ARCHIVES OF ENVIRONMENTAL PROTECTION
ARCHIWUM OCHRONY ŚRODOWISKA

vol. 26 no. 2 pp. 7–19 2000

PL ISSN 0324-8461

THE MEASUREMENT OF TROPOSPHERIC OZONE CONCENTRATIONS IN SOUTHERN POLAND USING THE PASSIVE SAMPLERS AND PLANT BIOINDICATORS

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Keywords: tropospheric ozone, passive samplers, tobacco plants, Southern Poland, national parks, Kraków Province.

POMIARY STĘŻENIA OZONU PRZYZIEMNEGO W POLSCE POŁUDNIOWEJ PRZY UŻYCIU PRÓBNIKÓW PASYWNYCH ORAZ BIOWSKAŹNIKÓW ROŚLINNYCH

Stężenia ozonu przyziemnego mierzono w okresie od początku maja do końca września 1996 i 1997 roku na 14 stanowiskach w województwie krakowskim i w pięciu karpackich parkach narodowych (PN). Stężenia te określano za pomocą próbników pasywnych. W dwóch stanowiskach (Kraków-centrum) i w Szarowie (30 km na wschód od miasta) eksponowano roślinne biowskaźniki ozonu (*Nicotiana tabacum* L.). Najwyższe średnie stężenie ozonu dla dwutygodniowego okresu ekspozycji w rejonie Krakowa zanotowano w 1996 roku. Wynosiło ono 137,7 $\mu g \cdot m^{-3}$. W obrębie województwa krakowskiego obserwowano znaczne zróżnicowanie stężeń ozonu między poszczególnymi stanowiskami: najniższe stężenia notowano w Krakowie i na obszarze położonym na zachód i północny zachód od miasta, najwyższe natomiast w obszarach położonych na północ i południe od Krakowa. Najwyższe stężenia ozonu wśród stanowisk zlokalizowanych w górskich parkach narodowych notowano w Bieszczadzkim i Babiogórskim PN. W Karpatach średnie dwutygodniowe stężenia ozonu w 1997 roku były wyższe niż w roku 1996.

Statystycznie istotnie wyższe uszkodzenia liści tytoniu (odmiany wrażliwej na ozon – Bel-W3) obserwowano u roślin eksponowanych poza Krakowem niż w centrum miasta.

Summary

Concentrations of ground level ozone were measured from May to September 1996 and 1997 in 14 localities throughout the Kraków Province and in five Polish national parks (NP) located in the Carpathians. Ozone concentrations were measured using passive samplers. At two sites (Kraków – city and Szarów – 30 km east of the city) ozone bioindicator tobacco plants (*Nicotiana tabacum* L.) were also exposed in the field. The highest forthnightly O_3 mean values in Kraków Province were recorded in 1996 and they reached 137.7 $\mu g \cdot m^{-3}$. Great variations in the tropo-

spheric O_3 concentrations were recorded from different exposure sites in Kraków Province: the lowest concentrations were recorded from Kraków and the areas west and north-west of the city, and the highest concentrations from areas north and south of the city. Among mountain exposure sites the highest ozone concentrations were recorded in Bieszczady NP and Babia Góra NP. The two-week long average concentrations in the Carpathians were higher in 1997 than in 1996.

Significantly greater damage to tobacco Bel-W3 leaves occurred in the plants exposed at Szarów than in plants exposed in the city centre.

INTRODUCTION

The concentrations of ozone in the troposphere are now two- and threefold when compared to the measurements taken in the last century [2, 22]. During the last ten years the increase in O_3 concentration ranged from 1.3 to 2.6%, depending on the location of the station [6, 7, 16, 17], due to the higher production of ozone precursors from combustion of gasoline and other fuels [5].

Ozone is the major compound of photochemical smog [11]. Southern Poland is the most polluted area due to the high emission of sulphur, nitrogen oxides and heavy metals from local and remote sources [19]. The Kraków agglomeration and the Upper Silesian Industrial Region are the major sources of ozone precursors in this part of Poland [12]. Recent studies performed in southern Poland (Upper Silesia Region, Beskidy Mountains, Kraków Region) have shown that during the summer, one-hour average ozone concentrations may reach 230 $\mu g \cdot m^{-3}$ [3, 10].

