The NO concentration began to increase almost at once after reaching the maximum concentration by ozone.

The association between high pressure system and the formation of photochemical oxidants has been widely recognized and discussed [15], [16]. In high pressure

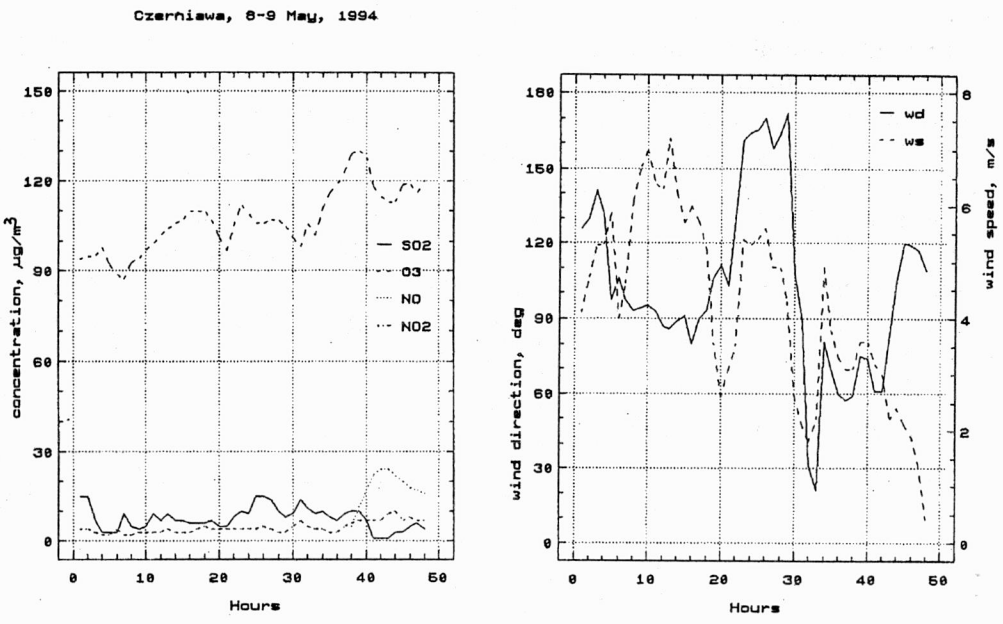

Fig. 7. Hourly variations in pollutant concentrations
as well as in wind direction (wd) and wind speed (ws) for May, 8–9, 1994, Czerniawa

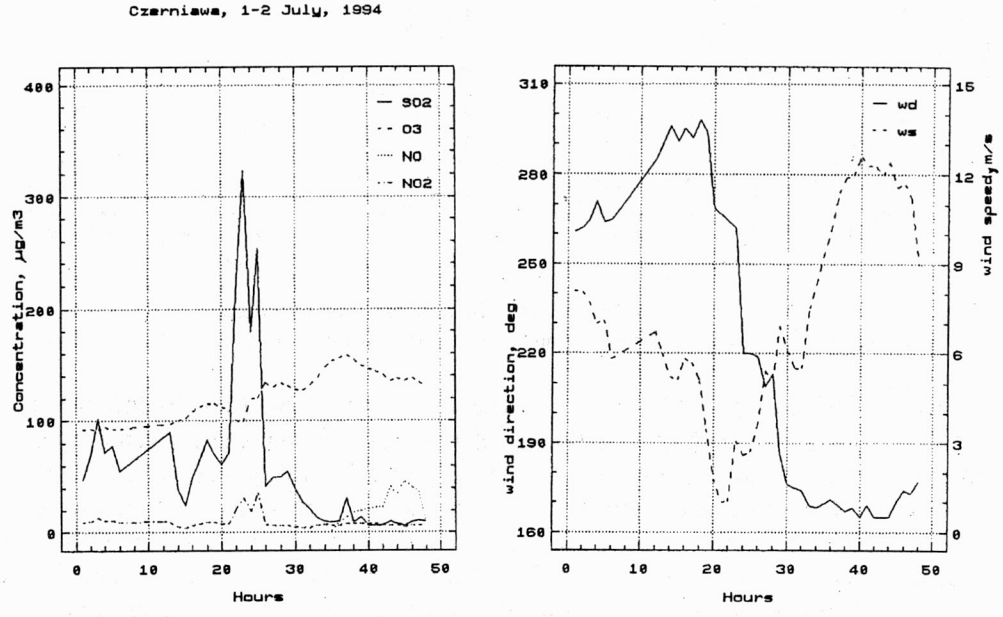

Fig. 8. Hourly variations in pollutant concentrations
as well as in wind direction (wd) and wind speed (ws) for July, 1–2, 1994, Czerniawa

