BIOMETEOROLOGICAL DETERMINANTS OF THE TROPOSPHERIC OZONE CONCENTRATION IN THE SUBURBAN CONDITIONS OF WROCLAW, POLAND

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Abstract

Our aim was to determine relationships between the tropospheric ozone concentration and the nitrogen dioxide concentration, selected meteorological factors and radiation-effective temperature in the suburban area of the city of Wroclaw. Hourly data from 2006-2011 used in the study included concentrations of tropospheric ozone, nitrogen dioxide concentrations, air temperature, air humidity, wind velocity and total irradiance of the sun. Radiation-effective temperature (TRE) was determined on the basis of the four meteorological elements. Concentrations of ozone in southwest Poland show annual, seasonal and daily variation. The highest values occur in the warmest time of the year and day, while the lowest ones are noted in the cold season of the year, in October and in the morning. The variation of ozone concentrations was found to be shaped by changes in nitrogen dioxide concentrations, meteorological elements and the bioclimatic indicator. Ozone and nitrogen dioxide were mutually most strongly correlated in the winter season. In the warm season of the year, from April to October, the ozone concentration was mainly shaped by air temperature, solar radiation, wind velocity and TRE (positive effects), and by relative humidity of the air (a negative influence). The multiple regression equation best described the ozone concentration variation in May. Increased concentrations of ozone were found to appear during thermal stress, expressed as a sensation of warmth/heat, but could also occur under thermal comfort conditions.

Keywords: evaluation of human warmth/hot feeling, radiation-effective temperature, southwest Poland.

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INTRODUCTION

Tropospheric ozone is of interest to many researchers because it is harmful to human health (HAŁUSZKA et al. 1998, LI et al. 2011), while being widespread in many European countries (REID et al. 2012, EEA 2014, MADRIGANO et al. 2015, SOER 2015). An important research issue concerns the circumstances in which photochemical episodes occur. In order to explain their occurrence, it is useful to specify the variation of ozone concentrations on different spatial and temporal scales. The ozone concentration variation depends on both the concentration of NO_2 and variable weather conditions (GIRGZDIENE, SAKALAUSKIENE 2007, PAWLAK, JAROSŁAWSKI 2015). High ozone concentrations usually occur in the warm season of the year, over a wide area, often spreading across state borders (STRUŻEWSKA et al. 2012). The main stimulus is a conducive pattern of meteorological conditions.

Meteorological elements directly influence the course of chemical processes in the atmosphere. For instance, a relationship has been confirmed between solar radiation and photo-dissociation reactions of NO_2 and O_3 (JACOBSON 2005), as well as the cloud cover and concentration of ozone in the cloud and its surroundings (LIU et al. 1997). The process of ozone production and particularly the reaction speed are directly affected by temperature. There is an indirect effect as well, because biogenic emission decreases at low temperature (SILLMAN, SAMSON 1995). At higher temperatures, there is an elevated emission of VOC, especially isoprene, which plays an important role in ozone production. An increasing ozone concentration is clearly observed in the Mediterranean region (PAWLAK, JAROSŁAWSKI 2015).

The water vapour content, too, has a significant impact on ozone production. Namely, a lower vapour content leads to the reduction of sources of radicals that control the ozone generation (STRUŻEWSKA et al. 2012). Biogenic hydrocarbons (e.g. isoprene – C_5H_8) may contribute significantly to the formation of ozone in urban areas. These compounds, issued mainly by deciduous trees, are moved by advective transport, and their contribution to the production of ozone reaches approx. 25% during the most severe emissions (CHAMEIDES et al. 1988). The variation of ozone concentrations is rarely applied as an indicator in biometeorological studies (KALBARCZYK et al. 2015). Relationships between indicators of thermal discomfort, PM_{10} and general quality of air were studied by PALIATSOS and NASTOS (1999), PAPANASTASIOU et al. (2009) and MAVRAKIS et al. (2012).

The aim of the present work was to determine relationships between tropospheric ozone concentrations and nitrogen dioxide imission, selected meteorological elements as well as radiation-effective temperature and certain characteristics of the biometeorological conditions in the suburban zone of Wroclaw.

MATERIAL AND METHODS

The study was based on hourly data from January to December, collected at a station of the Lower Silesian Regional Inspectorate of Environment during six consecutive years: 2006-2011. The imission station was situated at the Agricultural Experimental Station, located in Bartnicza Street, in the eastern outskirts of Wroclaw (λ = 17°08′28″ E, φ = 51°06′58″ N, hs = 120 m above sea level).

The input information consisted of gas air pollution values as well as data about the meteorological elements: tropospheric ozone concentration (O_3, µg m^{-3}), nitrogen dioxide concentration (NO_2, µg m^{-3}), air temperature (Ta, °C), relative humidity (Rh, %), wind velocity (v, m s^{-1}) and total irradiance of the sun (Kglob, W m^{-2}). Missing Kglob values were retrieved from Wroclaw--Karlowice Station (λ = 17°01′46″ E, φ = 51°07′46″ N, hs = 121 m above sea level), which is the nearest to the experimental site and, simultaneously, best at reflecting solar conditions of the suburban zone of Wroclaw.

Based on the four meteorological elements: Ta, Rh, Kglob and v reduced to the level of 2 m above the sea, radiation-effective temperature (TRE, °C) was determined, using an application included in BioKlima 2.6., which had been developed and made available for the study by Błażejczyk (2004).