According to the measurements taken using analyzers at the two stations in Kraków between 1992 and 1996, the annual mean ozone concentrations ranged from 41 to 49 $\mu g \cdot m^{-3}$ (Table 1). During those years the mean concentrations in winter were between 22 and 38 $\mu g \cdot m^{-3}$, whereas in summer they ranged from 52 to 59 $\mu g \cdot m^{-3}$. Maximum 30-minute concentrations were higher than 200 $\mu g \cdot m^{-3}$, and 24-hour concentrations higher than 100 $\mu g \cdot m^{-3}$ (Table 1). In addition they are higher than background O_3 levels [11, 20].

Table 1. Ozone concentrations ($\mu g \cdot m^{-3}$) in Kraków during the period 1992–1996 (measurements using analyzers) [21]

Year	Max 30-min	Max 24-hour	Annual mean	Winter mean	Summer mean
1992	414	176	43	27	59
1993	269	115	43	33	53
1994	230	113	41	22	56
1995	205	103	49	38	56
1996	227	117	41	29	52

Little is known about ozone concentrations in the Carpathian Mountains. In the Ukrainian part of the Carpathians, two-week average ozone concentrations measured using the passive method in August 1995, ranged (depending on the exposure site) from 52 to 100 $\mu g \cdot m^{-3}$ [1]. Ozone concentrations in the Beskidy Mountains (Carpathians) in Poland exceeded potentially phytotoxic levels [10].

This paper discusses the results of a two year (1996 and 1997) ozone study in the Kraków Province and in Polish national parks located in the Carpathians, in which passive samplers were used. In Kraków Province the measurements were aimed at assessing the effect of ozone on bioindicator plants. The studies conducted in national parks were also aimed at designated potentially ozone-sensitive species of native plants.

STUDY AREA AND METHODS

Measurements of ground level ozone concentrations were taken in Kraków Province and in five mountain national parks. Ozone concentrations were measured during the period from May (June) to September 1996 and 1997.

The Kraków Province covers an area of 3254 km^2. Measurements of ozone were taken at 14 exposure sites: 3 were located within the city of Kraków (in the western, central, and eastern parts of the city), whilst the remaining 11 stations were evenly distributed throughtout the Kraków Province (Fig. 1).

