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CHANGES IN SULPHUR DIOXIDE CONCENTRATIONS IN THE ATMOSPHERIC AIR ASSESSED DURING SHORT-TERM MEASUREMENTS IN THE VICINITY OF OLSZTYN, POLAND

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Abstract

Sulphur dioxide is one of the principal gases responsible for the quality of atmospheric air. Air pollution, even relatively low one, is not indifferent to human health. In 2007-2009, an investigation was carried out to follow changes in the concentration of sulphur dioxide in atmospheric air, in different time intervals, depending on the air temperature and humidity. This paper draws on results of continuous measurements taken at the Station of Monitoring the Immission of Air Pollutants in Olsztyn-Kortowo. The measured concentrations of SO_2 were presented graphically as means for different time intervals. It has been demonstrated that changes in SO_2 concentrations were mainly induced by drops in temperatures, emissions and other industrial actitivities, as well as the course of the growth and development of plants. The highest concentrations appeared at noon and the lowest ones were at night. Analysis of similarities verified hourly variations in the SO_2 concentrations in air. Four distinc groups were distinguished: nocturnal (1:00-7:00 a.m.), mid-day (10:00 a.m.-3:00 p.m.), a group with two time intervals (8:00-9:00 a.m.; 4:00-7:00 p.m.) and late evening (8:00 p.m. - midnight). Considering the aerial concentration levels of this gas in monthly intervals, high similarity was observed between the early spring versus the autumn and winter seasons. Dependences between SO_2 concentrations and temperature were confirmed by analysis of the correlation coefficients and linear regression equations. Likewise, relationships between SO_2 concentrations and air humidity were analyzed in different time intervals and presented both graphically and statistically, by computing correlations and regression equations. Irrespective of the source of emission and meteorological con-

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ditions, the SO_2 concentration remained on a low level and did not exceed the permissible threshold either in the vicinity of Olsztyn or in the whole region of Warmia and Mazury.

Key words: SO_2, hourly, daily, monthly and annual cycles, temperature, humidity.

ZMIANY STĘŻENIA DWUTLENKU SIARKI W POWIETRZU ATMOSFERYCZNYM W POMIARACH KRÓTKOOKRESOWYCH W REJONIE OLSZTYNA

Abstrakt

Dwutlenek siarki jest jednym z ważniejszych gazów odpowiedzialnych, za jakość powietrza atmosferycznego. Zanieczyszczenia powietrza nawet na stosunkowo niskim poziomie nie są obojętne dla zdrowia człowieka. W latach 2007-2009 badano zmiany stężenia dwutlenku siarki w powietrzu atmosferycznym – w różnych przedziałach czasowych – w zależności od temperatury i wilgotności powietrza. W pracy wykorzystano wyniki z pomiarów ciągłych Stacji Monitoringu Imisji Zanieczyszczeń Powietrza w Olsztynie-Kortowie. Określone stężenia SO_2 przedstawiono graficznie jako średnią z różnych okresów pomiarowych. Wykazano, że na zmiany stężenia wpływały głównie spadki temperatury, działania emisyjne i gospodarcze oraz wynikające z przebiegu wegetacji roślin. Najwyższe wartości stężeń stwierdzono w godzinach południowych, najniższe w nocnych. Analiza podobieństw potwierdziła zróżnicowanie godzinowe stężenia SO_2 w powietrzu. Wydzielono wyraźnie 4 grupy: nocną (1:00-7:00), okołopołudniową (10:00-15:00), grupę w 2 przedziałach czasowych (8:00-9:00; 16:00-19:00) oraz wieczorną (20:00-24:00). Rozpatrując stężenie tego gazu w powietrzu w zależności od stężenia miesięcznego, znaczne podobieństwo wykazano w okresie wiosenno-letnim oraz jesienno-zimowym. Zależności między stężeniem SO_2 a temperaturą potwierdzono, dyskusyjnie, współczynnikami korelacji i równaniami regresji liniowej. Podobnie, zależności między stężeniem SO_2 a wilgotnością powietrza analizowano w różnym przedziale czasowym w ujęciach graficznym, a także statystycznym, wykazując związki korelacyjne i równania regresji. Niezależnie od źródeł emisji i warunków meteorologicznych, stężenie SO_2 utrzymywało się na niskim poziomie i nie przekraczało dopuszczalnych norm w rejonie Olsztyna i na całym obszarze Warmii i Mazur.

Słowa kluczowe: SO_2, cykle godzinowe, dobowe, miesięczne, roczne, temperatura, wilgotność.

