

5.04 PM₁₀, CO AND NO_x CONCENTRATIONS IN THE TUHOBIĆ ROAD TUNNEL, CROATIA

Ivan Bešlić1, Krešimir Šegal, Anica Šišović1 and Zvezdana Bencetić Klaić2

¹ Institute for Medical Research and Occupational Health, Zagreb, Croatia

² Andrija Mohorovičić Geophysical Institute, Faculty of Science, University of Zagreb, Zagreb, Croatia

INTRODUCTION

Airborne pollutants, and in particular, particulate matter (PM) continue to give a rise to concern as a result of their adverse effects on human health (D'Amato et al., 2001; Fisher et al., 2004; Stedman, 2004). A number of studies suggest that not all particles are equal with respect to health effects (e.g., Schwartz et al., 1996, Englert 2004). These findings have led to the introduction of size specific particulate mass concentrations, where PM₁₀ and PM_{2.5} correspond to thoracic and alveolar particles, respectively.

In urban areas, vehicular transport is recognized as an important source of, among other, PM₁₀, PM_{2.5} and trace gases, such as, carbon monoxide (CO) and nitrogen oxides (NO_x) (Gertler et al., 2000; Querol et al., 2001; Paoletti, 2003; Rodriguez et al., 2004). Therefore, various recent studies focus on emission factors, i.e. amounts of species emitted per vehicle km driven or per volume of fuel consumed (Abu-Allaban et al., 2003; Hausberger et al., 2003; Kristensson et al., 2004). In this study we investigated air quality conditions found in the in real-world traffic situations during a summertime touristic season, which is characterized with an intensive vehicular transport. Namely, we collected and analyzed data for a tunnel located on the main road connecting Croatian inland with the Northern Adriatic. Specifically, we examined a dependence of pollutant levels (PM₁₀, CO, NO and NO₂) on traffic density. Additionally, we investigated a relationship between PM₁₀ and trace gases concentrations. Weekend and weekdays data were analyzed separately due to noticeably different traffic densities.

EXPERIMENTAL

PM₁₀ particle fraction and trace gases CO, NO and NO₂ have been sampled during 6 weeks (21.07.2002 - 01.09.2002) in the third longest tunnel in Croatia (Tuhobić, 2140 m). It is located on the Zagreb-Rijeka road, with an inlet at $\varphi = 45^{\circ}19'39.5''$ and $\lambda = 14^{\circ}39'42.1''$. The tunnel has two lanes separated by the centreline, one for each direction. Eight mini portable air samplers (Air Metrics, USA) were placed in the centre of the tunnel, at the height of 1.70 m above the ground. PM₁₀ samples were collected on a Whatman quartz fibre filters with the diameter of 47 mm. Gases were collected in Tedlar bags. The ventilation system was operating during the measurement.

Weekend samples (from Friday at 8 LST to Sunday at 24 LST) were collected for 8-hour sampling intervals. During the weekdays, 4-hour cumulative samples were collected throughout the whole week. In other words, pollutant concentration for weekday for the sampling interval from 0 to 4 LST, for example, corresponds to an average 0-4 LST concentration for five consecutive days (Monday-Friday). For every sample corresponding number of vehicles was recorded. Data on particular vehicle specification and speed were not available. Out of the total 82 PM₁₀ samples 46 and 36 were collected during weekends and the weekdays, respectively. The total number of trace gases samples was 62, where 34 were collected during weekends.

RESULTS

A summary of the measurements is given in Table 1. They show that concentrations of all pollutants are on the average higher during the weekend compared to weekdays. The same is also valid for the diurnal variations of pollutant concentrations (Figure 1). This may be attributed to substantially higher average traffic density compared with the weekdays (about 730 and 420 vehicles per hour, respectively). Since concentrations at any time depend on the concentrations and other events that have taken place at previous time intervals (*Abu-Allaban et al.*, 2003), this implies lower background concentrations for the weekdays. In addition, for PM this also implies less intensive resuspension caused by vehicular traffic. The effects of the ‘history’ on pollutants concentrations are the most clearly illustrated with the pattern of the diurnal variation for the weekdays (Figure 1, left), where a time interval with maximum traffic density (16-20 LST) precedes the interval with maximum concentrations per vehicle (20-24 LST). Similarly, the time interval with minimum traffic density (0-4 LST) is followed by the interval with minimum concentrations of trace gases per vehicle and with substantial decrease in PM₁₀ concentration per vehicle (4-8 LST).