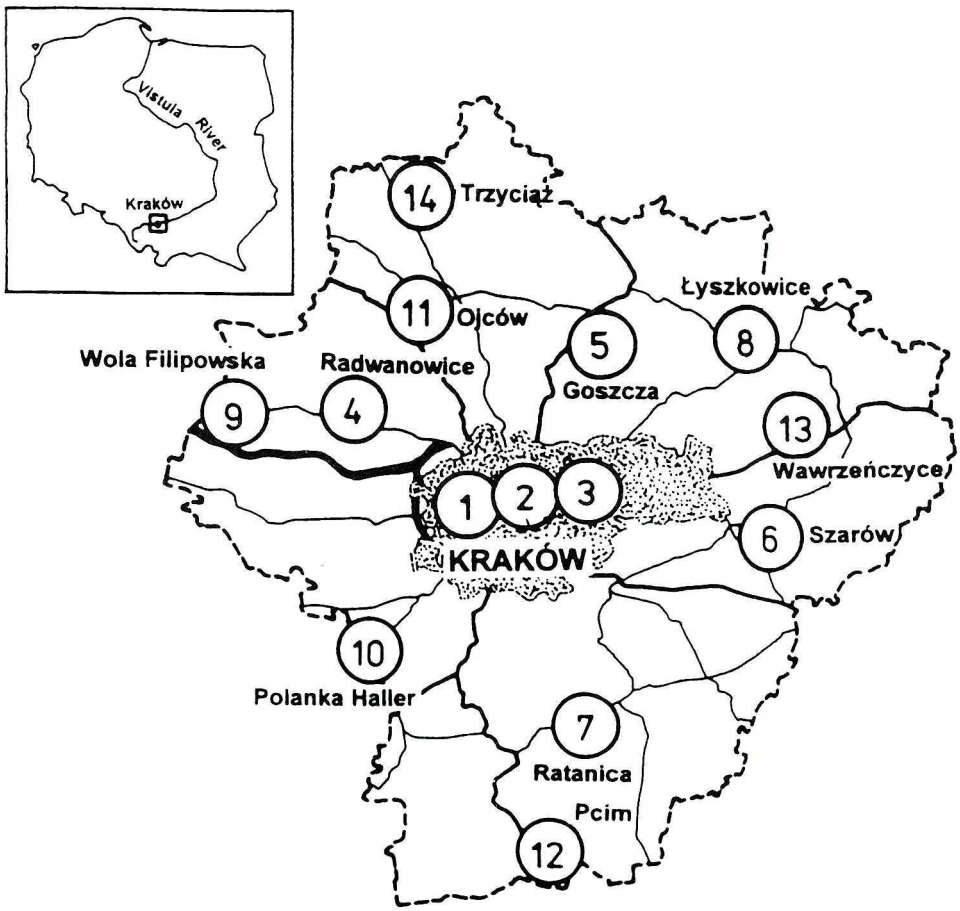

Fig. 1. Location of the ozone measuring sites in Kraków Province

Measurements of tropospheric ozone concentrations were also made in five Polish national parks in the Carpathians: Babia Góra NP, Tatra NP, Pieniny NP, Magura NP, and Bieszczady NP. The measurement stations located in the national parks differed in their altitude and specific composition of tree stands (Table 2).

All measuring stations were equipped with Ogawa passive samplers (Ogawa & Co, Pompano Beach, FL) [14]. Passive samplers were exposed from May (June) to September in two-week intervals. In all the mesurement stands throughtout Kraków Province passive samplers were located in an open area, 2 – 2.5 m above the ground. The measuring stations in three mountain national parks (Babia Góra NP, Tatry NP, Bieszczady NP) were installed at the high of about 980 – 1200 m, and in the remaining two (Pieniny NP and Magura NP) at approximately 600 m a.s.l. (Table 2). In 1997 the exposure site in the Tatra NP was moved to a lower site (1080 m a.s.l.). In all locations priority was given to open areas with western exposure, receiving the flow of westerly winds that prevail in southern Poland.

The "Ogawa" sampler consists of two separate nitrite coated cellulose filters between two discs of stainless steel gauze. The nitrite coated filters were oxidized to a nitrate ion in the presence of ozone [14]. Two blank filters refrigerated in sealed containers were used for each exposure. After exposure in the field the filters were extracted in ultrapure water and analyzed for nitrate by ion chromatography (Dionex-100). The calculation of the value of nitrates into the concentration of ozone was made in relation to the data obtained from the active monitor (Thermo Environmental Model 49 UV-photometric ambient O_3 analyzer, Franklin, MA, USA), installed in Kraków (Balicka street in western part of the city) by the formula given by Koutrakis et al. [14]. This station belongs to the Voivodeship Inspectorate of Environmental Protection in Kraków.

Following Manning and Feder [18], Heggestad [13], Krupa et al. [15] two cultivars of tobacco (*Nicotiana tabacum* L.) – Bel-B (ozone tolerant) and Bel-W3 (ozone sensitive) – were chosen as bioindicator plants for assessing the effect of ozone. They were exposed in the field from June to September 1996 and 1997 (for one-week periods) at two sites located in the Kraków Province: at Lubicz Street (the centre of the city), and in Szarów (30 km east of Kraków, a rural site). The plants used for the exposure had been cultivated in a greenhouse in charcoal-filtered air. The plants were grown in a mixture of peat, bog-moss, and perlite. The plants were watered daily and fertilized (Peter's professional) once a week. When the plants had reached the stage of four leaves, ten specimens of each cultivar of tobacco (Bel-W3 and Bel-B) were planted at each of the two stations. Leaf injury was measured according to the procedure described by Manning and Feder [18].

The differences in the leaf injury of the tobacco cultivars exposed at Kraków and Szarów were tested using the Friedman's two-way analyses of variance method. All analysis were performed using Statgraphics Version 5.0

(Statgraphics 1991). A map showing the distribution of O_3 concentrations in the Kraków Province was made using Surfer 5.0 (Golden Software). The graphic description was made using CorelDraw 3.0 (Corel Canada).

