

MONITORING OF PM_{2.5} PARTICLE FRACTION IN ZAGREB AIR

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ABSTRACT

Investigations of PM_{2.5} in Zagreb ambient air started in autumn of 1998 by developing and testing sampling equipment and analytical procedures. For a period of five years samples were collected at only one sampling site. Parallel measurements of total suspended particulates (TSP), PM₁₀ and PM_{2.5}, as well as heavy metal and anion content in particle fractions were performed. Later step was the investigation of seasonal and space differences in PM_{2.5} particle fraction performed at four sampling sites, for a month period in summer and again in winter period. The results of these investigations, together with the conclusions of several recent WHO meetings resulted in PM_{2.5} Zagreb network implementation in year 2005.

Key Words: PM_{2.5} particle fraction, Concentration Distribution, Proposed Limit Values

1. INTRODUCTION

Investigation of particulate pollution in Zagreb air started in 1972 by monitoring the concentrations of total suspended particulate matter (TSP) and their heavy metal content. Because of better relation to the health outcomes (ISO 1995, WHO 1998) in 1999 parallel PM₁₀ and PM_{2.5} particle fraction monitoring started at one sampling location representing northern, residential part of Zagreb. Monitoring included determination of lead, manganese and cadmium, as well as anion content (sulphates, nitrates and chlorides) (Čačković 2004) and PAHs in both particle fractions (Šišović 2004). Health outcomes based on measured concentration data were evaluated by means of AirQ computer program (Šega 2001). Concentrations of PM_{2.5} particle fraction were related to TSP and PM₁₀ fraction concentration. In order to get the insight in seasonal and spatial distribution of particle concentrations in Zagreb area a pilot investigation was performed at four sampling sites located at different parts of the town, for a month period during summer and again in winter season. The results of these investigations, together with the conclusions of several recent WHO projects and WHO meetings (WHO 2000, WHO 2002, WHO 2004) resulted in PM_{2.5} Zagreb network implementation. Samples are collected on daily basis and additional analyses of lead, cadmium, manganese, sulphates, nitrates, chlorides are performed, as well as the determination of sample total β -activity.

Clean Air For Europe (CAFE) programme Second Position Paper on Particulate Matter (2004) states that there is strong evidence to conclude that fine particles (PM_{2.5}) are more hazardous than larger ones (coarse particles) in terms of mortality and cardiovascular and respiratory endpoints in panel studies. Epidemiological studies on large populations have been unable to identify a threshold concentration below which ambient PM has no effect on health. WHO developed new exposure response relationship for PM_{2.5}. Paper recommends a range of values (12 to 20 µg/m³) for the integrated assessment procedure to identify an appropriate PM_{2.5} annual average limit value. Recommends value for PM_{2.5} around 35 µg/m³ (not to be exceeded more than 10% of the days of the year) as a starting point for consideration.

In this paper, the results of PM_{2.5} mass concentration obtained at Zagreb monitoring network during 2000-2005 period are presented and assessed.

2. MATERIALS AND METHODS

For the 1999-2004 period monitoring was performed at one sampling site located at northern residential part of town. Daily PM_{2.5} particle fraction samples were collected on 47 mm diameter Whatman Quartz filters from 55 m³ of ambient air by means of low volume samplers LVS3. Filters were preconditioned for 24 hours before and after the sampling in desiccators for low humidity. Particle mass is determined by weighing on microbalance Mettler Toledo MX-5. Concentration results are normalized to the air pressure of 1 bar and air temperature of 20 °C.

In 2005 PM_{2.5} monitoring started at five sampling sites. Sampling site located in the town center is characterized by dense traffic in street canyons and numerous individual heating appliances. Northern residential sampling site is characterized by medium traffic density and individual heating, western residential-industrial by dense traffic and individual heating, eastern residential-industrial, while southern sampling site is located in residential area characterized by dense traffic and district heating. For 2005 monitoring campaign filters are kept for 48 hours under ~50% relative humidity and temperature of 20 °C.