INTRODUCTION

Sulphur dioxide is a widespread atmospheric air contaminant, emitted from a variety of sources, mainly from industrial plants (KOPER, SIWIK-ZIOMEK 2005). Sulphur compounds, according to KOMARNISKY et al. (2003), are among the most ubiquitous atmospheric pollutants, playing a crucial role in the chemistry of the Earth's atmosphere. For example, they strongly affect the climate and weather (CHARLSON et al. 1992, ROBOCK 2000). Large amounts of contaminants reach the troposphere from natural and anthropogenic sources (SPEIDEL et al. 2007). The volume and range of variation of actual concentrations of pollutants are strongly dependent on the current meteorological conditions, including ambient temperature (ROGALSKI, LENART 2011), and on changes occurring in the atmopshere (CZARNECKA et al. 2007, YOUNG-MIN et al. 2002). Larger quantities of sulphur dioxide in wintertime together with its weaker transformation into other sulphur compounds are responsible for a higher fluctuation in the concentrations of SO_2.

Sulphur dioxide in the air is responsible for interfering the proper functions of a healthy human (Atkinson et al. 2001, Tunnicliffe 2001). The noxious character of this compound intensifies as the SO_2 concentration in the air goes up (Ibald-Mulli et al. 2001, Sarnat 2005, Peden, Reed 2010). The atmopsheric concentration of sulphur dioxide also differentiates phytotoxic effects of the compound on plants and in soil, in which both excess and deficit of sulphur ought to be avoided (Rogalski 2006, Skwierawska et al. 2014). The mechanism through which SO_2 spreads in the air is very complex. Emission from point sources depends on such factors as climatic conditions, land relief, height and construction of chimneys, land development, etc. Thus, research and observations of SO_2 distribution must be carried out continuously and meticuously (Young-Min et al. 2002, Rogalski, Lenart 2004).

Considering the versatility of deletrious effects produced by sulphur dioxide in the environment, this study has have been undertaken to demonstrate changes in concentrations of this compound measured in several time intervals, whose momentary fluctuations resulted in specific effects.

MATERIAL AND METHODS

In 2007-2009, observations were conducted on the variation in sulphur dioxide concentrations in the air, which were based on results of measurements taken at the Station of Monitoring the Immission of Air Pollutants, managed by the Chair of Air Protection and Environment Toxicology, at the University of Warmia and Mazury in Olsztyn. The station is situated in the southern outskirts of Olsztyn, on Kortowskie Lake, at a site located at the following geographical coordinates φ = 53°27′N and λ = 20°27′E, 104 m above sea level. The station lies about 600 m from the largest housing estates of Olsztyn, a town with the population of 174,941 (www.polskainfo.pl). There are some industrial facilities in the near vicinity of the station, about 1000 m away, including a petrol station and a municipal thermal power plant. At a distance of 400 m to 600 m from the station there is a dual carriageway and some town buildings. With this localization it was possible to measure concentrations of the air contamination with sulphur dioxide in the context of spatial impact. The localization of the station and the collection methods comply with the requirements set in the Regulation of the Minister of the Environment (2002). The air sucked in at the height of 4 m above the ground level is directed into an air-conditioned contains an immission analyzers.

The concentration of SO_2 was measured by an MLU 100E fluorescence analyzer. The data were subjected to microprocessing, which generated the results as means from the measurements. The information collected at the station was transmitted at one-hour intervals to the main computer, where it was stored and processed in a CS 5 system. The results of measurements underwent statistical elaboration with Statistica v. 10, while the classifica-

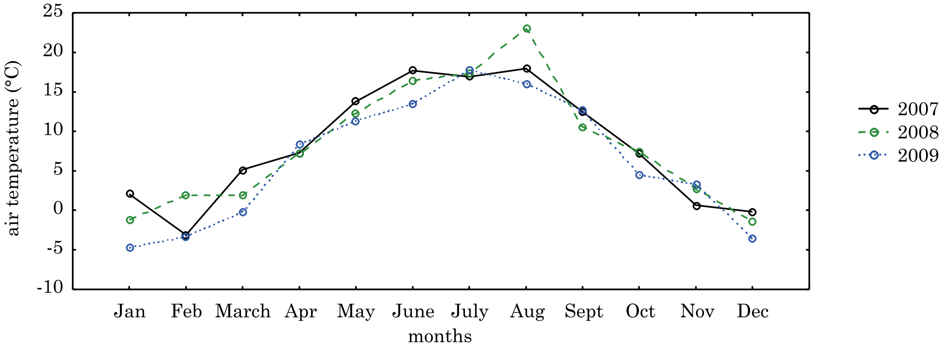


Fig. 1. Mean monthly air temperatures at the monitoring station in Olsztyn-Kortowo in 2007-2009

tion and principal component analyses were performed with the MVPS v.3.1 programme. Relationships between the air temperature and relative humidity versus concentrations of sulphur dioxide in the air were uidentified by the Spearman's rank correlations (rs) and Pearson's linear correlation at $p = 0.01$ (Gołaszewski et al. 2003). Parallel to the analysis of sulphur dioxide concentrations, the air temperature was measured using a MICROS STEP air temperature sensor, set in the range of -50°C to +80°C and at sensitivity of 0.01. The air humidity was determined with a MICROS SRH humidity sensor, composed of a plate with a variable absorbtion coefficient.