TRE temperature describes the heat sensation by a human staying in an open area exposed to the sun. The evaluation of thermal sensation felt by people by the TRE indicator was achieved on the Mikhailov scale (Błażejczyk 2004). For each TRE temperature class, the mean biometeorological and aerosanitary conditions were estimated, and their coincidence with hourly values of O_3 and NO_2 was assessed and calculated.

The overall effects of biometeorological conditions on the O_3 concentration were identified through a stepwise regression analysis. Initially, all the variables, considered for each of the calendar month of a year, regardless of whether their impact on the concentration was statistically significant or negligible in a single regression, were submitted to analysis. Eventually, a set of variables was identified that had played the most important role in shaping the volume of the current imission of the analyzed gas pollution. Parameters of the regression function were determined using the least squares method. The overlap of the functions with the empirical data was assessed with an adjusted coefficient of determination (R^2) and the one describing differences between the standard deviation of the dependent variable and the error of the regression equation ($|SD-Sy|$), while the presence of autocorrelation of random components was evaluated with the Durbin-Watson test. Partial correlation analysis was used (r^2) in order to determine the contribution of each of the selected factors in predicting ozone concentrations.

RESULTS AND DISCUSSION

During the six-year period, concentrations of both ozone and NO_2 varied in successive months and years (Figure 1). The highest average monthly concentrations of ozone and NO_2 occurred in 2006 (for O_3 in July, for NO_2 in January), and the lowest ones were in 2009 (O_3 in December) and 2010 (NO_2 in May). An almost reverse relationship between concentrations of ozone and nitrogen dioxide was confirmed. Ozone concentrations were at approximately the same high level for a third of the year, from April to July.
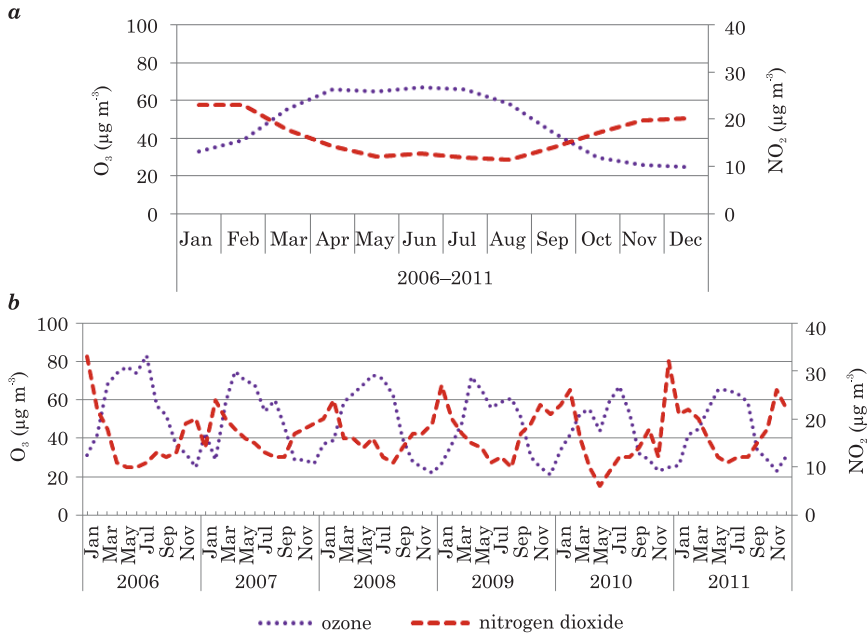

Fig. 1. Average monthly concentrations of tropospheric ozone and nitrogen dioxide in multiple years (*a*) and in following years (*b*). The years 2006-2011

The highest ozone concentrations occurred in urban areas of Warsaw in July, and in the suburban and rural areas of central Poland in May and April (ROZBICKA, ROZBICKI 2014, PAWLAK, JAROSŁAWSKI 2015). Simultaneously, the lowest average NO_2 concentrations occurred in May through August, i.e. with a one-month delay compared to the ozone maximum concentration. The lowest ozone concentrations occurred at the end of the year, and the highest concentrations of NO_2 were recorded at the beginning of the year (Figure 1*a*).

Relationships between the ozone and nitrogen dioxide concentrations were confirmed statistically for all months, seasons and mean annual values. The strongest relationship was demonstrated for concentrations in the late winter and early spring, and the weakest dependence was attested to during the summer (Table 1). This may implicate the dominant role of the interac-

Table 1

Evaluation of linear relationships between concentration of tropospheric ozone and nitrogen dioxide in 2006-2011