areas there is often subsiding air that inhibits cloud formation which results in widespread sunshine and high temperatures. On the one hand, the light winds allow accumulation of those precursors from which ozone is formed, but on the other hand when the subsidence occurs the transport of ozone from the stratosphere takes place. Hence, high ozone concentrations are often found in high pressure areas. The peroxy radicals and ozone are the principal oxidizing agents for NO in the lower layers of atmosphere. Therefore, in the afternoon, when the photochemical processes slow down and the concentrations of oxidizing agents of NO decrease, the concentration of this gas increases. Low concentrations of SO_2 and NO_2 can be explained either by the inflow of clearer masses of air or by intense chemical transformations of these compounds.

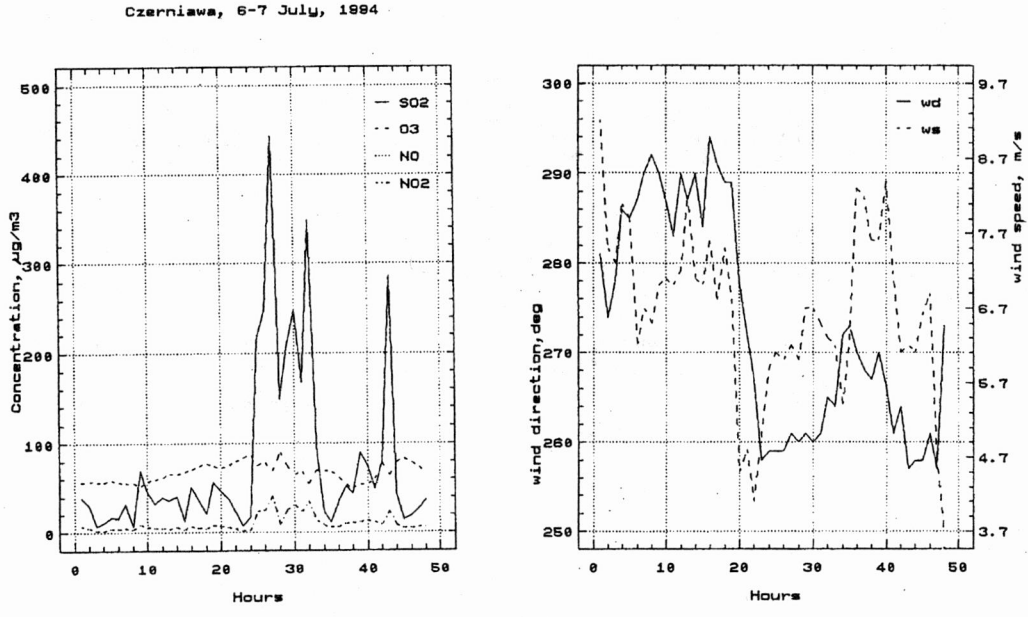

Fig. 9. Hourly variations in pollutant concentrations as well as in wind direction (wd) and wind speed (ws) for July, 6–7, 1994, Czerniawa

Two other examples of the episodes which occurred on June, 1–2nd and 6–7th are shown in figures 8 and 9. On the 1st of June the Sudeten were affected by a high pressure system with a center over Germany. This high pressure system tended to weaken and the Sudeten were in the area of weak gradient pressure. Winds were light and changed their direction from NW to SE. The air masses stagnating over Germany began to reach the mountains at about 11 p.m. The concentrations of SO_2 decreased if the wind blew from the SE direction and if its speed increased. The example given in figure 9 shows the behaviour of pollutants in the vicinity of weather

front (warm). This front finishes the period of anticyclone weather created by the high pressure system with the center over the middle Europe. A common feature of the front is the presence of ascending air currents near the front. Several authors have published descriptions of these fronts from the meteorological point of view [17], [18]. Ahead of this front the polluted masses of air which stagnate in the central Europe are transported over the mountains. Figure 10 shows another, similar example of high SO_2 concentrations associated with the weather front (warm). The front was oriented towards west-east so that the low-levels winds ahead of the front continued to accumulate pollutants as the air was moving over the regions of high emission rate. The masses of air were highly polluted which is also confirmed by very low concentrations of ozone.

