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TOTAL SUSPENDED PARTICULATE MATTER CONCENTRATIONS IN ZAGREB DURING THE 1975-1993 PERIOD

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Daily average mass concentrations of total suspended particulate matter were measured at three sampling sites in Zagreb, and evaluated for the period April 1975 – March 1993. Each sampling site represented a different town area (residential, business and administrative, industrial) with different traffic density and type of energent used for space heating. The time trends of concentration levels could, to a certain extent, be attributed to traffic flow modification in the vicinity of the sampling sites, introduction of natural gas in dwellings and degree of energy consumption influenced by the standard of living. Periodograms show a well pronounced seasonal dependence of total suspended particulate matter concentrations, with high concentrations during winters. Analysis of the results in respect to the European Community air quality limits (1) and the levels of other pollutants (SO_2 and smoke) leads to the conclusion that particulates being a persistent permanent problem have become a major issue concerning ambient air pollution in Zagreb.

Key terms:
air quality limits, periodogram, seasonality, suspended particulate matter levels, urban area

Measurements of total suspended particulate matter (TSP) were performed in Zagreb, capital of Croatia, city of about 1,000,000 inhabitants, administrative, cultural and industrial centre of the region. The main ambient air pollution sources are fossil fuel combustion for heating, energy production, transport and industry.

While SO_2 and "smoke" concentrations have considerably decreased (three to four times) in the past 25-30 years (2), due to substitution of coal for liquid fuels and gas, and propagation of district heating systems, the TSP concentration levels did not follow the same trend and have remained almost on the same level.

The purpose of this investigation was to describe possible time trends and seasonal dependences of TSP concentrations and to compare the measured concentrations with the proposed air quality limits (1). The results should be used to initiate and promote measures for solving the TSP problem.

MATERIALS AND METHODS

Three sampling sites were chosen for a long-term TSP investigation, representing the city centre (business/residential), and the city western (industrial/residential), and northern parts (residential). Samples were collected on a daily basis (noon to noon), at least 200 samples per site, distributed during seasons. Each sampling cycle started on April 1st and lasted until March 31st (2). At all three sampling sites, sampling equipment was situated on a flat roof, approximately 3-4 m above the ground level, relatively close to the street (5-15 m). The inlet of the sampling device was constructed to cut off the particles at a mass median diameter (MMD) of approximately 20 μm. Particulate matter was collected on membrane filters, 102 mm in diameter, at an average flow rate of 150 $Lmin^{-1}$. Prior to mass determination, before and after sampling, filters were preconditioned in a desiccator to a constant humidity for a period of 24 hours (3). Comparison with other gravimetric methods gave satisfactory results (4).

RESULTS AND DISCUSSION

Annual averages and maximum TSP concentrations, for three sampling sites during the 1975-1993 period, are shown in Table 1. It can be seen by comparison with the European Community limit values that the concentrations were frequently higher (except for the northern residential part, during the time period after the year 1990), as it was shown earlier in comparison with other cities (5). The seasonal and spatial differences in TSP concentrations could be partially explained by particle size distribution, since the transport of larger particles (predominant mass contribution to the overall concentration) is much less pronounced compared to the small ones (6).

Levels describing C_{avg} (average concentration), C_{max} (maximum concentration), C_{50} (median), and C_{98} (the concentration 98% of the results are below) are shown through the years in Table 1. Concentrations equalling or exceeding 50 and 98% of the results collected during a one year period (C_{50} and C_{90}) were compared to the proposed EC limits of 150 and 350 gm^{-3} respectively. The concentration values exceeding the proposed limits are underlined in Table 1.

Table 1 *Annual averages, maximum, C_{50} and C_{98} TSP concentrations in Zagreb during the 1975–1993 period (μgm^{-3})*