Indicator	Month												Year	Season			
	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec		XII-II	III-V	VI-VIII	IX-XI
r	-0.56	-0.68	-0.65	-0.59	-0.55	-0.58	-0.49	-0.52	-0.49	-0.55	-0.53	-0.61	-0.55	-0.55	-0.59	-0.52	-0.54
t	-42.3	-57.1	-53.6	-47.1	-42.5	-37.9	-36.6	-39.8	-34.8	-39.7	-37.9	-42.3	-140.5	-66.7	-80.6	-63.5	-66.6
p	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01
SD O_3	17.8	20.4	24.9	31.1	33.6	35.3	36.3	30.0	26.4	19.3	17.1	15.0	31.0	19.0	30.5	34.1	22.4
SD NO_2	16.2	13.3	11.9	12.2	10.2	10.2	8.2	8.4	9.4	9.6	11.1	9.5	11.8	13.6	11.7	8.8	10.3

r – Spearman's rank correlation coefficient, t – t-Student test, p – significance level, SD – standard deviation (μg m^{-3}), XII-II – winter, III-V – spring, VI-VIII – summer, IX-XI – autumn

tion between anthropogenic origin emissions and transport of pollutants in a cool season, which is important for shaping the level of ozone (GODŁOWSKA 2004, GODŁOWSKA, TOMASZEWSKA 2006). During the spring and summer seasons, relationships between the ozone and nitrogen dioxide concentrations may be weaker due to the imission of hydrocarbons (PAWLAK, JAROSŁAWSKI 2015). The suburban location of the measuring station allowed us to identify just this one factor. Negative correlations between ozone and NO_2 concentrations have also been found by MARĆ et al. (2014), ROZBICKA et al. (2014). Different results and positive correlations have been reported in India by SAINI et al. (2008).

For the time period considered, reverse relationships between concentrations of ozone and NO_2, although less strong, were also visible in the average daily concentrations by months (Figure 2). The lowest ozone concentrations
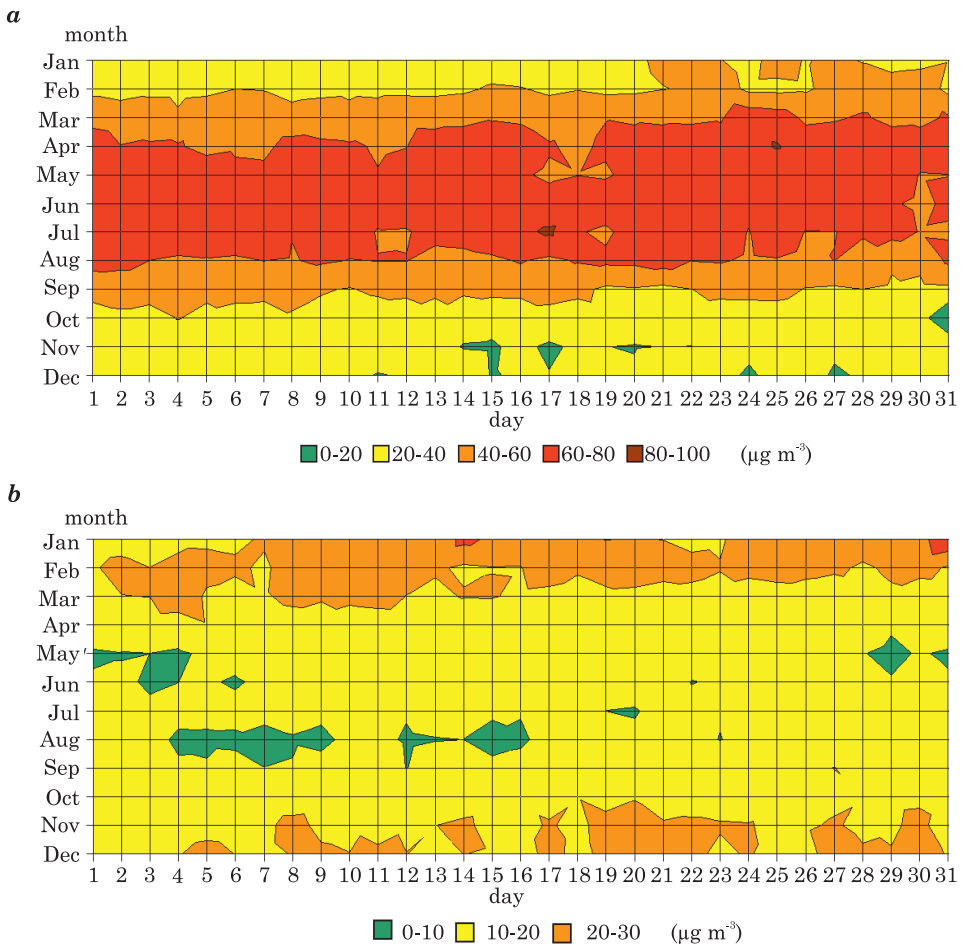
a

b

Fig. 2. The time distribution of average daily concentrations of tropospheric ozone (*a*) and nitrogen dioxide (*b*) by months. The years 2006-2011

could be observed most often in the second decade of November. The average daily concentrations of ozone exceeded the value of 80 µg m^{-3} only twice. Regarding average daily concentrations of NO$_2$, its increased values (>20 µg m^{-3}) occurred subsequently to instances of high ozone concentrations. High concentrations of NO$_2$ are associated mainly with the cool season. The highest values of NO$_2$, >80 µg m^{-3} were also recorded only twice, in January. In turn, the lowest values occurred in the May-August period.