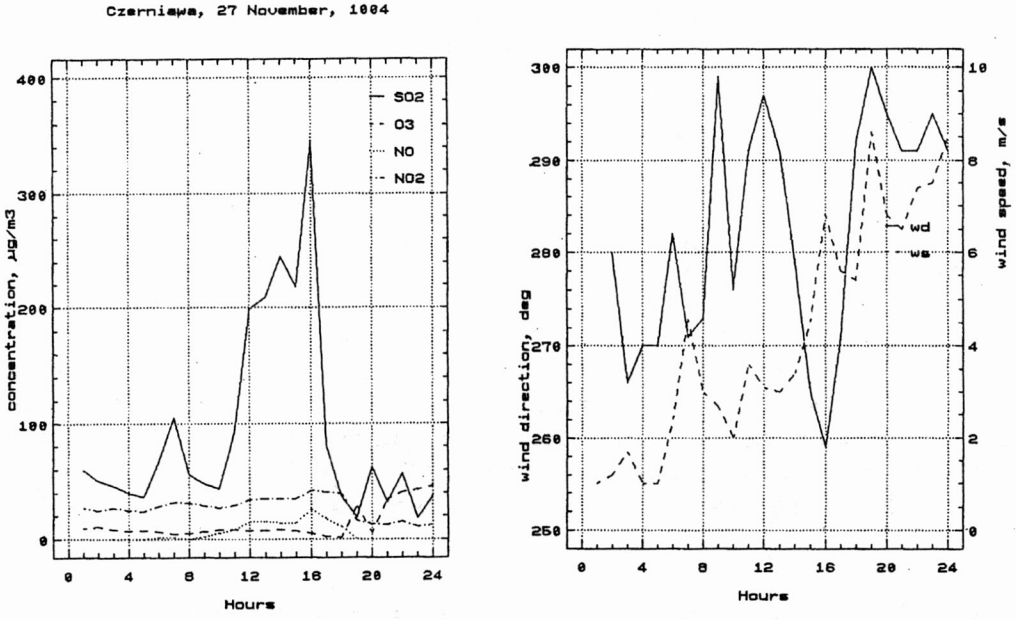

Fig. 10. Hourly variations in pollutant concentrations as well as in wind direction (wd) and wind speed (ws) for November, 27, 1994, Czerniawa

An unusual episode occurred at 3 a.m. on 10th December (figure 11). The concentrations of SO_2, NO_2 and NO increased from 35 to 775 $\mu g/m^3$, from 10 to 74 $\mu g/m^3$ and from ca. zero to 19 $\mu g/m^3$, respectively. The behaviours of ozone and NO_x in plumes produced by electric power plants are the same. The location of the ozone depression corresponds closely with the NO_x peak due to the depletion of ozone which reacts with freshly emitted NO in the plume [19]; in this case with freely advecting NO in polluted air masses. In this case, the highest hourly concentrations of SO_2 and NO_2 in 1994 were recorded. The episode lasted only 1 h (from 3 a.m. to 4 a.m.). The