Table 1. Summary of measurements. E and D correspond to weekend and weekday samples (i.e. 8-hour and 20-hour sampling interval, respectively), N is the number of samples and nv is the average number of vehicles per sample.

	SAMPLE	N	MEAN	MEDIAN	MIN.	MAX.	ST. DEV.
PM ₁₀ (μG/M3)	E	46	272.3	285.8	35.8	749.2	140.4
	D	36	244.5	217.3	17.9	504.6	133.6
CO (MG/M3)	E	34	6.5	5.6	0.0	17.5	4.6
	D	28	4.1	4.4	0.0	11.3	3.4
NO (μG/M3)	E	34	513.7	485.1	233.2	1002.3	185.7
	D	28	308.4	303.5	175.5	466.3	87.7
NO ₂ (μG/M3)	E	34	1007.0	911.2	110.7	2498.9	645.5
	D	28	846.3	843.6	51.3	1873.7	522.9
NV (PM ₁₀)	E	46	5850.1	5865.5	1596.0	10267.0	2104.5
	D	36	8469.9	10316.0	2295.0	12700.0	3334.8
NV (TRACE GASES)	E	34	5846.1	5842.5	1596.0	10267.0	1994.1
	D	28	8310.3	9163.0	2536.0	11847.0	3286.2

To examine the relationship between the traffic density and pollutant concentrations, an approach of *Kukkonen et al.* (2001) was employed, where near-ground-level concentrations originate from local traffic and stationary sources, background contribution and the resuspension of particles from street surfaces. For the tunnel we may neglect the contribution of stationary sources. Further, we assume a linear dependency between the pollutant concentrations due to the local traffic and the traffic density:

$$C_i = A_i N + C_i^{\text{BG}}, \quad (1A)$$

$$\text{PM}_{10} = B N + \text{PM}_{10}^{\text{RES}} + \text{PM}_{10}^{\text{BG}}, \quad (1B)$$

where C_i and PM_{10} are the total (measured) near-ground-level concentrations of i -th trace gas and thoracic particles, respectively, n is the number of vehicles per hour, while a_i and b are constants to be determined empirically. The superscripts ‘bg’ and ‘res’ refer to the background pollution and resuspension of particles from street surfaces, respectively. A set of

corresponding regression equations is listed in Table 2, while a scatter plot for PM₁₀ vs. traffic density for all samples is illustrated in Figure 2.

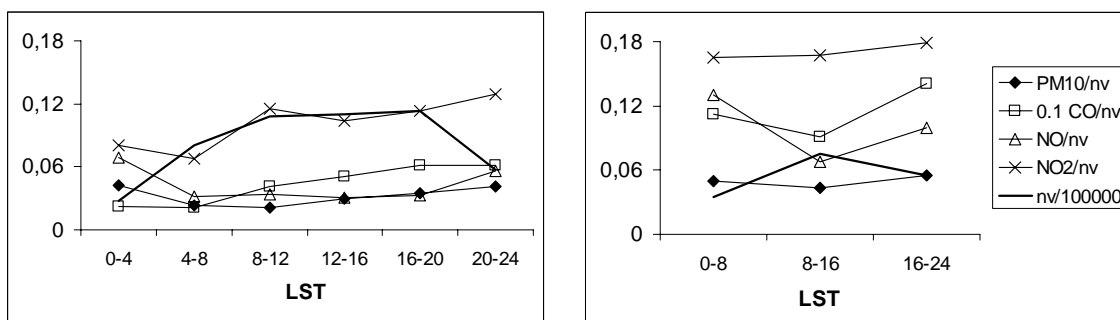


Figure 1. Diurnal variations of pollutant concentrations per vehicle ($\mu\text{g}/\text{m}^3$) and number of vehicles (nv) for weekdays (left) and weekend (right).