The average air temperature for the whole period of observations was 6.92°C, although it was higher in 2007 and in 2008, when it reached 8.5°C and 7.28°C, respectively, with an exceptionally warm winter in 2008 (Figure 1). In that year, the air temperature dropped below 0°C only in January and December, when it was still four-fold higher than in the analogous time in the previous year. An indicator suggesting high variability of the weather condtions in the analyzed winter seasons was the number of days with sub-zero temperatures. In 2007 and 2008, the number of such days was similar: 59 and 53, respectively. The year 2009 was the coldest, with the average temperature of 6.31°C, and freezing temperatures persisting until April. January 2009 was an extremely cold month, with the monthly mean temperature of -4.76°C and the lowest temperature of -20.7°C. There were 92 days with freezing temperatures throughout the winter season.

During the research period, the air humidity remained on a high level (above 85%), peaking to 93.7% in 2008, being slightly lower in 2007 (87.9%) and declining to 80% in 2009 (Figure 2). It should be added that the measured relative air humidity was characterized by a considerably large number of extreme results.

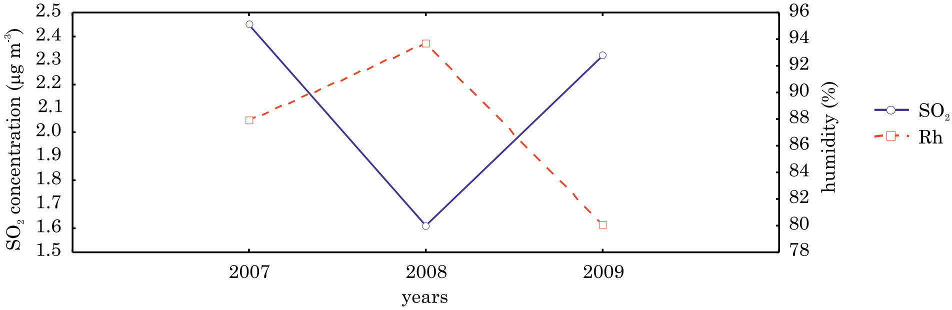


Fig. 2. Relationships between SO_2 concentration and relative humidity in 2007-2009

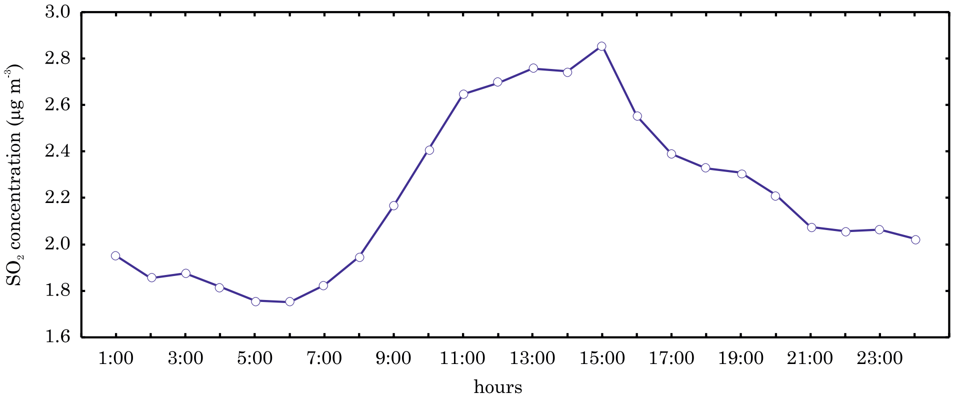


Fig. 3. Daily courses of mean one-hour concentrations of sulphur dioxide in the air in Olsztyn-Kortowo in 2007-2009