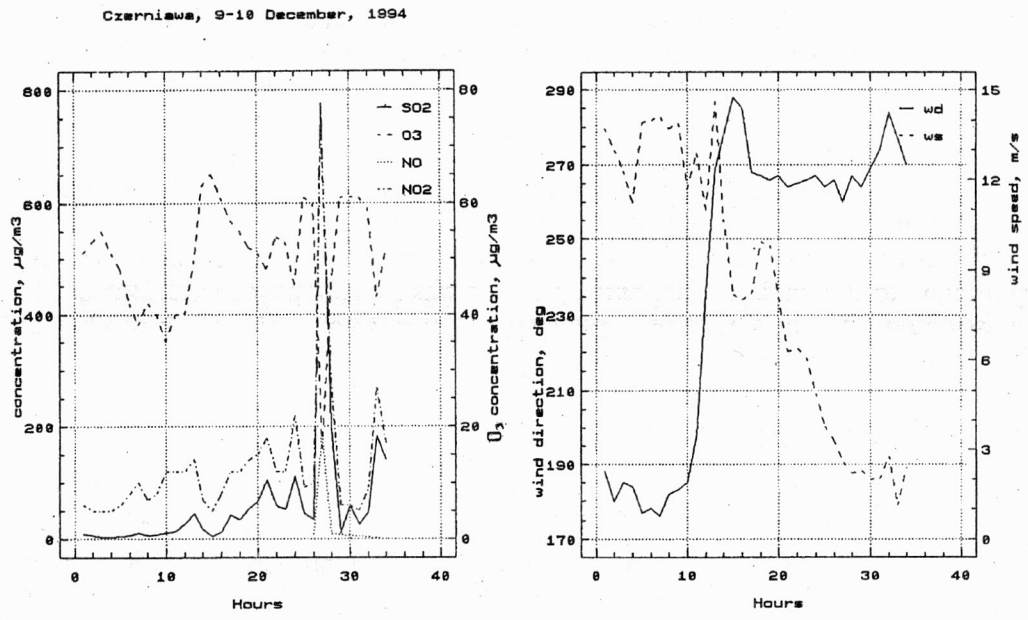

Fig. 11. Hourly variations in pollutant concentrations
as well as in wind direction (wd) and wind speed (ws) for December, 9–10, 1994, Czerniawa

simultaneous increase in the concentrations of the three pollutants characteristic of burning and the decrease in ozone concentrations (from 60 to 19 $\mu g/m^3$) must exclude the measuring error. During the episode, the Sudeten were in the polar maritime masses of air which were preceded by a warm front with rains, and then by a cold front. The winds from west dropped at 2 a.m. and remained weak during following hours. The episode occurred in the first hour after "stopping" of air masses.

3.3. RELATIONSHIPS BETWEEN BACK TRAJECTORIES AND POLLUTANT CONCENTRATIONS

In the previous part of the paper, the variability both in the pollutant concentrations and in the synoptic scale circulation described by weather fronts and pressure systems were discussed. Now the back trajectories are used for this purpose. Trajectories describe the path followed by a parcel of air over a period of time. Back trajectories follow an air parcel backward in the time from the site receptor (Mount Szrenica) to potential source region. Back trajectories have been determined for the succeeding days of two measurement periods in July and October. Some examples are presented in figures 12–14. It should be stressed that these periods were characterized by totally different meteorological conditions.

