# URB-URBAN AND RURAL AIR QUALITY

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## URBAN AND RURAL AIR QUALITY

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# SOURCE APPORTIONMENT OF VOLATILE ORGANIC COMPOUNDS IN 3 FRENCH SITES BY DESCRIPTIVE ANALYSES AND RECEPTOR MODELLING

**Fabien Troussier, Nadine Locoge and Jean Claude Galloo**

Ecole des Mines de Douai, Department of Chemistry and Environment, and Central Laboratory of Air Quality Survey (LCSQA), France, troussier@ensm-douai.fr

## ABSTRACT

Large quantities of volatile organic compounds (VOC) are emitted into the atmosphere by different sources such as the automobile traffic, residential and biogenic sources or industrial emissions. Consequently, it is interesting to evaluate the temporal and spatial contribution of the different sources to the ambient concentrations. In France, VOC measurements are carried out by air quality monitoring networks. Continuous hourly measurements of 31 VOC from $C_2$ to $C_9$ were performed since 2001 in 3 regional networks (Marseille, Strasbourg and Grenoble). Preliminary results show that Marseille site present an average VOC concentration two to three times higher than the other sites and Grenoble site seems to be the most sensitive to seasonal variation. Simple mathematical and descriptive analyses (daily variation and wind roses) have been made and, so, a great majority of the compounds seems to result from the sources related to the urban traffic. Some compounds have singular behaviour: notably, for Marseille site, benzene and cyclohexane concentrations are very influenced by a far chemical factory emission. Lastly, receptor modelling results confirm the substantial prevalence of the urban traffic sources among the involved sources whatever the site (between 35 and 60% of total emissions) and some sources such as automobile exhaust source have a clear daily variation.

**Key Words:** volatile organic compound, source attribution, data analysis, receptor models

## 1. INTRODUCTION

Appendix VI of the European directive on ozone 2002/3/EC specifies that the measurement of the ozone precursors has to relate to a "suitable" list of Volatile Organic Compounds (a list of 31 VOC is recommended). VOC coupled with $NO_x$ are the most important precursors of tropospheric ozone formation and VOC are especially important in urban areas because their reaction with the hydroxyl radical (OH) plays a critical role in atmospheric photochemical reactions. Many studies present several sites (Colòn and al., 2001; Tanaka and Samukawa, 1996) or long measuring campaigns (Hakola and al., 2003) but few have three different sites at three different latitudes with hourly measurements since many years as describing below. Since 2001, the hourly measurement of the NMHC has been started in several

French air quality monitoring networks and consequently a large VOC concentration databases is now available. Each site presents a specific typology: urban, periurban and rural site under industrial influence for Marseille, Strasbourg and Grenoble networks respectively. Three databases were made up both with VOC data concentrations and weather data collected on the three sites. The mains sources influencing the ambient VOC concentrations at each site of measurement were systematically indexed. To date, the three databases join more than 1.200.000 data of VOC concentration. For this study, we have done in first time descriptive analyses and in second time receptor modelling analysis.

## 2. SAMPLING AND CHEMICAL ANALYSIS METHODS

The analytical method used for VOC measurements was developed in the Chemistry and Environment department of the Ecole des Mines de Douai (EMD) in the framework of Central Laboratory of Air Quality Survey (LCSQA). It is based on:

1-preconcentration of volatile organic compounds of ambient air sample on a trap filled with a mixture of adsorbents maintained at - 30°C thanks to a system of Peltier cooling, followed by a thermal desorption until a temperature of 300°C (Turbo Matrix Auto System provided by Perkin Elmer)
2-analysis by gas chromatography (GC Perkin Elmer) using a dual capillary columns system (CP Sil 5CB, 50 m x 0,25 mm, and Plot $Al_2O_3/Na_2SO_4$, 50 m x 0,32 mm)
3-detection by FID (flame ionisation detector) whose answer is proportional to the effective carbons number of the detected molecule (Tranchant et al., 1995).

For more explanation, the analytical procedure was described in details in a previous paper (Badol et al., 2004).

The measuring device produces large numbers data each day (744 per 31 measured VOC). It is essential to apply check procedures quality in order to validate these data:
1-Checking of the identification of the peaks
Measurements on the long term often involve a drift of the retention time, which can imply misidentifying. Thus peaks assignment is checked for any chromatogram while representing the evolution of the peaks retention time according to time. A rupture in the curve of retention times is due to a misidentifying (Veillerot et al., 1998).
2-Reproducibility
The study of reproducibility is done by the follow-up of the evolution of the peaks surfaces of the target compounds during the checking of the calibration (analyses of a standard mixture every two weeks). It is generally not tolerated a difference higher than 10% between the unit of the values of the surfaces of each injection compared to a reference value. Beyond the tolerance of 10%, the analytical device is examined in order to locate the dysfunctions (Badol et al., 2004).
3-Intercomparison
Each year, canisters containing a gas mixture of VOC are analysed and circulate between the networks and the “Ecole des Mines de Douai” whose values are taken as reference. Any important difference between measurement and the actual value in

the canisters can be proposed. Thus the networks can validate their results and locate possible dysfunctions of their analytical device (Borbon et al., 2003).

## 3. FIELD STUDIES AND SAMPLING SITES

For this work, we systematically listed the whole of the sources of VOC likely to influence the three receptor sites located near 3 large French cities: Marseille, Strasbourg and Grenoble.

For Marseille, VOC monitoring is led on the station of Prado (urban station, 43°27'N, 5°13'E) located in the Southern half of the city near a busiest artery of Marseille (27000 vehicles per day). Near the site a lot of trade such as laundries or petrol stations can influence ambient VOC concentrations. In addition, two far important factories sites in terms of VOC emissions will be likely to have a sufficient influence to be detected on the sampling site:

- a chemical factory specialized in the manufacture of an amino acid (approximately 10 km in the East of the site)
- the port of Marseille which extends between 3 and 11 km in the North-West from the receptor site and which has an varied activity like naval repair or freight transport

For the zone covered by Strasbourg network, the monitoring of the VOC is led in Schiltigheim (48°21'N, 7°25'E), town of approximately 31000 inhabitants being next to Strasbourg (North-West). It is a periurbain station located in office and school zone. Near the site, few specific local sources seem likely to influence VOC measurements. However, the presence in the vicinity (approximately 50 meters) of the monitoring site of a small gasoline depot is likely to influence VOC concentrations. Strong sources of VOC (such as oil or chemical industries, printing works, etc) but relatively far away from the site of measurement (up to 10 kilometers) are likely to be able to influence the measured concentrations. Thus, some industrial emissions seem likely to have a sufficient contribution to be measured when the dominant winds blows in direction of the sampling site.

For the zone covered by Grenoble network, the 31 VOC monitoring is led on a rural station under industrial influence: the station of Champagnier (45°06'N, 5°43'E). The monitoring site can be described as slightly influenced. Indeed, it does not have activities recognized like strongly transmitting of VOC (laundry, garage, petrol station…) except the relatively limited automobile traffic (proximity of a little attended road). Consequently, of share its geographical situation, the station could be subjected mainly to the action of a near factory site which produces mainly polychloroprene rubber. According to the direction of wind, the site could be also influenced by the emissions of a chemical pole (South-eastern) or the emissions of far important industries (North-western, about 5 kilometers).

## 4. CHEMICAL MASS BALANCE RECEPTOR MODEL

For this study, we have used the Chemical Mass Balance receptor (CMB, Watson et al., 1994, 1998, 2001). It consists of a least-squares solution to a set of mass-balances equations that express each receptor chemical concentration as a linear sum of products of source profile abundances and source contribution. Mathematically the system is written:

$$C_i = \sum_{j=1}^{J} F_{ij} \cdot S_j, \quad i = 1,...,I \tag{1}$$

where $C_i$ is the concentration of compound $i$, $F_{ij}$ is mass fraction of compound $i$ from source $j$ (weighting percentage), $S_j$ is the mass concentration from source $j$ (µg/m$^3$), J is the number of sources and I is the number of compounds.
In contrast to other receptor models (PMF and UNMIX for example), which extract source compositions from the data, CMB requires the user supply source profiles. Also in contrast to the other models, CMB is applied separately to each observation, rather than operating on the data set as a whole. CMB is largely used for environmental data and produce generally good results for VOC data (Vega et al, 2000; Miller et al., 2002). Consequently, according to our good knowledge of field studies (see section 3), we have chosen to use this receptor model.

## 5. RESULTS

### 5.1. Preliminary results

First, for each site, the sum of the medians and means of the 31 VOC were calculated (see table 1). Thus the exposure of sites to the VOC can be quantified. The site of Marseille presents an average VOC concentration two to three times higher than the sites of Grenoble and Strasbourg in agreement with its urban localisation. Strasbourg and Grenoble show similar lowest concentrations. For the first quoted, the explanation lies in the fact that it is located in a little village subjected to few VOC emissions sources. For Strasbourg, the site area corresponds in a fairly inhabited zone, made up of offices, small companies and school establishments. The site of measurement is thus far away from the principal sources of VOC and will be subjected punctually to the emissions of distant industries.

Table 1: Sums of medians and means (µg/m$^3$) of the 31 targets VOC

| Marseille | | Strasbourg | | Grenoble | |
|---|---|---|---|---|---|
| median | mean | median | mean | median | mean |
| 66.22 | 94.76 | 26.05 | 39.67 | 22.71 | 36.00 |

Secondly, a substantial difference in the distribution of the involvement of the measured total concentrations of each VOC family is noticed from the winter months to the summer ones (see table 2). For the site of Grenoble, an increasing of 11% for aromatics family and a decreasing of 9% for alkenes family is observed during the

passage of the winter to the summer months and, so, it is possible to affirm that this site is the most sensitive to seasonal variation. Its rural aspect and the proximity of mountainous massif responsible for very cold winter temperatures (about 1°C on average for a winter day) require higher winter heating. The reduction of olefinic hydrocarbons percentage (25% in winter and 16% in summer) would be directly related to the residential heating whose emissions profiles show a prevalence of the alkenes family (GENEMIS profile available on www.ier.uni-stuttgart.de). Likewise the increase of aromatics compounds percentage can be see as the increase of solvent evaporation source which profile is made up almost exclusively by aromatics compounds (Passant, 2002). For Strasbourg site, we have almost the same phenomenon: decrease of alkenes percentage and increase of aromatics compounds percentage during the passage of the winter months to the summer ones. The same argument as Grenoble can be developed but we can also think that important variation of wind direction with season is also an important factor. In fact, if winter and summer wind roses of Grenoble are almost similar, Strasbourg ones are very different.

Table 2: Seasonal variation of medians sums (µg/m$^3$) of each family compound

| Family compound | Marseille | | Strasbourg | | Grenoble | |
|---|---|---|---|---|---|---|
| | Winter | Summer | Winter | Summer | Winter | Summer |
| Alkenes + acetylene | 13% | 11% | 23% | 17% | 25% | 16% |
| Alkans | 39% | 43% | 51% | 50% | 43% | 41% |
| Aromatics | 48% | 46% | 26% | 33% | 32% | 43% |

### 5.2. Spatial and temporal analysis

The analyses of the daily and seasonal variations and concentration roses were then carried out for the whole of the compounds. Generally, a great majority of compounds exhibit a shape with a “double wave”: first sharp maximum in the morning and a second one in the afternoon both corresponding to traffic rush hours (see figure 1, ethylbenzene measured on Strasbourg site). Thus, the urban traffic seems to be the principal source of the majority of compounds.

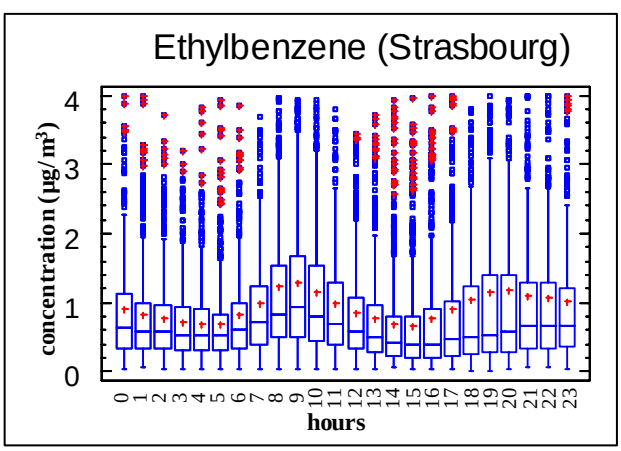


Figure 1: “Double wave” profile of majority of compounds

However, on each site, some compounds showed a singular behaviour (in this study only one singular behaviour were developed). For example, daily variation analyses show high night values for benzene and cyclohexane measured on the site of Marseille (see figure 2, benzene profile). According to this very singular profile (see majority of compounds profile), winds roses were looked with much meticulousness. These ones exhibit high concentrations for a wind direction between 50° and 80°. To explain these observations, it is necessary to know the localisation of Marseille: in the South of France and near the Mediterranean Sea. During the night, the ground cools quickly while the sea keeps approximately the same temperature. It results a land breeze, which blows from East to West on Marseille. Consequently, in the light of these information, the high night values of benzene and cyclohexane can be allotted to emissions related to a factory specialized in the chemistry located in the East of the city (see field studies and sampling sites section).

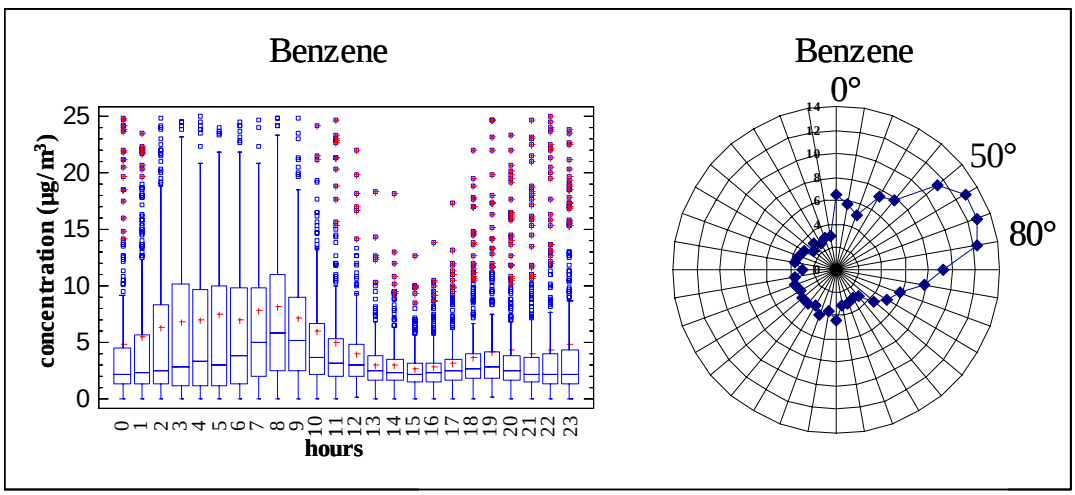


Figure 2: Daily and concentration roses analysis of benzene measured on Marseille site

### 5.3. Receptor modelling analysis

These basic descriptive analyses seen above have been only the first stages necessary to the attribution of the emissions sources for the measured concentrations. Thus CMB and PMF models were applied on the three databases. In this study, only CMB results are shown. Discriminations between summer and winter data have been done previously. Thus monthly, summer and winter runs are carried out for the three sites. For CMB modelling, the whole of sources profiles likely to influence concentrations measured on the three sites (see section 3) were tried. Whatever the season and the site, very few sources seem able to influence ambient concentrations. There are 5 common sources (vehicle exhaust, gasoline evaporation, leaks of natural gas and GPL, biogenic and domestic heating) to which we must add:

- rubber factory source for Grenoble
- printing works and solvent sources for Strasbourg
- printing works/ink, solvent and amino-acid factory sources for Marseille

For the three sites, during the passage of the summer to the winter months, we can observe a variation in the percentage explained by each emission source to the total concentration. Whatever the site, vehicle exhaust is the most important source (35 to 61% of the total concentration depending of the season and the site, see table 3). Despite Marseille site is the most submit to urban traffic (27000 vehicles per day), surprisingly, relative percentage of vehicle exhaust source of Marseille is the lowest of the three sites. Relative percentage of vehicle exhaust source depend not only of urban traffic intensity but also of the number and the intensity of the others sources on which the receptor site is submitted. Thus, the urban situation of the Marseille site, with much of small punctual sources, makes that this one presents the lowest relative percentage for vehicle exhaust source.

Table 3: Seasonal relative percentage of vehicle exhaust source on the three sites

| Marseille | | Strasbourg | | Grenoble | |
|---|---|---|---|---|---|
| summer | winter | summer | winter | summer | winter |
| 35.4% | 41.1% | 46.0% | 53.8% | 58.9% | 61.4% |

Concerning the daily evolution of the vehicle exhaust source, CMB modelling exhibit a shape with a “double wave” with first sharp maximum in the morning and a second one in the afternoon for each site, whatever the season (see figure 3).

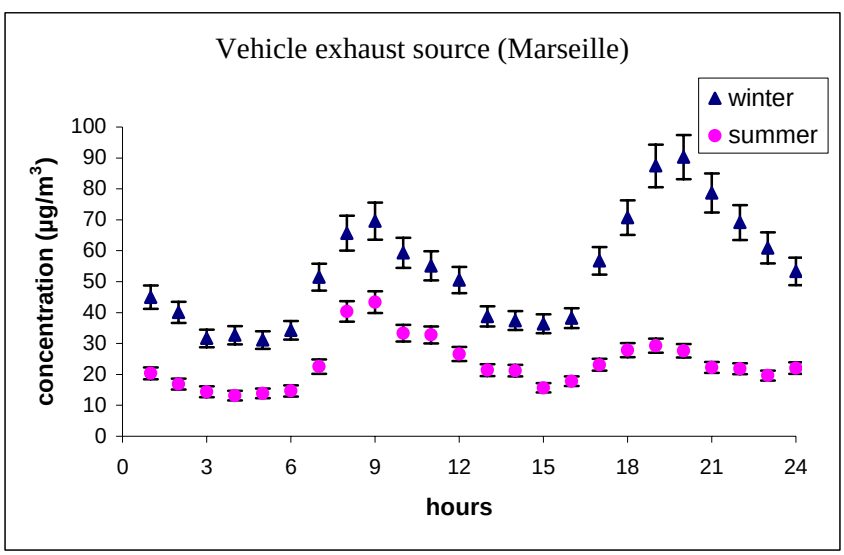


Figure 3: Summer and winter daily concentration variation of vehicle exhaust source of Marseille site

Only this source presents this behaviour. The same daily variation could be characterised for the daily variation of the majority of compounds concentrations. This result confirms the assumptions of section 5.2: majority of compounds stems from urban traffic and particularly from the vehicle exhaust (notably for Grenoble and Strasbourg site). For Marseille site, this assertion seems more debatable because

the part of the vehicle exhaust source decreases until 35.4% in summer. Consequently, the importance of this source remains sufficiently large to influence daily profile of the majority of compounds. But, we can see from this result that the only one examination of the daily profile is not sufficient to affirm that the majority of compounds comes principally from urban traffic and particularly from the vehicle exhaust. Winter modelled concentrations of vehicle exhaust source are stronger than the summer ones and the summer and winter profiles show a sensible difference for the second wave (see figure 3). Besides, the hourly source variation, scattering dispersion can have also an important role for the scale of measuring concentrations. Thus, the seasonal difference observed can be explained by a night boundary layer for winter decreasing mixing layer and increasing the concentrations and a diurnal boundary layer for summer increasing mixing layer and, so, decreasing the concentrations (Seinfeld and Pandis, 1997).

Daily evolution of the whole of source can be represented according to CMB modelling. Each of them has a singular evolution. For example, the source representing the amino-acid factory of Marseille has a singular profile with high night and weak constant diurnal concentrations (see figure 4). A very similar profile has been shown in section 5.2. for two compounds (benzene and cyclohexane). Consequently, according to their lifetime (4.7 days and 20 hours for benzene and cyclohexane respectively) and their transport time (about one hour for land breeze of 2m/s) these compounds are principally allotted to emissions related to the factory specialized in the chemistry located in the East of Marseille.

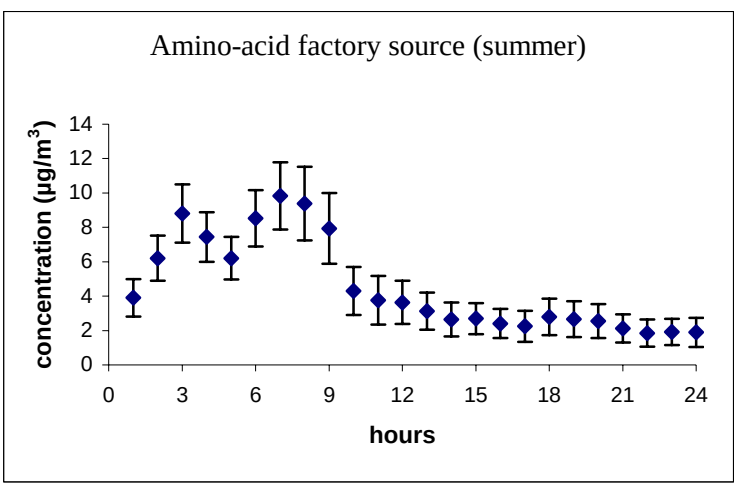


Figure 4: Daily concentration variation of amino-acid factories source on Marseille site

Lastly, for Grenoble site, we can see the increase of evaporative source (EVAP + RUBBER) during the passage of the winter to the summer months and the lost of heating source (see figure 5). The disappearance of this source characterised by a strong prevalence of olefinic family confirms the assumption given to explain “preliminary results” (section 5.1)

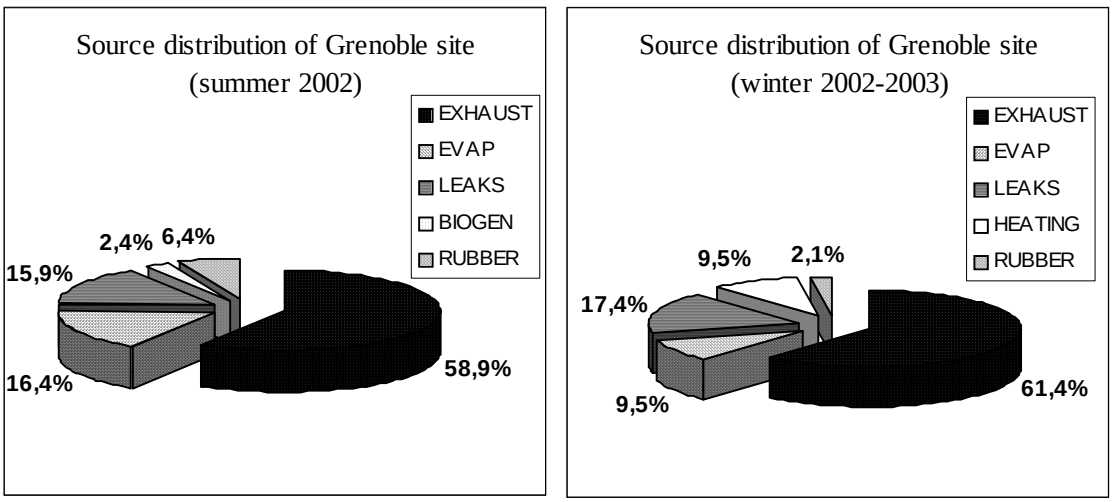


Figure 5: Seasonal variation of source distribution of Grenoble Site. Sources: EXHAUST = vehicle exhaust, EVAP = gasoline evaporation, LEAK = leak of natural gas + liquefied petroleum gas vapour, HEATING = domestic heating, BIOGEN = isoprene emission from biogenic sources, RUBBER = rubber factory

## 6. CONCLUSION

Many analyses have been made on three VOC databases coming from three French regional networks. Preliminary, spatial and temporal analyses have been made. Seasonal study of relative percentage variation of each family compound allows affirming that the site of Grenoble is the most sensitive to seasonal variation. Most of compounds seem to come, for the most part, from vehicle exhaust source. In addition, we see that the simple descriptive mathematical and statistical methods allow a partial attribution of the VOC sources in particular for the compounds resulting mainly from the industrial sources. VOC daily and seasonal distributions are controlled by a combination of emission factors, dispersion conditions and respective lifetime of the compounds. Thus, meteorological conditions can have an important effect and singular or exceptional wind direction can bring back far factories emissions. Receptor model have also been used. The results confirm the substantial prevalence of the urban traffic source (especially for Grenoble site) among the involved sources. In addition, in the summer months, evaporation sources can have a great importance (up to 50% of total VOC emission according to the receptor site). Lastly, some sources have a clear daily and/or seasonal variation. Thus, some assumptions formulated with preliminary, temporal and spatial analysis could be checked.

## 7. ACKNOWLEDGEMENTS

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# PM2.5 COMPOSITION IN MILAN (ITALY)

**G. Lonati, M. Giugliano and S. Ozgen**

Department of Environmental, Hydraulic, Infrastructures and Surveying Engineering
Politecnico of Milan, 20133, Milan, Italy

## ABSTRACT

In sampling campaigns -carried out by means of a high-volume gravimetric sampler- performed between August 2002 and December 2003, 24-h PM2.5 samples have been collected at an urban background site in downtown Milan and analysed for elemental and organic carbon, ionic species (i.e., chloride, nitrates, sulphates and ammonium) and some elemental species. Chemical speciation data are evaluated also in terms of primary and secondary components of fine particulate matter, and in particular SOA and the primary contribution from traffic are estimated.