RESULTS AND DISCUSSION

Different emission sources and volumes of emitted pollutants, mostly affected by anthropogenic factors, constantly modify concentrations of sulphur dioxide in the atmospheric air. Conditions like these were also responsible for the aerial levels of SO_2 in and around Olsztyn-Kortowo (Figure 3). There were distinct differences during the day, e.g. lower and more stable concentrations at night hours, higher concentrations in the afternoon hours, with the peak around 3.00 p.m. followed by decreasing levels in the late afternoon until late at night. The meteorological conditions played an important part. Other significant factors included anthropogenic conditions such as increasing economic activity, traffic flow, heating of houses, etc. A similar course of daily fluctuations was reported by ANDRZEJEWSKA, OLSZEWSKI (2008) and PEREZ (2001). Considering daily mean values measured in individual months and years, the achieved variation of SO_2 concentrations was closer to the actual state owing to a smaller number of averaged values. The mean daily concentration of sulphur dioxide in the air was low (2.34 µg m^{-3}), but the maximum value reached 9.02 µg m^3. However, daily values in individual

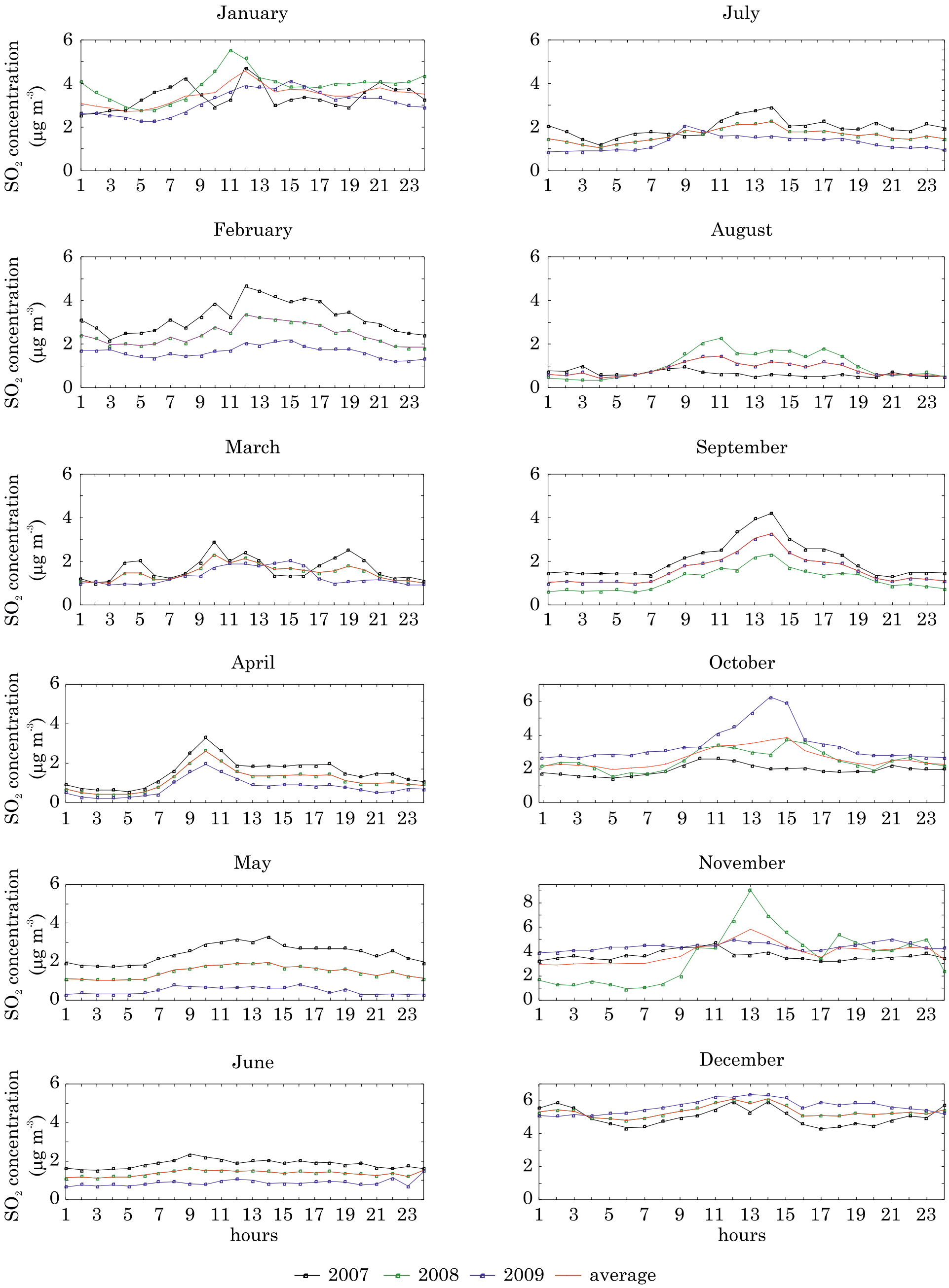


Fig. 4. Daily courses of mean one-hour SO_2 concentrations in the air in Olsztyn-Kortowo in individual months during the years 2007-2009