Hourly concentrations of ozone and NO$_2$ took an almost opposite course. The highest average hourly concentrations of ozone were between the hours of 10:00 to 18:00 during the months of April to July (Figure 3). During the same period, and even until August, there were the lowest NO$_2$ concentra-
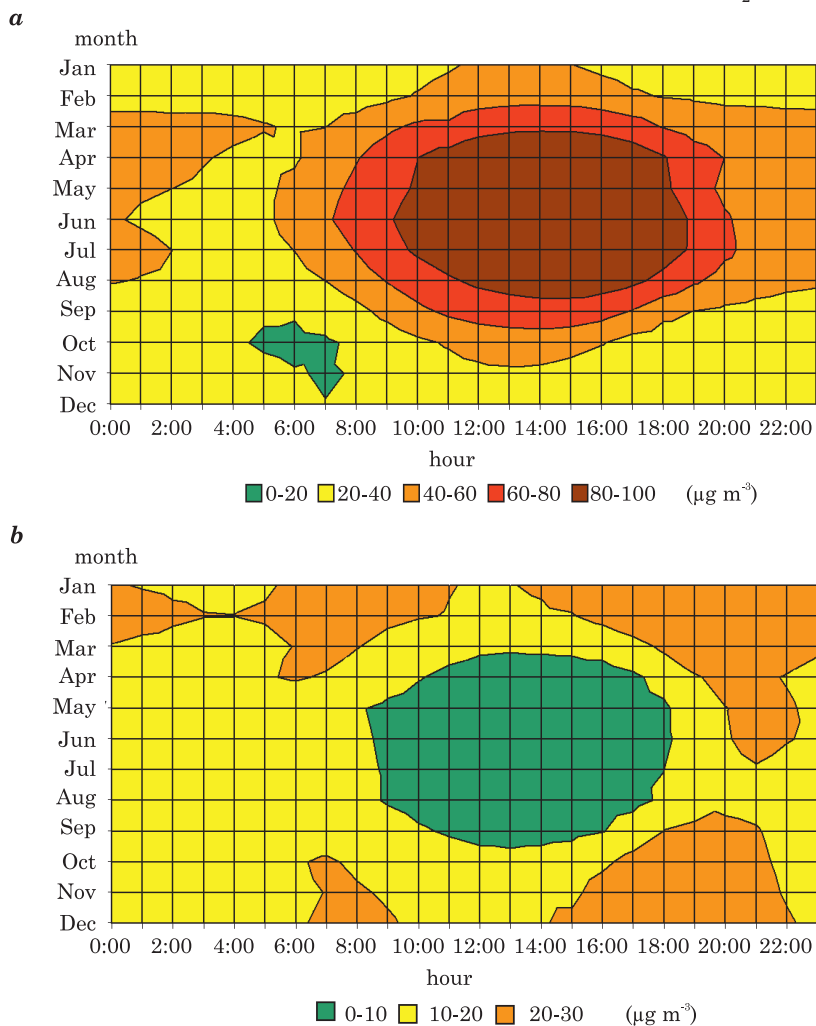

Fig. 3. The time distribution of average hourly concentrations of tropospheric ozone (*a*) and nitrogen dioxide (*b*) by months. The years 2006-2011

tions. The lowest average daily ozone concentrations occurred in October and morning hours. The highest daily concentrations of NO_2 occurred in the evening and morning. The variation of ozone concentrations documented here, with a pronounced seasonal and diurnal maximum and minimum, is typical of ozone concentrations in Europe, but has been observed in other parts of the world (GIRGZDIENE, SAKALAUSKIENE 2007, SAINI et al. 2008, CHEN et al. 2014).

Our analysis of the incidence of hourly ozone concentrations within the adopted ranges confirmed their marked seasonality. Although the whole-year average is most often in the range 20-60 μg m^{-3}, concentrations in the seasonal pattern are within the range of 40-60 μg m^{-3} in the spring and summer, while concentrations in the range of 0-40 μg m^{-3} prevail in the autumn and winter (Figure 4). In individual months, the low concentrations (0-20 μg m^{-3}) were most often (over 42% of the days) observed in November and December.
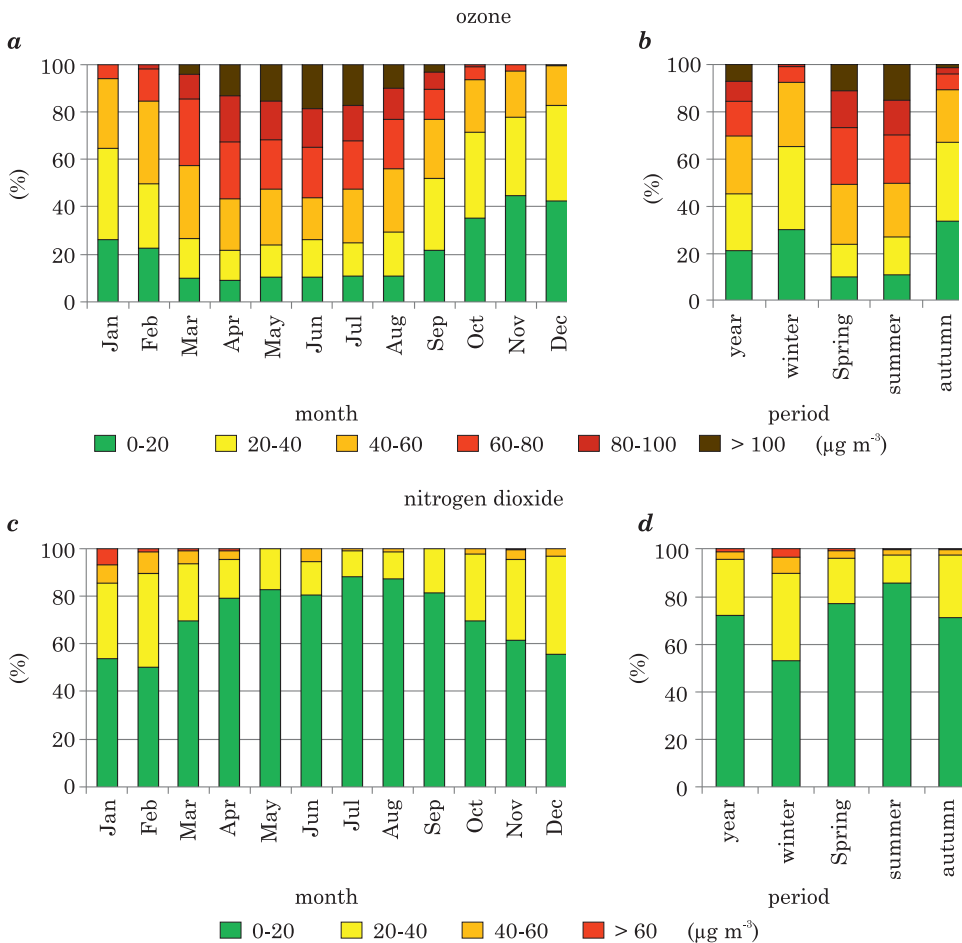