Table 2. Main characteristics of O_3 measuring stations in national parks in Poland

National park	Latitude	Longitude	Altitude (m a.s.l.)	Exposure	Angle of slope	Forest composition
Babia Góra NP	49°35'18"N	19°35'12"E	980	SW	25°	90% *Picea abies*, 10% *Abies alba*
Tatra NP – 1996	49°16'16"N	20°02'33"E	1200	N	10°	100% *Picea abies*
Tatra NP – 1997	49°17'42"N	20°06'55"E	1080	W	15°	98% *Picea abies*, 2% *Sorbus aucuparia*
Pieniny NP	49°25'12"N	20°20'35"E	580	W	18°	60% *Picea abies*, 20% *Fagus sylvatica*, 20% *Abies alba*
Magura NP	49°31'43"N	21°30'28"E	540	W	10°	15% *Pinus sylvestris*, 80% *Fagus sylvatica*, *Alnus glutinosa*, *Abies alba*
Bieszczady NP	49°06'55"N	22°36'30"E	980	SW	25°	95% *Fagus sylvatica*, 5% *Picea abies*, *Abies alba*, *Salix caprea*, *Sorbus aucuparia*, *Acer pseudoplatanus*

RESULTS

OZONE CONCENTRATIONS IN KRAKÓW PROVINCE

According to the measurements taken in 1996, the mean ozone concentrations varied markedly depending on the location and the time of year (Table 3). The highest ozone concentrations were recorded in May and in the first half of June (Table 3). The maximum average concentration for a two-week exposure period was 137.7 $\mu g \cdot m^{-3}$ at the Trzyciąż station in the northern part of the province. Concentrations had been reduced by 50% by the second half of June and continued to fall slowly in July and August. Still lower ozone concentrations were recorded in September (Table 3). The highest ozone concentrations were recorded in Goszcza, Ratanica, Polanka Haller, Łyszkowice, and Trzyciąż. Among the three locations in Kraków, the highest concentrations were recorded as a rule at Balicka Street (western part of the city). The lowest concentrations occurred at Ojców, Pcim, Wola Filipowska, and in the city centre at Lubicz Street (Table 3).

In 1997 ozone concentrations in the period from May to September were less differentiated among the fortnightly exposure periods than in 1996. The highest ozone concentrations reached 93 $\mu g \cdot m^{-3}$. The highest ozone concentrations were recorded in May and June and the lowest ones in the second half of September (Table 4). In 1997, the highest and the lowest concentrations

Table 3. Mean ozone concentrations ($\mu g \cdot m^{-3}$) in Kraków Province in 1996 (measurements using passive samplers)

Exposure site	Exposure period										
	30/04–15/05	15/05–31/05	31/05–16/06	16/06–01/07	01/07–16/07	16/07–30/07	30/07–13/08	13/08–02/09	02/09–16/09	16/09–30/09	Mean value
Kraków-city											
1. Balicka Street	75.6	79.4	50.3	39.3	36.5	27.5	27.1	27.4	22.1	14.3	40.0
2. Lubicz Street	63.7	55.8	56.5	18.2	20.7	16.8	20.6	15.1	11.6	8.6	28.8
3. Żeromski hospital	74.0	69.3	83.8	34.8	32.6	24.8	28.1	24.6	15.9	9.6	39.8
Kraków Province											
4. Radwanowice	77.9	75.8	75.1	39.1	36.6	27.3	27.5	26.8	19.8	15.4	42.1
5. Goszcza	92.0	94.9	101.1	44.7	46.4	37.6	38.1	40.3	27.9	20.5	54.4
6. Szarów	82.4	85.0	88.2	45.3	41.4	32.3	29.0	28.7	21.1	15.5	46.9
7. Ratanica	88.0	90.7	94.9	46.5	42.8	35.5	33.3	31.9	25.3	15.8	50.5
8. Łyszkowice	76.5	90.0	87.7	41.2	36.9	32.2	35.6	35.4	25.8	21.0	48.2
9. Wola Filipowska	72.5	67.8	72.8	28.9	27.4	19.1	23.7	20.4	15.5	14.2	36.2
10. Polanka Haller	83.9	89.4	94.3	42.5	42.9	34.6	31.8	33.0	22.6	14.1	48.9
11. Ojców	74.1	67.4	66.3	29.0	18.0	23.0	19.5	15.0	12.9	12.3	33.8
12. Pcim	38.9	69.5	71.9	30.6	27.3	22.8	21.3	21.2	16.0	10.3	33.0
13. Wawrzeńczyce	90.6	78.5	77.2	38.7	32.9	26.1	24.1	19.4	18.4	13.0	41.9
14. Trzyciąż	39.9	137.7	90.7	43.7	39.9	33.5	31.5	33.0	23.6	18.3	47.4