months were varied (Figure 4). The graphic presentations of changes in the SO_2 air concentrations, shown hour by hour, imply considerable shifts in daily sets. In 2007, larger dispersion of concentrations appeared in winter months, especially in January, February and March, with the peak concentrations between 9 a.m. and 1 p.m., and in December during night and afternoon hours. Similar variations occurred in 2008, with the maximum values determined in November and December. In the coldest year, 2009, the highest concentrations of SO_2 in air began to be noted as early as October and continued until December, mostly in afternoons. The course of daily fluctuations in SO_2 concentrations, presented by total values gathered during the

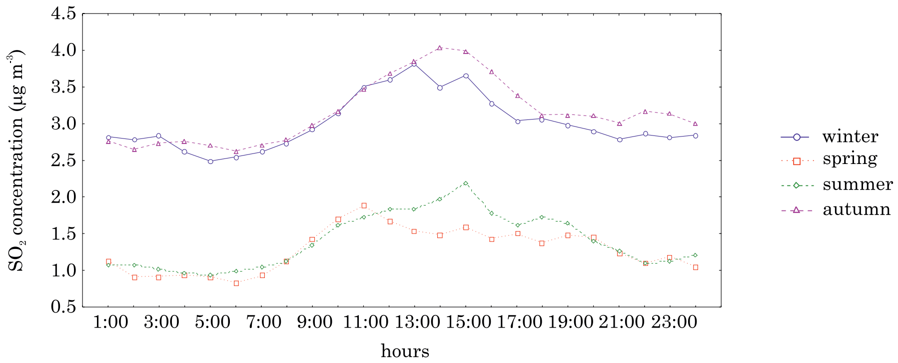


Fig. 5. Averaged courses of mean one-hour concentrations of sulphur dioxide in seasons of the year during the years in Olsztyn-Kortowo

research, support our previous observations that SO_2 concentrations fluctuated more in 2007 than in 2008-2009. The summarized data give us a complete picture of greater variability of sulphur dioxide concentrations in winter than in summer.

When analyzing SO_2 concentrations in the seasons of the year (Figure 5), it was demonstrated that daily fluctuations in winter and autumn were characterized by an approximately same course of curves, and that those curves assumed a shape similar to the ones achieved in summer and in spring. Presumably, an increase in the sulphur dioxide concentrations was mostly due to emission from the thermal heating of houses in residential estates near the monitoring station (suburbs like Dajtki, Słoneczny Stok). Similar results were reported by KOLBARCZYK, KOLBARCZYK (2007), who conducted observations in several localities in north-western Poland. There, the highest SO_2 concentrations were recorded between 11.00 a.m. and 4.00 p.m., with an evident peak noticed at 3.00 p.m. Thus, daily fluctuation cycles in SO_2 concentrations were largely shaped by a set of meteorological conditions. Road transport is thought to be another important source of atmospheric pollutants (LIPING, YAPING 2005). The highest SO_2 concentrations in the summer between 3 and 4.00 p.m. is attributed to a more intensive traffic flow, because most of the through roads near Olsztyn cross the city *(www.gospodarka.olsztyn.eu)*. The concentration of SO_2 measured at weekends was lower than on weekdays, and showed a stronger relationship with the vehicle traffic. Similar results were obtained by VANDAELE et al. (2002), MIESZALSKI (1981) showed that under daylight the diffusion of sulphur into plant leaves is more rapid than in the dark. Plants involved in phytoremediation contributed heavily to the daily changes in SO_2 concentrations in the atmosphere. Decreased sulphur emmission into the air may lead to the depletion of this element from soil, thus depressing crop yields and bioavailability of other nutrients, which invariably deteriorates the quality of yields (JANKOWSKI et al. 2008, 2014). In turn, the form of sulphate or elemental sulphur applied as fertilizer modifies the content of total carbon in soil (MAJCHEREK et al. 2013).

It is worth mentioning that the decomposition of organic matter under anaerobic conditions leads to the generation of hydrogen sulphide. The released hydrogen sulphide, which is an unstable compound, is quickly broken down to the final product, i.e. SO_2 (SYED et al. 2006, TAMMINEN, ANDERSEN 2007). Another factor which could have induced differences in daily concentrations of sulphur dioxide in the air is the woodland surrounding the monitoring station (an urban park and a forest). Deciduous trees dominant in those tree communities are able to absorb more SO_2 than conifers.