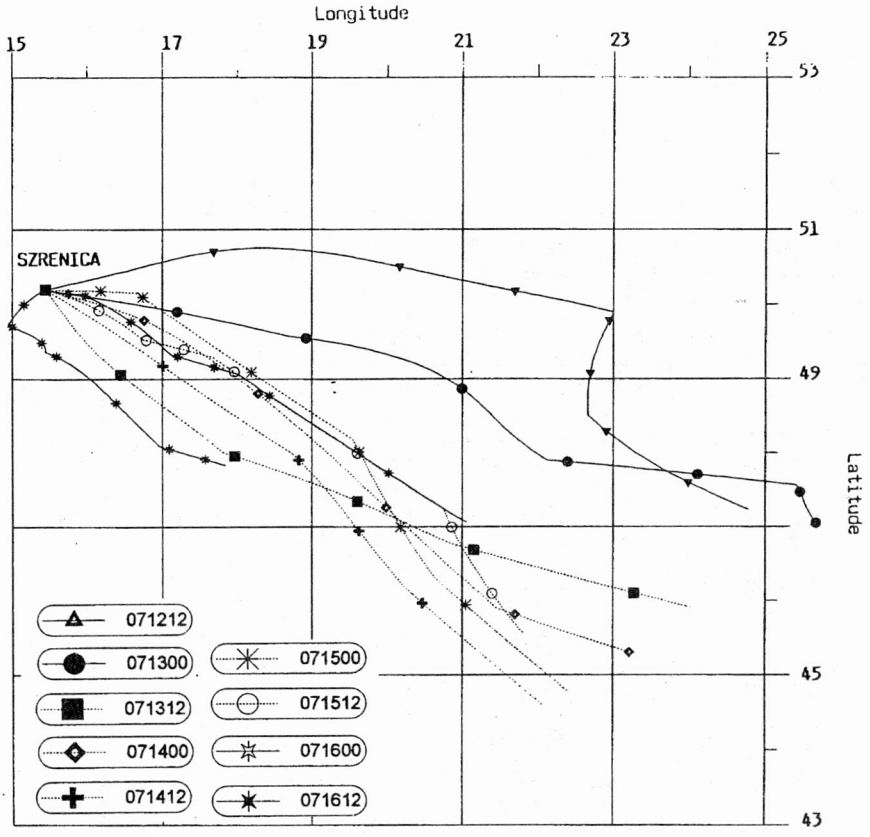

Fig. 12. 48th back trajectories for July, 12–16, 1994, Szrenica

In July, the anticyclone weather prevailed. This type of weather with exceptionally high temperatures was created by the high pressure system with the center over the south Baltic. Over this period of measurements, the inflow of the polar continental air masses was dominant. In October, the weather became unsettled. At the beginning of the cycle (19–22), the anticyclone weather prevailed, and then (23/24, 26, 28) we dealt with the cyclone weather with fronts.

In July, the concentrations of SO_2 were very low, below the detection limit, whereas those of total sulphur were high, especially on 13–15, 22–24, 27–28 July. This fact could prove that an intense conversion of SO_2 into sulphate ions took place. Unfortunately, it may not be discussed now due to lack of data on precursor species in the oxidation of SO_2. However, the high pressure systems, which dominated during that period, are often associated with subsidence, clear skies, high temperatures and stagnation of air masses which leads to high levels of radicals and ozone produced photochemically. These react with SO_2 in the atmosphere. In October, there was