Table 4. Mean ozone concentrations ($\mu g \cdot m^{-3}$) in Kraków Province in 1997 (measurements using passive samplers)

Exposure site	Exposure period										
	29/04 – 15/05	15/05 – 28/05	28/05 – 16/06	16/06 – 30/06	30/06 – 15/07	15/07 – 30/07	30/07 – 14/08	14/08 – 02/09	02/09 – 16/09	16/09 – 30.09	Mean value
Kraków-city											
1. Balicka Street	63.0	93.0	64.1	56.3	56.7	45.1	45.4	53.7	36.7	30.4	55.4
2. Lubicz Street	44.9	38.8	32.6	38.3	33.2	28.0	30.8	35.9	22.3	15.1	32.0
3. Żeromski hospital	58.0	49.8	24.2	35.0	52.2	40.8	38.2	42.3	31.6	23.1	39.5
Kraków Province											
4. Radwanowice	49.4	52.6	52.8	51.7	50.0	44.9	48.3	57.0	40.6	33.3	48.1
5. Goszcza	79.3	71.2	83.0	72.6	65.6	59.9	68.6	74.5	54.7	51.3	68.1
6. Szarów	71.3	59.3	69.2	60.0	54.6	48.7	47.7	53.6	44.1	36.7	54.5
7. Ratanica	83.6	67.8	60.1	75.4	60.9	60.5	58.3	74.2	60.8	50.8	65.2
8. Łyszkowice	74.5	70.5	68.3	68.1	58.5	54.2	60.4	69.4	52.3	45.9	62.2
9. Wola Filipowska	48.9	40.5	39.1	45.0	34.4	58.9	35.7	43.7	32.1	21.9	40.0
10. Polanka Haller	55.0	43.7	34.0	41.3	34.7	28.1	28.8	35.8	27.9	23.1	35.2
11. Ojców	54.9	38.1	45.2	46.3	38.3	32.6	38.4	43.0	35.3	23.4	39.6
12. Pcim	63.2	58.1	58.7	61.7	55.0	45.1	51.3	55.8	41.7	29.7	52.0
13. Wawrzeńczyce	70.8	62.2	70.0	63.3	52.4	47.5	56.7	62.3	54.5	42.5	58.3
14. Trzyciąż	69.1	65.1	67.0	66.9	57.1	31.2	54.5	67.1	56.4	49.8	58.4

were as a rule recorded at the same exposure sites as recorded in 1996. Ozone concentrations in the Kraków Province are generally high. Previous studies in this region have found that ozone concentrations in this region are higher than levels accepted as background [3, 8, 9].

All the sets of measurements have a similar pattern of spatial differentiation of ozone concentration in the Kraków region, irrespective of the period or the year (1996, 1997). The lowest concentrations occurred in an area extending from the centre to the west and north-west of Kraków and the highest were recorded in the areas to the north and south of the city (Fig. 2). Ozone concentrations in Kraków were lower than those outside the city due to ozone reaction with nitric oxide which is produced by vehicles in the city [4].