A study by ROGALSKI and LENART (2011) proved that the power of dependence between the concentration of SO_2 and the air temperature (T) was expressed by the highest values of correlation coefficients (rs), which were statistically significant only in the months:

February: $SO_2 = 1.6811 - 0.1980 \times T$ $rs = -0.4606$

April: $SO_2 = 0.3633 + 0.2672 \times T$ $rs = +0.4274$

June: $SO_2 = 0.2091 + 0.0726 \times T$ $rs = +0.3869$

In February, growing temperature caused a decrease in the concentration of SO_2 in the air. In April and June, the effect was opposite, most probably due to interactions with other meteorological elements or an influx of contaminated air.

The analysis of similarities in SO_2 concentrations in the atmospheric air in Olsztyn-Kortowo, in 2007-2009, shown in hourly intervals, revealed four distinct groups of concentrations related to different times of the day (Figure 6). The night and early morning hours (from 1.00 to 7.00 a.m.) reached a 97% similarity level. Another branch consisted of morning hours (8.00-9.00 a.m.) and afternoon hours (4.00-7.00 p.m.). In this group, the following afternoon hours: 4.00-5.00 p.m. and 6.00-7.00 p.m., were most similar (97%). The third cluster consisted of late evening and night hours (8.00 p.m. to midnight), where close similarity in terms of sulphur dioxide concentrations was noticed between 10.00 and 11.00 p.m. (98%). The fourth group was made up of the midday hours (10.00 a.m. to 3.00 p.m.). Ranges of SO_2concentrations in the atmospheric air in those hours overlapped each other to a large extent, thus verifying their high similarity.

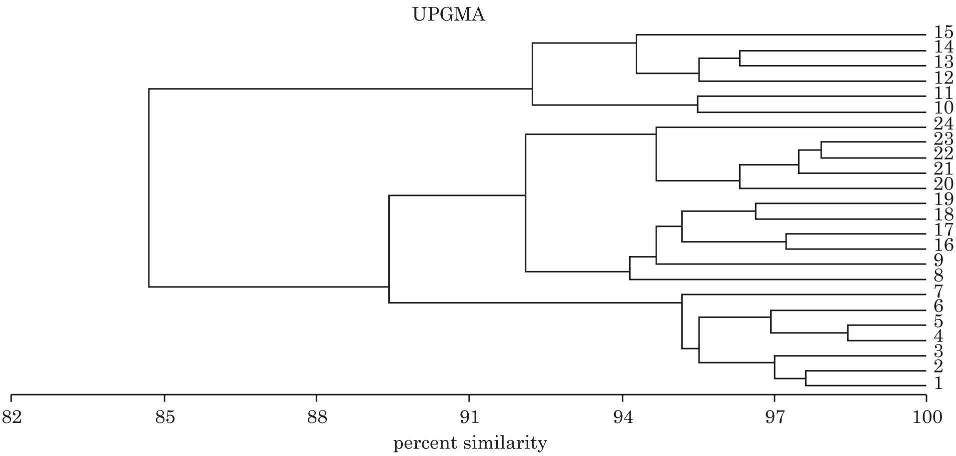


Fig. 6. An hourly dendogram of the similarity of SO_2 concentrations in the air in Olsztyn-Kortowo in 2007-2009

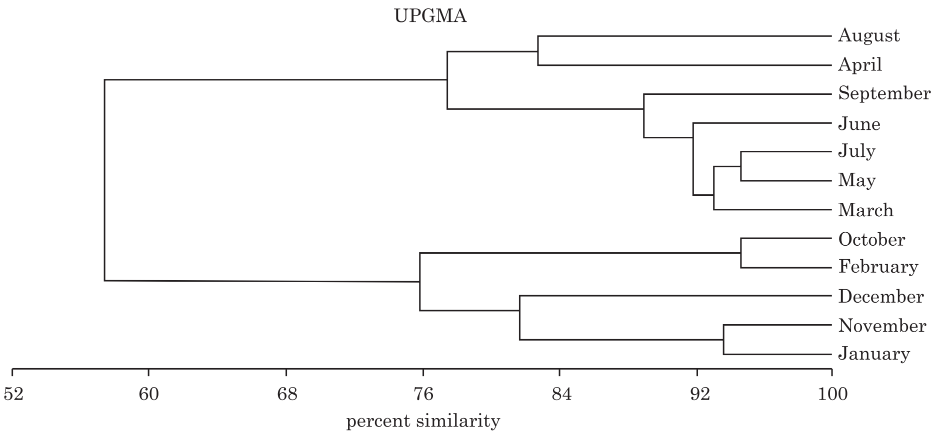


Fig. 7. Dendogram of monthly similarities in SO_2 concentrations in the atmsopheric air in Olsztyn-Kortowo in 2007-2009