Season	Centre-west				City centre				Residental area north			
	C_{avg}	C_{max}	C_{50}	C_{98}	C_{avg}	C_{max}	C_{50}	C_{98}	C_{avg}	C_{max}	C_{50}	C_{98}
75/76	155	507	144	340	185	623	171	456	135	483	110	310
76/77	141	579	138	305	164	450	150	363	123	332	185	271
77/78	160	506	151	306	160	362	148	342	132	400	111	358
78/79	151	461	138	330	162	587	147	417	137	375	131	289
79/80	139	498	130	408	133	469	120	416	138	350	127	281
80/81	137	571	120	355	130	830	107	332	143	488	124	417
81/82	119	526	102	307	112	473	96	260	158	466	139	393
82/83	114	524	105	312	121	380	102	338	133	418	107	369
83/84	125	488	104	296	131	733	115	292	127	375	111	292
84/85	132	417	107	355	142	519	118	365	131	471	111	350
85/86	95	333	88	260	136	529	126	271	120	294	113	258
86/87	43	505	122	324	147	736	126	393	130	519	119	327
87/88	126	381	115	260	123	352	123	276	107	424	98	212
88/89					156	431	142	376	92	300	81	236
89/90					124	363	115	305	79	277	64	213
90/91					145	577	139	316	67	215	64	195
91/92					130	401	113	342	54	174	47	134
92/93					93	284			51	218		

Monthly mass averages of TSP and their polynomial smoothing, are presented in Figures 1–3. Clearly shown seasonal dependence of TSP mass concentrations was justified by periodogram calculations. No other significant periodic dependence was found. Polynomial smooths represent concentration trends over the years.

In the city centre this trend can be explained by the introduction of natural gas in this part of the town (years 1975–1979), traffic density change as a consequence of traffic flow modification in neighbouring streets (years 1984–1989), and decline of standard of living during the war and in the post-war period (years 1990–1993) which resulted in lower energy consumption and lower traffic density. In the northern residential area, the rise of TSP concentrations in the early eighties is a result of large dwelling colony construction in the vicinity of the sampling site, while the decrease shown in the later period can be attributed

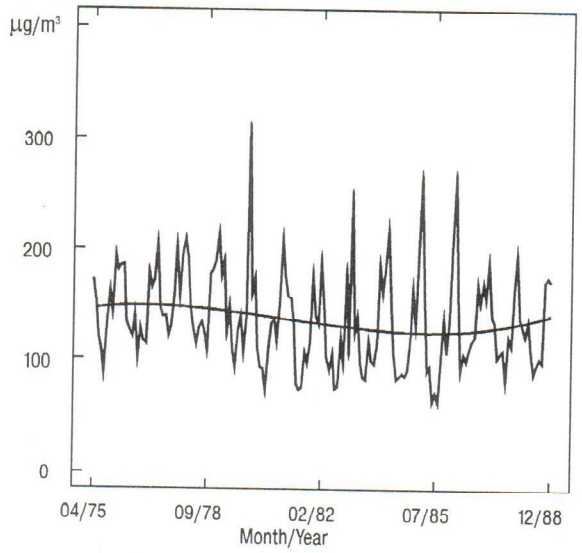

Figure 1 *Time series and polynomial smoothing of TSP concentrations in Zagreb centre-west*

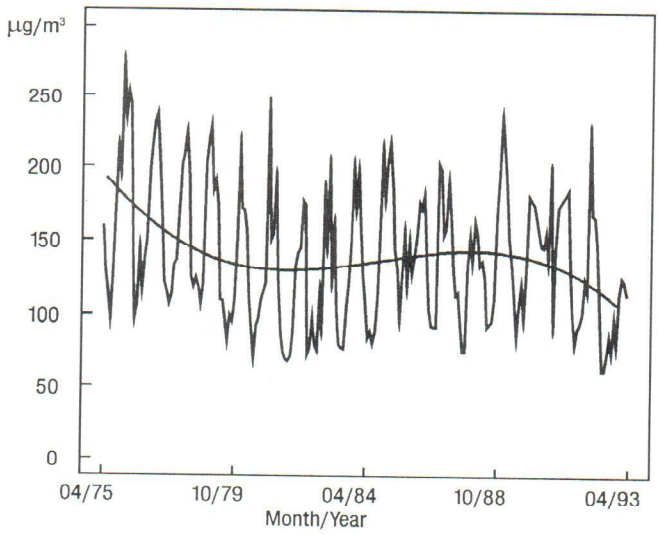

Figure 2 *Time series and polynomial smoothing of TSP concentrations in Zagreb city centre*