Fig. 4. The incidence of hourly values for concentrations of tropospheric ozone and nitrogen dioxide by months (*a*, *c*), seasons and years (*b*, *d*). The years 2006-2011

The incidence of higher concentrations gradually increased in the consecutive, warmer months. As for nitrogen dioxide, concentrations within the lowest range of 0-20 µg m^{-3} prevail throughout the year. The highest concentrations can be observed most frequently in winter and in January.

The variation of ozone concentrations depends on both the concentration of NO$_2$ and variable weather conditions (GIRGZDIENE, SAKALAUSKIENE 2007, PAWLAK, JAROSŁAWSKI 2015). Of the weather elements affecting the ozone concentration variation, solar radiation and air temperature are attributed the most significant role (LI et al. 2011, MARĆ et al. 2014). The relationship between ozone concentration and high air temperature has been confirmed in many studies (ROZBICKA et al. 2014, PAWLAK, JAROSŁAWSKI 2015). In this work, positive correlations were determined between the concentration of ozone and air temperature, radiation, radiation-effective temperature (TRE) value from April to October, and the velocity of wind, while a negative one was confirmed between ozone and relative humidity of the air. The strongest relationship occurred between ozone and air humidity.

The strongest correlations between ozone and air temperature were confirmed only in two summer months, whereas in winter ozone correlated most strongly with winds. Low temperature usually leads to distinctly lower biogenic emission (SILLMAN, SAMSON 1995). Water vapour content has a significant impact on the process of ozone production, i.e. less vapour causes a reduction in the sources of radicals that control the production (STRUŻEWSKA et al. 2012). Moreover, the strong correlation between the concentration of ozone and humidity can be explained by the comprehensive nature of this meteorological element (ROGALSKI et al. 2014).

Positive correlation between ozone concentration and solar radiation was confirmed in earlier studies in north-western Poland (KALBARCZYK, KALBARCZYK 2009). Radiation and air temperature have an effect on the course of the photo-dissociation reaction of NO$_2$ and O$_3$ (JACOBSON 2005). The high level of ozone concentration, found to persist in south-western Poland for up to a third of the year, may be the result of a specific impact of the distribution of sums of radiation in Poland, with the maximum in May, while the air temperature is usually the highest in July (LORENC 2005). The positive role of wind arises from the importance of airborne transport of contaminants to suburban areas and further away from cities.

Among independent variables affecting the variability of ozone concentrations, particularly important seem to be relative air humidity (Rh), wind velocity (v) and NO$_2$, which significantly interacted in all months of the year. However, different factors had the greatest impact on ozone concentrations in individual months. The strongest dependence of ozone concentrations is confirmed to be on NO$_2$ (negative) in winter and early spring, Rh (negative) in spring and autumn, and air temperature (Ta, positive) in summer. The regression equation best described the variation in ozone concentrations in May. PAWLAK and JAROSŁAWSKI (2015) cliamed that the inclusion of meteoro-

logical elements made it possible to explain 75% of this variation in summer and 70% in winter. Lɪ et al. (2011) reported 40-70% of the variation explained. In respect of NO_2, PAWLAK and JAROSŁAWSKI (2015) concluded that this component allowed them to explain 89% of the variability of ozone concentrations.