3. RESULTS AND DISCUSSION

Statistical parameters of PM_{2.5} concentrations for the 2000-2004 monitoring period are presented in table 1. Capture rate for 2000-2003 period was very high, though enabling good description of the results, while during the year 2004, due to the equipment failure and service, a number of concentration data for summer (5 June - 19 July) and again winter (12-31 December) are missing. No significant difference was found between measuring periods (one way ANOVA). Average concentrations ranged between 24.8 µg m⁻³ and 28.8 µg m⁻³, while interquartile range was between 14 µg m⁻³ and 36 µg m⁻³. Concentration distributions are skewed towards higher concentrations resulting in medians lower compared to the average concentrations.

Table 1. Statistical parameters of PM2.5 particle fraction concentrations (μgm^{-3})

| Year | 2000 | 2001 | 2002 | 2003 | 2004 |
|------------------------|-------------|-------------|-------------|-------------|-------------|
| N | 364 | 364 | 364 | 364 | 301 |
| C_{avg} | 27.2 | 26.8 | 26.5 | 28.8 | 24.8 |
| Median | 21.3 | 20.0 | 22.7 | 21.4 | 20.2 |
| C₂₅ | 14.1 | 14.4 | 15.0 | 13.9 | 14.3 |
| C₇₅ | 31.9 | 31.4 | 31.7 | 36.0 | 30.9 |
| C₉₈ | 80.3 | 82.6 | 79.2 | 96.0 | 69.7 |

To avoid the influence of accidental and/or unusual situations, 98th concentration percentiles are shown instead their maximum concentrations. As it is shown in Figures 1-5 distributions of yearly concentration sets could be best fitted to lognormal distribution (Kolmogorov-Smirnov), as is the usual case with air pollutant concentrations.

Mass ratio of PM2.5/PM10 was in the range 30-100 % and their distributions could be fitted to normal distribution. Variations are caused by weather conditions (rain, snow, wind, washout, air temperature and pressure) and show no seasonal dependence.

In table 2. average values and attributable standard deviations of PM2.5 proportions in PM10 for the year periods are given.

Table 2. Average values and standard deviations of PM2.5/PM10 ratio (%)

| Year | 2000 | 2001 | 2002 | 2003 | 2004 |
|----------------|-------------|-------------|-------------|-------------|-------------|
| N | 364 | 364 | 364 | 364 | 301 |
| Average | 68.5 | 72.9 | 71.3 | 71.3 | 73.3 |
| Std | 15.5 | 12.6 | 14.0 | 14.3 | 11.9 |

Results show that approximately 70 % of PM10 mass contribute particles in PM2.5 size range. Since the correlations of PM2.5 to health endpoints are much stronger compared to PM10 and almost all PAHs, heavy metal and anion content is found in PM2.5 particle fraction, monitoring of PM2.5 will probably replace PM10 monitoring in the future.