**Key words:** PM2.5, SOA, OC/EC, crustal matter

## 1. INTRODUCTION

In Lombardy, Italy's most industrialized region, air quality standards for PM10 are frequently exceeded, especially in Milan, the main city of the region, and in its metropolitan area, where about three millions inhabitants live. The first Daughter Directive (1999/30/EC) to the Air Quality Framework Directive (96/62/EC) that lays down limit values for PM10 does not establish a limit value for PM2.5 but prescribes the installation and the operation of measuring stations to supply data on PM2.5 concentrations. CAFE (Clean Air For Europe) Working Group on particulate matter reconsidered in the II Position Paper on Particulate Matter the existing limit values for PM10 set by the First Daughter Directive. The Working Group recommended the use of PM2.5 rather than PM10 as the principal metric for assessing exposure to particulate matter in the light of World Health Organisation's (WHO) statement that "fine particles (commonly measured as PM2.5) are strongly associated with mortality and other endpoints such as hospitalization for cardiopulmonary disease". The Working Group suggested an annual average PM2.5 limit value in the range of 12-20 $\mu g\ m^{-3}$ and a 24-h average limit value around 35 $\mu g\ m^{-3}$ not to be exceeded more than 10% of the days of the year. As at the present moment in Milan information on the fine fraction of PM10 is scarce, the goal of this study is to define the composition and the contributions of various species to PM2.5 bulk mass in Milan.

## 2. PM2.5 SAMPLING

The monitoring site was Via Messina downtown Milan (UB site) which can be considered an urban background site not directly exposed to traffic emission. Sampling campaigns were performed from August 2002 through December 2003: 118 24-h samples have been collected and analyzed for elemental and organic

carbon, ionic species (i.e., chloride, nitrates, sulfates and ammonium) and elemental species (sulfur, potassium, iron, copper, zinc, lead, titanium, vanadium).

The sampling of PM2.5 has been carried out by means of a high-volume (30 $m^3$ $h^{-1}$ flow rate) gravimetric sampler DIGITEL DA-80H, equipped with PM2.5 cut-off inlet and 150 mm quartz filters (www.digitel-ag.com). Specific experiences with high-volume samplers support the hypothesis, assumed in this work, that negative and positive artifacts coexist with no significant variation in 24-h mass concentration (Tsai and Perng, 1998).

The PM2.5-loaded filters have been punched in three portions for separate analytical determination of ionic, carbon and elemental species. The carbon species, elemental carbon (EC) and organic carbon (OC), have been determined by Thermal-Optical Transmission (TOT) method (Birch and Cary, 1996; Schauer et al., 2003) at the Sunset Laboratory (www.sunset.com). The ionic components ($Cl^-$, $NO_3^-$, $SO_4^{2-}$, $NH_4^+$) have been ultrasonically extracted from the filter sample with 20 ml of deionized water for 30 minutes and then determined by means of high pressure liquid chromatography technique (HPLC). Elemental composition was determined by Energy Dispersive X-Ray Fluorescence (XRF) spectrometer SPECTRO X-LAB 2000 (www.spectro.com). Different targets (Mo, $Al_2O_3$, Highly Pyrolytical Orientated Graphite - HOPG) were used as polarizer or secondary target.

## 3. RESULTS AND DISCUSSION

At the UB site 24-h concentration levels of PM2.5 are in the 7.8-133.4 µg $m^{-3}$ range, with an annual average of 40.0 µg $m^{-3}$. The analysis of seasonal data points out relevant differences in concentration levels between cold and warm seasons (October to March and April to September, respectively); the highest concentrations are usually observed in the cold season, while the lowest concentrations tend to occur in the warm one.
The seasonal distributions differ significantly (Table 1): during the warmer months 59% of the PM2.5 concentration data are within the range of 20 to 40 µg $m^{-3}$, almost all the data are lower than 60 µg $m^{-3}$ ($98^{th}$ percentile = 46 µg $m^{-3}$) and none exceeds 80 µg $m^{-3}$. Conversely, 72% of the colder months' observations are higher than 40 µg $m^{-3}$ (39 samples out of 54) and 10% exceeds 100 µg $m^{-3}$.

Table 1 Percentiles of 24-h average PM2.5 concentrations (µg $m^{-3}$ @ 20°C and 101.3 kPa)

| | Annual | Warm | Cold |
|---|---|---|---|
| Minimum | 7.8 | 7.8 | 12.2 |
| $25^{th}$ percentile | 22 | 19 | 37 |
| $50^{th}$ percentile | 30 | 23 | 56 |
| $75^{th}$ percentile | 54 | 28 | 69 |
| $90^{th}$ percentile | 72 | 37 | 100 |
| $98^{th}$ percentile | 111 | 46 | 129 |
| Maximum | 133.4 | 66.6 | 133.4 |

The distribution of PM2.5 24-h concentrations is well fitted by a lognormal model (Figure 1), characterized by a geometric mean of 33.2 µg $m^{-3}$ and by a geometric standard deviation of 1.83. On seasonal basis, lognormal fitting results in a geometric mean of 22.8 µg $m^{-3}$ and in a geometric standard deviation of 1.47 for the warm season; corresponding figures for the cold season data set are 51.8 µg $m^{-3}$ and 1.66.

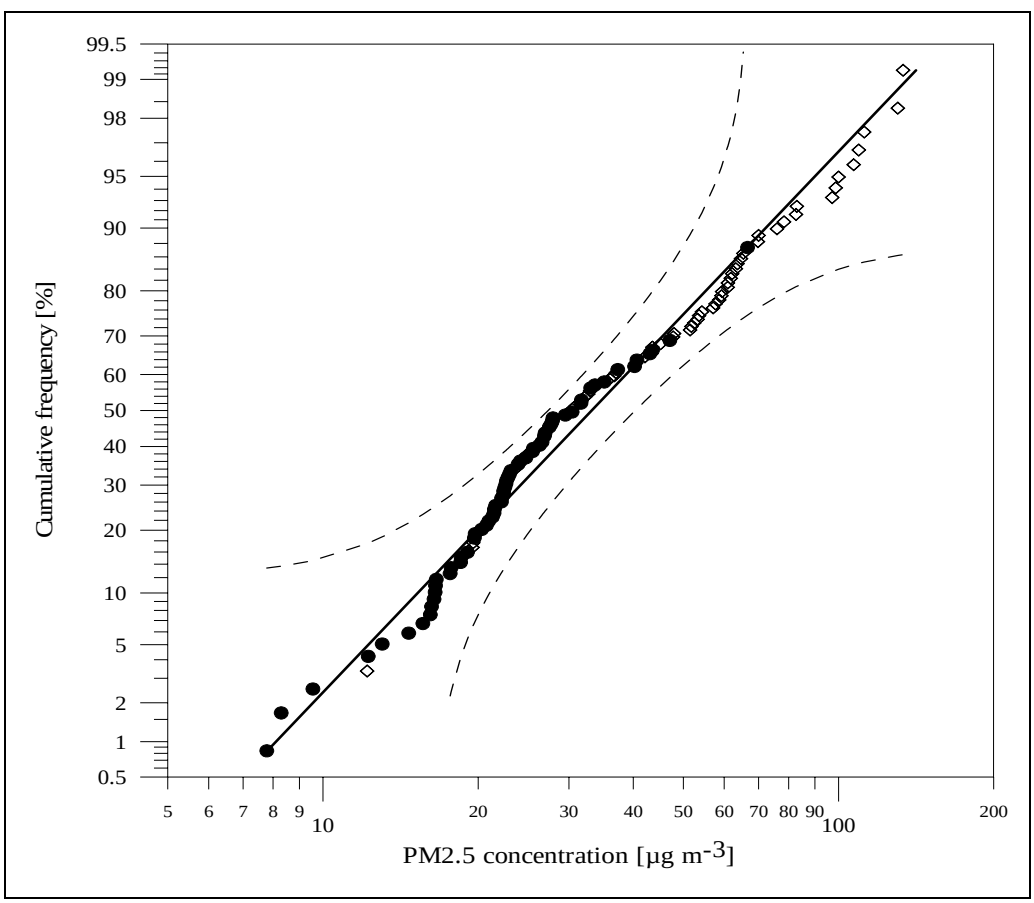


Figure 1 Distribution of PM2.5 24-h concentration data (Warm season data: white diamonds; Cold season data: black dots; Lognormal distribution model: solid line; 95 % confidence region for the cunulative distribution function of the PM2.5: dashed lines).

From the analysis of major chemical species (Table 3) it is seen that on the annual basis a substantial fraction (65.8%) of the fine particle mass is constitued by particulate organic matter (POM) (12.9 μg $m^{-3}$), nitrate (8.7 μg $m^{-3}$) and sulfate (4.7 μg $m^{-3}$). In addition to carbon, POM includes other atomic constituents that make up organic molecules, mainly hydrogen, oxygen, and nitrogen: to account for the presence of these latter in the organic compounds, OC concentrations were multiplied by a factor of 1.4 to estimate the organic mass (US EPA, 2004). POM levels are more than twice (19.7 μg $m^{-3}$) in the cold season than in the warm season (7.2 μg $m^{-3}$).

Table 3 Average mass composition (mass ± std. dev.; μg $m^{-3}$) of PM2.5 samples and relative contributions (%) to PM2.5 mass

| | **Year** | | **Warm season** | | **Cold season** | |
|---|---|---|---|---|---|---|
| Samples | **118** | | **64** | | **54** | |
| Parameter | Concentration | Relative contribution | Concentration | Relative contribution | Concentration | Relative contribution |
| Elemental Carbon | **1.4 ± 0.7** | 3.5 | **1.2 ± 0.5** | 5.0 | **1.6 ± 0.8** | 2.8 |
| POM | **12.9 ± 10.1** | 32.3 | **7.2 ± 3.9** | 29.4 | **19.7 ± 11.0** | 33.7 |
| Chloride | **0.3 ± 0.6** | 0.9 | **0.1 ± 0.1** | 0.2 | **0.7 ± 0.7** | 1.2 |
| Nitrates | **8.7 ± 11.2** | 21.7 | **3.2 ± 3.6** | 13.0 | **15.2 ± 13.5** | 26.1 |
| Sulfates | **4.7 ± 2.9** | 11.8 | **4.2 ± 2.1** | 17.2 | **5.3 ± 3.6** | 9.1 |
| Ammonium | **3.0 ± 2.8** | 7.5 | **1.7 ± 1.3** | 6.8 | **4.5 ± 3.2** | 7.8 |
| Crustal Elements | **1.5 ± 1.1** | 3.8 | **1.6 ± 0.9** | 6.6 | **1.3 ± 1.3** | 2.3 |
| Metallic Oxides | **0.3 ± 0.2** | 0.8 | **0.1 ± 0.1** | 0.5 | **0.3 ± 0.2** | 0.6 |
| Reconstructed Mass | **32.9 ± 24.3** | 82.2 | **19.3 ± 9.1** | 78.7 | **48.7 ± 26.8** | 83.5 |
| Unidentified Mass | **7.1 ± 7.2** | 17.8 | **5.2 ± 4.3** | 21.3 | **9.6 ± 9.2** | 16.5 |
| Total Measured Mass | **40.0 ± 26.4** | - | **24.5 ± 9.9** | - | **58.3 ± 28.0** | - |

Sulfate average concentration (4.2 μg $m^{-3}$) is greater than nitrate concentration (3.2 μg $m^{-3}$) in the warm season, nevertheless, nitrate concentrations in cold season (an average about 15.2 μg $m^{-3}$) are much higher than those of sulfate (around 5.3 μg $m^{-3}$ on average). This may be due to the fact that ammonium nitrate formation is favoured under conditions of high relative humidity and low temperature, frequently occurring in the winter period (US EPA, 2004), and that the instable atmospheric conditions occuring in summer period facilitate the aqueous phase formation of sulfate in the interstitial spaces of clouds and fog droplets. On some days with high PM2.5 concentrations nitrate concentrations overtake POM concentrations (i.e., on 13 days out of 33 when PM2.5 concentrations were greater than 50 μg $m^{-3}$). Ammonium average concentration is greater in the warm season (1.7 μg $m^{-3}$) than in the cold season (4.5 μg $m^{-3}$), with an annual average around 3.0 μg $m^{-3}$. Elemental carbon levels do not change very much throughout the year (1.2 μg $m^{-3}$ and 1.6 μg

$m^{-3}$ in warm and cold season, respectively) with an average concentration of 1.4 μg $m^{-3}$ on the annual basis. The generic "crustal matter" component of PM2.5 samples has been estimated based on Al, Si, Ca, Fe, K and Ti oxides by the following formulation (Marcazzan et al., 2001):

$$\text{Crustal matter} = 1.89\text{Al}+2.14\text{Si}+1.4\text{Ca}+1.36\text{Fe}+1.2\text{K}+1.67\text{Ti} \quad (1)$$

where only a fraction of iron and potassium concentration of natural origin is considered. These fractions have been estimated based on crustal enrichment factors (Mason, 1966): iron of natural origin is about 50% and 30% of the total iron, respectively in warm and cold season; corresponding figures for potassium are 100% and 60%. The total concentration of oxides of metallic elements is composed of heavy metal oxides (CuO, ZnO, $PbO_2$, $V_2O_5$) and of the anthropogenic component of iron oxide ($Fe_2O_3$). The total amount of crustal matter and metal oxides average concentrations does not exceed 2 μg $m^{-3}$ neither on seasonal nor on annual basis. Unidentified mass determined from mass closure analysis is constituted mainly by oxides of unaccounted elements and water.

Concerning carbon species, organic carbon is largely dominating and accounts for about 81% and 90% of the total carbon (OC+EC), in warm and cold season, respectively. Poor correlation is observed between organic carbon and elemental carbon suggesting lack of a single dominant source in the cold season and the secondary nature of a great part of organic carbon in the warm season. The OC/EC ratio ($[OC/EC]_{cold}$ = 8.6; $[OC/EC]_{warm}$ = 4.2), which is usually employed to estimate the extent of secondary organic aerosol formation, turns out to be larger than those registered for other UB sites around the world ($[OC/EC]_{cold}$ = 2.4-3.5; $[OC/EC]_{warm}$ = 1.3-3.9), and comparable to those noted for rural sites ($[OC/EC]_{rural}$ = 4.6-8.1). Inspection of elemental and organic carbon concentrations shows that the high ratios observed in this study can be attributed more to low elemental carbon levels than to elevated OC concentrations. The amount of secondary organic aerosol is estimated using elemental carbon as a tracer of primary organic carbon (EC tracer method):

$$\text{OC}_{\text{PRIMARY}} = \text{OC}_{\text{NC}} + \left[ \text{EC} \cdot \left( \frac{\text{OC}}{\text{EC}} \right)_{\text{PRIMARY}} \right] \quad (2)$$

$$\text{OC}_{\text{SECONDARY}} = \text{OC} - \text{OC}_{\text{PRIMARY}} \quad (3)$$

$$\text{SOA} = \text{OC}_{\text{SECONDARY}} \cdot 1.4 \quad (4)$$

where $OC_{NC}$ represents the non-combustion POM (e.g., OC derivating from biogenic source), $(OC/EC)_{PRIMARY}$ is the ratio for the local primary source affecting the measured concentrations (Turpin and Hunzicker, 1995; Cabada and Pandis, 2002). The primary contribution from traffic is determined using the OC/EC ratio ($[OC/EC]_{PRIMARY\text{-}trf}$ = 0.67) observed at a kerbside position in a road tunnel site in Milan. This latter value can be assumed as a suitable estimate of the OC/EC ratio characteristic of the real traffic source as the monitoring site is not affected by other emissions active in the area and is directly exposed to the traffic emissions. This ratio is in good agreement with the findings of Gillies et al. (2001) who found a ratio of 0.76 and with the primary OC/EC ratio that can be estimated based on OC and EC

emission factors of the circulating vehicles (Cabada and Pandis, 2002; Kleeman et al., 2000).

As there was no pollution source other than traffic to generate organic carbon, the amount of secondary organic aerosol in the warm season (6.1 μg $m^{-3}$) is obtained by subtracting the primary particulate organic matter due to traffic from the total particulate organic matter mass. In the warm season the secondary organic aerosol constitutes about 83.9% of the particulate organic matter mass and 24.7% of the PM2.5 bulk mass (Figure 2). Due to the impossibility of discriminating between condensed semivolatile organic matter and primary particulate organics collected on filter samples, the ratio, obtained from the ambient measurements of carbon species at the UB site, can not be employed to estimate a primary OC/EC ratio representative of the area inclusive of all sources. The presence of pollution sources other than traffic, for example domestic heating, makes impossible the prediction of the amount of primary organic carbon directly from the elemental carbon concentrations, and therefore, the estimation of secondary organic aerosol in the cold season. However, it is possible to confirm that in the cold season secondary formation of organic carbon by photooxidation is expected to be negligible given the scarce insolation, low temperature and ozone levels.

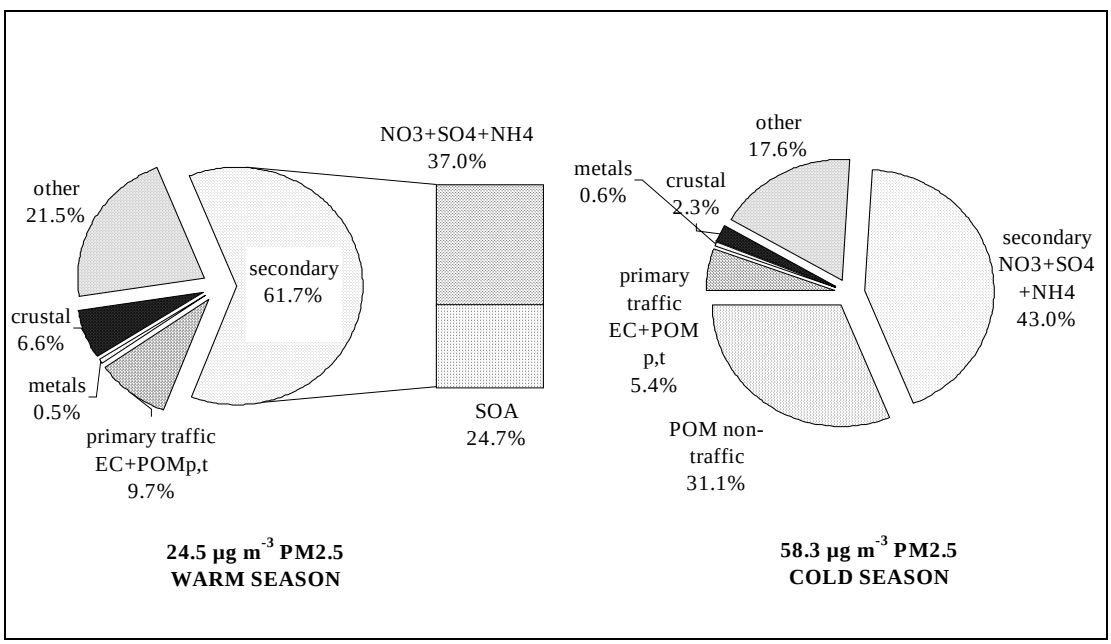


Figure 2 Primary and secondary contributions to PM2.5 bulk mass in warm and cold season.

With regard to elements, there is a strong enrichment of S and some metals (Cd, Pb, Zn and Cu) in the PM2.5 mass, whereas the enrichment factor of the other elements are next to unity (Table 2).

Table 2 Crustal enrichment factors for various elements

| | S | K | Fe | Cu | Zn | Pb | V |
|---|---|---|---|---|---|---|---|
| Warm season | 1283 | 1 | 2 | 17 | 94 | 757 | 2 |
| Cold season | 2647 | 2 | 6 | 175 | 872 | 5228 | 4 |
| Year | 1607 | 1 | 2 | 43 | 260 | 1964 | 2 |

According to a suggested classification criteria for crustal enrichment factor (Biegalski et al., 1998), an enrichment factor value lower than 3 is expected for an element of crustal origin, while one greater than 50 for an element of anthropogenic origin. Thus K, Fe and V appear to have a crustal origin, whilst the remaining elements are of anthropogenic origin. In particular, enrichment factors of S, Pb, Cu and Zn are much above 50; copper's enrichment factor results to be less than 50 in the warm season, but in any case above the stated limit for crustal origin.
Ionic composition of PM2.5 has been also evaluated with regard to the balance between the major anions ($NO_3^-$, $SO_4^{2-}$, $Cl^-$) and ammonium ($NH_4^+$). Although there is some scatter, data points generally fall above the 1:1 line, indicating that the ammonium is entirely taken up in neutralizing the anions and it is generally not enough to balance the negative charges of nitrate, sulfate and chloride; only very few samples are characterized by an excess of positive ammonium charges. (Figure 3)

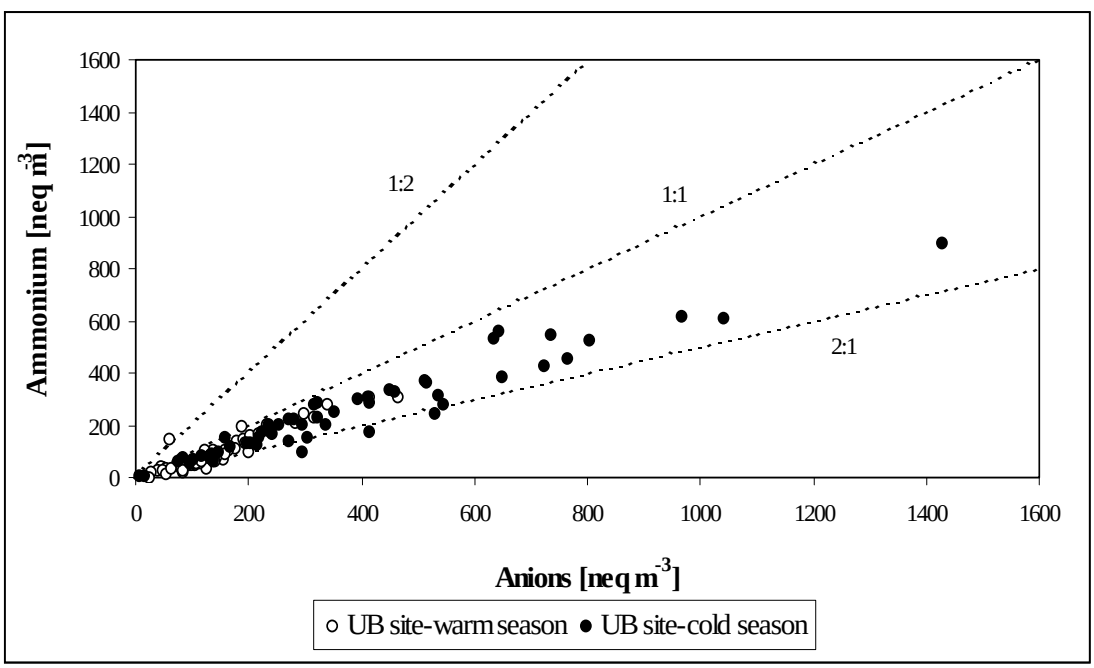


Figure 3 Ionic balance for PM2.5 24-h samples.

Linear correlation analysis between PM2.5 bulk mass and the analyzed species points out that:

- EC, V and crustal species such as Ti, Al, Ca, Si, show no relationship with the PM2.5 bulk mass on seasonal or annual basis;
- POM and secondary inorganic species such as nitrate and ammonium, are well correlated with PM2.5 bulk mass on both seasonal and annual bases;
- Sulfate, a secondary inorganic species, shows less correlation with PM2.5 mass in the cold season than in the warm season, while elemental sulfur shows more correlation in the cold season than in the warm season;

while the linear correlation analysis between the major species points out that:

- Secondary inorganic species (i.e., nitrate and sulfate) have strong relationship with ammonium, especially in the cold season;
- Concentrations of elements of crustal origin, such as Ti and K, have a high correlation;
- POM has a good relationship with all metals (except V) in the cold season;
- EC and OC concentrations show no correlation.

Comparing the the annual average PM2.5 concentration found in this study with levels for other countries in Europe given in literature (Querol, 2004), it is seen that PM2.5 levels in Milan (40 μg $m^{-3}$) are far greater than those observed at urban background stations in Northern and Southern Europe (8-15 μg $m^{-3}$ and 19-25 μg $m^{-3}$, respectively), and slightly greater than those observed in Central Europe (16-30 μg $m^{-3}$). On the other hand it is noted that the PM2.5 levels in Milan are comparable to those of the traffic exposed sites in Central and Southern Europe (22-39 μg $m^{-3}$ and 28-35 μg $m^{-3}$, respectively).

Particulate carbon levels (14.3 μg $m^{-3}$), expressed as the sum of POM and EC concentrations, are also comparable to those of the traffic exposed sites in Central and Southern Europe (8-16 μg $m^{-3}$ and 8-12 μg $m^{-3}$, respectively).

Sulfate, nitrate and ammonium ion concentrations (16.4 μg $m^{-3}$) are much higher with respect to the concentration levels recorded in other countries either at urban background or roadside stations, which do not exceed 11 μg $m^{-3}$.

In terms of relative contributions to PM2.5 mass the findings of this study (35.8% and 41%, for carbonaceous matter and secondary inorganic aerosol respectively) are consistent with the figures reported in literature (Querol, 2004).

## 4. CONCLUSIONS

Based on 118 mass samples collected at the urban background site in Milan from August 2002 through December 2003, it is found that the PM2.5 mass concentration which varies between a warm-season minimum of 7.8 μg $m^{-3}$ and a cold-season maximum of 133.4 μg $m^{-3}$, has an average value of 40 μg $m^{-3}$, and does not meet the limit values proposed for long-term and short-term in the II Position Paper. The threshold of 35 μg $m^{-3}$ is exceeded 47 times in the 2003 and the annual average is two times greater than the upper limit value (20 μg $m^{-3}$). The average value observed in this study is far greater than the annual average PM2.5 levels in the majority of European urban background sites and comparable to those of the traffic exposed

sites. The analysis of seasonal data points out relevant differences in concentration levels between cold and warm seasons; the highest concentrations (an average about 58.3 μg $m^{-3}$) are usually observed in the cold season, while the lowest concentrations (around 24.5 μg $m^{-3}$) tend to occur in the warm one. The presence of many emission sources and the meteorological conditions unfavorable for dispersion of the pollutants are responsible for the high levels observed in winter time.