The inclusion of the biometeorological index into the equation did not improve the accuracy of the description of the ozone concentration variation. The radiation-effective temperature and NO_2 considered in conjunction (Table 2) best described the variability in ozone concentrations in spring, but worst in

Table 2

Evaluation of the dependence of tropospheric ozone concentrations on the TRE indicator and NO_2 concentration in individual months

| Month | Regression equation | | | N | F | \|SD-Sy\| (μg m^{-3}) | Sy (μg m^{-3}) | R^2 |
| | intercept | regression coefficient | | | | | | |
		TRE (°C)	NO_2 (μg m^{-3})					
Jan	46.78	-0.26 (-0.12***)	-0.79 (-0.58***)	2940	767.9	4.0	13.8	0.343
Feb	61.67	n.s.	-1.02 (-0.67***)	3190	1345.1	5.8	14.6	0.457
Mar	76.24	-0.75 (-0.22***)	-1.31 (-0.701***)	1921	1051.2	9.4	15.5	0.522
Apr	76.80	2.303 (0.503***)	-1.45 (-0.57***)	3971	3053.6	11.6	19.5	0.6059
May	56.94	2.88 (0.58***)	-1.501 (-0.48***)	3039	2351.3	13.2	20.4	0.6074
Jun	48.11	3.038 (0.55***)	-1.606 (-0.45***)	2389	1643.0	13.0	22.3	0.579
Jul	38.42	3.24 (0.55***)	-1.71 (-0.38***)	4104	2437.1	11.8	24.5	0.542
Aug	32.82	3.15 (0.57***)	-1.49 (-0.42***)	4051	2905.2	10.8	19.2	0.589
Sep	38.16	2.67 (0.59***)	-1.25 (-0.44***)	3696	2745.3	9.7	16.7	0.598
Oct	47.46	0.96 (0.302***)	-1.11 (-0.55***)	3510	1158.1	4.2	15.1	0.397
Nov	42.31	0.22 (0.085***)	-0.809 (-0.53***)	3512	655.9	2.8	14.3	0.272
Dec	44.59	0.18 (0.079***)	-0.98 (-0.62***)	2881	916.7	3.3	11.7	0.389

In brackets – partial correlation coefficient, N – number of degrees of freedom, F – F-Snedecora test, \|SD-Sy\| – difference between a standard deviation of a dependent variable and a standard error of equation estimation (μg m^{-3}), Sy – standard error of equation estimation (μg m^{-3}), R^2 – determination coefficient, *** significant at $p \leq 0.01$ n.s. – linear and curvilinear relationship non-significant at $p \leq 0.1$, TRE – radiative-and-effective temperature (°C)

November. Nitrogen dioxide was more strongly correlated with ozone than TRE during the longer, cooler part of the year. The strongest negative correlation was confirmed between the ozone values and NO_2 in March. The TRE index and ozone concentration were most strongly positively correlated in the warm half-year, particularly in September. TRE was more strongly correlated with ozone than NO_2 in spring and summer.

In a comprehensive approach, when many elements were considered concomitantly (Table 3), no relationships were verified between ozone concen-

Table 3

Evaluation of the dependence of the concentration of tropospheric ozone and weather elements on the concentration of NO_2 in individual months

Month	Regression equation						N	F	|SD-Sy| ($\mu g \cdot m^{-3}$)	Sy ($\mu g \cdot m^{-3}$)	R^2
	intercept	Regression coefficient									
		Ta (°C)	Rh (%)	RAD (W m^{-2})	v (m s^{-1})	NO_2 ($\mu g \cdot m^{-3}$)					
Jan	64.39	n.s.	-0.33 (-0.23***)	0.043 (0.14***)	5.16 (0.28***)	0.019 (-0.41***)	2940	841.8	6.1	11.7	0.533
Feb	73.11	n.s.	-0.26 (-0.18***)	0.034 (0.16***)	3.34 (0.18***)	-0.804 (-0.52***)	3190	1021.8	7.2	13.2	0.561
Mar	96.63	0.13 (0.028*)	-0.52 (-0.43***)	0.0098 (0.068***)	4.87 (0.25***)	-0.9001 (-0.48***)	1921	1295.0	14.2	10.7	0.771
Apr	107.92	1.26 (0.22***)	-0.66 (-0.50***)	n.s.	3.18 (0.11***)	-0.99 (-0.39***)	3971	4156.6	17.4	13.7	0.8072
May	97.71	1.76 (0.28***)	-0.74 (-0.53***)	n.s.	4.99 (0.14***)	-0.87*** (-0.28***)	3039	4481.4	21.2	12.4	0.855
Jun	96.73	1.807 (0.27***)	-0.76 (-0.51***)	n.s.	3.93 (0.11***)	-0.96 (-0.27***)	2389	2557.0	20.3	15.0	0.8106
Jul	5.71	4.14 (0.61***)	-0.17 (-0.11***)	n.s.	5.86 (0.15***)	-0.95 (-0.21***)	4104	2424.9	16.5	19.8	0.7025
Aug	23.94	3.61 (0.56***)	-0.28 (-019***)	n.s.	3.86 (0.11***)	-0.91 (-0.26***)	4051	3068.1	15.0	14.9	0.751
Sep	59.39	2.41 (0.44***)	-0.52 (-0.37***)	n.s.	3.14 (0.11***)	-0.63 (-0.22***)	3696	3843.6	14.8	11.6	0.8061
Oct	70.091	0.57 (0.14***)	-0.49 (-0.38***)	0.0098 (0.061***)	5.58 (0.27***)	-0.62 (-0.38***)	3510	1417.8	8.1	11.3	0.668
Nov	99.69	0.16 (0.047***)	-0.78 (-0.49***)	0.0089 (0.035***)	4.13 (0.27***)	-0.55 (-0.37***)	3512	1283.3	7.1	10.0	0.646
Dec	93.43	n.s.	-0.69 (-0.43***)	n.s.	4.22 (0.24***)	-0.012 (-0.43***)	2881	1828.0	6.2	8.8	0.655

Explanations see Table 2

trations and radiation in the warm half-year, or between ozone concentrations and air temperature in the winter months. The lack of statistical confirmation of the correlation between ozone concentrations and total radiation in the warm half-year probably results from a stronger interaction of its de-

rivatives, i.e. air temperature and air humidity. With respect to NO_2 concentrations, they were most strongly correlated with the velocity of wind, whose increase significantly reduced the level of air pollution.