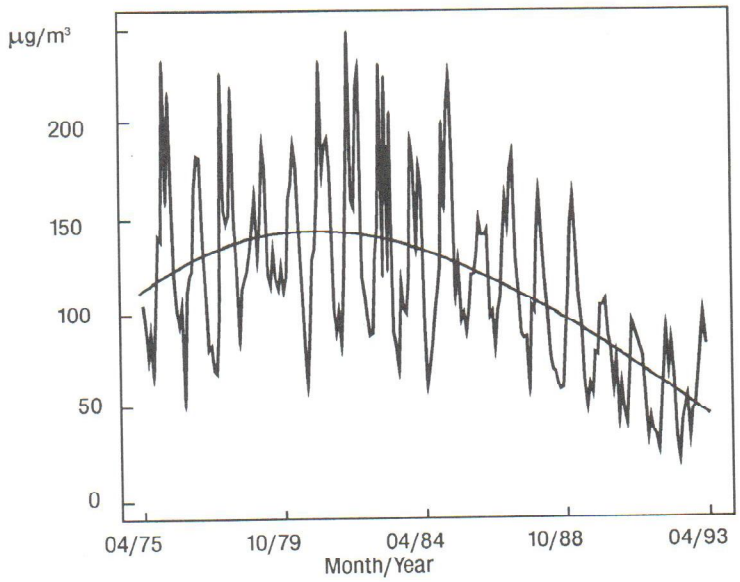

Figure 3 *Time series and polynomial smoothing of TSP concentrations in Zagreb northern residential area*

to the end of building activities as well as to the same reasons as described for the city centre. Concentrations at the centre-west location show a less pronounced trend, but a slight increase can be observed since 1985, when traffic became denser following the construction of a subway under a nearby railroad. Measurements stopped at the end of 1988, so the third period of interest described by a decline in the standard of living during the war and in the post-war period could not be evaluated.

Pronounced seasonalities and seasonal subseries of monthly TSP concentration averages for the three sampling sites are presented in Figures 4-6. Concentration levels during winter are 1.8-2.0 times higher compared to the summer ones. This could be attributed to high energy consumption for space heating, as well as to climate-related frequent temperature inversions and high relative air humidity favouring particle coagulation and agglomeration. However, these seasonal differences are less pronounced than for SO_2 and smoke concentrations because of the sources that are active throughout the year, like redispersion of settled dust by traffic and wind, and poor maintenance (unsatisfactory road cleaning and building repair). Subseries for individual months over the years show patterns similar to the polynomial smooths (trends) described above for the whole period of investigation. That is in good agreement with possible causes mentioned earlier (building constructions, changes in traffic density, decline in standard of living) since all these emission sources are active all year long.