Analyzing in terms of primary and secondary components it is seen that the primary component of fine particulate matter is mainly composed of elemental carbon, crustal matter, metal oxides and the primary fraction of particulate organic matter, on the other hand the secondary component is constituted by some ionic species (i.e., nitrate, sulfate, ammonium) and secondary organic aerosol. High PM2.5 episodes are principally due to secondary species, in particular due to increasing nitrate concentrations in winter time, and increasing particulate organic matter, which is mostly secondary in nature, in summer time. Secondary species account for 61.7% of the bulk mass in the warm season. In the cold season secondary inorganic species constitute the 43.0% of the bulk mass. The primary contribution from traffic is about 9.7% and 5.4% of the total PM2.5 mass in warm and cold season, respectively. The contribution of crustal elements and metal oxides is 7.1% in cold season, and 2.9% in warm season. The unidentified mass constitutes 21.5% and 17.6% of the bulk mass, in warm and cold season, respectively.

The temporal pattern comparisons of PM2.5 mass concentrations and ambient levels of nitrogen oxides, sulfur dioxide, ozone shows that these gases play a significant role in the particulate matter formation. Thus, intervention strategies other than on primary PM emissions, must also control the emissions of precursors gases, namely nitrogen and sulphur oxides and ammonia. However, the relationships between PM2.5 mass and individual species of precursor gases are not linear: thus a decrease in emissions does not necessarily brings an equal decrease in PM2.5 concentrations levels. Therefore, an exhaustive study on the secondary formation mechanisms of both inorganic and organic aerosol must be done when deciding short-term measures as well as elaborating long-term policies to control the ambient fine particulate matter levels.

A critical point is the exigency of a categorisation of volatile organic compound emissions as primary particles or as gases, which would make possible the determination of the real magnitude of the secondary organic aerosol also in winter time. Moreover, the present legislation estabilishes limit values for benzene alone (the Second Daughter Directive [2000/69/EC]), so this lack of information on volatile organic compounds' ambient air levels hinders the realization of air quality models.

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# ORIGIN OF DIURNAL VARIATION OF SURFACE OZONE IN KOLA PENINSULA AND FINLAND

**Sergey Rumyantsev[1], Valentin Roldugin[1] and Tuomas Laurila[2]**

[1]Polar Geophysical Institute, Fersman str., 14, Apatity, Murmansk region *Russia 184209* rumyantsev@pgi.kolasc.net.ru, roldugin@pgi.kolasc.net.ru
[2]Finnish Meteorological Institute, Air Quality Research, Sahaajankatu 22E SF-00880 Helsinki, Finland, Tuomas.Laurila@fmi.fi

## ABSTRACT

The monthly average daily courses of surface ozone concentration (SOC) at the Lovozero in the Kola peninsula and at four stations in Finland are investigated. The seasonal behavior of the amplitude of SOC daily variations are determined. The role of factors, having an influence on the ozone daily variations, has been investigated by numerical modeling using the simple one–box photochemical model. It is shown that mainly the average SOC daily variation is caused by dry deposition and destruction of ozone by nitrogen oxide at night, and by changing both photodissociation of nitrogen dioxide and concentration of the organic peroxyradicals.

**Key Words:** surface ozone, diurnal course, ultraviolet, peroxyradicals, mixing layer.

## 1. INTRODUCTION

Surface ozone can strongly influence the state of biological and technical objects. The ozone near terrestrial surface originates in transport from the upper layers of the atmosphere, where it is formed by rigid solar ultra-violet radiation (UVR), and in local generation by soft solar UVR from the ozone precursors. As shown at airplane investigation (Mauzerall et al., 1996), in summer in northern region the local photochemical processes give the main part of the ozone, contained in the boundary layer of the atmosphere; the second important source is the transfer from the upper troposphere.

Variations of SOC are caused by both change of solar radiation and by change of conditions of ozone penetration from the upper troposphere to the surface. The ground SOC observations give information on the proportion of two sources on the surface through a daily course of SOC. In Northern Europe the diurnal variations have been studied experimentally in Finland (Hakola et al.,1991; Laurila and Lättilä,1994; Laurila, 1996, 1999) and the Kola peninsula (Larin et al.,1997). In these studies, no theoretical investigations of the problem, permitting to connect experimental characteristics of the variations with parameters of atmospheric environment, have been carried out. In the present study, the SOC in the Kola peninsula and Finland is investigated.

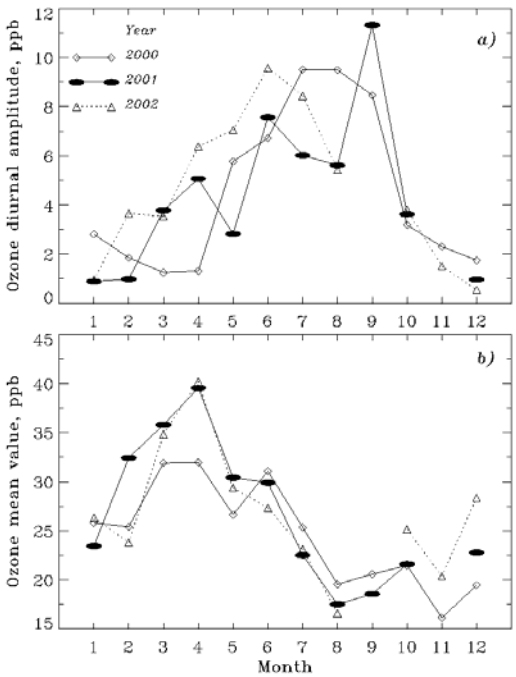


Figure 1. Monthly mean values of amplitude of ozone diurnal variation (top) and its average value (bottom) in Lovozero.

## 2. OBSERVATIONS

The results of SOC measurements in Lovozero in the Kola peninsula in 2000-2002 are used for the research. There is no industrial activity near the Lovozero settlement. The every minute ozone measurements are made by DASIBI - 1008 AH device and are registered to a data gathering system together with meteoparameters. The SOC data, measured at the station, represent background values for the high-latitude atmosphere with a small level of industrial pollution.

The found daily curves for all months are approximated by the sum of two first diurnal harmonics. The amplitude of daily variation, defined as difference of the approximation for concrete month, is shown in Fig. 1a. The daily variation is maximal in June – September in different years, amounting to 10 ppb, and is minimal in winter months, 1-3 ppb. The seasonal variation of mean SOC value, defined as half-sum of the maximum and minimum values of the approximation, is shown in Fig. 1b. As a whole, the monthly change of a daily variation corresponds to the change of solar light exposure.

## 3. THE PROBABLE REASONS OF DAILY VARIATIONS

The content of the surface ozone undergoes the influence of many factors. A daily SOC variation may be caused by: a) the daily change of solar UV intensity, b) the change of ozone precursors concentration; c) the change of thermodynamic parameters of air; d) the change of height of mixing layer; e) the daily change of dry deposition velocity, most appreciable in summer time; f) the daily variations of ozone transport rate. The role of these factors is apparently different. The results of (Belan and Sklyadneva, 2001) indicate a significant role of solar radiation flux in the formation of SOC daily course. As it is qualitatively noted in (Rovinskii and Egorov, 1986), the height change of mixing layer in the atmosphere, connected with sunrise and sunset, can cause daily SOC changes. It is shown in (Roldugin et al., 2004), that the change of ozone precursor concentration from spring to autumn results in a significant change of daily SOC variation amplitude. There are only theoretical estimations of the daily change of dry deposition velocity in summer in the high latitudes (Ganzveld and Lellieveld, 1995), and there is known practically nothing about daily variations of ozone transport.

To find out the contribution of the a-e factors into daily SOC variation, we use numerical modeling of generation and destruction processes of ozone.

## 4. MODEL CHARACTERISTICS

To solve this problem, we use the simple one–box photochemical model (Rumyantsev and Roldugin, 2003), which suggests that the concentrations of trace gases are homogeneous in the mixing layer. This model describes adequately some basic characteristics of ozone distribution in the background and polluted conditions (Roldugin et al., 2003).

The model characteristics are given in (Rumyantsev and Roldugin, 2003) in a most detailed form. The model describes the chemical transformation of 9 trace gases: $O_3$, NO, $NO_2$, $NO_3$, $N_2O_5$, $HNO_3$, $CH_2O_3$, PAN and $HO_2$ in the mixing layer, where the concentrations of these gases are supposed constant in height. They react with each other and with solar UVR in 33 reactions, presenting the basic interactions, which are realized in the near-surface atmosphere. It is supposed that the formed atomic oxygen transforms to ozone right away. The following gas species take into account as given: molecular hydrogen, aqueous vapor, hydroxyl, carbon oxide, formaldehyde, methane, organic peroxyradicals $RO_2$, including peroxyacetyl radical $CH_3C(O)O_2$. The formative ozone compounds, named as ozone precursors, include the hydroperoxyl $HO_2$ and organic peroxyradicals. The sources of $HO_2$ are reactions of hydroxyl OH. Methylperoxy radical $CH_3O_2$ is formed at the oxidation of methane by hydroxyl. Sources of $HO_2$ and $CH_3O_2$ are known more or less.

The photodissociation rates of $NO_2$, $NO_3$, the concentrations of hydroxyl OH and important ozone precursors $CH_3C(O)O_2$, $RO_2$ are given as the following function of local time: positive branch of sinusoidal function, when Sun is above the horizon, and it is equal to zero, when Sun is below the horizon. In the light period the bulk of the volatile organic compounds (VOC) is injected, and hydroxyl is generated. Organic peroxyradicals are originated in reactions of hydroxyl with VOC (Isidorov, 2001). Dry deposition velocities for different species are taken from the references. They are independent from daytime and season except for ozone. For ozone the velocities are taken from (Ganzveld and Lellieveld, 1995) and (Markova, 2002). In summer months, the model for Alaska, depending on daytime, was used too with the minimal value $v(O_3) = 0.004$ m/s at night, and maximal value 0.01 m/s at daytime using the results of (Ganzveld and Lellieveld, 1995). The simple model of the mixing layer height $h$, typical for the middle latitude atmosphere (Honore et al., 2000), is used: $h = h_{min}$ for $t \le t_r$; $h = h_{min} + (h_{max} - h_{min})(t - t_r)/t_0$ for $t_r \ge t \ge (t_r + t_0)$; $h = h_{max}$ for $(t_r + t_0) \le t \le t_d$; $h = h_{min}$ for $t \ge t_d$. Here $t_r$ and $t_d$ – the times of sunrise and sundown, $t_0 = 6$ hours, $h_{min}$, and $h_{max}$ – minimal and maximal values of the mixing layer height. The maximal value of the photodissociation rate of nitrogen dioxide, that has been taken equal to $6 \cdot 10^{-3}$ $s^{-1}$, resulted from the measurements in Spitzbergen (Beine et al., 1999).

The model permits to research diurnal SOC variations, caused by factors a – e.

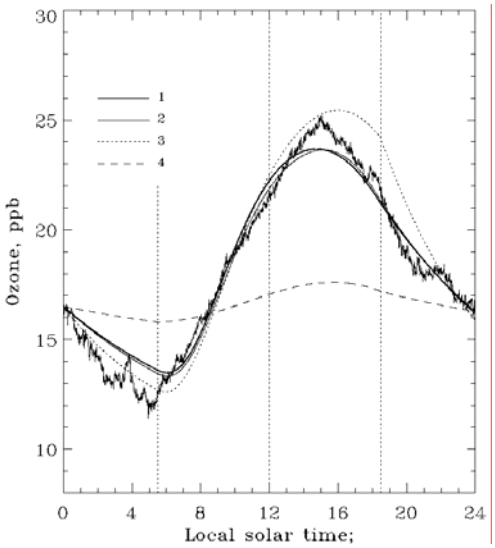


Figure 2. Mean daily variation of surface ozone concentration in Lovozero in September 2001 (uneven curve) and simulation curves 1 – 4, calculated for different chemical and meteorological conditions, see the text. The vertical dotted lines show sunrise, noon and sunset times.

## 5. COMPARISON OF MODELING WITH EXPERIMENT

The described model calculations have been compared with the experimental data obtained in Lovozero. The diurnal SOC variations, averaged for one month, are taken as experimental data. The modeling was carried out with alternating change of parameters, managing by the factors a – e, to single out the parameters, whose changes cause diurnal SOC variation. At first, the calculations were realized for conditions, when the temperature is taken equal to a mean value for the month; then the calculations are made for the case with observed daily distribution of temperature. Further, the SOC is calculated in the situation, when the concentrations of organic peroxyradicals $CH_3C(O)O_2$ and $RO_2$ are equal to zero. In this case the ozone generation takes place only from the precursors $HO_2$ and $CH_3O_2$, which are always present in the model and are being for it as the components of internal origin in contrast to $CH_3C(O)O_2$ and $RO_2$, taken from outside. At these calculations, the height of the mixing layer has always been accepted $h_{max} = h_{min} = 1000$ m. Then the calculations were carried out for a varying configuration of the mixing layer with $h_{min} = 300$ m, $h_{max} = 1000$ m. In summer months, the SOC was calculated for time-dependent dry deposition velocity with height of the mixing layer $h = 1000$ m. At such sequence of calculations the role of the factors, influencing the ozone content, is ascertained in turn: photodissociation rate, temperature, concentration of organic peroxyradicals, configuration of the mixing layer and changing of dry deposition velocity.

In Fig. 2 the monthly mean experimental values of diurnal SOC variation for September, 2001 and the results of calculations with average temperature of air is 4.5º C and the maximal value of $NO_2$ photodissociation rate is equal to 0.003 $s^{-1}$. The maximal concentration of the basic ozone precursors, organic peroxyradicals $RO_2$, is equal to 0.03 ppb. Line 1 shows the result of computation for the case, when solar UVR and organic peroxyradical concentration change only. All other parameters, which can determine the SOC are constant during the day. The daily SOC variation in the calculations is produced by those parameters, which vary with time, i.e. by photodissociation intensity and concentration of the ozone precursors in this case. Line 2 shows the results for the case, when the temperature of air varies within the day according to observations. It is obvious, that the distinction of this case from the previous one (line 1), is insignificant. Line 3 describes the case of varying mixing layer height: $h_{min} = 300$ m, $h_{max} = 1000$ m with other parameters as for the line 2. The

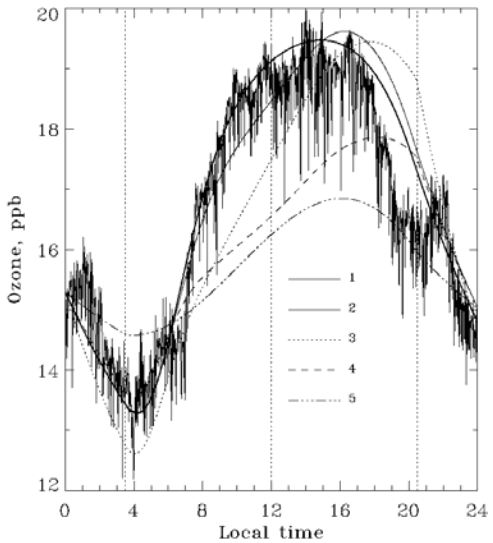


Figure 3. As in Fig.2 but for August 2002.

incorporation of the dependence of the mixing layer height on time *h(t)* increases ozone absorption due to dry deposition. Line 4 represents the results of calculations, when the concentrations of $RO_2$ and $CH_3C(O)O_2$ are equal to zero, and the ozone generation occurs only from the $HO_2$ and $CH_3O_2$ precursors at $NO_2$ photodissociation. One can see, that the amplitude of diurnal variation strongly decreases, the concentrations of generated hydroperoxyl and of peroxymethyl radical are insufficient for an explanation of the observable change of SOC variation. The comparison of curves 1 and 3, 1 and 4 shows, that the influence of change of the mixing layer height on SOC is appreciable enough, but strongly concedes to the influence of the organic peroxyradicals content. As it follows from computation, the night decrease of SOC in September, caused by dry deposition and destruction of ozone by nitrogen oxide, is ceding place, approximately at 5 - 6 h of local solar time (LST), to the growth caused by the strengthening of photodissociation at the increase of solar UVR. The beginning of growth approximately coincides with the sunrise. At about 14 h LST the recession of SOC (caused by reduction of photodissociation) starts, which is influenced insignificantly by the collapse of the mixing layer at 18.5 hours.

In Fig. 3 the mean SOC values in August, 2002 are given. Line 1 represents the results of modeling for the average temperature of this month 7.5º C. Values of other parameters are the following: the maximal $RO_2$ concentration is equal to 0.02 ppb, the maximal $NO_2$ photodissiciation rate is equal to $6 \cdot 10^{-3}$ $s^{-1}$. Line 2 illustrates the calculation for the observed change of diurnal temperature in August. The appreciable difference of daily ozone variation (lines 1 and 2) in the results of modeling during 08 - 20 h LST in this case is caused by the growth of air temperature in daytime, which brings changes of chemical reactions rates and reduction of ozone loss. Line 3 represents the case of varying mixing layer height with $h_{min}$ = 300 m, $h_{max}$ = 1000 m, and with values of other parameters as for line 2. The time distribution of SOC in this case appreciably differs from the one, presented by lines 1 and 2 because of additional ozone absorption at dry deposition. Line 4 illustrates calculation for the changing mixing layer and varying dry deposition velocity, taken, as in (Ganzveld and Lellieveld, 1995), instead of average dry deposition velocity, used at computations of lines 1 - 3 in this Figure. It is clear from comparison of the curves 3 and 4, that the difference between these cases is essential, except for the night hours. At last, line 5 shows what the daily variation would be in the absence of organic peroxyradicals, provided other conditions are the same, as for line 2. In this case SOC variations are noticeably less than for lines 1-3, reflecting the fact, that the ozone generation is weak at small amount of the precursors. The

influence of change of the parameters of the mixing layer and dry deposition velocity on diurnal SOC variation can be appreciable for the chosen conditions.

In Fig. 4 the comparison of the experimental data with the calculated ones for the period with a small daily variation about 4 ppb in March, 2001 is shown. Lines 1 and 2 show results of calculations for constant temperature T = -14° C and for variable atmospheric temperature correspondingly. The modeling curves pass closely to each other and to experimental data. Curve 3 illustrates the results of computations under condition of equality to zero of organic peroxyradicals concentration, when only photodissociation change takes place.

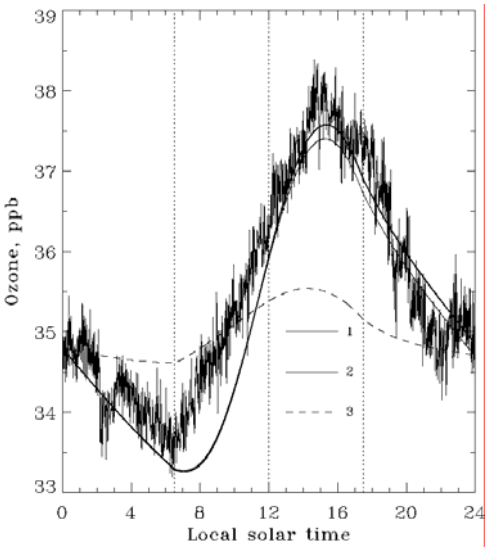


Figure 4. As in Fig.2 but for March 2001.

Thus, it is seen from figures 2 - 4 that the basic variation of the ozone content at this time is caused by the dry deposition, destruction by nitrogen oxide and joint change of solar UVR and concentration of organic peroxyradicals. The influence of changes of the mixing layer height is most appreciable in the periods, when the light and dark periods of day are close in duration, then the small heights of mixing layer and, as a consequence, large values of dry deposition rate, result in appreciable variations of ozone concentration. In summer months, the role of changes of temperature within a day becomes more appreciable.

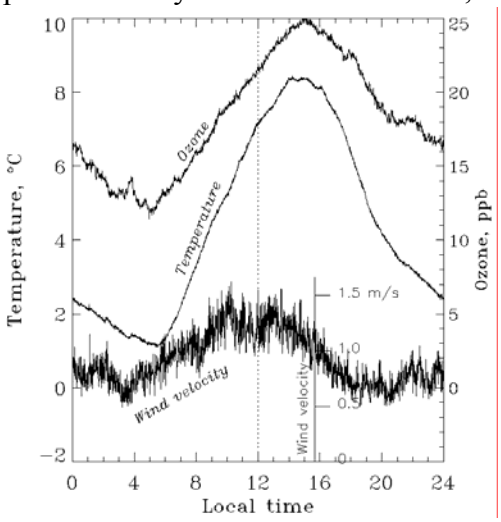


Figure 5. Mean daily variations of surface ozone concentration, temperature and wind velocity in Lovozero in September, 2001.

As it is noted, one of the reasons of diurnal SOC course can be the daily variation of velocity of ozone transportation, both vertical, and horizontal. First of all this transportation is connected with wind, which not only transfers ozone from other areas, but also mixes the air along vertical.

In Fig. 5, as a typical example, the diurnal variations in September, 2001 of SOC, already considered in Fig. 2, and of wind, and also of temperature are shown as functions of LST. One can see the distinct daily course of wind, but it differs from the ozone daily course: the maximum is achieved near the local noon, but not 3 hours after it, as for ozone, there is no characteristic minimum at 5 hour. Therefore, it is possible to make a conclusion, that wind does not play a determining role in the formation of the diurnal course of SOC.

## 6. DIURNAL SOC VARIATION IN FINNISH OBSERVATORIES

The ideas, used above for the explanation of daily SOC course in Lovozero, were applied for analysis of daily courses at four Finnish stations: Oulanka, Ähtäri, Virolahti and Utö. The most northern station Oulanka settles near the Polar circle in a wood at the height of 300 m, far from sources of pollution. The station Ähtäri is located in the central Finland, at the height of 180 m, also in a woody district. Virolahti lays on the coast of the Finnish gulf eastwards of Helsinki, and the air is subjected to industrial pollution from large seaports of the gulf. The station Utö is located on a rocky woodless island in the Baltic sea at the height of 7 m. The atmosphere in Finland is exposed to the influence of pollution both from their own industry, and from transboundary transport of industrial air pollution from Western Europe.

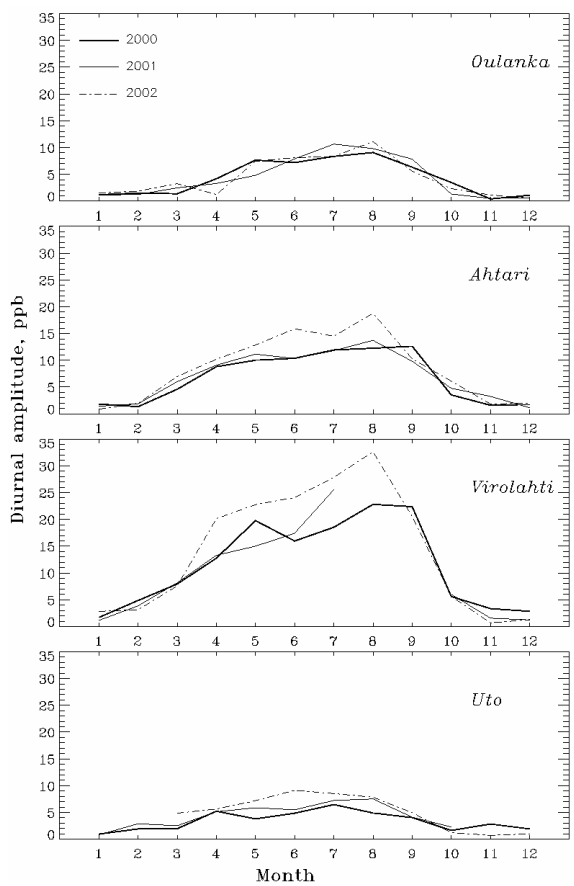


Figure 6. Monthly mean values of diurnal amplitudes of surface ozone variations in 2000 – 2002 at four Finnish observatories.

The hourly average ozone data for these stations for 1997 - 2002 are taken on the site http://www.nilu.no/projects/ccc/mnetwork/index.html.

Comparing the daily courses of the geographically closest stations Lovozero and Oulanka, a close resemblance of both forms and amplitudes of daily courses for most of months is detected. The SOC values in Lovozero are a little bit lower, than in Oulanka, that is, probably, connected to the prevalence of westerlies, and to industrial air pollution from the Western Europe.

The amplitudes of diurnal SOC variations for different months for finnish stations are found and shown in Fig. 6. The maximal amplitudes of average diurnal variations in Lovozero and Oulanka, equal at these stations, make up about 10 ppb, see Fig. 1a and Fig. 6. The results of paper (Laurila, 1999) for 1990-1996 are the similar.

The comparison of results of observations in Lovozero and at the Finnish stations shows, that the distribution of the daily variation of surface ozone within a year and in different geographical areas is determined by the basic rules about generation, absorption and transport of ozone, which are known now and realized in the submitted model.