Interesting results were obtained by juxtaposing the values of tropospheric ozone concentrations, nitrogen dioxide concentrations and selected meteorological elements in the various classes of the human feel of heat (Figure 5). Conditions evaluated as comfortable in months from different seasons of the year differed not only in the average air temperature, amount
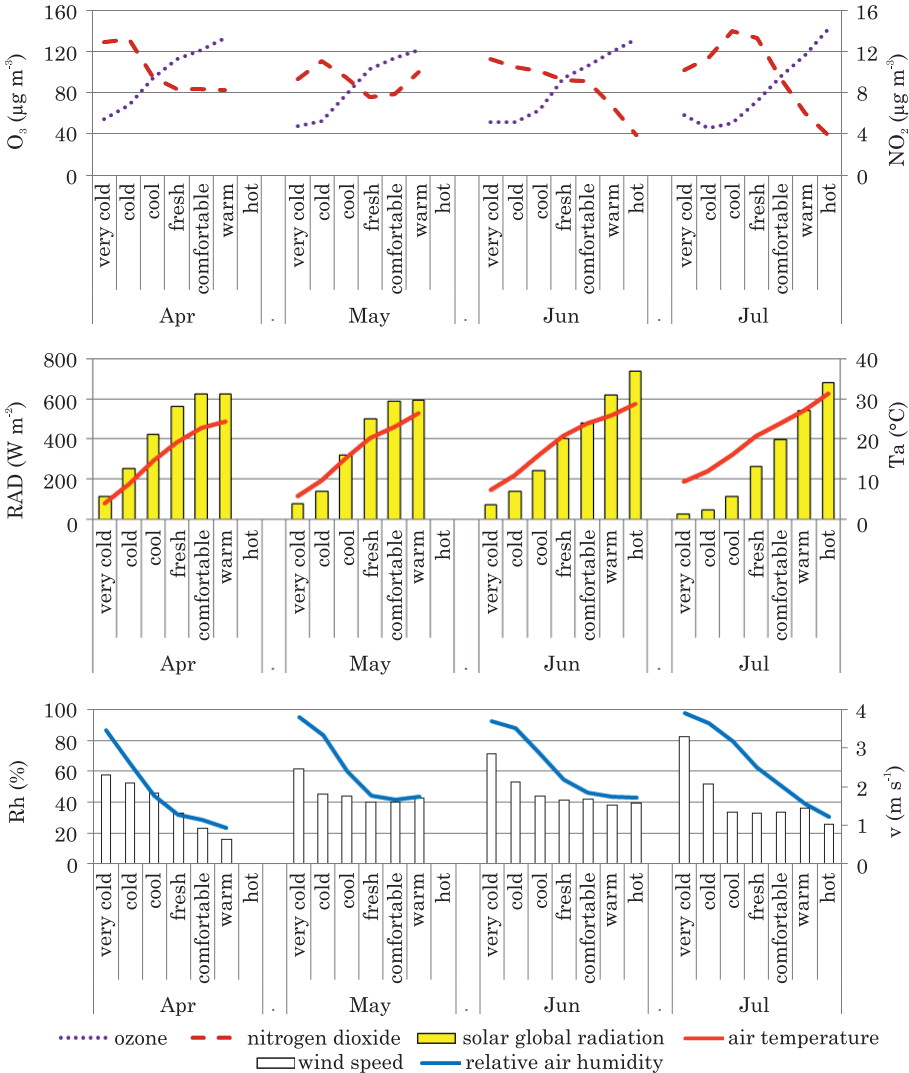