Significantly higher damage to tobacco Bel-W3 leaves were found in plants exposed at Szarów, outside the city limits (for data obtained in 1996 $F = 54.08$, $df = 1$, $p < 0.001$; for data obtained in 1997 $F = 14.00$, $df = 1$, $p < 0.001$). The greatest damage occurred in June, smaller in July – August, and comparatively

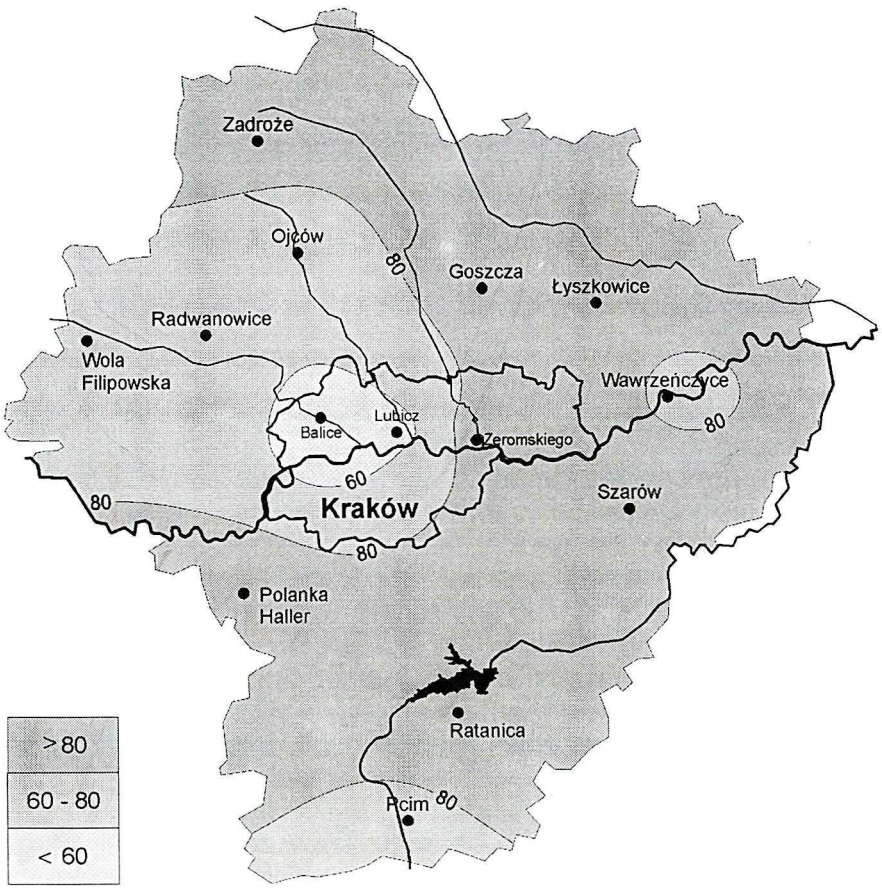

Fig. 2. Ozone concentrations ($\mu g \cdot m^{-3}$) in Kraków Province (exposure period: 31 May – 16 June 1996)

slight in September (Fig. 3). In plants exposed at Szarów, the greatest damage to single leaves of the ozone-sensitive tobacco cultivar (Bel-W3) covered from 50 to 60% of the leaf blade. Plants exposed in the city in most cases showed little or no noticeable ozone damage (a very small percentage at most). In ozone tolerant (Bel-B) tobacco plants exposed in the field, no damage to the leaves was observed at any station.