The analysis of clusters based on the per cent similarity of SO_2 concentrations in a monthly cycle (Figure 7) showed a considerably distinct character of these values in the atmopsheric air in Olsztyn-Kortowo, in 2007-2009. The autumn and winter months were distincly different from the spring and summer ones. In the former group of months, the highest similarity (95%) with respect to the atmospheric SO_2 concentrations in Olsztyn-Kortowo was detected for October and February. The SO_2 concentrations similar to the ones in that group were demonstrated in December and in November and January, which additionally formed a sub-group of their own, with a similarity level of 94%. Another group (95%) consisted of the SO_2 concentrations recorded in July and May, to which the concentrations of atmospheric sul-

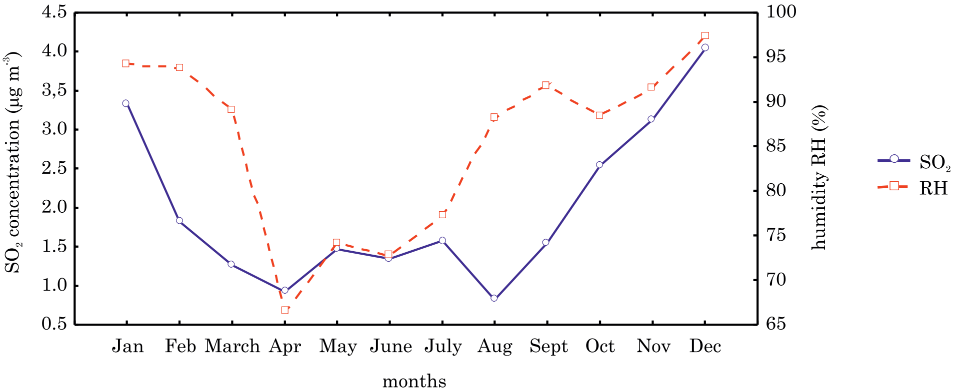


Fig. 8. Relationships between SO_2 concentration and humidity in individual months measured in Olsztyn-Kortowo, in 2007-2009

phur dioxide were most similar in March (93%) and June (92%). The least similar level of SO_2 in the atmospheric air was characteristic for the group composed of August and April (77%), versus the group made up of March, May, July, June and September.

The annual course of air humidity, in monthly intervals, against the backdrop of SO_2 concentrations, is illustrated in Figure 8. From April to July, the air humidity was not shown to have any impact on modifications in the concentration of SO_2 in air. This might have been caused by higher temperatures prevailing in that season or else the occurrence of some other ele-