Fig. 5. Concentrations of tropospheric ozone and nitrogen dioxide and the values of meteorological elements (Ta, Rh, RAD, v) in the various classes of the human feel of heat according to the Mikhailov scale in selected months. The years 2006-2011
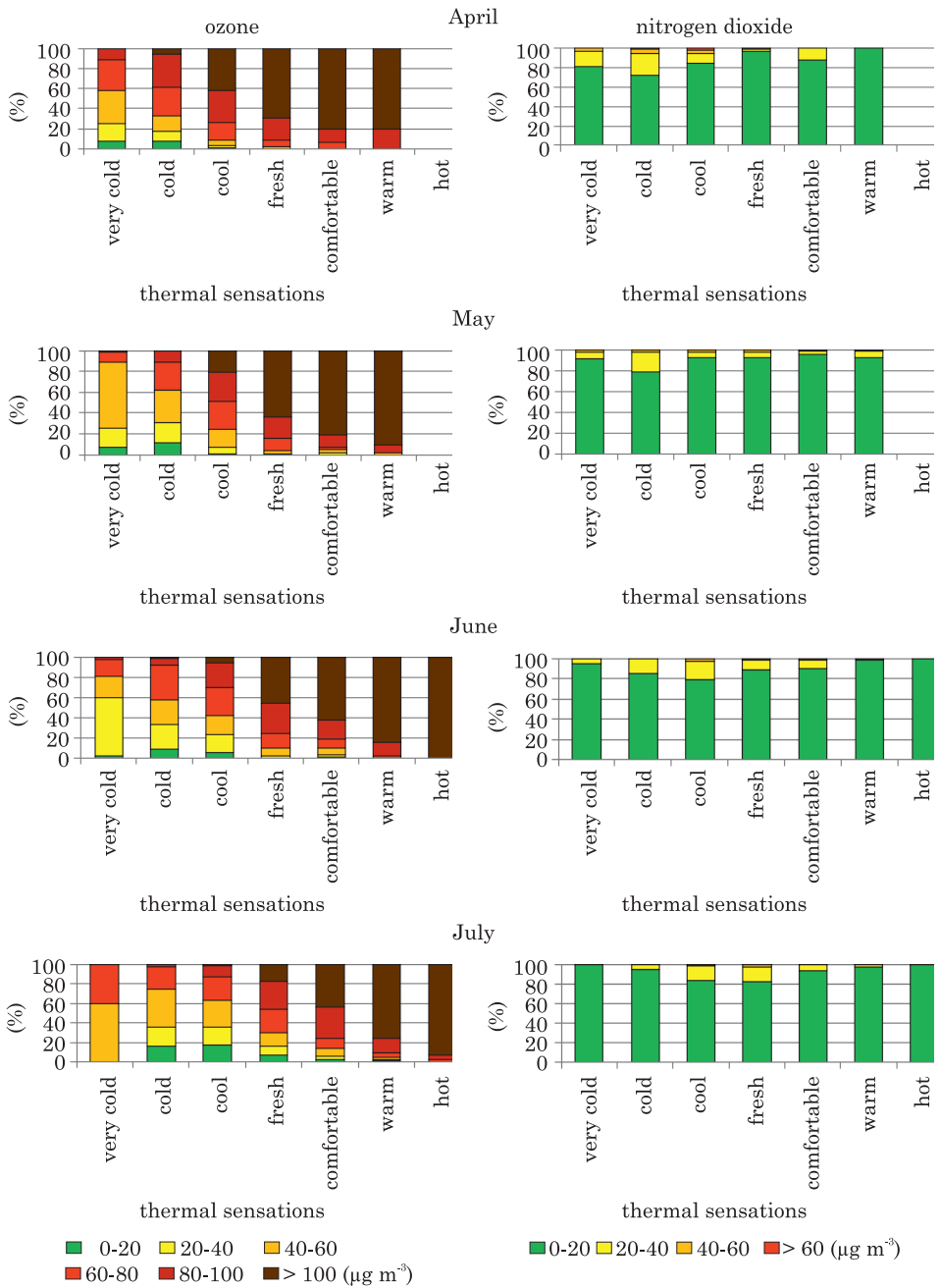
Fig. 6. The incidence of hourly values for concentrations of tropospheric ozone and nitrogen dioxide in the various classes of the human feel of heat according to the Mikhailov scale. The years 2006-2011

of radiation and other meteorological elements, but also in the average concentrations of ozone and NO_2. The lowest ozone concentrations, according to the feel heat classes, attained similar values in individual months. They occurred when the conditions referred to as very cold and cold (in July) prevailed. The highest ozone concentrations were characteristic for the highest thermal feel classes, that is warm in spring and hot in summer. The difference between the ozone concentration in the warmest thermal sensation class in July and May exceeded 20 µg m^{-3}. In turn, the lowest NO_2 concentration occurred in summer, when it was hot. The highest concentrations of NO_2, according to months, were observed in different albeit but always chilly classes of heat feel. In spring, the highest concentration of NO_2 occurred in the feel class cold, in June - in the very cold class, and in July - in the cool class.

The conditions experienced as hot and warm were oppressive not only due to the lack of comfort, but also because of the highest incidence of high ozone concentrations (Figure 6). Likewise, conditions identified as comfortable on the scale of thermal sensations, were characterized by the predominance of the largest ozone concentrations in the months of spring and summer. In the case of hourly nitrogen dioxide concentrations, a slightly different regularity emerged. The largest incidence of higher NO_2 concentrations in spring occurred during the cold class, but cool in June, and fresh in July. Elevated ozone concentrations coincided with the thermal stress identified as heat, but they also occurred during spells of the weather described as bioclimatic comfort. A more frequent occurrence of heat stress in town centres was confirmed, both in summer and winter (BŁAŻEJCZYK et al. 2014).

CONCLUSIONS

1. The concentrations of ozone in southwest Poland show annual, seasonal and daily variations, typical of Central Europe.

2. The variation of ozone concentrations was affected by the nitrogen dioxide concentration, meteorological elements and the bioclimatic index. The correlation between ozone and nitrogen dioxide was the strongest in the winter half-year, while the influence of meteorological elements dominated in the warm season of the year.

3. Among the meteorological elements, the ozone concentration variation was most strongly correlated with relative humidity of air (negatively, in spring and autumn), and, subsequently, with air temperature and solar radiation (positively, in the summer) and wind velocity (positively, in winter).

4. In suburban areas, above-average ozone concentrations occur most often in conditions of heat stress, but can also take place in conditions of thermal comfort.

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