The best correlation between the PM₁₀ and trace gases was obtained for NO₂ (Figure 3), where correlation coefficients for all, weekend and weekday samples were following: 0.85, 0.83 and 0.91. Corresponding figures for CO were 0.53, 0.51 and 0.60, while for NO they were 0.25, 0.15 and 0.63, respectively. Further, assumed linear dependency between the concentrations of PM₁₀ and NO₂ ($\text{PM}_{10} = k \text{NO}_2 + \text{PM}_{10}^{\text{bg}} + \text{PM}_{10}^{\text{res}}$) yielded to $k = 0,19, 0,18$ and $0,22$ for all, weekend and weekdays, which agrees reasonably with the value $0,11$ obtained for urban NO_x caused by vehicular transport (Kukkonen *et al.*, 2001). Background PM₁₀ values for the tunnel were 101 (all), 108 (weekend) and 84 $\mu\text{g}/\text{m}^3$ (weekdays) again suggesting the influence of the pollution history on background levels.

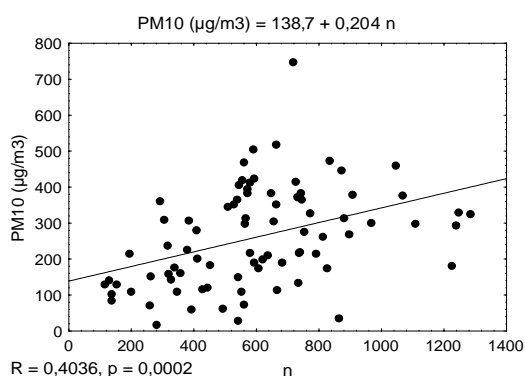


Figure 2. PM₁₀ vs. number of vehicles per hour (n) for all samples.

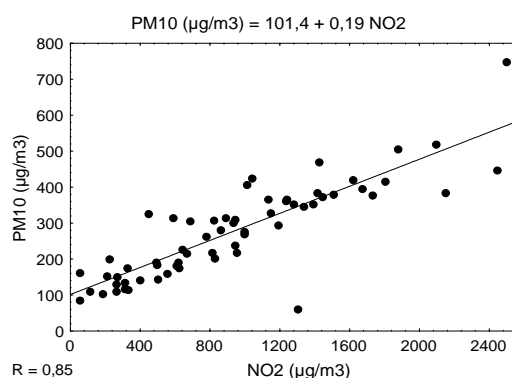


Figure 3. PM₁₀ vs. NO₂ for all samples.

Table 2. Dependence of pollutants concentrations ($\mu\text{g}/\text{m}^3$) on the traffic density for all (A), weekend (E) and weekday samples (D). N is the number of samples and n is the number of vehicles per hour. Correlation coefficients (R) which are significant at the level $p = 0.05$ are underlined.

SAMPLE	N	R	REGRESSION EQUATION
A	82	<u>0.40</u>	$\text{PM}_{10} = 138.7 + 0.204 N$
E	46	<u>0.37</u>	$\text{PM}_{10} = 129.0 + 0.196 N$
D	36	<u>0.56</u>	$\text{PM}_{10} = 53.7 + 0.451 N$
A	62	<u>0.37</u>	$\text{CO} = 1984.5 + 5.800 N$
E	34	0.14	$\text{CO} = 4716.4 + 2.500 N$
D	28	<u>0.53</u>	$\text{CO} = -509.9 + 11.000 N$
A	62	<u>0.42</u>	$\text{NO} = 251.7 + 0.288 N$
E	34	0.04	$\text{NO} = 490.1 + 0.032 N$
D	28	<u>0.46</u>	$\text{NO} = 206.6 + 0.245 N$
A	62	<u>0.36</u>	$\text{NO}_2 = 463.7 + 0.800 N$
E	34	0.21	$\text{NO}_2 = 596.0 + 0.562 N$
D	28	<u>0.65</u>	$\text{NO}_2 = -16.2 + 2.076 N$

CONCLUSIONS

Tunnel measurements offer the possibility of investigating real-world conditions, since they are representative for a huge variety of vehicles. Results of this study suggested a correlation between the pollutants concentrations and the traffic density, which is in contrast with the results *Abu-Allaban et al.* (2003) and *Martuzevicius et al.* (2004). However, apart from the fact that both above studies investigated open road data, in the first study a real-time concentrations, with the time resolution of 1 s, were measured, and, thus, they exhibited a strong ‘memory’ of previous concentrations and vehicle counts. On the other hand, second study employed a daily values, which were accompanied with low variations of traffic intensity.

Though correlation between pollution levels and traffic density was generally weak for all pollutants, it was stronger for weekday samples, i.e. for lower traffic densities. The latter, together with the pattern of diurnal variations of pollutants concentrations strongly suggests influence of pollution history on background values if the timescale < 1 day is considered. For the 8-hour samples investigated in this study, background contributions for PM_{10} , CO and NO_2 were of the same order of magnitude as the contributions from traffic, while for NO they were even about 20 times higher. This implies that a time-series data analysis is needed in further investigation.