A daily ozone variation is weak from November till February at all stations, it becomes appreciable in March and grows, reaching the maximum in June - September, and then goes down to small values in October (Fig. 6). In various years, the maximal value of a daily variation is reached in different months. In March - September the solar UV flux and intensity of VOC emissions vary strongly during an intensive growth and decay of vegetation. Increase and decrease of UVR flux and of VOC concentration results in increase and decrease of the contents of the ozone precursors. The ozone concentration increases and decreases during this time accordingly. The structure of the observed daily variations is the same at examined stations: a drop in night hours with a minimum in the morning, caused by night ozone destruction by trace gases and by dry deposition, then growth with a maximum at afternoon hours, caused by increase of a solar UV flux and VOC concentration at the sunrise, and a reduction connected with the lowering of the Sun and ozone generation weakening. A certain contribution into amplitude of the daily variation can be brought by anthropogenic pollution of air, which strengthens or decreases the ozone generation by UV radiation.

The amplitude of daily variation increases from north to south for continental stations (except for Utö), i.e. the ozone generation amplifies in this direction, in which both UVR flux and VOC emission are increased. It is evident from Fig. 6, in which the maximal values of average diurnal amplitude amount to 10 ppb for Oulanka, 19 ppb - for Ähtäri , 32 ppb - for Virolahti ; 9 ppb - for Utö. The Utö sea station surroundings are poor in vegetation and, as a consequence, the VOC concentration is lower there, than in woody areas of Finland

## 7. CONCLUSIONS

The average daily SOC variation in Lovozero is caused by dry deposition and destruction of ozone by nitrogen oxide at night, and by change of both photodissociation of nitrogen dioxide and concentration of the organic peroxyradicals during daylight hours. Amount of hydroperoxyle and peroximethyl radical, formed in the atmosphere and being a part of precursors, is not enough for explanation of the observed SOC variability. The peroxyradical concentrations, which are necessary for SOC variations, have to been created by growth in the vegetation period. The contribution of change of a mixing layer height and variability of dry deposition velocity is a small value and can be noticeable only in summer months. The contribution of the horizontal ozone transport into the appearance of diurnal variation seems to be not to amount to much.

The temporary distribution of SOC at four stations in Finland has been investigated, the average SOC values and the amplitude of diurnal variations of ozone concentration are found. The increase of the amplitude of diurnal SOC variation from north to south at continental stations of Finland is obtained. The comparison with results of observations in the Kola peninsula is made. On the basis of the considered chemical model, which has been applied quantitatively to the Lovozero data, the qualitative explanation of behaviour of diurnal variations in Finland is given.

**8. ACKNOWLEDGEMENT**
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# ANALYZING AIR CANALS OVER GREATER TEHRAN AREA FOR DILUTION OF AIR POLLUTANTS

**Z. Jahangiri, A. Asgari[*] and A. Sedaghatkerdar[**]**

Senior Expert of Atmospheric Science & Meteorological Research Center (ASMERC), Tehran, Iran, z_jahangiri@yahoo.com
[*] Faculty Member of ASMERC, asgari-a@irimet.net
[**] Director ASMERC, aseda@irimet.net

## ABSTRACT

Because of the massive global environmental impacts of fossil fuel use, decision makers seriously want to find ways to reduce air pollution in their own countries. Moreover environmental concerns are the driving force behind the development of clean and renewable energy sources. There are two basic methods to reduce air pollutants. First is emission reduction from fossil fuel energy sources and second is use of appropriate meteorological approaches for diluting air pollutants. In this paper, air canals over greater Tehran area are analyzed. Since this mega city is surrounded by mountains and is almost in the form of a big valley; air pollutants are trapped in boundary layer. Therefore wind could be of too much importance. In this regards relatively strong winds make urban atmosphere turbulent and pollutants are transported to the upper urban atmosphere. In this work, we firstly determine wind field and wind rose over greater Tehran area. In the next stage, air canals are specified too. At the end, we present some suggestions for strengthening these canals such as plantation of trees in appropriate locations, selection of suitable building's orientation against the wind and so on.

**Key Words**: Air pollutants, Deposition, Disperse, High and Low Pressure, Turbulence

## 1. INTRUDUCTION

The atmosphere has served as a sink for emissions of volcanoes and a variety of geographical processes, forest and grassland, and decomposition and other biological processes for hundreds of millions (if not billions) years. It has also served as a sink for pollutants generated by human activities, proceeding from man's first use of fire to the smelting of metal ores and use of fossil fuels such as coal, oil, and natural gas to motor vehicle and other emissions from our every industrialized and technologically advanced modern times (Arya, 1999).

Despite its vastness, the atmosphere (at least in the short term) is not perfect sink. Its ability to carry away (transport), dilute, (disperse), and ultimately remove (deposition) waste products released to it is limited by various atmospheric motion phenomena. Pollutant concentrations may reach unacceptable levels as a result of local or regional overloading of the near-surface atmosphere, topographical barriers,

and micro-, meso-, and macroscale air motion phenomena. The atmosphere serves as a medium for atmospheric chemical reactions that ultimately serve to remove contaminants. These reactions may produce pollutants that may themselves pose significant environmental concerns. Levels of long-lived pollutants such as methane ($CH_4$), nitrous oxide ($N_2O$), and carbon dioxide ($CO_2$) may increase, causing global warming and in the case of halogenated hydrocarbons, stratospheric ozone ($O_3$) depletion (Godish, 2004).

Because of technological and economic limitations, we have little choice but to use the atmosphere for the disposal of airborne wastes. Like other natural resources, our use of the atmosphere has to be a wise one, recognizing its limitations and using in a sustainable way.

## 2. WIND

Wind is a meteorological variable commonly used to describe air movement in the horizontal dimension. Wind result from difference in air pressure that are caused by unequal heating of the earth's surface. In the absence of friction and the earth's rotation, air would flow from areas of high to low pressure. The direction of airflow is controlled by a combination of the pressure gradient force, Coriolis Effect, and friction. Horizontal winds are characterized by both wind speed and direction. Wind speed is also affected by horizontal pressure and temperature gradients and friction, which is proportional to the roughness of earth's surfaces (surface roughness). For continuously emitting stack sources, dilution begins at the point of release. This plume dilution is inversely proportional to wind speed; i.e., by doubling wind speed, pollutants concentration is decreased by 50% of its initial value. The effect of wind speed is to increase the volume of air available for pollutant dispersal. Urban areas are characterized by relatively high surface roughness and, as a consequence, diminished wind speeds. This is ironic in the sense that urban areas, because of their relatively high pollutants emissions, are in greater, not lesser, need of being ventilated by wind. Directional aspects of global winds include the Prevailing northeasterly flows in the subtropics, southwesterly flows in the middle latitudes, and easterly flows at high latitudes in the northern hemisphere. They also include the cyclonic (clockwise) and anticyclonic flows associated with migrating low- and high-pressure systems. Because flows are somewhat circular, wind direction will depend on one's position in the circulating pressure cell. It also depends on local topography. At night in river valleys, airflows are downslope and downriver; they are upslope during daylight hours. Along sea and lake coasts, winds during clear weather flow inland during the day and waterward at night (Sorbjan, 2003).

Wind direction is quite variable, with large changes often occurring over relatively shorts periods of time. A change in wind direction of 30˚ or more in 1 h is not uncommon. Over a period of 24 h it may shift by 180˚. Seasonal factors may cause wind direction variations of up to 360˚. Wind direction and variability can have significant effects on air quality. Areas downwind of point sources where winds are relatively persistent may experience relatively high ground-level concentrations compared to other areas at similar distances. If the wind is more variable, pollutants

will be dispersed in larger volume of air and be more equally distributed around the source; ground-level concentrations are therefore likely to be lower. Wind direction is particularly important in the transport and dispersion of pollutants over large geographical areas. For instance, it is southwesterly airflows that carry acid precursors from the U.S. Midwest to the northeastern states and southeastern Canada. Similar flows have transported pollutants from countries of southeastern Asia to the West Coast of U.S. [1]. Although global winds are important in determining the prevailing winds in a given area, local climatic conditions may have an influence on the most common wind directions. Local winds such as sea and land breeze are always superimposed upon the layer scale wind systems, i.e. the wind direction is influenced by the sum of global and local effects. When larger scale (synoptic patterns) winds are light, local winds may dominate the wind patterns. Mountain regions display many interesting weather patterns. One example is the valley wind which originates on south-facing slopes (north-facing in the southern hemisphere). When the slopes and the neighboring air are heated, the density of the air decreases, and the air ascends towards the top following the surface of the slope. At night the wind direction is reversed, and turns into a downslope wind. Figure 1 shows schematic diagram of mountain winds. These local winds may have important role on air quality, i.e. these may caused dilution of pollutants or concentrating of them [URI1].

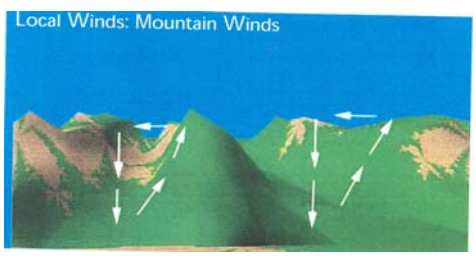


Figure 1. Mountain Winds.

## 3. DATA

In this study, we used 9 synoptic and climatological stations in Great Tehran and neighboring areas. Table 1 shows type and geographical specification of the stations under study.

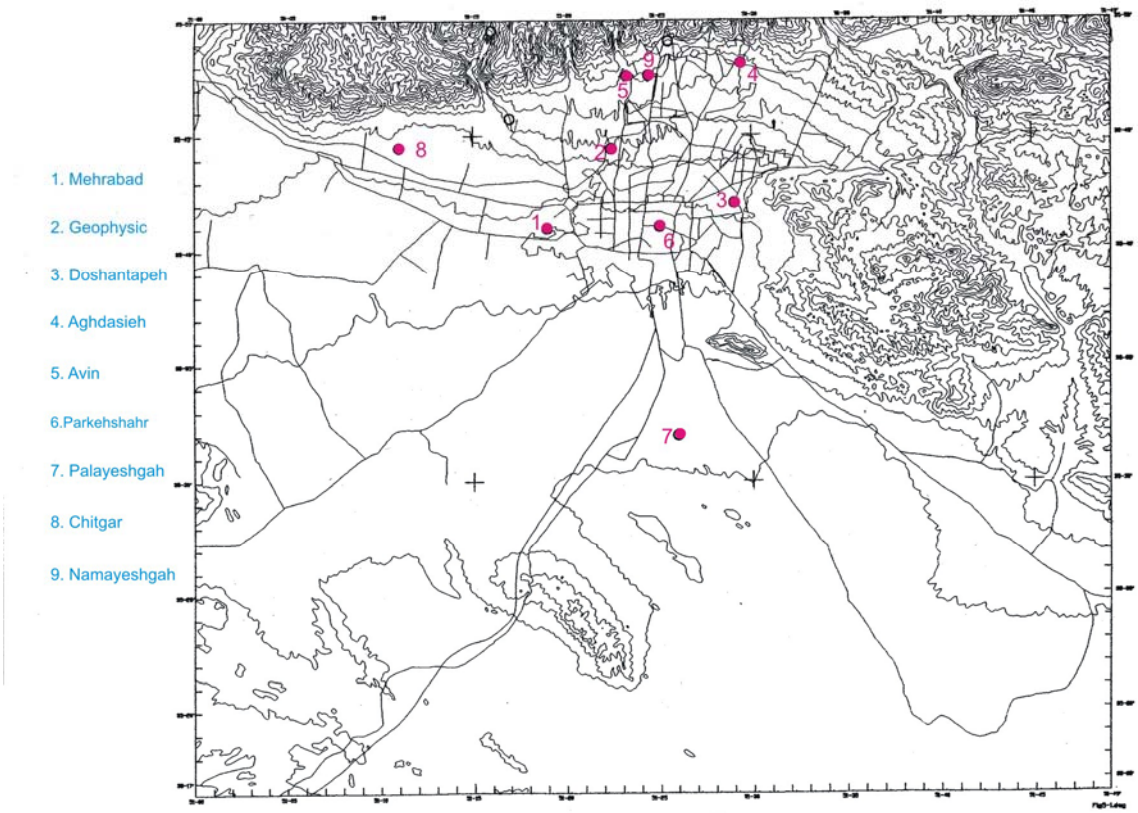


Figure 2.Topographical map of Tehran area and also location of studied meteorological stations.

Figure 2 shows studied stations network which superimposes upon topographical map of Tehran and its surroundings. Scale of map is 1/250000. Geographical area is limited to 51˚ to 51˚ 45′ of longitude and 35˚ to 35˚, 30′ of latitude. Interval in both axes of the map is 5 minutes (Research and…, 1995)

Table 1. Type and geographical characteristics of 9 meteorological stations.

| | Name | Type | Latitude ° ′ | Longitude ° ′ | Height over mean sea level (m) |
|---|---|---|---|---|---|
| 1 | Mehrabad | Synoptic | 35 , 41 | 51 , 19 | 1191 |
| 2 | Geophysics | Synoptic | 35 , 44.5 | 51 , 22.5 | 1260 |
| 3 | Doshantapeh | Synoptic | 35 , 42 | 51 , 29 | 1209 |
| 4 | Aghdasieh | Synoptic | 35 , 48 | 51 , 29 | 1548 |
| 5 | Evin | Climatological | 35 , 47.5 | 51 , 23.5 | 1520 |
| 6 | Parkeshahr | Climatological | 35 , 41 | 51 , 25 | 1210 |
| 7 | Palayeshgah | Climatological | 35 , 32 | 51 , 26 | 1160 |
| 8 | Chitgar | Synoptic | 35 , 42.5 | 51 , 13 | 1270 |
| 9 | Namayeshgah | Climatological | 35 , 47.5 | 51 , 24.5 | 1541 |

Source: IRIMO

## 3. TOPOGRAPHICAL CHARACTRISTICS OF TEHRAN

Tehran's **t**opographical structure plays a significant role on air pollution. This city is surrounded by mountains from northern and eastern sides. Because of the horseshoe shape of mountains around the Tehran, local winds can not transport pollutants far from the city; therefore pollutants move between north and south of area. In the same region, prevailing winds and synoptic patterns appoint final situation of air quality.

## 4. WIND ROSE

You will notice that strong winds usually come from a particular direction. To show information about the distributions of wind speeds, and the frequency of the varying wind directions, one may draw a so-called wind rose on the basis of meteorological observations from sufficiently long period. We have divided the compass into 8 sectors, one for each 45 degrees of the horizon. A wind rose may also be drawn for 12 or 16 sectors. For instance Figure 3 shows wind rose of Geophisics station for 5 years period (1991-1995. Width of the rectangle is directly related to the wind speed, by increasing wind speed, its width increases (scale of wind speed varies 1 to 37 knots). Length of rectangle indicates percentage of wind directions frequency, in other words, the longer length shows more percentage of wind directions frequency. Percentage of calm (wind speeds less than 0.5 meter per second) is showed in center of wind rose diagram. Mean wind speed is superimposed upon scale of wind speed percentage (uniform circles) as a closed curve. Figure 3 shows wind rose of months of April, May, and June and whole season of spring in the Geophysics station. It is clear that the prevailing wind directions are north and northeast with speeds in the range of 4 to 6 knots, and fastest winds have speeds in the range of 28-33 knots.

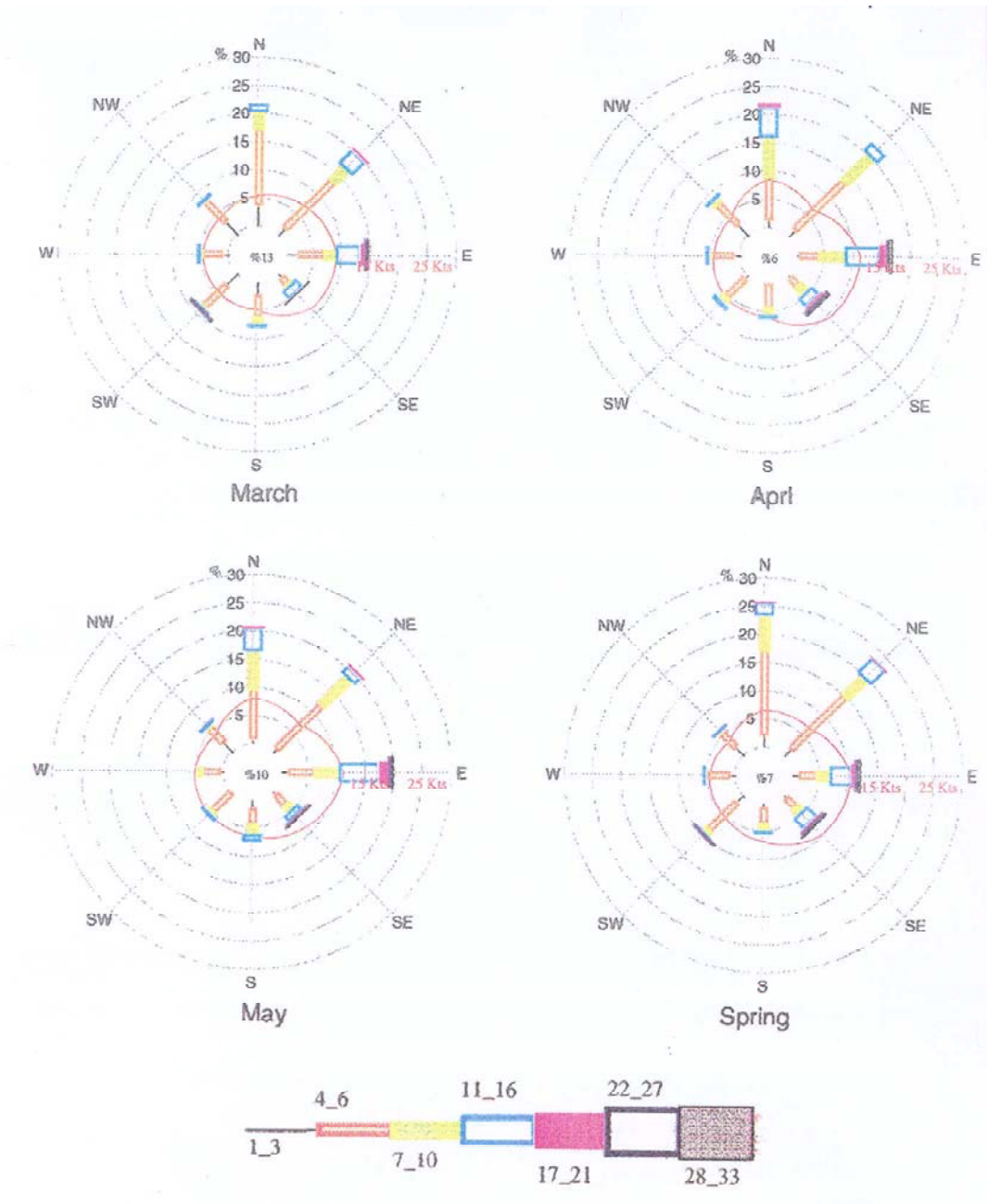


Figure 3. Wind rose of Geophysics station in 3 months and season of spring (1991-1995).

## 5. ANALYSIS of AIR CANALS OVER TEHRAN AREA

During spring, Prevailing winds enter metropolitan area of Tehran from west and northwest directions. Figure 4 shows the pattern for Greater Tehran Area on a topographic map. The red circles indicate studied stations over Great Tehran Area and the red arrows represent prevailing wind in the stations. The green and blue arrows respectively reveal wind vectors and streamliners over studied region.

In summer, prevailing winds are blown from southwest and divided to two branches. First have southern direction and the other passes southern and central parts of Tehran and then exits from eastern and southern edges of the city. Figure 5 illustrates whole situations for summer season.

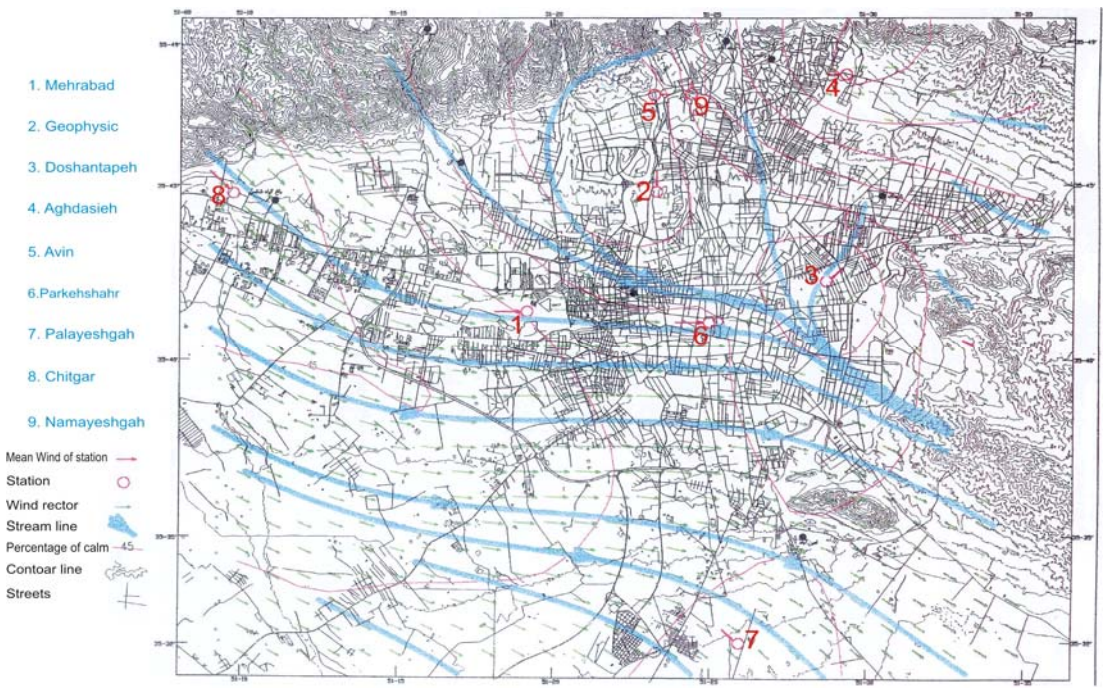


Figure 4. Air canals over Tehran in spring time for 1986 to 1995.

In autumn, winds are frequently western (Figure 6) and Winds mostly are blow from northwest to the southeast in winter season (Figure 7).

We superimposed 4 figures (4-7) relating to the 4 seasons and found that direction of prevailing winds is western whole of year. Figure 8 indicates this results and streamlines (dark blue tick lines) have western direction.

## 6. STRENGTHING OF AIR CANALS OVER TEHRAN

There are different methods for strengthen of air canals that here we briefly introduce some of them). One of them is movement industrial centers from west to the east of Tehran. The other method is creation of appropriate vegetation cover in the windward edge of the city, especially in the northwest parts of the city (Melaragno, 1980). Height and width of buildings, orientation of them against the wind, and orientation of the streets are known as control factors to air flows in urban internal parts of Tehran (Jenab, 1984). The existing of tall buildings cause air pollution to concentrate in downtown of the city. It is suggested that tall buildings are exposed to the prevailing wind by small side. In such cases, we would have better movement of air throughout city (Kasmaee, 1984).

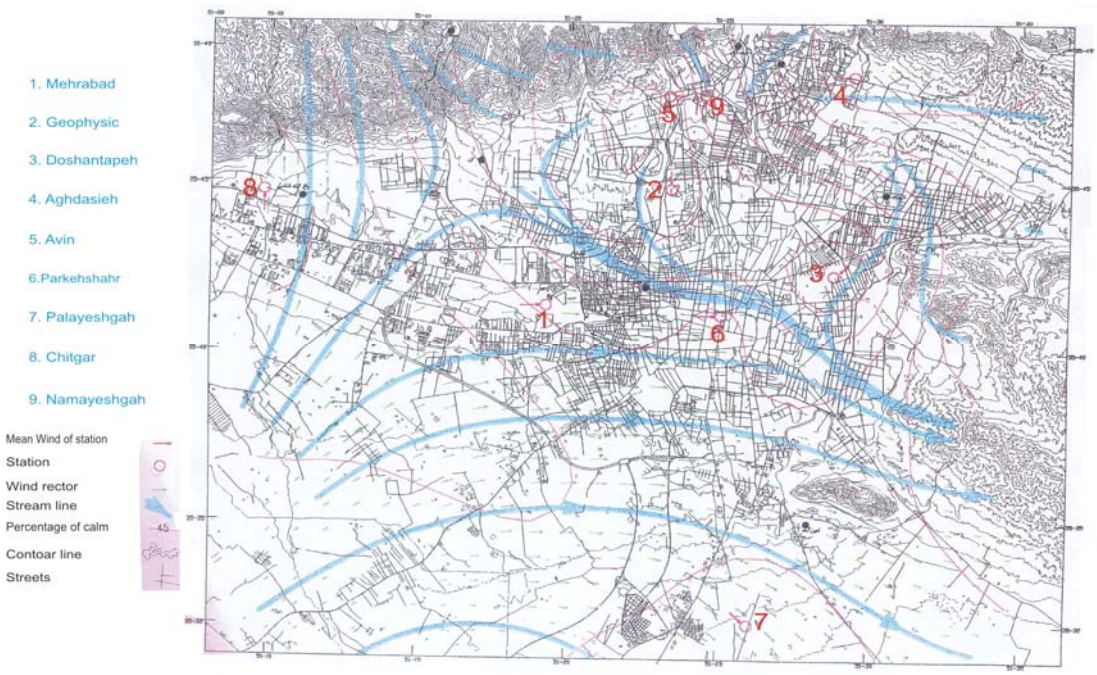


Figure 5. Air canals over Tehran in summer time for 1986 to 1995.