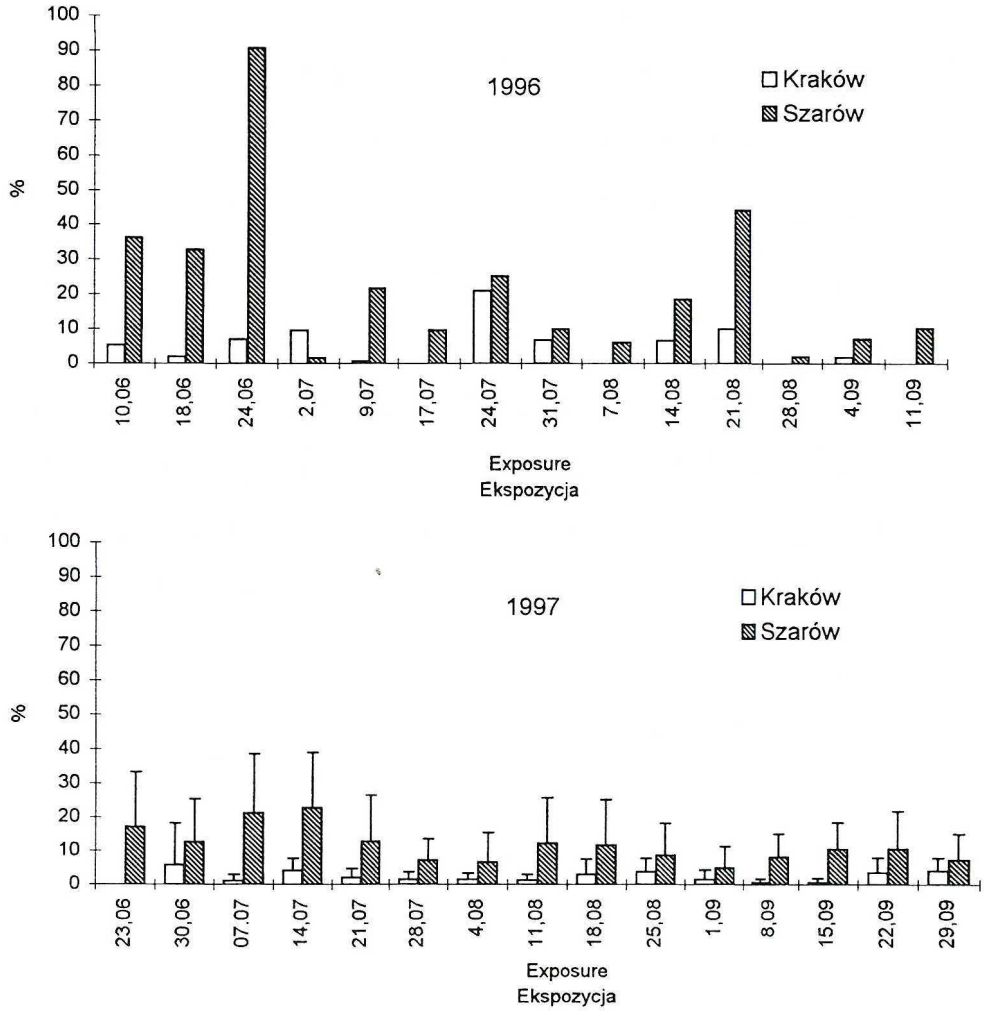

Fig. 3. Leaf injury to the Bel-W3 tobacco plants exposed at two sites in Kraków Province in 1996 and 1997. Percentage of leaf area damaged presented as mean sum of four leaves damaged on each of ten plants

OZONE CONCENTRATIONS IN POLISH MOUNTAIN NATIONAL PARKS

Over the sampling period in 1996, the highest ozone concentrations in particular measurement periods were recorded in Bieszczady NP ($29.4-100\ \mu g \cdot m^{-3}$), followed by Babia Góra NP ($26.7-88.0\ \mu g \cdot m^{-3}$) (Table 5).

The lowest concentrations of ozone were recorded in the Tatra NP (15.6 – 41.9 $\mu g \cdot m^{-3}$). The intermediate concentrations were noted in Pieniny NP (19 – 77.7 $\mu g \cdot m^{-3}$) and Magura NP (22.3 – 71.4 $\mu g \cdot m^{-3}$) (Table 5). Differences in mean fortnightly ozone concentrations among all measuring stations located in the Carpathians, were statistically significant (in decreasing order as follows: Bieszczady NP > Babia Góra NP > Pieniny NP > Magura NP > Tatra NP) ($F = 25.94$, $df = 4$, $p < 0.001$; LSD test, $p \leqslant 0.05$) (Table 5). It seems that differences in the ozone concentrations among the stations are mainly associated with the altitude and location of the samplers. Ozone concentrations were higher at stations located at higher altitudes in the Bieszczady NP and Babia Góra NP. The Babia Góra NP is one of the most polluted parks, and the elevated ozone levels could stem from this. These high concentrations of ozone in the Bieszczady NP could be assiociated with "low sources" of emissions (including ozone precursors) coming from charcoal burners. Despite the installation of samplers in the Tatra NP at altitudes similar to those in the Bieszczady NP and Karkonosze NP, the ozone concentrations there were markedly lower. This was probably caused by the location of the samplers which had been installed in a small glade surrounded by upper montane spruce forest and shielded on three sides by high cliffs. As a result, the air flow was markedly lower than at the other sites.