PM_{10} concentrations exhibited the strongest correlation with NO_2 (better than with NO_x). Assumed linear dependency $\text{PM}_{10} = k \text{NO}_2 + \text{PM}_{10}^{\text{bg}} + \text{PM}_{10}^{\text{res}}$ resulted in $k \approx 0.2$, which reasonably agrees with the results of *Kukkonen et al.* (2001).

ACKNOWLEDGEMENTS

This study was done under the ongoing SILAQ project (Sofia Initiative for Local Air Quality, initiated in 1995).

REFERENCES

Abu-Allaban, M., J. A. Gillies, and A. W. Gertler, 2003: Application of a multi-lag regression approach to determine on-road PM_{10} and $\text{PM}_{2.5}$ emission rates. *Atmos. Environ.*, **37**, 5157-5164.

- D'Amato, G., G. Liccardi, M. D'Amato and M. Cazzola, 2001: The role of outdoor air pollution and climatic changes on the rising trends in respiratory allergy. *Respir. Med.*, **95**, 606-611.
- Englert, N., 2004: Fine particles and human health-a review of epidemiological studies. *Toxicol. Lett.* (in press).
- Fisher, P. H., B. Brunekreef and E. Lebret, 2004: Air pollution related deaths during the 2003 heat wave in the Netherlands. *Atmos. Environ.*, **38**, 1083-1085.
- Gertler, A. W., J. A. Gillies and W. R. Pierson, 2000: An assessment of the mobile source contribution to PM₁₀ and PM_{2.5} in the United States. *Water Air Soil Pollut.*, **123**, 203-214.
- Hausberger, S., J. Rodler, P. Strum and M. Rexeis, 2003: Emission factors for heavy-duty vehicles and validation by tunnel measurements. *Atmos. Environ.*, **37**, 5237-5245.
- Kristensson, A., C. Johansson, R. Westerholm, E. Swietlicki, L. Gidhagen, U. Wideqvist and V. Vesely, 2004: Real-world traffic emission factors of gases and particles measured in a road tunnel in Stockholm, Sweden. *Atmos. Environ.*, **38**, 657-673.
- Kukkonen, J., J. Harkonen, A. Karppinen, M. Pohjola, H. Pietarila and T. Koskentalo, 2001: A semi-empirical model for urban PM₁₀ concentrations, and its evaluation against data from an urban measurement network. *Atmos. Environ.*, **35**, 4433-4442.
- Martuzevicius, D., S. A. Grinshpun, T. Reponen, R. L. Gorny, R. Shukla, J. Lockey, S. Hu, R. McDonald, P. Biswas, L. Kliucininkas and G. LeMasters, 2004: Spatial and temporal variations of PM_{2.5} concentration and composition throughout an urban area with high freeway density – the Greater Cincinnati study. *Atmos. Environ.*, **38**, 1091-1105.
- Paoletti, L., B. De Berardis, L. Arrizza, M. Passacantando, M. Inglessis and M. Mosca, 2003: Seasonal effects on the physico-chemical characteristics of PM_{2.1} in Rome: a study by SEM and XPS. *Atmos. Environ.*, **37**, 4869-4879.
- Querol, X., A. Alastuey, S. Rodriguez, F. Plana, C. R. Ruiz, N. Cots, G. Massague and O. Puig, 2001: PM₁₀ and PM_{2.5} source apportionment in the Barcelona Metropolitan area, Catalonia, Spain. *Atmos. Environ.*, **35**, 6407-6419.
- Rodriguez, S., X. Querol, A. Alastuey, M.-M. Viana, M. Alarcon and E. Mantilla, 2004: Comparative PM₁₀-PM_{2.5} source contribution study at rural, urban and industrial sites during PM episodes in Eastern Spain. *Sci. Total. Environ* (in press).
- Schwartz, J., D. W. Dockery and L. M. Neas, 1996: Is daily mortality associated specifically with fine particles? *J. Air Waste Manage.*, **46**, 927-939.
- Stedman, J. R., 2004: The predicted number of air pollution related deaths in the UK during the August 2003 heat wave. *Atmos. Environ.*, **38**, 1087-1090.