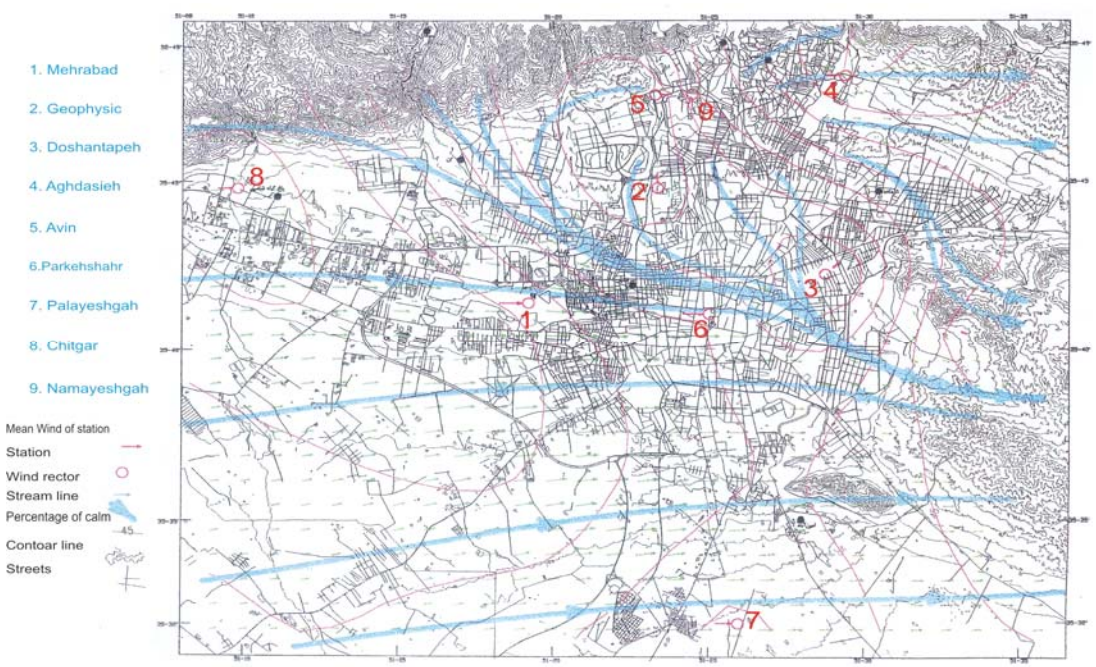


Figure 6. Air canals over Tehran in autumn time for 1986 to 1995.

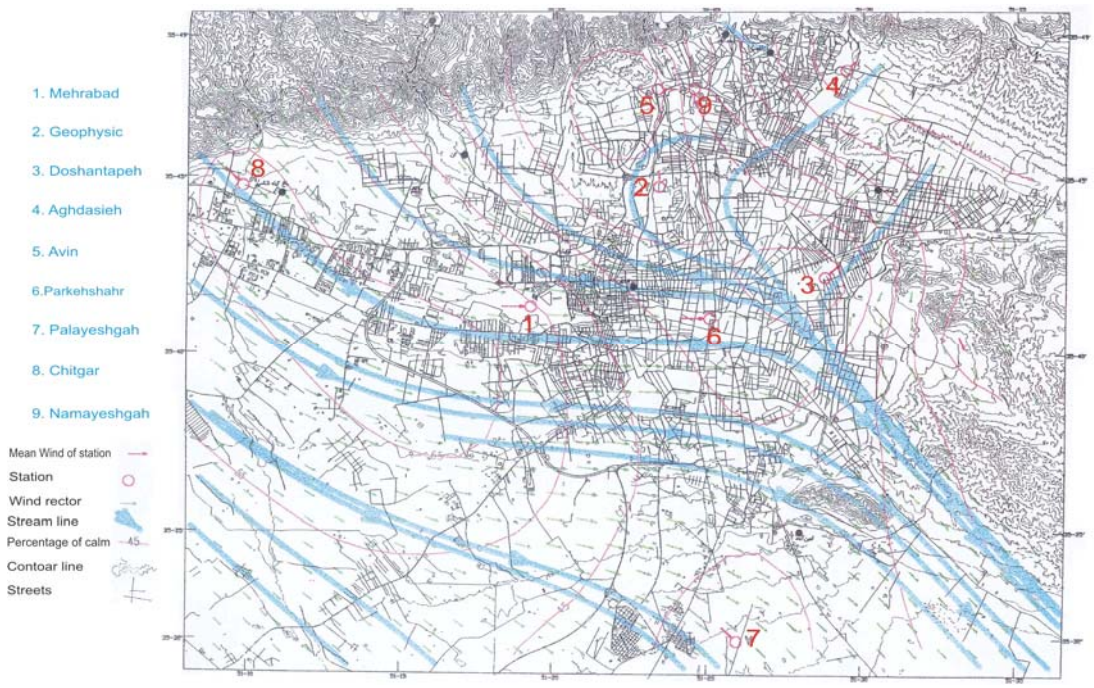


Figure 7. Air canals over Tehran in winter time for 1986 to 1995.

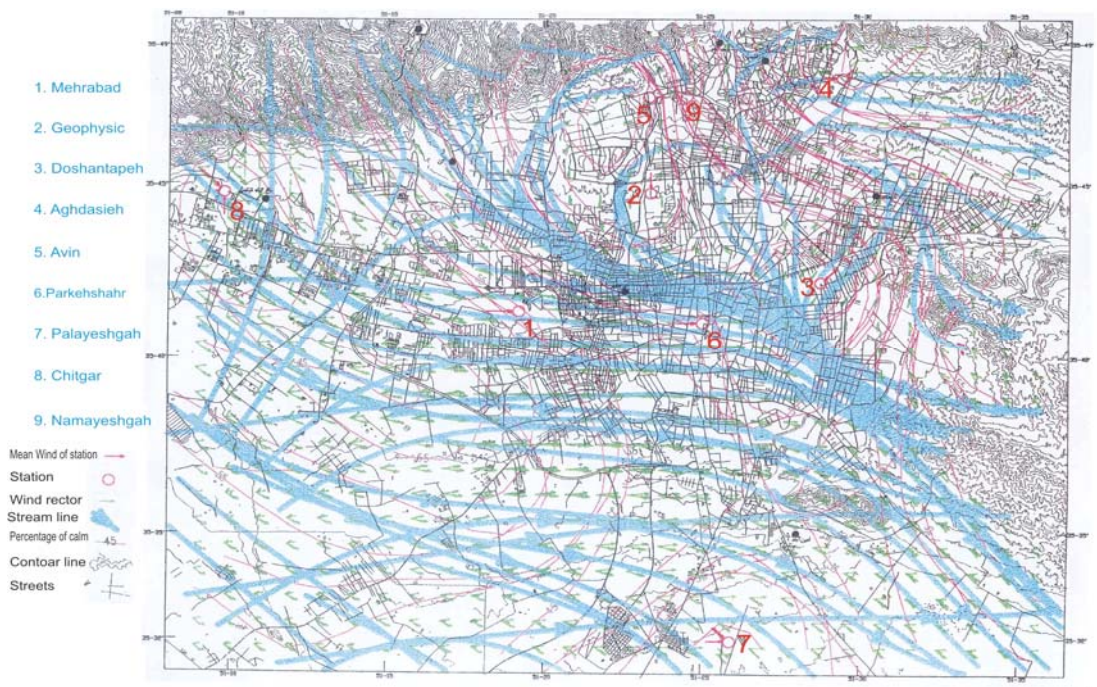


Figure 8. Annual air canals over Tehran for 1986 to 1995.

## 7. CONCLUSION

In this work, we understand that winds over Greater Tehran Area are very complicated from season to season and prevailing winds mainly are western. As Tehran has its specific complex topography and almost located in a valley, it is vulnerable to air pollution. Our obtained results show that we have to be very careful in establishment of factories and workshops (point sources) in the city. At the moment, most of pollution producing industries is located in the west of Tehran and pollutants are easily transported to the city and remain in there for considerable period of time (few hours to few days). These situations especially occur in the late autumn and early winter.

## 8. **ACKNOWLEDGEMENT**

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# PM 10 CONCENTRATIONS AND HUMAN ACTIVITIES: A STUDY IN CATANIA

**M. Ferrante, R. Fallico, G. Mirata and P. Di Mattia**

Department "G.F. Ingrassia" – University of Catania – Sicily (Italy)

## ABSTRACT

The WHO estimates that every year in the Italian cities thousands of peoples die because of very high concentration of thin powders; many more are the illnesses linked to their presence.

The PM10 is a mixture of particles of different nature and composition which, due to its small size (< 10μm), can easily disperse in the athmosphere and, as a consequence, it reaches an almost uniform value on the whole urban conglomeration.

Through this study we want to draw the picture of the problem in the city of Catania in the year 2004 from the environmental point of view; data obtained were also compared to known data from other Italian cities.

Many are the Cities where legal limits are not respected, and many more will be in the future, since those limits will be put at lower levels up to 2010. Many cities, however, pushed by the national norms, have taken measures in order to oppose such a pollution. Unfortunately, efficacy of those interventions are limited and insufficient to bring back pollution within legal limits. To protect peoples' health we need major structural interventions which would change, among other factors, the urban mobility.

In order not to go on living in a continuous state of emergency, it is necessary that the problem of pollution by PM10 becomes a priority for public administrations, and a point of reference not only for actions which concern the environment as such, but also for all actions concerning economy and city planning.

Filters were situated in special stations displaced by the Catania city council in different areas of the city; they were collected over the whole spingtime-summer period. Filters were of two types: cellulose ester made, and glass fibers made; after being metallized, they were examined at the scanning electron microscope (SEM) which had linked a Xrays detector. Data analysis shows nine main types of samples taken from the different sites: particles rich in: carbon, carbonates, soyates, silica, silicates, iron, metals, vulcanic ashes, carbon with heavy metals.

In this study we tried to make clear which characteristics of PM10, beyond differences of values due to environmental concentration, are related to the specific type of the sample site and to human activities performed there.

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# COMPARING URBAN AIR QUALITY IN EUROPE IN REAL TIME
# A REVIEW OF EXISTING AIR QUALITY INDICES AND THE PROPOSAL OF A COMMON ALTERNATIVE

**Sef van den Elshout[1], Karine Léger[2] and Fabio Nussio[3]**

[1] DCMR EPA Rijnmond, PO Box 843, 3100 AV Schiedam, The Netherlands
[2] AIRPARIF, Rue Crillon 7, F-75004, Paris, France
[3] STA Spa, Via Ostiense 131/L, I-00154, Rome, Italy. jel@dcmr.nl.

## ABSTRACT

The EU directives on air quality force member states to inform the public on the status of the ambient air quality. The internet is commonly used for this purpose and often air quality is being presented as an index ranging from good to bad. A review of existing websites and air quality indices shows that the way air quality is interpreted differs considerably.

The paper presents a new air quality index. The index is part of a project to develop a website dedicated to comparing air quality in European cities. The common air quality index (CAQI) is not aimed at replacing existing local indices. The CAQI is a set of two indices: one for roadside monitoring sites and one for average city background conditions. Differentiating between roadside and general city conditions is a first step in assuring consistence in the parameters that are being compared.

**Key Words**: urban air quality, air quality indices, public communication

## 1. INTRODUCTION

The Framework directive and associated daughter directives on air quality in the European Union not only force member states to monitor and report on their air quality but also to actively inform the public on the status of the ambient air quality (EU1, www). The Aarhus convention (ratified by the EU in 2005) further enforces the concept that citizens have the right to be informed on the environmental conditions they live and work in (EU2, www). Over the past years a good number of cities and countries have started to display monitored or modelled air quality data on the internet. For most of the monitoring organisations, the internet is the easiest way to meet the dissemination of information requirements of the European (and/or national) legislation. The fact that so much air quality information is available on the internet makes it tempting to compare different cities in different countries. This proves particularly difficult. Apart from the European Environmental Agency's ozone website there are no possibilities to compare cities/countries side by side

(EEA, www). Even if one surfs from one site to the other, comparison is not easy: air quality is presented in different ways using different interpretation criteria and a different typology of stations, which is usually not clearly explained.

The most widespread way to interpret air quality on the internet is the use of an index ranging from good to bad to make the detailed measurements in micrograms more understandable for the general public. A review of existing websites and the associated air quality indexes shows that the way air quality is interpreted differs considerably across the world. More surprisingly, even amongst the EU member states who share common legislation, the indices used do vary. There are a number of reasons to explain these differences. Some of them are historical and conceptual: the index existed before the EU regulations came into force and the index was based on health and exposure criteria, e.g. the UK index (DEFRA, www). The fact that air quality problems (sources, meteorological conditions, etc.) tend to differ is also one of the reasons. The indices tend to be calibrated to the local situation to make sure that there is some variation in the index from day to day (to make it entice repeated visits to a website) and that the typical range of pollutant conditions occurring locally is being covered.

To facilitate the international comparison of near real time air quality the CITEAIR project[1] is developing a common operational website (COW) where cities can display their air quality information side by side. The project aims at making air quality comparable across Europe and the COW will be open for any city to join. The COW is planned to be available by the end of 2006. The common website needs a common air quality index (CAQI). The CAQI is *not* aimed at replacing existing local indices. This would be an unrealistic ambition as in many cities the public has got used to the local, tailor-made index, and the CAQI will be, by the nature of the fact that is common, a non-specific compromise. CITEAIR envisages that there is room for two sources of air quality information on the internet: a local website, in the national language with a dedicated presentation (using a well established and known local index relying on more detailed air quality information); and a common website aimed at comparing - in near real time - the air quality in your own city to the air quality in other European cities. For this website a specific index had to be developed.

## 2. REVIEW OF AIR QUALITY INDICES

There are a substantial number of different ways to interpret air quality in near real time. The most common way to do so is the use of an index, generally based on a number of sub-indices for individual pollutants[2]. There is a wealth of indices and even countries who share the same legislation, or sometimes areas/cities within the same country have different indices. Some of the differences can be explained by the local differences in the nature of the air quality problems. Some other differences are

[1] CITEAIR is an initiative of the cities of Leicester, Paris, Prague, Rome and Rotterdam. Information on CITEAIR is available at http://citeair.rec.org

[2] A version of this paper with a list of indices collected in the course of the development of the CAQI annexed will be available on http://citeair.rec.org

due to fundamentally different approaches. The UK index (and US-EPA) for example is strongly related to perceivable effects. The bands in the index are explained in health terms. This implies that the index covers a very wide range of concentrations and that actual concentrations are very often in the "good" or "moderate" end of the scale. Air quality in Europe, fortunately, is rarely poor enough to cause acute health effects so any index based on health impacts tends to trail at the lower end of the scale for most of the time.[1]

Other indices take a different approach. For example the ATMO index, a countrywide standard in France (Airparif, www) has bands that are somehow linked to values that are also used in the current EU directives. The alert thresholds in the directives tend to define the higher end of the scale. In these cases the top end of the index scale ends somewhere in the middle of the health effect based scales. For example the worst end (very poor) of the $NO_2$-index in France corresponds to 400 μg/m3. In the UK this is in the lower end of the "moderate" band and in the US it is even considered too low to calculate an index value.

Communication-wise the health-based indices have both a clear advantage and a disadvantage. The advantage is that the index value displayed at the website is easy to interpret: it does or does not cause health effects. The disadvantage is that the index is almost always indicating that air quality is good and pollution is low whereas the limit values for long-term exposure are often exceeded. This leads to an apparent paradox: a citizen regularly checking the local air quality website will always get the message that the air quality is good whereas at the end of the year local government puts out a report that he or she is living in a hotspot area for which an action plan is required. This is the paradox between short- and long-term air quality criteria. The criteria for short-term exposure are often met except for episodes, like for example in the summer of 2003. The criteria for long-term exposure are often not met, in Europe's urban areas. The ATMO-type of indices provide some differentiation at the lower end of the scale to assure that the air quality is not always "good". However in this case it is very difficult to attach some kind of health interpretation to the index.

The differences between the two approaches vary from one pollutant to the other. On ozone, the agreement tends to be quite reasonable but for $NO_2$ and $SO_2$ the differences are substantial. For $PM_{10}$ the picture is mixed partly because the way $PM_{10}$ affects health and on what timescale this occurs is still subject to a lot of research. This implies that during typical summer episodes the indices tend to agree more or less. On days with less air pollution the interpretation gaps widen.

The long-term ↔ short-term paradox, and the problem to communicate it, typically occurs on the internet. In an annual report the focus is on long term air pollution. On text TV pages dedicated to smog warnings the focus and interpretation is based on health effects. However, internet presentations often serve multiple roles: informing the public, but also making the public aware of air quality issues. In this case the

[1] It should be note that there is increasing evidence indicating that $PM_{10}$ has both short and long-term effects even at moderate concentration levels (WHO, www).

paradox is difficult to resolve: highly variable hourly (or daily) data is being presented to assure an attractive and frequently changing situation that encourages repeated visits. On the other hand, the most challenging limit values appear to be the criteria for the year average so interpreting commonly occurring hourly values in terms of good or bad is fairly arbitrarily. They are not bad from the short-term exposure point of view but might be bad from the long-term exposure point of view. An attempt to overcome this was described by Van den Elshout (2004). For $NO_2$ and $PM_{10}$ an expected hourly pattern is established for a whole year, based on historic data. This pattern is scaled (up or down) in such a way that it provides a reference pattern that would lead exactly to the limit value. In this way a, be it hypothetical, identification of hourly values that contribute to the exeedance of the year average limit value can be made.

## 3. COMPARING CITIES ON THE INTERNET

Apart form the fact that the bands differ from one country/city/area to the other, the data behind the index also differ. Whereas most websites have a page explaining how the index is calculated, other methodological aspects are generally not explained. Does the index represent measurements at background stations, traffic stations, a mixture? And in case of PM, how is it monitored, if automated equipment is used is it corrected? In the UK the index for PM depends on the monitoring method (DEFRA, www) but in most cases there is no way of knowing how PM concentrations were established.

CITEAIR aims to provide *one* index and make a difference between background and traffic stations. The potential of having one index will be apparent from the following example in which we try to compare air quality at a given day in four cities. The indices system is described in table 1.

Table 1: Indices used on the internet in Paris, Leicester, Rome and Rotterdam

| **ATMO Paris** | ozone-1h | PM10-24h | NO2-1h | index |
|---|---|---|---|---|
| very good | 29 | 9 | 29 | 1 |
| | 54 | 19 | 54 | 2 |
| good | 79 | 29 | 84 | 3 |
| | 104 | 39 | 109 | 4 |
| average | 129 | 49 | 134 | 5 |
| mediocre | 149 | 64 | 164 | 6 |
| | 179 | 79 | 199 | 7 |
| poor | 209 | 99 | 274 | 8 |
| | 239 | 124 | 399 | 9 |
| very poor | >=240 | >=125 | >=400 | 10 |

| **UK** | ozone-8h | PM10-24h | NO2-1h | index |
|---|---|---|---|---|
| low | 32 | 21 | 95 | 1 |
| | 66 | 42 | 190 | 2 |
| | 99 | 64 | 286 | 3 |
| moderate | 126 | 74 | 381 | 4 |
| | 152 | 86 | 477 | 5 |
| | 179 | 96 | 572 | 6 |
| high | 239 | 107 | 635 | 7 |
| | 299 | 118 | 700 | 8 |
| | 359 | 129 | 763 | 9 |
| very high | >=360 | >=130 | >=764 | 10 |

| **Rome** | ozone-1h | PM10-24h | NO2-1h | index |
|---|---|---|---|---|
| good | 90 | 100 | 100 | 50 |
| moderate | 135 | 150 | 150 | 75 |
| mediocre | 180 | 200 | 200 | 100 |
| unhealthy | 360 | 400 | 400 | 200 |
| very unhealthy | > 360 | > 400 | > 400 | >200 |

| **Rotter-dam*** | ozone-1h | PM10-24h | NO2-1h | index |
|---|---|---|---|---|
| good | | 20 | 100 | - |
| moderate | 180 | 40 | 200 | - |
| bad | 240 | 60 | 400 | - |
| very bad | >240 | >60 | >400 | - |
| | | | | |

* Ozone classification from the national smog pages, other classes from a local traffic website.

Three out of four cities have an index, two indices range from 1 to 10, the other from 1 to 200. Two cities have 10 classes, one has 5, one has 4. Two describe air quality in terms of good and bad, one in terms of health and the fourth in terms of pollution levels. The class boundaries are very different. If someone would want to compare these four cities at a given moment he or she would not only have to visit four websites but also be faced with four completely different presentations and qualifications.

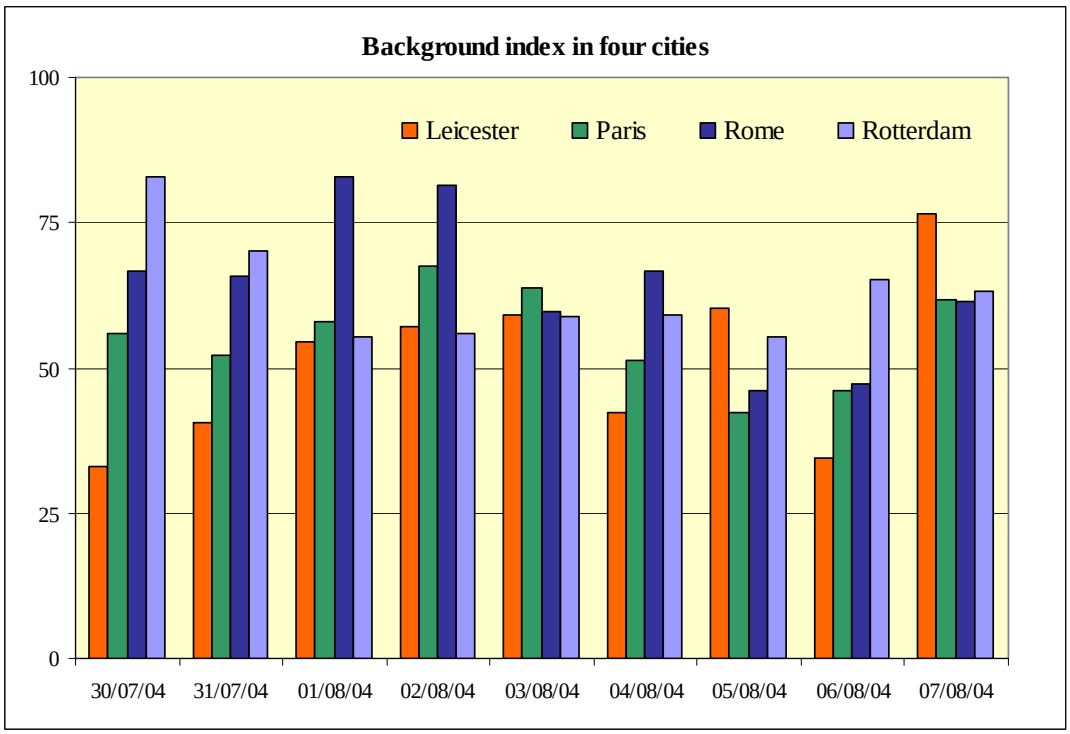


Figure 1: The CAQI applied to background stations in four cities July-August 2004

As an example, look at the period end of July - early August 2004 (Figure 1). The background index was quite high in all cities. On the 3rd of August the cities would have had a similar CAQI value. The cause of the elevated background concentrations was different though: $PM_{10}$ in Leicester and Paris, and ozone in Rome and Rotterdam. If someone had looked at the four different websites he or she would have had no possibility of comparing the information. See Table 2. Paris looks worse than Leicester as both seem to have a similar scale (1 to 10); and how to compare the score of 79 of Rome to the others: is it safe to assume that 79 out of 200 equals 4 on a 1 to 10 scale?

Table 2: the CAQI and the local indices on a day with above average concentrations

| | CAQI | Pollutant | Own city index | Pollutant | Own city classification |
|---|---|---|---|---|---|
| Leicester | 59 | $PM_{10}$ | 4 | Ozone | low-moderate |
| Paris | 64 | $PM_{10}$ | 6 | $PM_{10}$ | mediocre |
| Rome | 60 | Ozone | 79 | Ozone | mediocre |
| Rotterdam | 59 | Ozone | - | $PM_{10}$ | bad |

## 4. CITEAIR'S COMMON AIR QUALITY INDEX (CAQI)

The CAQI is calculated according the grid in table 3, by linear interpolation between the class borders. The final index is the highest value of the sub-indices for each component. As can be seen there are two CAQI-s: one for traffic monitoring sites and one for urban background sites. The traffic index comprises $NO_2$ and $PM_{10}$, with CO as an auxiliary component. The background index obligatory comprises $NO_2$, $PM_{10}$ and $O_3$, with CO and $SO_2$ as auxiliary components. In most cities the auxiliary components will rarely determine the index (that is why they are auxiliary) but in a city with industrial pollution or a seaport $SO_2$ might occasionally play a role.

Table 3: Proposed pollutants and calculation grid for the CAQI

| Index | Class | Traffic | | | City Background | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | | $NO_2$ | $PM_{10}$ | CO | $NO_2$ | $PM_{10}$ | $O_3$ | CO | $SO_2$ |
| Very low | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 |
| | 25 | 50 | 25 | 5000 | 50 | 25 | 60 | 5000 | 50 |
| Low | 25 | 50 | 25 | 5000 | 50 | 25 | 60 | 5000 | 50 |
| | 50 | 100 | 50 | 7500 | 100 | 50 | 120 | 7500 | 100 |
| Medium | 50 | 100 | 50 | 7500 | 100 | 50 | 120 | 7500 | 100 |
| | 75 | 200 | 75 | 10000 | 200 | 75 | 180 | 10000 | 300 |
| High | 75 | 200 | 75 | 10000 | 200 | 75 | 180 | 10000 | 300 |
| | 100 | 400 | 100 | 20000 | 400 | 100 | 240 | 20000 | 500 |
| Very High | > 100 | > 400 | >100 | >20000 | > 400 | >100 | >240 | >20000 | >500 |

| | |
|---|---|
| $NO_2$, $O_3$, $SO_2$: | hourly value / maximum hourly value in μg/m3 |
| CO | 8 hours moving average / maximum 8 hours moving average in μg/m3 |
| $PM_{10}$ | hourly value / daily average in μg/m3 |

Comparing air quality in different cities is a tricky issue: is the air quality being determined in the same way (this mainly applies to particulate matter) and at comparable locations? This is not an issue that we, as the CITEAIR project and the proponents of the QACI, can solve. The website will take for granted whatever a city supplies as input in either category. However, as a first step to improve comparability, the index will be reported both for roadside and city background locations. This is considered an important improvement over city averages: some monitoring networks are designed to monitor, or spot areas of poor air quality (with possibly a high number of roadside stations) whereas others are aimed at providing an average city picture.