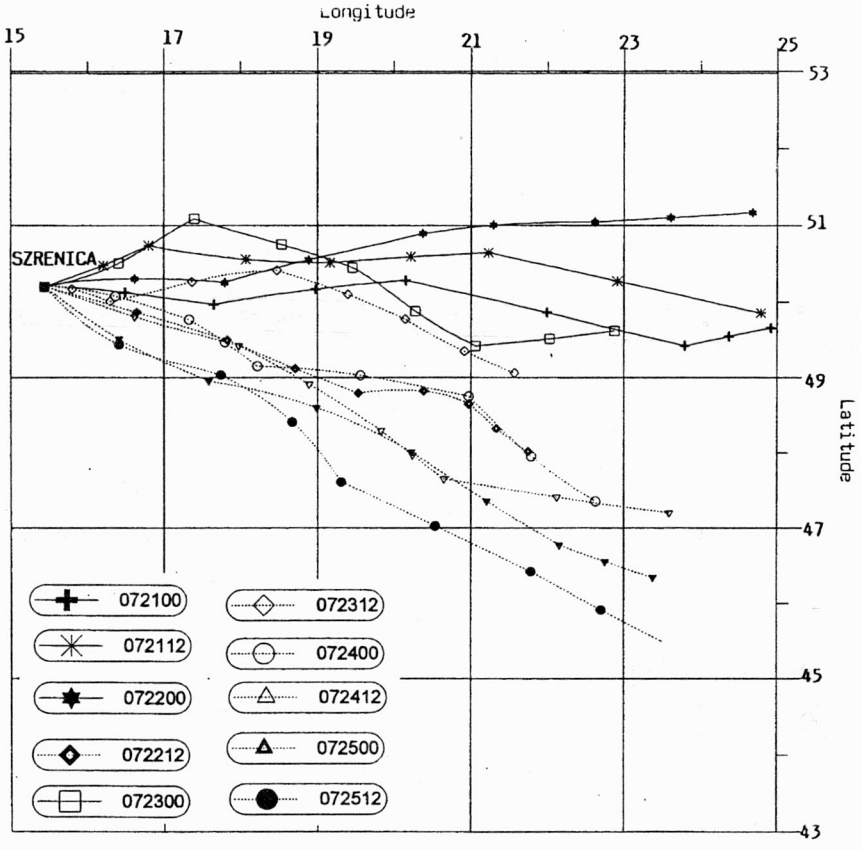

Fig. 13. 48th back trajectories for July, 21–25, 1994, Szrenica

a considerable variation in the daily concentrations of SO_2. The highest concentrations were observed in the days when the fronts moved, while the greatest differences in the concentrations of SO_2 and total sulphur were monitored in the days preceding the front movement. The similar regularity was also observed earlier [5], [12].

The extreme of total sulphur concentrations and very low SO_2 concentrations were observed more frequently during the day in advance of a frontal system approaching the Sudeten from west. At that time the transport of pollutants occurs under unstable cyclonic conditions which facilitate the incorporation of them into frontal system prior to their removal in a precipitation event.

There are known two approaches to testing the relationship between chemical observations and synoptic-scale circulation, expressed by backward trajectories. In the first approach, clean and dirty air masses are separated on the basis of the differences in pollutant concentrations characteristic of each type of trajectory. In the second approach, the statistical method of clustering allowing us to form the groups of distinct circulation patterns is used [20], [21]. In this paper, we use the first

approach because the insufficient data makes it impossible for us to employ the second approach. It will be adopted in the future.

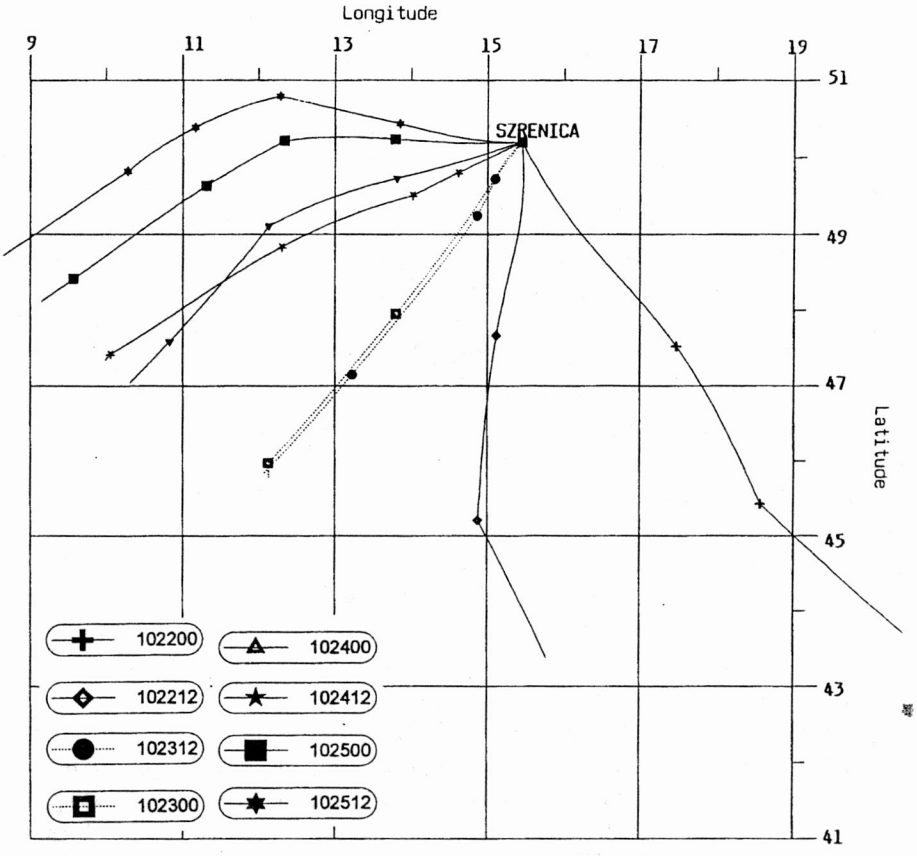

Fig. 14. 48th back trajectories for October, 22–25, 1994, Szrenica

Figures 12–14 show the dirty and clean episode back trajectories for the periods studied (the dotted line represents the dirty episodes). We selected two distinct classifications for our study. The air masses are dirty if the concentration of total sulphur is higher than 20 $\mu g/m^3$. This assumption was accepted for the sampling period in July only. In all the cases, except for July 27, the high concentrations of total sulphur were recorded at the back trajectories from the SSW–SE direction. It also happened that at the same type of back trajectories the clean air masses arrived.