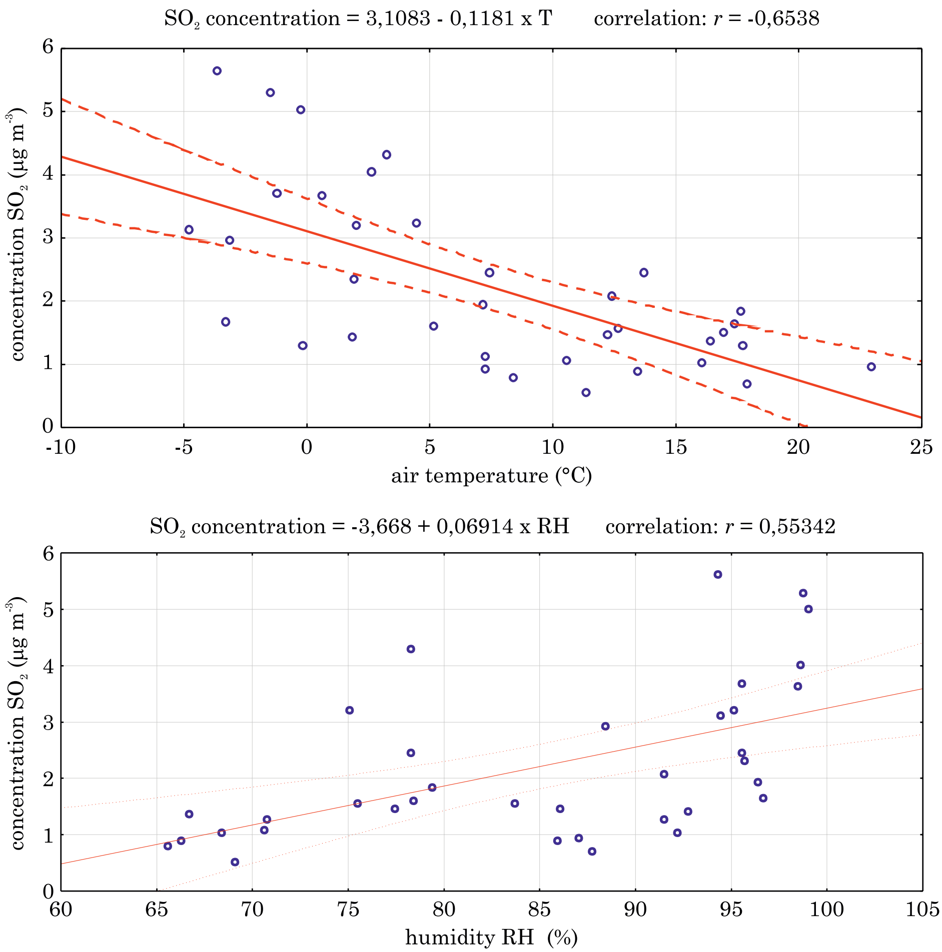
Table 1

Relationships between SO_2 concentrations and humidity from January 2007 to December 2009 in Olsztyn-Kortowo

Test period	Spearman's rank-order correlation		Regression equation
	r_S	p	
January	-0.228313	0.050405	SO_2= 10.6493– 0.0782 x RH
February	-0.227116	0.066670	SO_2= 6.7037– 0.0498 x RH
March	-0.072198	0.583557	SO_2= 0.58 + 0.0088 x RH
April	-0.462549	0.000257	SO_2= 2.8621– 0.0289 x RH
May	0.241314	0.022718	SO_2= 0.3887 + 0.0151 x RH
June	0.075987	0.476571	SO_2= 1.1159 + 0.0034 x RH
July	0.096585	0.403365	SO_2= 1.3966 + 0.0028 x RH
August	-0.111528	0.380278	SO_2= 84.4197 + 0.0007 x RH
September	-0.072223	0.498738	SO_2= 2.1251 - 0.0066 x RH
October	-0.298917	0.006714	SO_2= 6.3823 - 0.0429 x RH
November	-0.346826	0.001412	SO_2= 5.2664 - 0.0215 x RH
December	-0.045240	0.684658	SO_2= 10.4166 - 0.0626 x RH

RH – relative humidity of the air (%)

ments stabilizing the characteristics of air. In order to describe dependences between SO_2 concentrations and air humidity, the results of measurements were submitted to analysis of correlation rs and regression equations were developed (Table 1). Our analysis of the issuing data implies that the highest values of the correlation coefficient, which were statistically significant at the same time, occurred in the following months: April, October and November. The correlation and regression variations may have been due to less efficient SO_2 oxygenation and, consequently, a higher level of this gas in the air. A similar observation was reported by Kleniewska (2004). Besides, the aggregated effect of meteorological elements could have led to some synergistic interactions between the air temperature and humidity. Regardless of the above, the air flowing to Olsztyn might have demostrated elevated sulphur dioxide contamination. Based on the regression equations, it can be estimated that



Rys. 9. Relationships between SO_2 concentration versus the air temperature and humidity in individual months measured in Olsztyn-Kortowo, in 2007-2009

the amount of SO_2 in the atmospheric air depends on the known meteorological elements. The graphic depiction of values (Figure 9), including the dispersion of data, defined the character of relationships between the variables. According to IGRASHI et al. (2006), to assess the influence of humidity on the SO_2 concentration in air one would have to further analyze relationships of other gases and ions present in the atmosphere with air humidity.

CONCLUSIONS

1. The variation in the concentrations of sulphur dioxide in the air is the function of aggregated dependences between the abiotic variables, emission processes caused by anthropogenic activity and – to a large extent – the growth and development of plants. Regardless of the above relationships, the effect of air temperature and humidity on SO_2 concentrations in the air has been proven.

2. Daily concentrations of sulphur dioxide in the air showed some coincidence with periodic drops in ambient temperature and with rush hours causing more intensive traffic in Olsztyn.

3. The highest values of sulphur dioxide concentrations occured at noon, lower in the morning and afternoon hours and the lowest ones were at night.

4. In autumn, relative humidity revealed a stronger influence on the concentration of sulphur dioxide in the air. In the other seasons, the correlation of these variables was weak or very weak.

5. The nature of the dependence, identified by the regression equation, between the concentration of sulphur dioxide and temperature was that of a reversely proportional relationship, while that between the SO_2 concentration and air humidity was a proportional one.

6. Irrespective of the source of emission and influx of air masses, the concentration of sulphur dioxide in and around Olsztyn-Kortowo remained on a low level, never exceeding the thresholds, not even duirng extremely high fluctuations.

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