The CAQI is used both for a daily index and for an hourly index. In the website the daily index will be shown for the past day (D-1). For the current day, the past 24 values of the hourly index will be available, to be updated every hour. A daily index for today would need forecasting or 'nowcasting' a facility that is not available in each city with a monitoring network, hence the option of an hourly index. The hourly index is also a reasonably dynamic parameter, enticing repeated visits to a website.

Participating cities are advised to submit average data from the stations they qualify as city background and traffic. The use of average data leads to better representativity and less missing data. However, if a city wants to select (or only has) one station in each category, that data will be used.

The choice of the classes in the CAQI is heavily inspired by the EU legislation and based on a compromise between the participating cities. The dividing line between medium and high is often linked mainly to the values mentioned in the directives: alert thresholds ($SO_2$, $NO_2$, $O_3$) or air quality objectives when available on a daily basis (CO and $PM_{10}$). Class borders were regularly spaced for the main components. $PM_{10}$ is an exception. To avoid that the CAQI is completely dominated by $PM_{10}$ the value of 50 μg/m3 as a daily average was positioned as the bordering line between low and medium. For the setting of the CO and $SO_2$ borders additional inspiration was sought from the DAPPS index which aims to define the component sub-indices based on the relative risks attributed to each component (Cairncross and John, 2004).

The CAQI resembles the ATMO index discussed above and it differs substantially from, for example, the UK and US-EPA indices. It therefore shares the drawbacks of the ATMO (no clear link with health effects, fairly arbitrarily quality interpretation for short-term exposure). But it also shares its advantage: frequently changing index values that capture the hour-by-hour changes and make a website dynamic. The latter was of overriding importance as raising awareness is a key objective of the common website.

As cities join the website the exact bands might need reassessment to maintain an attractive comprise. The current classes were derived based on the episodes of august 2003 and a recent year of data (April 2004 - March 2005) for the cities of Leicester, Paris, Rome and Rotterdam.

Table 4: Percentage of hours that a pollutant determines the final index (2004-2005)

| Traffic Index | Leicester | Paris | Rome | Rotterdam |
|---|---|---|---|---|
| $NO_2$ | 85 | 53 | 31 | 49 |
| $PM_{10}$ | 14 | 47 | 69 | 51 |
| CO | 1 | 0 | 0 | 0 |
| | 100 | 100 | 100 | 100 |

| City background Index | Leicester | | Paris | | Rome | | Rotterdam | |
|---|---|---|---|---|---|---|---|---|
| | main | main + auxiliary | main | main + auxiliary | main | main + auxiliary | main | main + auxiliary |
| $NO_2$ | 30 | 33 | 35 | 35 | 24 | 24 | 21 | 20 |
| $PM_{10}$ | 24 | 25 | 27 | 27 | 36 | 35 | 46 | 45 |
| $O_3$ | 46 | 42 | 38 | 38 | 40 | 42 | 34 | 26 |
| CO | | 0 | | 0 | | 0 | | 0 |
| $SO_2$ | | 0 | | 0 | | 0 | | 9 |
| | 100 | 100 | 100 | 100 | 100 | 100 | 100 | 100 |

The tables show that in these four cities CO almost never plays a determining role in neither the traffic nor the background index. For the second auxiliary variable $SO_2$ the situation is slightly different. In Rotterdam, with a seaport and a petrochemical industry, in 9 % of the hours $SO_2$ would have determined the index[1].

Figure 2 shows the daily indices in the four test cities for a period of twelve months. The Rome background index shows a distinct seasonal pattern. In summer the background index is mainly determined by ozone, in winter by $PM_{10}$ and, to a lesser extent, $NO_2$. The seasonal pattern is absent in the other cities, though the shift in pollutants determining the index is fairly identical. The winter of 2004/2005 was rather mild so only some days with a higher index can be seen. The winter doesn't show up clearly. The traffic index is significantly higher than the background in Rome and Paris. This was to be expected in large cities with a big vehicle fleet, typical street-canyons, large ring roads, etc. In the much smaller city of Rotterdam the traffic index is only slightly higher than the background index. Leicester provides a mixed picture. With $NO_2$ being the dominant traffic pollutant in Leicester, the traffic index is relatively low in summer and higher in winter.

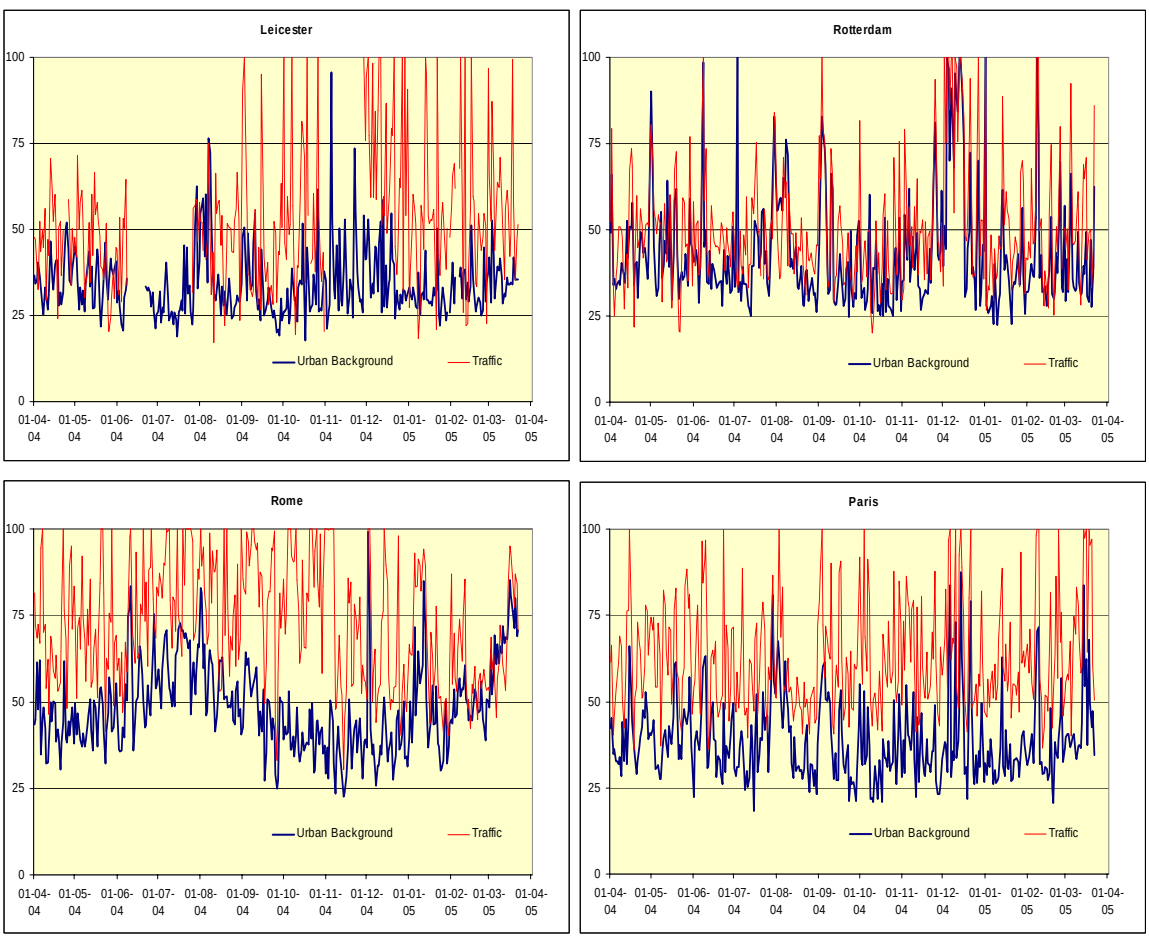


Figure 2: The CAQI (traffic and background) in four cities

[1] In fact even in Rotterdam this is exceptional. $SO_2$ determined the index in a short period with flares due to unexpected maintenance in a petrochemical plant and otherwise low concentrations.

The usefulness of a separate background and traffic index can be seen from figure 3 showing the daily index in Paris in August 2003. August 2003 was characterised by hot weather and poor dispersion conditions, leading to very high ozone concentrations. Except for a few days at the end of the month the background index was dominated by ozone. The traffic index was mainly determined by nitrogen dioxide with a few days of $PM_{10}$.

The poor dispersion conditions, combined with a large amount of imported ozone, are evident from the fact that the background index is similar or even higher than the traffic index, whereas normally (e.g. good dispersion conditions) there would be a gap of 15 to 25 index points between traffic and background. From the graph it can be seen that the traffic index drops in weekends (days labelled 6 and 7) whereas the background index rises. In this ozone dominated month, the relative lack of fresh exhaust (NO) emissions, leads to higher ozone concentrations in the weekend. This weekend ozone effect is well known (Lawson, 2003).

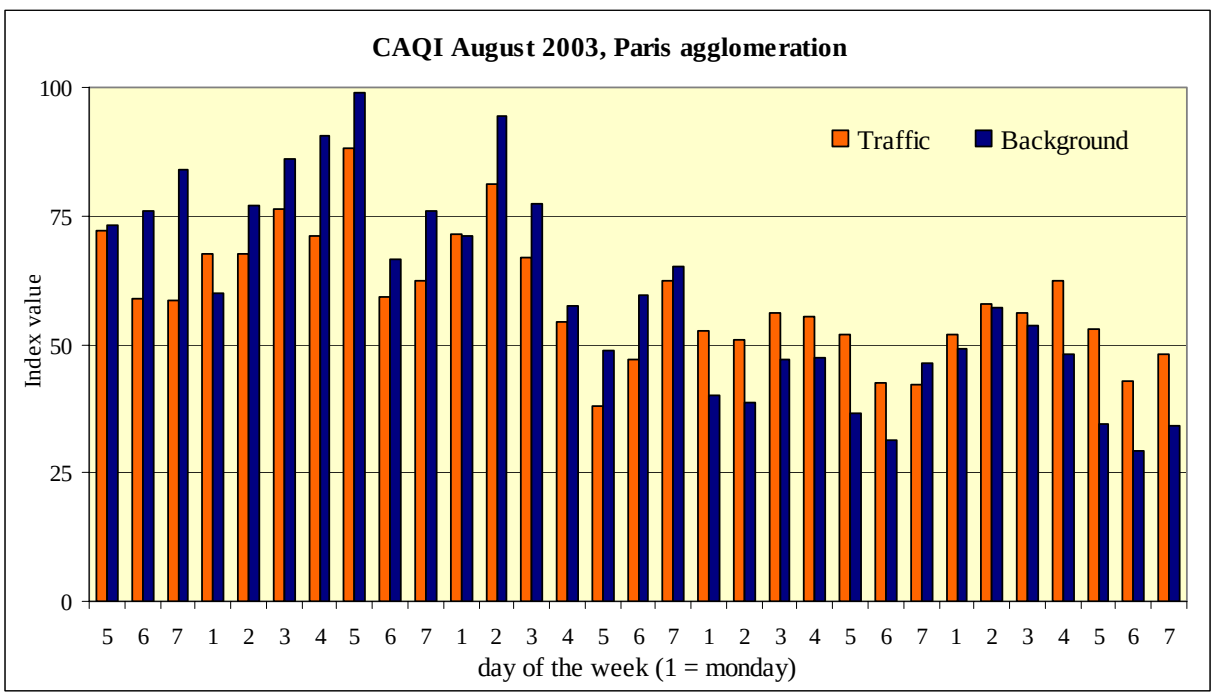


Figure 3: The traffic and background indices during an episode in Paris

## 5. CITEAIR AND THE COW

The CITEAIR common website (COW) will be launched in 2006, initially with contributions from the CITEAIR partners. Once the site is fully operational other cities will be invited to join and upload their data as well. The COW is meant to be an interesting *complement* to the cities' own websites. The COW and the CAQI are not launched to replace existing websites or indices. For those cities who are not yet on the internet, and/or do not currently use an index, the COW and the CAQI could

be their primary platform. It is envisaged that most calculations are done inside the COW. Cities only have to upload their data (through an agreed ftp format).

Cities engage in communication with the public, not only because of legal obligations but often also to raise awareness. This implies that air quality issues have to be presented in an attractive and educational way. The possibility to compare your own local air quality to a number of other European cities could be an asset in this respect. To make the COW attractive and to solicit repeated visits dynamic content such as an hourly updated index will be presented.

However, not every city has its own network or both traffic and background stations, and not everyone is able to deliver data in near real time. If cities want to participate in only one of the indexes, can only deliver data on a daily basis, or even only present year average data, they can still join (the concept of) the COW. Different sections of the website will provide a platform to compare different data (year average, daily, hourly) so participation is not limited to those with their own automated network.

## 6. ACKNOWLEDGEMENTS

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# EVALUATION OF SPATIAL VARIATIONS OF NON-ROAD AND AREA SOURCE EMISSIONS IN A SEMI-ARID COASTAL URBAN AIRSHED USING GIS TOOLS

**Kuruvilla John[1] and Zuber Farooqui[2]**

1. Frank H. Dotterweich College of Engineering, Texas A&M University-Kingsville, MSC 188, Kingsville, Texas 78363, k-john@tamuk.edu
2. Department of Environmental and Civil Engineering, Texas A&M University-Kingsville, MSC 213, Kingsville, Texas 78363, zfarooqui@even.tamuk.edu

## ABSTRACT

Emissions inventory needed for photochemical modeling are typically gathered at the county level. The spatial variations of non-road and area source emissions within a county have been largely ignored due to the difficulty in collecting, processing, and analyzing the data at smaller geographic scales. This method may affect regional photochemical modeling performance with respect to emissions inputs. This study adopted Geographical Information System (GIS) tools capable of managing spatially referenced data in order to evaluate the spatial variations of emissions from non-road and area sources within a semi-arid coastal urban airshed of Corpus Christi, Texas, based on activity, survey and surrogate data. The approach consisted of establishing spatial database, disaggregating county-wide emission into finer grids, and segregating geo-referenced emissions into the photochemical model grid. The spatial database included identification of facilities location and proper emission surrogates developed for allocation of these sources into proper grids. Key area source emission categories were considered as localized minor point sources wherever suitable such as airport location and oil/gas production activities. Locations of the sources were geo-referenced using GIS tools. A top-down approach was adopted for categories where locations of sources were unavailable. Population surrogates were used for all emissions related to human activities. County-wide emission estimates were then allocated to smaller grid resolution (1 x 1 km) based on surrogates, and spatial variations were then evaluated. Categories such as residential natural gas, liquefied petroleum gas, lawn and garden equipment and other residential related emissions were apportioned using housing surrogates. Finally, area source emissions that were geo-referenced and that had low amount of emissions, such as gasoline marketing, were aggregated into corresponding grid emissions. GIS was extensively used in the study to visualize process spatial data, and perform overlay operations to derive cell based inputs to integrate geo-coded air emissions data. The data generated provided spatially resolved gridded emissions useful for characterizing the impact of anthropogenic emissions in a photochemical model. This study provided the basis for the development of an environmental knowledge management system using GIS and it will be useful for air quality planners, policy-makers and analysts.

**Keywords:** Emissions inventory, photochemical model, GIS, urban airshed, spatial apportionment

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# MEASUREMENTS OF PARTICULATE MATTER $PM_{10}$ IN AIR UNDER LOW AIR TEMPERATURE CONDITIONS

**Igor Cuhalev and Roman Kocuvan**

Elektroinstitut Milan Vidmar, Hajdrihova 2, SI-1000 Ljubljana, Slovenia
igor.cuhalev@eimv.si, roman.kocuvan@eimv.si

## ABSTRACT

The paper deals with the issue of high $PM_{10}$ concentrations under low-temperature states in areas affected by thermal power plants as well as in urban environments. Under winter conditions, there are considerably increased $PM_{10}$ concentrations in air registered at the time of minimal concentrations of primary pollutants ($SO_2$ and $NO_x$). $PM_{10}$ concentrations are on the level of 80 $\mu gm^{-3}$ and more. This phenomenon has been noted on a rather vast area of Slovenia within the radius of 50 x 50 km. The characteristic pattern of the $PM_{10}$ concentrations at the time of low air temperatures is attributed to processes of secondary organic aerosols (SOA). This process is also connected with the temperature inversion phenomenon. Characteristic for a typical time pattern of its episodes is an increase in the $PM_{10}$ concentrations, and partly also in the $NO_x$ concentrations, followed by a distinct photochemical process with increased ozone and at the same time also $SO_2$ concentrations from the higher atmospheric layers. For the established organised air monitoring it is recommended to recognise conditions giving rise to SOA and to adequately notify the public about the phenomenon of high $PM_{10}$ concentrations and later also of the rest of photochemical oxidants.

**Key words**: Air Pollution, $PM_{10}$, SOA, Ozone, Low Air Temperatures

## 1. INTRODUCTION

The Milan Vidmar Electric Power Research Institute (EIMV) is responsible for performing continuous measurements of air quality at over 15 sites in areas affected by thermal power plants emissions all over Slovenia. Besides these measurements, it also measures air quality with its mobile measuring station for several other customers. Thus obtained measurement results are on an hourly basis available on the EIMV web pages as well as in its monthly and annual reports. At each measuring site there are measurements made of $SO_2$ concentrations and meteorological parameters and at some particular sites there are measurements of $NO_x$, $O_3$ and $PM_{10}$ made, too. The decision as to which parameters would be measured depends primarily on the type of the fuel used by a particular power plant.

When processing monthly measurement results obtained for various measuring sites, we quite often detect that time patterns of the pollutant concentration development deviate from the usual profession-grounded expectations. In such cases we analyse

the pollutant time pattern more in detail and take the steps as necessary to exclude any possible measurement error that might be attributed to the used measurement instruments or conditions at which measurements are taken.

Here below we show results of continuous measurements of $PM_{10}$ air concentrations made in February 2005 during a several days lasting period of extremely low air temperatures. These episodes became recognisable at -10ºC and they intensified as the temperatures continued lowering. At several measuring sites located all over Slovenia having different geographic and settlement characteristics as well as different pollution sources, we noted that they have a similar daily pattern of $PM_{10}$ air concentration variations. We detected that when air temperatures were low during a period of several days, $PM_{10}$ air concentrations reached the value exceeding $80\mu gm^{-3}$ (Fig.1). Our measurements were made on a continuous basis and the pertaining results were released at 30-minute intervals. In our analysis we focused on three measuring sites (A, B, C) for which we collected and measured all the samples ($SO_2$, $NO_x$, $PM_{10}$ and also meteorological parameters), and two measuring sites for which we were interested only in $PM_{10}$ concentrations. The A and B sites are located in an area affected by a thermal power plant, whereas the C site is in a minor urban settlement. The averaged distance between each of the observed sites is above 50 km.

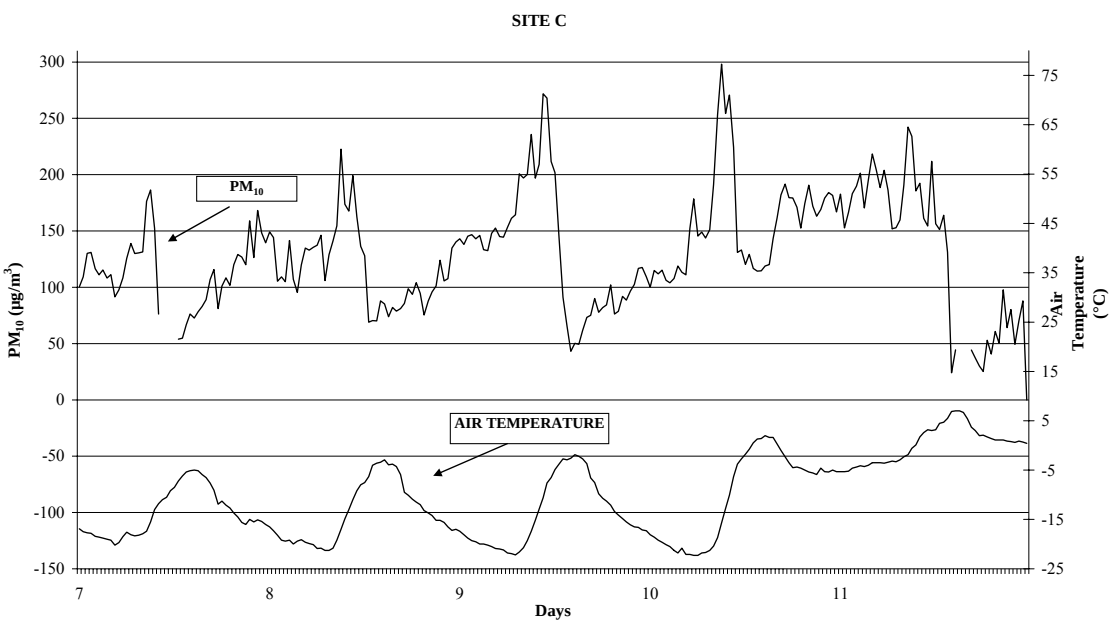


Figure 1: $PM_{10}$ concentrations in dependence of air temperature at site C

Presentation of the episode marked by increased $PM_{10}$ concentrations at low air temperatures is based on the following unexpected samples:

- $PM_{10}$ concentrations take place at low air temperatures and disappear after air temperatures increase,
- the time pattern for $PM_{10}$ concentrations in combination with low air temperatures occurs in a broader area within the radius of 50 x 50 km and more,

- concentrations of other pollutants ($SO_2$, $NO_x$ and $O_3$ ) are substantially lower and they only minimally follow the $PM_{10}$ time pattern.

As a result of the above basic observations, we found it necessary to exclude from the very start the possibility of existence of any measuring instrument error that may occasionally take place at low temperatures. The next to follow was a detailed comparative analysis between the $PM_{10}$ time samples and available meteorological parameters.

### 2.1 $PM_{10}$ Measuring Instrument Operation Analysis

Having compared two $PM_{10}$ measuring devices (Salminen and Karlsson, 2003), the authors established that at temperatures below the freezing point a higher concentration was systematically registered with the $PM_{10}$ measuring instruments together with β-attenuation unlike is the case with the referent gravimetric measuring instrument. In our case, we do not use the same type of the measuring instrument, but the air sample make-up method is the same. Thus measured increased concentrations were some 2 $\mu gm^{-3}$ higher. The value of concentration deviations of these two measuring instruments cannot be compared with our case in which concentrations increased above $80\mu gm^{-3}$.

For our measurements we used a $PM_{10}$ measuring instrument of the R&P (Rupprecht & Patashnick Co.) manufacture and of the TEOM 1400a type. It operates on the basis of the gravimetric principle and is suitable for continuous monitoring of mass concentrations of particulate matters. It uses the TEOM (Tapered Element Oscillating Microbalance) technology. Its operation follows the principle of indirect mass measurement by measuring the frequency of the used pendulum on which air particles are deposited.

When an air sample is placed in a heated pipe, it attains the set temperature which is higher than the ambient temperature, usually 40-50°C. By doing so, the effect of variable environmental conditions is reduced and the sample is freed from humidity. Consequently, the sample constantly has a low and stable relative humidity and its deposition on the filter is prevented. Correct operation of the measuring instrument is conditioned by its continuous temperature control a precise flow control.

We used the following two approaches to validate correct operation of the used measuring instrument:

- time samples of the obtained $PM_{10}$ measuring results were sent to a local agent authorized by the measuring instrument seller,
- a comparison was made between measuring results obtained with the $PM_{10}$ measuring instrument operating on the basis of 24-hour collection of samples on the filter and their weighing.

The local agent authorized by the measuring instrument seller advised us that their experts for the TEOM measuring instruments had examined the data and also directly contacted R&P to establish whether there was any possibility that measuring results were affected by low air temperatures. The reply they received was that

according to the R&P practice there was no connection between characteristics of the measuring instrument operation and low air temperatures.

The Slovenian Environmental Agency (ARSO) has presented us measuring results obtained with the Leckel measuring instrument collecting samples on the filter on a 24-hour basis whereupon these samples are weighed. Their instrument is qualified as a referent instrument (TEOM) used for determination of the constant correction for the TEOM measuring instruments. When these instruments are used at temperatures ranging from 40-50°C, some of the components evaporate. Comparative measurements made by the referent and the TEOM (ARSO) measuring instrument were time overlapped; they were both taken on the $10^{th}$ and $11^{th}$ February 2005. They were made at the distance of 1 km from site B. In Table 1 we show 24-hourly values for the two measuring instruments. As seen from them, the referent measuring instrument, too, measured in the time of our observation higher $PM_{10}$ concentrations. They are equal to those obtained with measurements made with TEOM. The reason for the difference between the measurements is air sample heating in the TEOM measuring instrument.