In 1996 the highest concentrations of ozone occurred in the first half of June (41.9 – 100 $\mu g \cdot m^{-3}$, depending on the station considered), falling to 38 – 68.5 $\mu g \cdot m^{-3}$ later in the month. The lowest concentrations were recorded at the end of the sampling season in the second half of September.

The average ozone concentrations were higher in 1997 compared to the 1996 season in all measuring stations in the Carpathians (Tables 5, 6). The highest O_3 concentrations were recorded (similar to 1996) in the Bieszczady NP (79.5 – 118.8 $\mu g \cdot m^{-3}$) and in the Babia Góra NP and the lowest in the Magura NP (48.5 – 84.1 $\mu g \cdot m^{-3}$). Differences in ozone concentrations among the mountain national parks were statistically significant (in decreasing order as follows: Bieszczady NP > Babia Góra NP > Tatra NP > Pieniny NP > Magura NP) ($F = 33.36$, $df = 4$, $p < 0.001$; LSD test, $p \leqslant 0.05$) (Table 6). The highest ozone concentrations were recorded in May and June and the lowest in the second half of September (Table 6).

Ozone concentrations in mountanous parts of Poland exceed potentially phytotoxic levels [10, 23]. Ozone-like injury symptoms were noted in a few plants native or common to Carpathians (*Sambucus racemosa*, *Corylus avellana*, *Alnus incana*, *Sorbus aucuparia*, *Ulmus glabra*, *Cornus sanguinea*, *Vaccinium myrtillus*, *Rubus* sp., *Impatiens parviflora*, *Astrantia major*, *Lapsana communis*, *Chaerophyllum aromaticum* and *Gentiana asclepiadea*).

CONCLUSIONS

According to the active and passive measurements of tropospheric ozone concentrations taken in southern Poland, ozone concentrations frequently ex-

Table 5. Mean ozone concentrations ($\mu g \cdot m^{-3}$) in mountain national parks in Poland in 1996 (measurements using passive samplers)

Exposure site	Exposure period								
	01/06–15/06	15/06–01/07	01/07–15/07	15/07–01/08	01/08–15/08	15/08–01/09	01/09–15/09	15/09–30/09	Mean value
Babia Góra NP	88.0	51.5	47.7	43.7	46.7	49.0	33.4	26.7	48.3
Tatra NP	41.9	38.0	31.2	26.8	27.6	22.0	24.2	15.6	28.4
Pieniny NP	77.7	48.2	41.7	35.7	43.9	39.7	27.4	19.0	41.7
Magura NP	71.4	45.5	40.9	33.2	34.7	34.5	25.5	22.3	38.5
Bieszczady NP	100.0	68.5	57.8	49.7	55.9	55.7	39.1	29.4	57.0

Table 6. Mean ozone concentrations ($\mu g \cdot m^{-3}$) in mountain national parks in Poland in 1997 (measurements using passive samplers)

Exposure site	Exposure period										
	01/05–15/05	15/05–01/06	01/06–15/06	15/06–01/07	01/07–15/07	15/07–01/08	01/08–15/08	15/08–01/09	01/09–15/09	15/09–30/09	Mean value
Babia Góra NP	119.2	75.6	107.9	99.3	84.5	80.7	99.5	94.5	85.7	65.0	91.2
Tatra NP	91.2	76.4	91.5	87.9	73.9	67.9	81.5	83.7	75.5	65.3	79.5
Pieniny NP	94.7	61.8	77.5	83.4	71.5	60.8	68.3	66.3	54.6	46.6	68.6
Magura NP	84.1	57.6	68.2	68.0	62.8	55.2	62.2	67.7	57.4	22.3	58.5
Bieszczady NP	118.8	91.8	100.3	101.2	87.9	86.8	87.4	94.9	79.5	29.4	93.0

ceed the generally accepted levels in that part of the country. Therefore, we can expect that ozone and sulphur dioxide and nitrogen oxides should share responsibility for the health condition of the forests in that region. Greater damage to ozone bioindicator plants exposed in rural, as opposed to urban, areas also indicates that ozone can affect the plant crops cultivated in the southern part of Poland.

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Received: November 20, 1998, accepted: December 7, 1999.