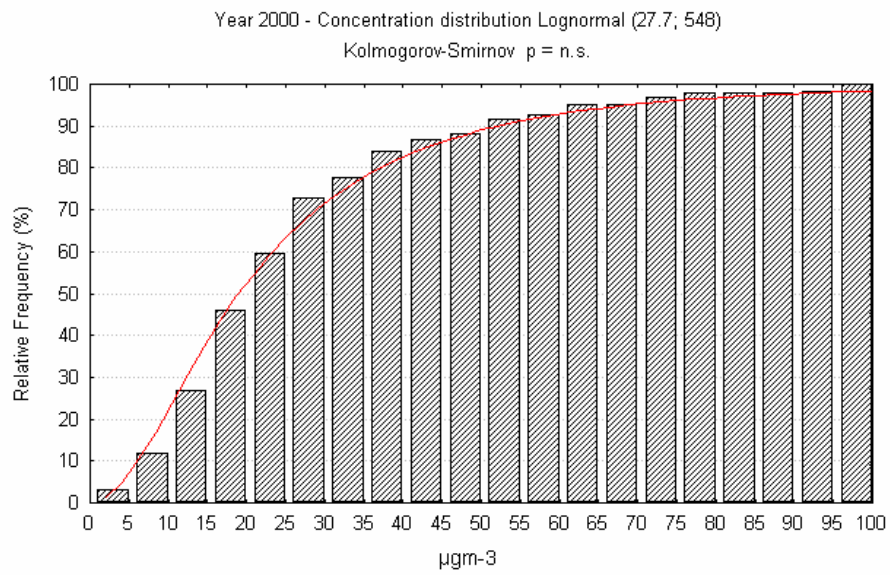


Figure 1. PM2.5 concentration distribution fitting – year 2000

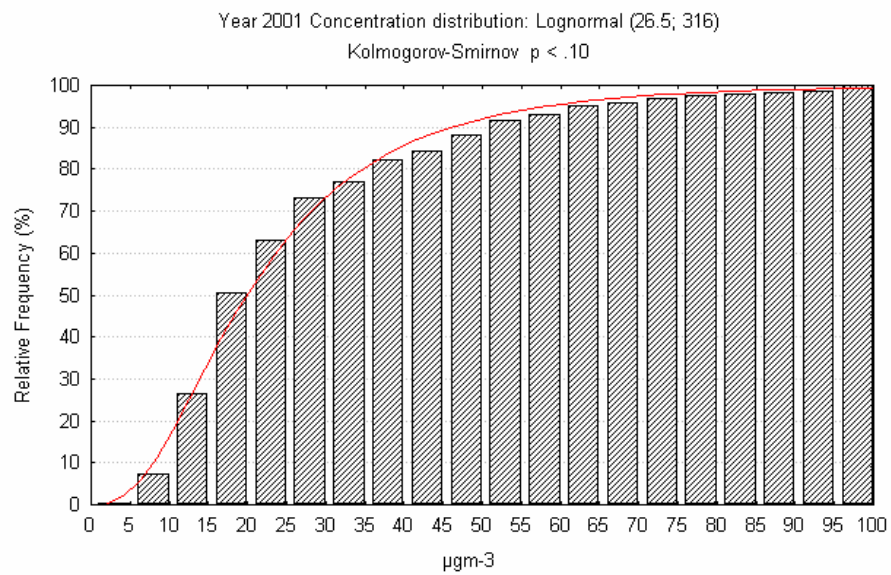


Figure 2. PM2.5 concentration distribution fitting – year 2001

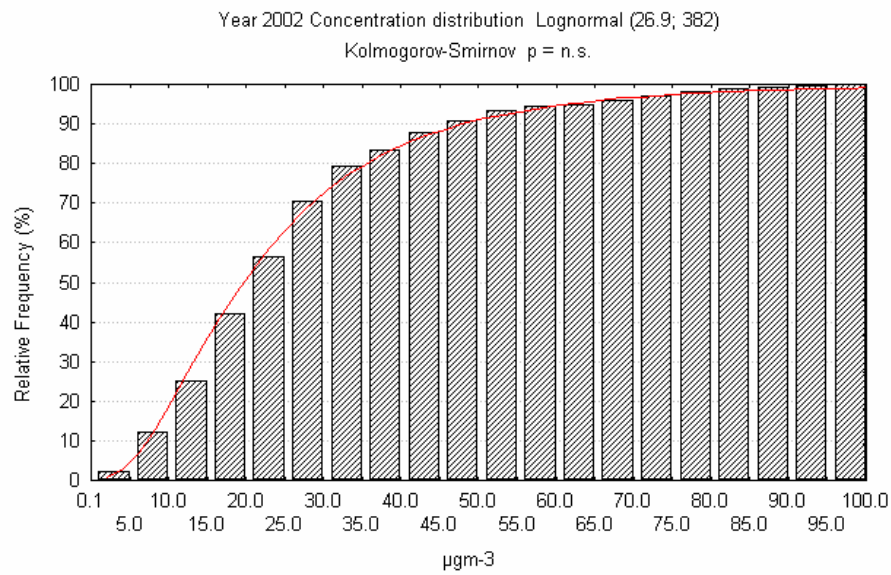


Figure 3. PM2.5 concentration distribution fitting – year 2002

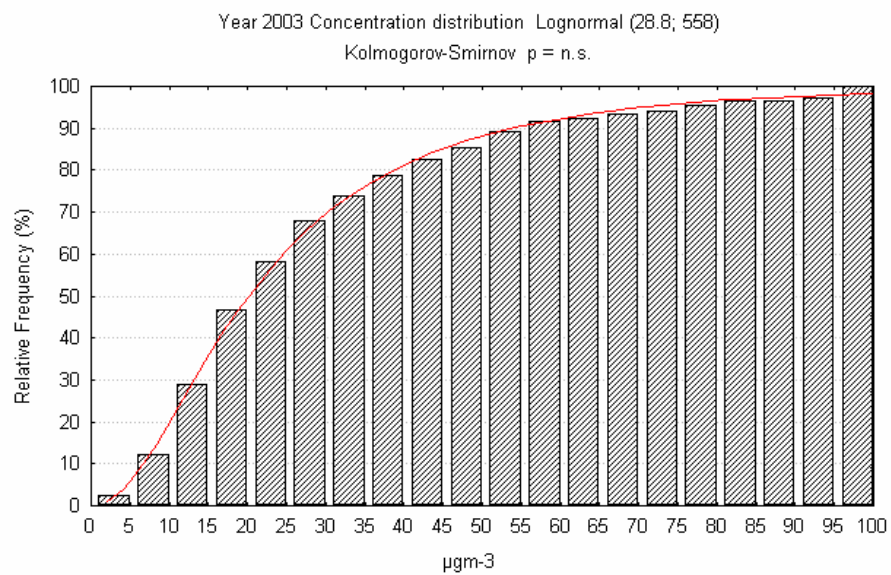


Figure 4. Concentration distribution fitting – year 2003

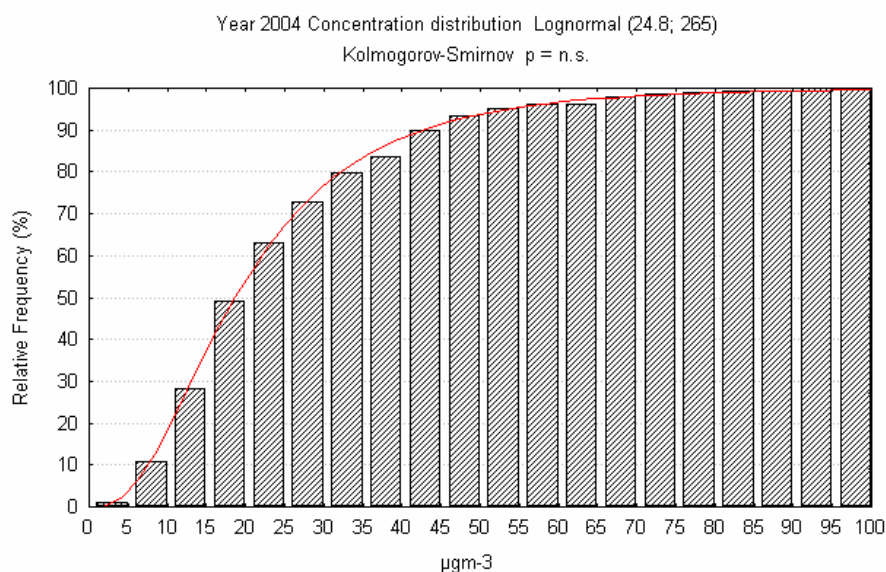


Figure 5. PM2.5 concentration distribution fitting – year 2004

Although there are no prescribed limit values, PM2.5 concentration levels measured in Zagreb could be compared to the proposed values (CAFÉ 2004). Annual averages for the whole monitoring period were much higher and out of proposed concentration range (12-20 μgm^{-3}). The percentages of measured concentrations higher than 35 μgm^{-3} are presented in table 3. Their frequencies are more than two times higher compared to the proposed one of 10 %.

Table 3. Percentage of the concentrations higher than 35 μgm^{-3} (%)

| Year | 2000 | 2001 | 2002 | 2003 | 2004 |
|------|------|------|------|------|------|
| % | 22 | 23 | 21 | 26 | 20 |

These comparisons allow us to conclude that PM2.5 concentration levels are elevated and the quality of Zagreb atmosphere regarding PM2.5 concentrations is not satisfactory.