Table 1: Comparison of 24-hourly $PM_{10}$ concentrations obtained with the referent (Leckel) and TEOM measuring instrument

| Date/Measuring instrument | Leckel ($\mu gm^{-3}$) | TEOM ($\mu gm^{-3}$) |
|---|---|---|
| $10^{th}$ February 2005 | 173.8 | 159.0 |
| $11^{th}$ February 2005 | 100.87 | 87.4 |

Our conclusion based on the above comparative measurements is that the obtained time sample of the increased $PM_{10}$ concentrations occurs under conditions of low air temperatures in both investigated types of the measuring instrument. Our position is that, as shown by the available data (comparative measurements), there is no connection between technical characteristics of the measuring instrument, in our case the TEOM measuring instrument, at the occurrence of increased $PM_{10}$ concentrations and low air temperatures. Below we present analyses of other physical and chemical processes in the atmosphere measured during the time of the observed episode.

### 2.2 Analysis Of Connection Between Gas Pollutants And $Pm_{10}$ Concentrations

The pollutant registered at the basic three locations is $SO_{2..}$ It is emitted either from the local thermal power plant or fire systems. As seen from the characteristics of the $SO_2$ concentration variations (Fig. 2), the atmosphere gets stratified at low air temperatures. This stratification is most likely connected also with the temperature inversion. At the ground level, $PM_{10}$ concentrations (measurements) constantly increase, whereas in higher layers the quantity of $SO_2$ increases (assumption). When the air temperature is above -5ºC and when there is wind that causes air mixing, $PM_{10}$ concentrations decrease as a result of such mixing and at the same time $SO_2$ is shifted from the upper layers thus making its concentrations to increase. After the temperature re-drops, the time patterns of $SO_2$ and $PM_{10}$ concentrations again take the reverse course.

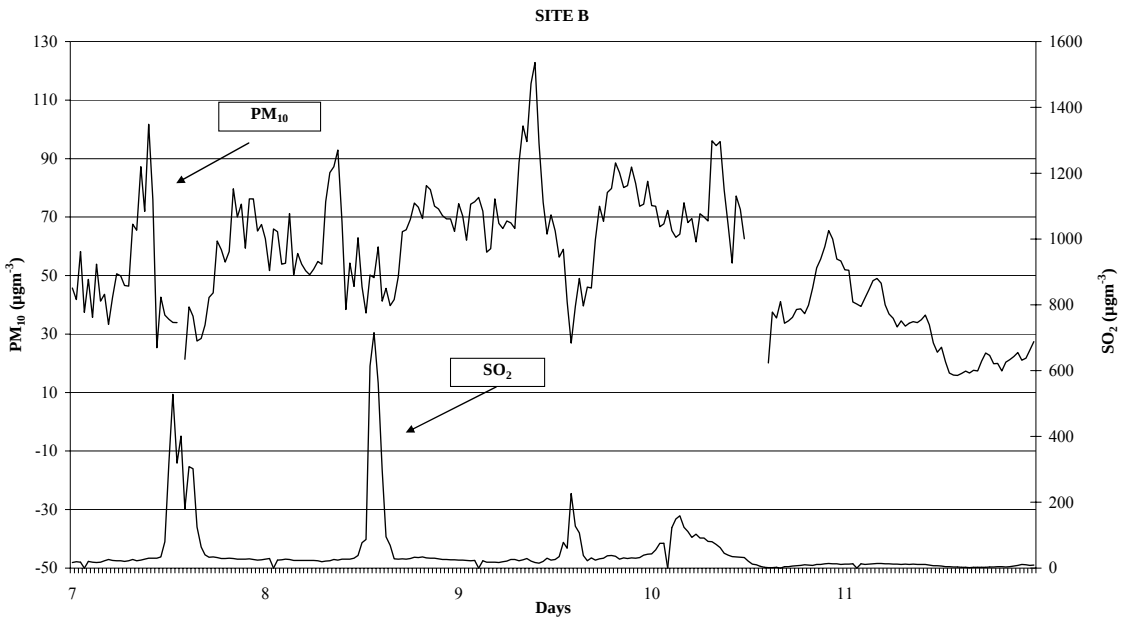


Figure 2: $PM_{10}$ concentrations in connection with $SO_2$ concentrations at location B

$NO_x$ concentrations partially follow the time pattern of $PM_{10}$ concentrations as it can be seen at site C (a minor fuel-oil heated urban settlement). $NO_x$ and $PM_{10}$ concentrations (Fig. 3) have similar time patterns.

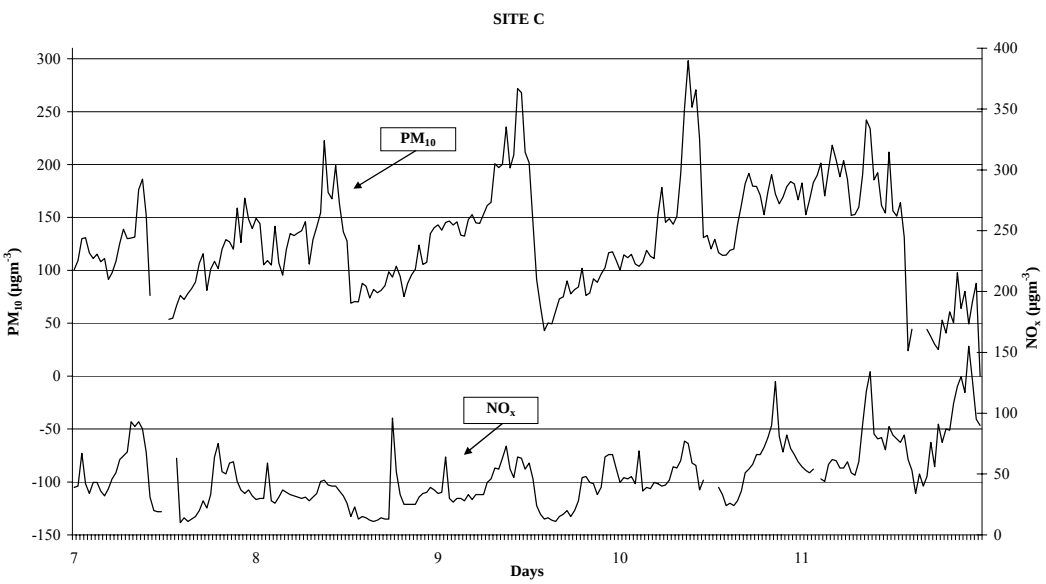


Figure 3: $PM_{10}$ concentrations in connection with $NO_x$ concentrations at site C

Episodes of increased ozone concentrations are for the observed season of the year and air temperatures surprisingly high; they exceed 100 $\mu gm^{-3}$. The ozone time pattern is similar to the $SO_2$ pattern and is evidently connected with deterioration of the inversion layer. When $PM_{10}$ concentrations increase, ozone concentrations

change minimally along with the simultaneously increasing of $NO_x$ concentrations. Yet, when inversion deteriorates, temperatures rise and when there is wind, ozone concentrations very much increase and at the same time $PM_{10}$ and $NO_x$ Concentrations decrease. Fig. 4 shows ozone concentrations at a location some 3 km distant from site A.

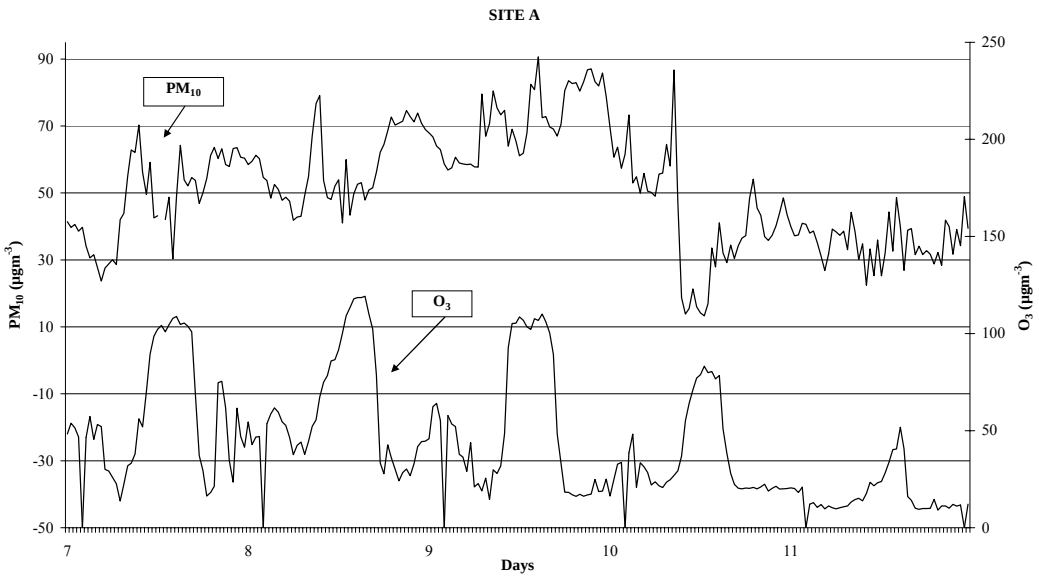


Figure 4: $PM_{10}$ concentrations in connection with ozone concentrations at site A

When connecting $PM_{10}$ time concentrations with gas pollutants ($NO_x$ and $O_3$) we can recognise the characteristic phenomenon of inversion and its deterioration and at the same time also the presence of photo-chemical processes reflecting themselves in the $NO_x$ and ozone behaviour. Time patterns of $PM_{10}$ and $NO_x$ concentrations match to some extent, whereas in $SO_2$ and ozone there are inversion patterns with regard to $PM_{10}$.

Being convinced that $SO_2$ is emitted from stationary sources (thermal power plant and minor fire systems) and is regarded as a primary pollutant, we can classify $NO_x$ as a pollutant of a secondary source as a result of the fact that its concentration time trend does not overlap with that of the $SO_2$ concentrations. For the same reason, $PM_{10}$, too, can in the observed case be classified as a secondary pollutant.

### 2.3 Analysis Of Dependence Of $Pm_{10}$ Concentrations On Meteorological Parameters

In our analysis we shall compare the basic three parameters: air temperature, relative humidity and wind speed. For the air temperature we already established above (Fig.1) a strong inversion connection with $PM_{10}$ concentrations.

Since the air temperature is connected with the wind speed, we expect there would be a strong connection between the two patterns. In Fig. 5 we can well see the connection between $PM_{10}$ concentrations and wind speed at the time of inversion deterioration taking place at site C.

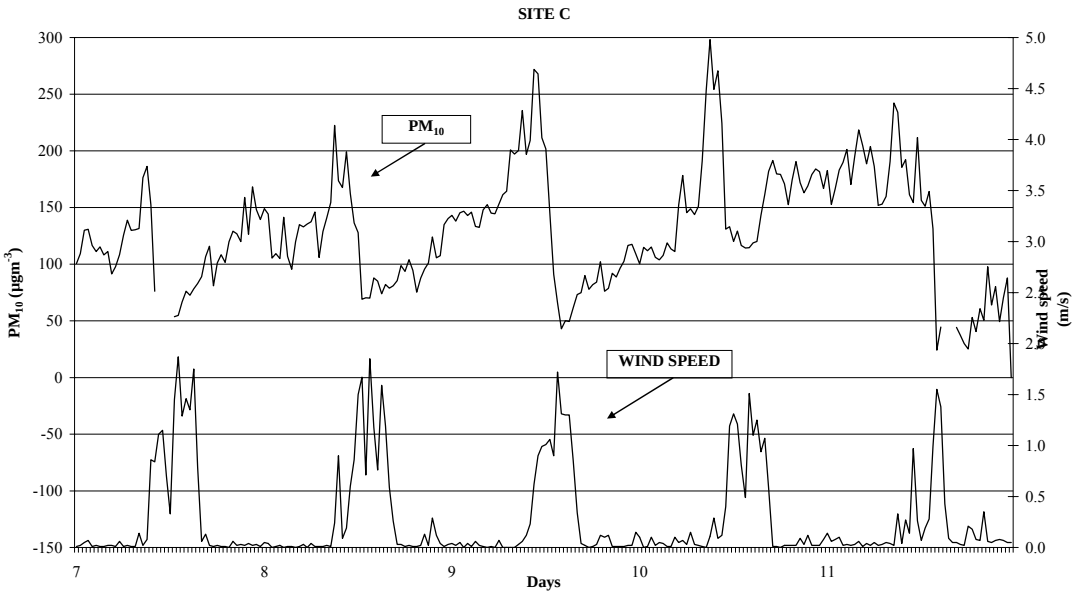


Figure 5: $PM_{10}$ concentrations in connection with the wind speed at site C

With regard to the recognised inversion dependence of $PM_{10}$ concentrations on air temperatures, we may expect that for relative humidity, too, there would be overlapping of time patterns of the $PM_{10}$ concentrations and the humidity content (%) in the air (Fig. 6), which is seen at site A.

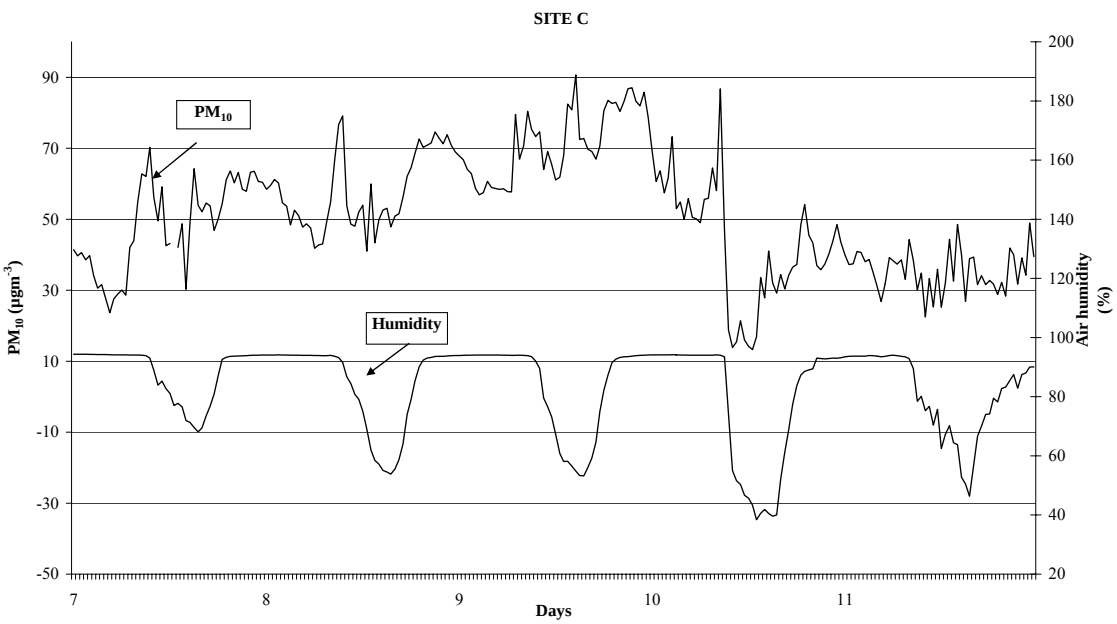


Figure 6: $PM_{10}$ concentrations in connection with relative humidity at site A

## 2.4 Discussion

Judging from the above data and analyses, we can conclude that in the observed time period in which air temperatures were low there was stratification of the ground-level layers of the atmosphere. With the increase in temperatures, this stratification

deteriorated. Having analysed the temperature and humidity values, our conclusion is that at each of the observed locations there was temperature inversion. This cannot be said for higher layers of the same locations since we do not possess for them data about temperature measurements.
From processes associated with air pollution we must mention ozone and nitrate oxides, which exhibit intensive photochemical processes, as well as increased $SO_2$ concentrations which also occur at inversion deterioration. Independently from these pollutants there are $PM_{10}$ concentration samples showing also a weak connection with the presence of nitrate oxides.

We assume that we have to deal with secondary organic aerosols (SOA) whose formation is described by (Chu at al, 2004). Conditions for the SOA formation are in our case the same as those defined by these authors, i.e. high humidity content, weak mixing of the atmosphere (inversion) and much of nitrate oxides. The only difference is that in our case SOA occur at low air temperatures while according to Chu et al. they occur at moderate winter temperatures. In both cases SOA occur in the winter season.

Low air temperatures give rise to an increased level of air emissions. Most affected are $PM_{10}$ concentrations whose share is several times greater than the one of gaseous pollutants. This means they do not increase proportionally with the overall emissions. The reasons for their increase are SOA which are described with physical-chemical processes taking place on active surfaces of $PM_{10}$:
- condensation, nucleation,
- absorption, adsorption,
- chemical processes, mostly oxidation.

Particles having a decisive role in formation of the above processes are:
- burnt particles – inorganic ash. In inorganic ash, which has hydrophil characteristics, there are neutralisation processes of acid components, in particular $SO_2$, $NOx_{,,}$ as a result of the alcalic character. As seen from the NOx and $O_3$ concentrations, in the observed period of time there are strong oxidation processes going on. They are favourable for formation of sulphates and nitrates which bind themselves together with the present water steam on the surface of the inorganic ash and thus increase the particle mass.
- »Black Carbon« particles, i.e. soot and smoke. These particles originate from incomplete burning and are the source for SOA formation. It is on these particles that absorption of mostly hydrophobe components takes place, this means various organic substances – carbon-hydrogens, vaporous organic compounds, dioxides, furans, aromatic compounds, which makes thus created SOA so extremely harmful for the human health.
- particles as remnants of non-burnt fuels whose impact on processes is not considerable.

As known from adsorption isotherms (Colls, 2002), adsorption of hydrophobic pollutants increases during the time of low air temperatures.

All the above processes give rise to an increase in the particle mass which - as a result of noxious substances – becomes on the surface the more and more aggressive and harmful for the human health.

## 3. CONCLUSION

Occurrence of increased $PM_{10}$ concentrations under conditions of low air temperatures in February 2005 can entirely be attributed to SOA. So far performed air pollution monitoring in areas affected by thermal power plant emissions ($SO_2$, $PM_{10}$ and rarely $NO_x$) has been based mostly on primary pollutants. Development of measuring instruments and increased scope of air quality measurements have revealed that secondary pollution is becoming the more and more prevailing: photochemical processes (ozone, $NO_x$) and SOA ($PM_{10}$). As seen from our analysis of February 2005, concentrations of primary pollutants ($SO_2$ and $NO_x$) were negligible. They can be associated with primary polluting originating from thermal power plants or local pollution sources. Despite their low level, they significantly contributed to high concentrations of ozone as well as $PM_{10}$, which was formatted with SOP, as a result of meteorological conditions (low temperatures, high humidity level, temperature inversion, minimal presence of primary pollutants.

The investigated time patterns of pollutant concentrations under conditions of low winter temperatures show very much unfavourable development scenarios of air pollution:

- a decrease in air temperatures and an increase at the ground level of $PM_{10}$ concentrations because of SOA,
- upon air inversion deterioration, at the ground level there is an increase in $SO_2$ concentrations accumulated from upper layers of the temperature inversion. At the same time, an intensified photochemical reaction of pollutants takes place at the ground level reflecting itself in increased ozone concentrations.

Increased $PM_{10}$ concentrations under low-temperature conditions in areas where primary pollutant sources already exist are the reason for the necessity of establishing an appropriate system for $PM_{10}$ concentration monitoring and informing the public about the air pollution rate as well as scenarios forecasting the expected increase in $SO_2$ concentrations and ozone during the time of temperature inversion duration or deterioration.

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# POLLUTED DAYS AND CLEAN DAYS: DIFFERENCES IN THE DILUTION POTENTIAL OF THE LOWER ATMOSPHERE AND IN THE MASS CONCENTRATION AND CHEMICAL COMPOSITION OF ATMOSPHERIC PARTICLES

**Cinzia Perrino, Maria Catrambone and Adriana Pietrodangelo**

C.N.R. Institute of Atmospheric Pollution
Via Salaria km. 29,300 - C.P. 10 - 00016 Monterotondo Stazione (Rome), Italy
perrino@iia.mlib.cnr.it

## ABSTRACT

The mass concentration and chemical composition of $PM_{10}$ and $PM_{2.5}$ has been determined daily during several field campaigns carried out in Central Italy. The studies were addressed to elucidate which chemical components were mainly responsible for the increase of particle concentration during pollution episodes.
During the studies, the dilution properties of the lower atmosphere have been estimated on the basis of natural radioactivity measurements. The results confirm that the atmospheric mixing plays a primary role in determining anthropogenic pollution episodes and indicate that during stability periods the increase in PM concentration is mainly due to carbon compounds and ammonium salts.

**Key words:** Atmospheric pollution, Particulate matter, Natural radioactivity, Atmospheric stability, Ammonium nitrate

## 1. INTRODUCTION

The limit values for $PM_{10}$ concentration, set in the European Union by the First Daughter Directive to the Air Quality Framework Directive, address both annual average concentration (40 ug/m$^3$) and daily average concentration (50 ug/m$^3$, to be exceeded no more than 35 times a year). In this perspective, the understanding of the different mechanisms that stay behind the increase in particle air concentration has become particularly important.
The comprehension of a pollution event requires not only the accurate determination of the chemical composition of the atmospheric particles but also the elucidation of the role played by the dilution properties of the lower atmosphere. The increase in the air concentration of a pollutant, in fact, is the result of the combination of the emission (and/or production) of the pollutants in the atmosphere and of the reduced capacity of the atmosphere to dilute them.
Information about the mixing properties of the boundary layer can be easily obtained by monitoring natural radioactivity due to Radon short-lived decay products. The emission flow of Radon from the ground, in fact, can be considered to be constant in the time and space scale of our observations; it follows that Radon air concentration at a given location only depends on the dilution factor. By determining the natural

radioactivity due to the Radon progeny attached to atmospheric particles we can obtain a reliable picture of the dilution properties of the lower atmosphere (Perrino et al., 2001, Vecchi et al., 2004).
The study of the mixing properties of the lower atmosphere allows us to identify periods characterised by a weak atmospheric mixing, which favour the occurrence of anthropogenic primary and secondary pollution events, as well as periods characterised by advection, during which natural pollution events may be recorded. The subsequent chemical analysis of the atmospheric particles allows a complete characterisation of the pollution event. The complex chemical nature of particulate matter prevents the determination of all the chemical species, but the bulk determination of the main compounds (elemental carbon, inorganic compounds – main ions and metals - and organic carbon as a total value) is sufficient to identify the natural or anthropogenic characteristics of each pollution episode, and to estimate the primary and secondary contribution to anthropogenic events (Chan et al., 1997, Marcazzan et al., 2001, Harrison et al., 2003, Harrison et al., 2004, Rees at al., 2004, Hueglin et al., 2005).

## 2. EXPERIMENTAL

The data reported in this paper have been collected during two field studies. The first one was carried out during the summer and the winter of 2003 in the framework of a research project funded by the Municipality of Rome and addressed to obtain a first characterisation of the chemical nature of particulate matter in the Rome area. The determinations were carried out at the urban background station of Villa Ada (VA), sited inside a park in the urban area of Rome, at a traffic station in Rome and at the semi-rural station of Montelibretti (ML) sited about 30 km NE of the centre of the city.
The second field study was carried in the framework of the Project “Fine Particles”, funded by the Lazio Region and aimed to characterise $PM_{10}$ and $PM_{2.5}$ pollution events in the area of the Region. It was carried out between October 2004 and July 2005 in six sampling stations, the VA station and a traffic station in Rome, the ML station, two urban stations in the cities of Viterbo and Latina and the regional background station of Fontechiari.
During both studies, natural radioactivity was measured by means of a automated stability monitor (PBL Mixing Monitor, FAI Instruments, Fontenuova, RM-I) that basically consists of a particulate matter sampler equipped with a Geiger–Muller counter for determining the total beta activity of the short-lived radon progeny. The instrument is automatic and operates on two filters at the same time: sampling is performed on the first filter for a 1-h sampling duration, then this filter undergoes the beta measurement phase while a second filter undergoes the sampling phase (residual radioactivity is taken into account by a software procedure). These instrumental features assure that the short-lived beta activity of the particles is determined continuously over an integration time of 1 h with a beta measurement period long enough to guarantee a good accuracy of the results.
Particulate matter concentration was determined on a daily basis by means of two beta monitors: SWAM5a (FAI Instruments, Fontenuova, RM-I) or SM200 (Opsis

AB, Furulund-S).
During the 2003 study EC/OC compounds were determined, over 6-h time periods, by means of a automatic monitor employing a direct-measuring thermal-$CO_2$ technique (Series 5400 Ambient Carbon Particulate Monitor, Rupprecht & Patashnick Co, NY-USA). During the 2004-2005 study, the EC/OC compounds were daily collected on quartz filters and then analysed by means of a thermo-optical analyser (OCEC Carbon Aerosol Analyser, Sunset Laboratory, OR-U.S.A.).
For the analysis of ions and metal content, particulate matter was daily collected on Teflon filters. These were analysed for their metal content (Al, Si, Fe, Mg, Ca, Ti, S) by energy-dispersion X-ray fluorescence (X-Lab 2000, Spectro Italia, MI-I), then extracted in a water solution and analysed for their anionic ($Cl^-$, $NO_3^-$, $SO_4^{=}$) and cationic ($Na^+$, $NH4^+$, $K^+$, $Mg^{++}$ $Ca^{++}$) content by ion chromatography (IC, DX100, Dionex Corporation, CA-U.S.A.).

## 3. RESULTS AND DISCUSSION

### Anthropogenic events

The time pattern of natural radioactivity measured at the urban background station of Rome during the campaigns of summer and winter 2003 is reported in Figure 1. The difference between the two graphs reflects the difference in the mixing properties of the lower atmosphere between warm and cold months: during the summer atmospheric stability during the night (high values of natural radioactivity) regularly alternates to convective mixing during the day (low values of natural radioactivity). During the winter, instead, advection periods, with constantly low values of natural radioactivity (December 7-9, 16 and 22-25) alternate to stability periods, with low mixing of the atmosphere during the night but also during the day (December 6, 14, 17-21).