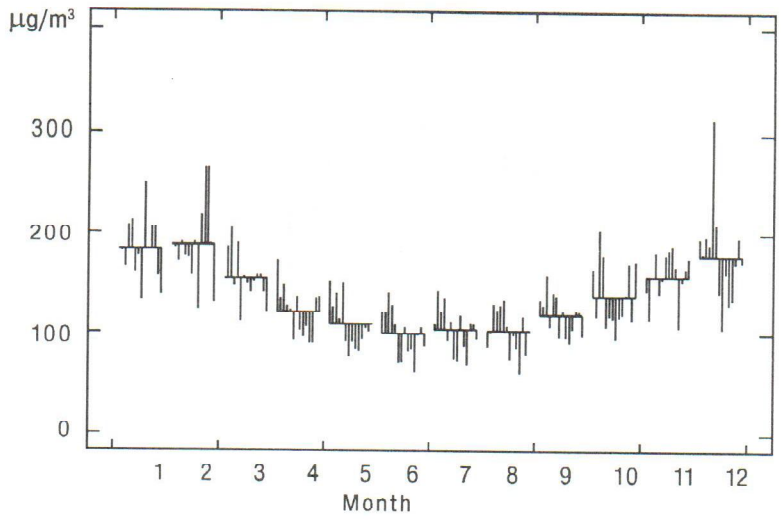

Figure 4 *Seasonal subseries of TSP concentrations in Zagreb centre-west*

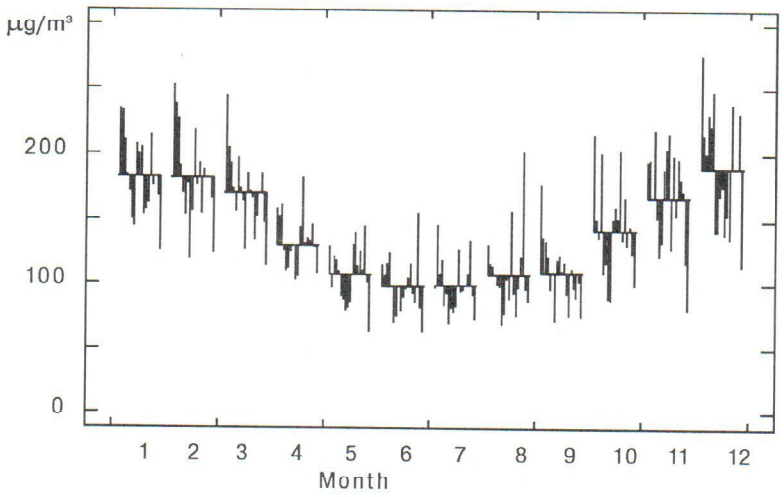

Figure 5 *Seasonal subseries of TSP concentrations in Zagreb city centre*

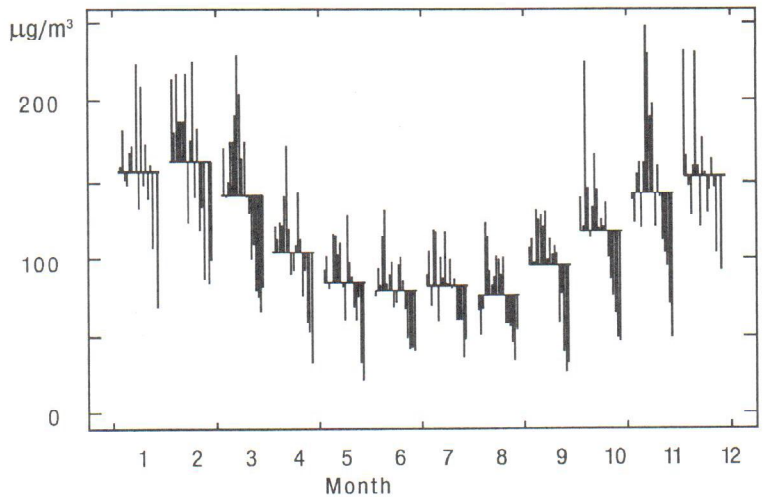

Figure 6 *Seasonal subseries of TSP concentrations in Zagreb northern residential area*

CONCLUSIONS

The TSP concentrations in Zagreb, for the period 1975-1993, were relatively high, especially in the city centre. They exceeded not only the guide values, but were frequently higher than the limit values as well. Particulates are a major and permanent air pollution problem in Zagreb ambient air.

Seasonal concentration dependence was obvious at all three sampling sites, primarily as a consequence of higher energy consumption during winter time.

The concentration trend over the years could be partly explained by changes in the type of fuel used for space heating, by changes in traffic density and/or building constructions in the surroundings of the sampling sites, as well as indirectly, by variations in the standard of living of Zagreb inhabitants.

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Sažetak

KONCENTRACIJE LEBDEĆIH ČESTICA U ATMOSFERI ZAGREBA U RAZDOBLJU 1975–1993.

Prosječne dnevne koncentracije ukupnih lebdećih čestica, mjerene na tri lokacije u gradu Zagrebu, prikazane su za razdoblje 1. travnja 1975. do 31. ožujka 1993. godine. Svako od mjernih mjesta predstavlja različito područje grada, s obzirom na gustoću prometa, vrstu energenta koji se upotrebljava za grijanje prostora, namjenu zgrada i sl. Vremenski trendovi koncentracija mogu se do stanovite mjere pripisati promjenama u režimu prometa u blizini mjernih mjesta, uvođenju plina na pojedinim područjima te razini korištenja energije ovisno o promjenama životnog standarda. Periodogrami pokazuju dobro izraženu sezonsku ovisnost koncentracija, s povišenim vrijednostima tijekom hladnog dijela godine. Usporedba dobivenih rezultata s preporučenim graničnim vrijednostima Europske zajednice vodi do zaključka da su lebdeće čestice najvažniji i stalni problem onečišćenja atmosfere u gradu Zagrebu.

Ključne riječi:
gradsko područje, periodogram, razine kakvoće zraka, sezonske promjene, razine lebdećih čestica

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