If we assume that in October our data are similar with those typical of July, then it is obvious that over the period of investigations we dealt with clean air masses only. Thus, it was quite reasonable to suppose that in October the air masses were dirty if the concentration of SO_2 exceeded the value of 10 $\mu g/m^3$. Such a case was recorded only three times and was associated with the trajectories from the

W direction. A more detailed analysis of the relationship between the origin of air masses and their chemical composition will be undertaken this and next years.

4. CONCLUSIONS

The results presented here show that the simple and most important factor in day-to-day variability of pollutant levels observed at the Szrenica and Czerniawa sites is the synoptic-scale circulation of the air masses prior to their arrival. Two general categories of meteorological conditions have been identified which could account for high concentrations of pollutants in the Sudeten. The first is connected with the high pressure system with light or moderate winds. The second is associated with weather fronts. We should note that not all high pressure systems and weather fronts necessarily are responsible for high levels of pollutants. There are certain conditions under which high concentrations are likely to be found. They are as follows: the slowly moving easterly anticyclonic system or the front shifting approximately from west to east ending anticyclonic weather.

It is well known that high pressure systems create favourable conditions for the formation of ozone. As ozone concentrations, in contrast to SO_2 and NO_x behaviour, were not correlated with man-made pollutants and wind direction, thus this lack of correlation seems the most reasonable explanation for the natural source of ozone in the mountains under study. Enhanced ozone concentrations during warm months, particularly in the days of high pressure system, are likely to be the result of photodissociation of molecular oxygen in the atmosphere as well as the downward transport from the stratosphere during subsidence of air masses. There is also an indication that NO_x is essential in controlling the amount of ozone in the atmosphere over the mountains. The small peaks in the SO_2 and NO_2 hourly profiles in the morning coincided with the small depletion of ozone. The location of ozone depression corresponded closely with the NO peak due to the inflow of polluted air masses (9/10 December) or the accumulation of the products of nitrogen dioxide photolysis typical of high pressure systems (the evening of 8/9 May).

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ANALIZA EPIZODÓW STĘŻEŃ ZANIECZYSZCZEŃ POWIETRZA W SUDETACH

Przedstawiono wyniki badania stężeń zanieczyszczeń powietrza z kilku stacji pomiarowych w Sudetach, aby udokumentować ich znaczną zmienność. Na podstawie przeprowadzonej analizy zdefiniowano warunki meteorologiczne, jakie mogą sprzyjać wysokim stężeniom ozonu, związków siarki i azotu w powietrzu. Do tego celu wykorzystano m.in. dane pomiarowe z automatycznej stacji monitoringu powietrza w Czerniawie oraz trajektorie napływu mas powietrza. Stwierdzono, że pojawienie się zanieczyszczeń w powietrzu w wysokich stężeniach następuje w okresach pogody antycyklonalnej lub okresach poprzedzających przejście frontów atmosferycznych, choć nie zawsze warunki te prowadzą do epizodów stężeń.

АНАЛИЗ ЭПИЗОДОВ КОНЦЕНТРАЦИЙ ЗАГРЯЗНЕНИЙ ВОЗДУХА В СУДЕТАХ

Представлены результаты исследования концентраций загрязнений воздуха из нескольких измерительных станций в Судетах, чтобы подтвердить их изменчивость. На основе проведенного анализа определены метеорологические условия, какие могут способствовать большим концентрациям озона, соединений серы и азота в воздухе. Для этого использованы м.др. измерительные данные из автоматической станции мониторинга воздуха в Черняве, а также траектории приплыва массов воздуха. Было установлено, что появление загрязнений в воздухе в больших концентрациях наступает во время антициклональной погоды или во время, опережающее проход атмосферных фронтов, хотя не всегда эти условия ведут к эпизодам концентраций.