Daily concentrations of PM2.5 particle fraction for the first six month period of year 2005 at all five sampling sites are presented at figure 6. Although the concentrations at all measuring sites follow the same trend, it could be noticed that somewhat higher concentrations were measured at sampling site south, and lower at the station located at the northern part of the town. It could be concluded that the fine particle fraction in Zagreb does not come primarily from the local source but that their transport over longer distances plays a significant role.

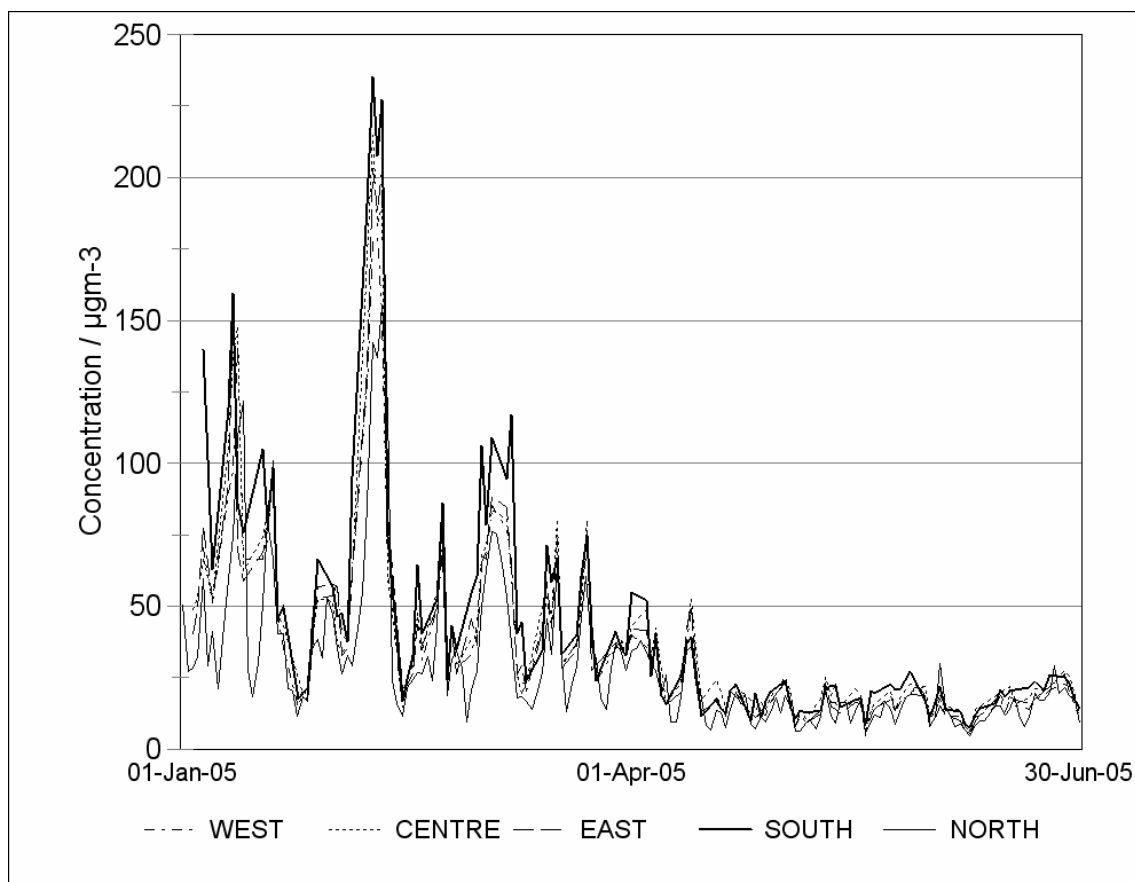


Figure 6. Trends of PM2.5 concentrations in Zagreb network (January – June 2005)

4. CONCLUSIONS

Concentration distributions and levels of PM2.5 particle fraction in Zagreb air do not differ significantly from year to year.

Air quality in Zagreb regarding PM2.5 particle fraction is unsatisfactory since the results obtained are higher compared to the proposed limit values.

Concentration levels do not depend on local pollutant sources only, and the transport of fine particles, because of their long airborne residence time, plays a considerable role, leveling the concentrations over a waste region.

In the future Zagreb monitoring network should abandon TSP monitoring and focus to PM10 and even more to PM2.5 particle fraction.

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