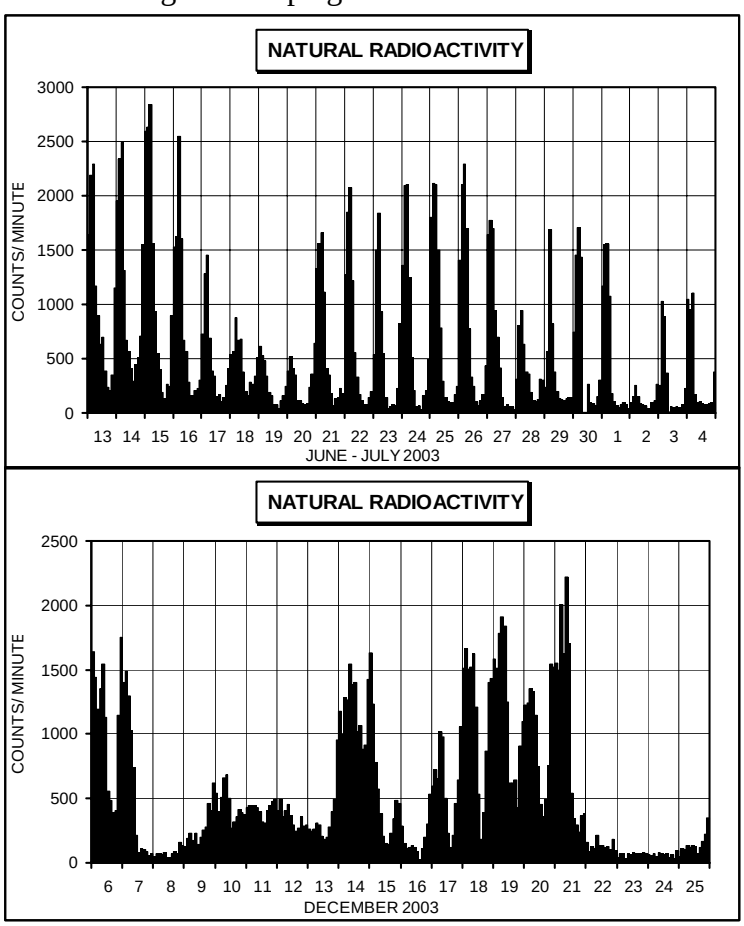


Figure 1: Time pattern of natural radioactivity during the summer (upper) and the winter (lower) campaign in Rome.

These differences in the

dilution properties of the lower boundary layer result in a different modulation of the particle concentration: although the average concentration of $PM_{10}$ during the two periods were quite similar (33.9 ug/$m^3$ during the summer campaign and 38.3 ug/$m^3$ during the winter campaign at the urban background station, 28.5 and 30.8 ug/$m^3$, respectively, at the rural station), the relative standard deviations are very different: around 20% for the summer period and more than 50% for the winter period. Considering the whole year 2003, about 70% of the exceedances of the limit value of 50 ug/$m^3$ at the urban background station were recorded during the winter period.
The time patterns of $PM_{10}$ and $PM_{2.5}$ at the VA and ML stations during the two field studies, reported in the left side of Figure 2, show that the concentration at the two sites were very similar. This finding indicates that the particulate concentration measured at the urban background station can be mostly attributed to regional-scale pollution. At the traffic station (right-hand side of Figure 2), during the summer campaign the level of $PM_{10}$ was very close to the VA and ML levels, while during the winter campaign we can distinctly observe higher values at the traffic station during the atmospheric stability periods, mainly during the multi-day period of December 17-21.

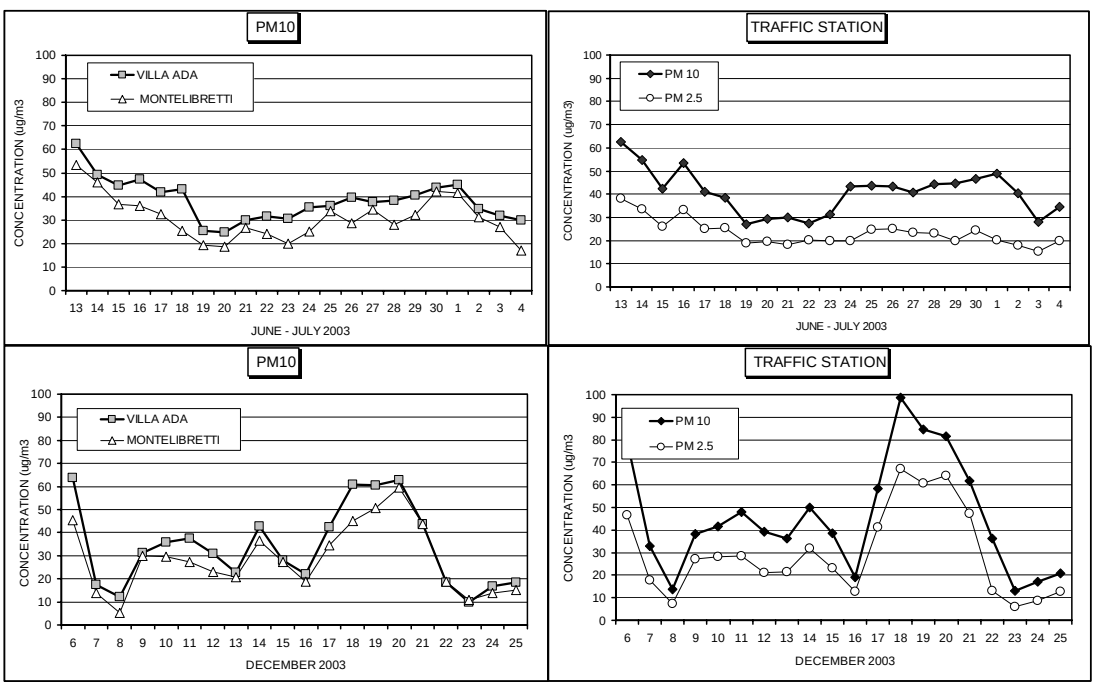


Figure 2 Left side: time pattern of $PM_{10}$ concentration at the urban background station of Villa Ada (VA) and at the rural station of Montelibretti (ML).
Right side: time pattern of $PM_{10}$ and $PM_{2.5}$ at a traffic station in the urban area of Rome.
Upper graphs refer to the summer campaign, lower graphs to the winter campaign.

The analysis of the chemical composition of atmospheric particles highlights that the increase of carbon compounds concentration is one of the main reasons for the increase of particle concentration during atmospheric stability periods. The graphs of Figure 3 show the comparison between the total concentration of carbon compounds and $PM_{10}$ (left graph) and the time pattern of elemental carbon and organic carbon (right graph) at the urban background station during the winter campaign. In these

conditions, the main increment in carbon concentration is clearly due to organic compounds.

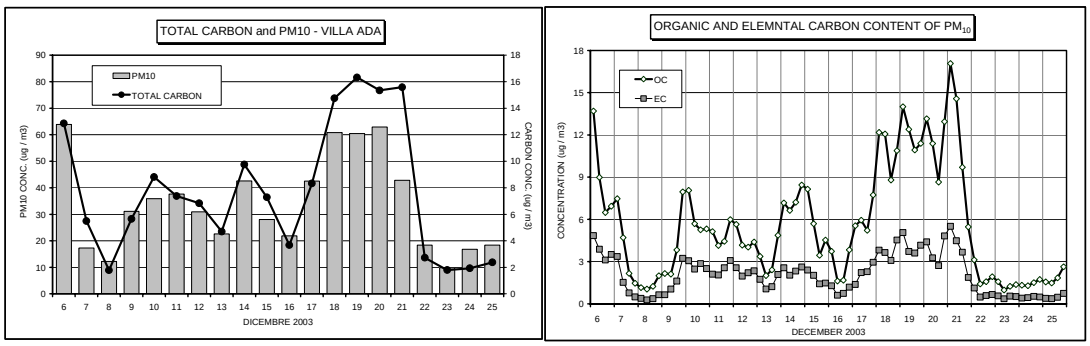


Figure 3: Comparison between $PM_{10}$ and total carbon compounds (left graph) and between organic carbon and elemental carbon (right graph) at the urban background station during the winter campaign.

By comparing elemental carbon and organic carbon concentration at the urban background station with the values obtained at the traffic station (Figure 4) we can observe that elemental carbon concentration at the traffic station is much higher that at the urban background station, showing the influence of the vicinity of the emission sources (mainly Diesel vehicles). Organic carbon concentration, instead, is more dependent on the aging of the air masses (relevance of the secondary organic compounds) and the values at the two stations are quite similar.

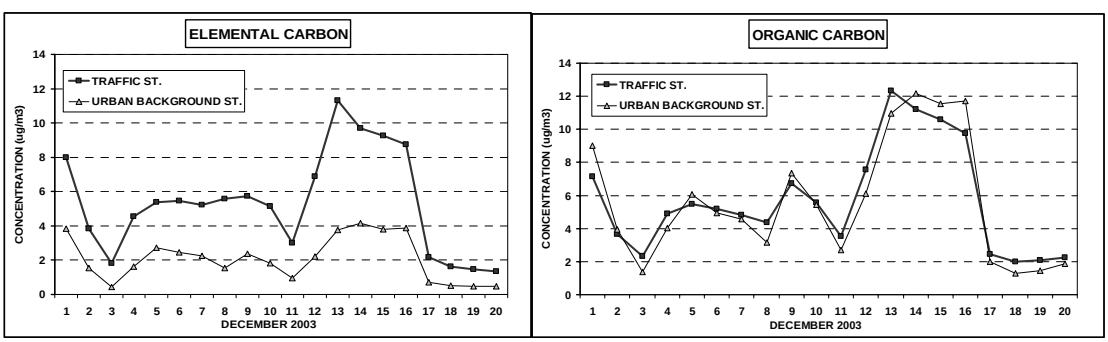


Figure 4: Comparison between elemental carbon (left graph) and organic carbon (right graph) at the traffic station and the urban background station during the winter campaign.

The other compound that can be considered as one of the main responsible for the increase of particulate matter concentration during atmospheric stability periods is ammonium nitrate. The increase of ammonium nitrate concentration at the background station and especially at the traffic station during the period 17-21 is evident from the data of Figure 5.

It should be stressed that when the collection of particles is carried out on a single filter membrane, as it is in most air quality network, the concentration of ammonium nitrate is generally underestimated, because of the negative bias due to the lost of the volatile fraction of ammonium salts during the sampling phase, the measurement phase and also during the period between the sampling and the chemical analysis (Rees et al., 2004).

An accurate determination of ammonium nitrate can be obtained only by using a filter pack composed of a Teflon filter followed by a Nylon (or an alkaline – impregnated) filter and an acid - impregnated filter for the recovery of nitric acid and ammonia evolved from ammonium nitrate salts. Two denuders must be placed upstream of the filter pack in order to remove, respectively, atmospheric nitric acid and ammonia.

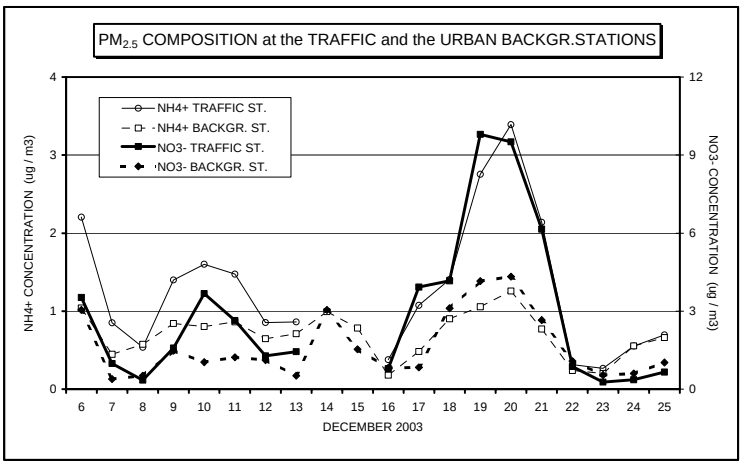


Figure 5: Ammonium and nitrate time pattern at the traffic and the urban background station during the winter campaign.

A field experiment was carried out during April 2005, aimed to investigate the influence of the internal temperature of the dust monitors on the recovery of ammonium nitrate. $PM_{2.5}$ was sampled at the Montelibretti station by means of the diffusion denuder / filter pack lines and by means of two co-located beta attenuation dust monitors. In the first instrument (SM200) the filter is heated at the temperature of 45°C during and after the measurement, until it is removed from the monitor. In the second instrument (SWAM5A), instead, the filter is kept at the local ambient temperature of 20°C. The results of the experiments, reported in Figure 6, show that in the case of a stable compound such as sulphate, the two monitors yield the same results, which are very close to the results yielded by the diffusion lines. In the case of nitrate, instead, less than 15% of the collected nitrate is, on average, lost by the 20°C monitor, and about 80% is, on average, lost by the

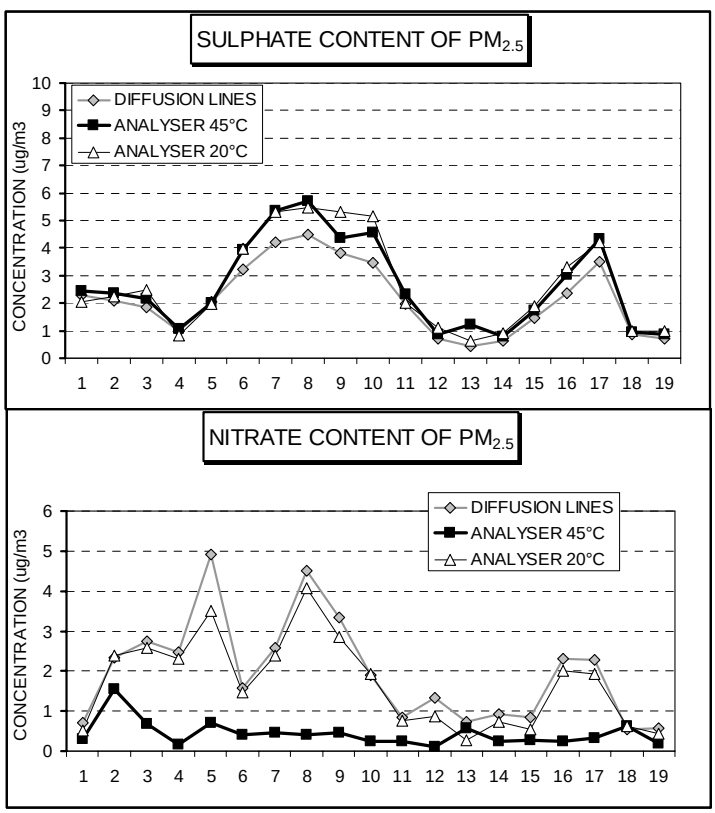


Figure 6: Comparison of sulphate and nitrate ion determined by denuder / filter pack diffusion lines and by heated and non-heated automatic monitors.

45°C monitor. These results indicate that the contribution of ammonium nitrate to PM concentration can be underestimated when a single filter is used, and that this underestimation may become severe in those automatic monitors where the filter membrane is heated.

**Natural events**

While anthropogenic pollution events are linked to a weak mixing of the lower atmospheric layers, natural events generally occur during advection periods, when the air masses may transport particles of natural origin from areas that can be also very far from the sites where the pollution events is recorded. In the area of Central Italy, two main types of natural events are frequently recorded: sea-spray transport from the Tyrrenian coasts and long-range transport of sand dust from North-African deserts. The occurrence of a natural event can be detected by the observation of low values of the natural radioactivity, which indicate advection, associated to an increase of the ratio between coarse and fine particles.

To detect the origin of a natural event and, in general, to obtain a reliable identification of the main sources of all particulate pollution events, it is important to carry out the analysis of the main components of atmospheric particles, that is ions, metals and EC/OC compounds.

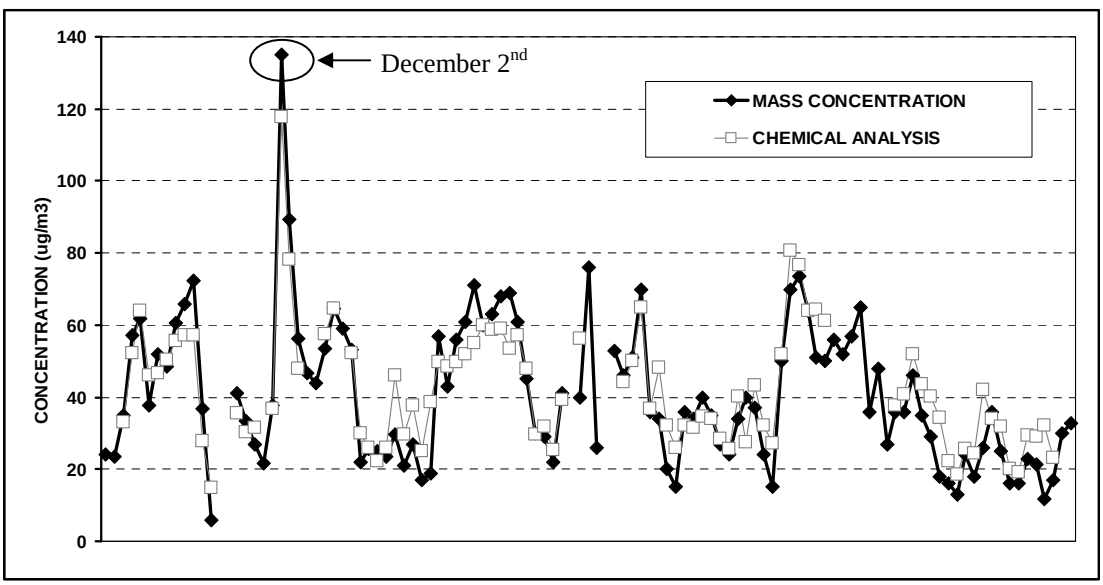


Figure 7: Comparison of the mass concentration of $PM_{10}$ in Latina as measured by the Air Quality Network and as reconstructed by the chemical analysis.

An example of the comparison between the mass concentration of $PM_{10}$ as measured by the local Air Quality Network and the sum of the chemical determinations carried out on the collected particles is reported in Figure 7. The data refer to the urban area of Latina, sited on the coast in the south of the Lazio region, and to 110 daily determinations carried out during the period October 2004 – April 2005. We carried out the reconstruction of the mass concentration from the chemical analysis by adding the contribution of anions (chloride, nitrate, sulphate), cations (ammonium, sodium, magnesium), metals (main components: aluminium, silicon, iron, calcium,

potassium), elemental carbon and organic carbon (without chemical speciation). Carbonate was calculated from the calcium and magnesium content; metal oxides were estimated as in Chan et al. (1997); the mass of organic material was calculated from the mass of organic carbon by multiplying OC by a factor ranging between 1.6 for urban traffic stations and 2.1 for rural station (Turpi and Lim, 2001; Rees at al., 2004). The very good agreement between the two data sets ($R^2$= 0.861) indicates that the quantitative analyses of the main components of the particulate samples give really reliable results.

The analysis of the crustal matter and of the sea spray aerosol for the six station participating in the "Fine Particles" project showed that both these type of events are common in Central Italy (Figure 8) and that during these events the natural components may reach up to 50% of the $PM_{10}$ mass concentration.

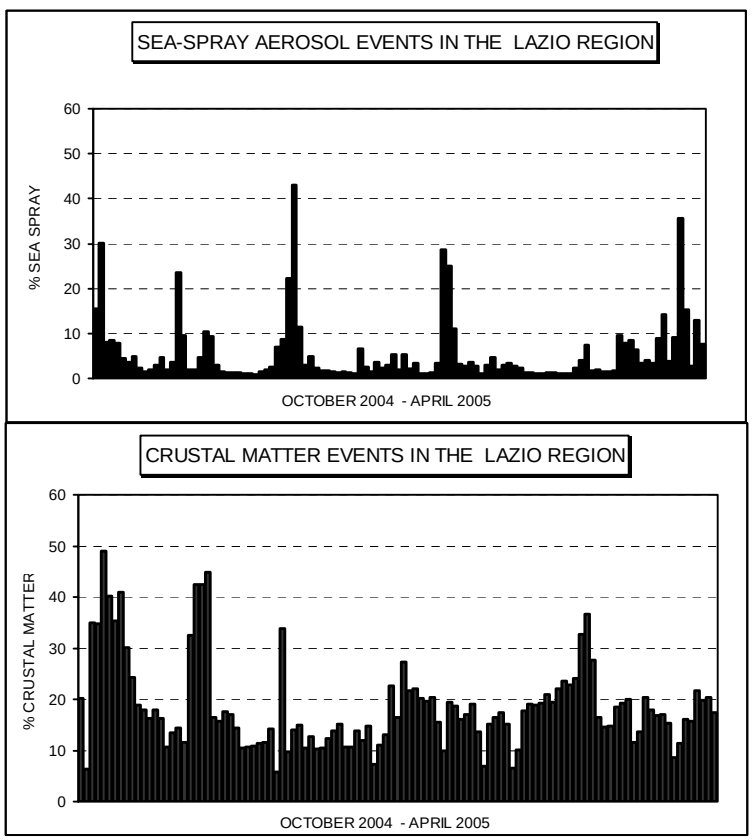


Figure 8: Per cent contribution of sea-spray and of crustal components to $PM_{10}$ concentration in the Lazio region.

However, sea-spray aerosol is generally a minor constituent of $PM_{10}$ (1-3%) and even during the transport of air masses from the sea the increase in $PM_{10}$ mass concentration due to these compounds does not generally exceed 10 ug/m$^3$. It follows that sea-spray events are generally "clean" events, with $PM_{10}$ concentration below the limit values.

Crustal components, instead, generally constitute 10-20% of the $PM_{10}$ concentration, and during sand events the contribution of these species to $PM_{10}$ may even exceed 100 ug/m$^3$. As a consequence, episodes of dust transport from the African desert, although not frequent, may easily cause exceedances of the limit values (in Figure 7, see the episode of December 2$^{nd}$, when the $PM_{10}$ concentration in Latina was close to 140 ug/m$^3$).

## 4. CONCLUSIONS

The joined use of the natural radioactivity monitoring for the evaluation of the mixing properties of the lower boundary layer and of the chemical analysis of the main constituents of atmospheric particulate matter constitute a sound and reliable

tool for the study of particulate pollution events and for the identification of natural episodes and of anthropogenic primary and secondary episodes.

## 5. AKNOWLEDGEMENTS

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# METEOROLOGICALLY ADJUSTED GROUND LEVEL OZONE TRENDS IN SOUTHERN TAIWAN

**Kuang-jung Cheng[1], Che-hui Tsai[1], Hsu-cherng Chiang[1] and Ching-wen Hsu[2]**

[1] Department of Water Resources and Environmental Engineering,
Tamkang University, Tamsui, Taipei-hsien, Taiwan
[2]Department of Environmental Engineering and Science,
Tajen Institute of Technology. Yanpu, Ping-tung, Taiwan

## ABSTRACT

Two methods were used to calculate the meteorologically adjusted ground level ozone trends in southern Taiwan. The first method utilized is a robust MM linear regression method. The second approach uses a multilayer perceptron (MLP) neuron network method. The observations obtained from 16 monitoring stations were analyzed and divided into six groups by hierarchical divisive clustering procedure. The daily maximum 1 h and 8 h ozone concentrations for each region are then calculated. The meteorologically adjusted trends obtained by linear regression method and MLP method are smaller than the unadjusted trends for all regions and average time. It indicts that the meteorological conditions in Taiwan tend to increase ambient ozone concentrations in recent years.

**Key Words :** linear regression, ANN, MLP, statistical analysis, long-term trend

## 1. INTRODUCTION

Photochemical air pollution in Taiwan is a serious environmental issue. In the past two decades, aggressive control strategies have been employed by the government to reduce the emissions of ozone precursor substances ($NO_x$ and VOCs). These efforts substantially reduced the ambient concentrations of $NO_x$ and VOCs, however, ground-level ozone concentrations still exhibit increasing trends in southern Taiwan (Chen et. al., 2004). Possible reasons of this problem include: change of emission patterns, modifications of land use, annual variations in meteorological conditions, and global warming. The objective of this study is to investigate the effects of the annual variations in meteorological conditions on long-term ozone trends in southern Taiwan.

It is well known that variations in meteorological conditions at different time scales can exert sufficiently large impacts on ozone concentrations. This makes measuring the effectiveness of a control program difficult. Great efforts have been made to separate the effects of meteorological conditions from the effects of emission reductions and other factors (Bloomfield et al., 1996; Cobourn and Lin, 2004; Cox and Chu, 1993, 1996; Fiore et al., 1998; Rao et al., 1995; Xu et al., 1996). Many statistical methods have been employed to investigate this problem, but no one method is most appropriate for all purposes and all meteorological scenarios

(Thompson et al., 2001). In this research, we used robust MM least square method and multilayer perceptron (MLP) artificial neural network (ANN) for analysis.
For linear regression model, ozone concentrations are expressed as linear functions of observed meteorological parameters and other factors. This approach is straightforward, however, it assume simple linear and additive associations between the variables are inadequate to capture interactions and nonlinearities in the ozone response. On the contrast, the ANN methods are more complex and flexible than linear statistical models. They are able to model strongly non-linear relationship between meteorological parameters and ozone concentrations. The main reason for selecting the MLP model for air quality prediction was its accuracy and reliability, compared with other available ANN model categories.

In this study, two methods were used to estimate the meteorologically-adjust ozone trends in Taiwan. In addition, the following factors were considered in this study:

(1) Because Daily maximum 1 h average concentration is the current ambient air quality standards adapted by many countries, trends associated with daily maximum 1 h concentrations are the focus of most statistical assessments. However, as noted by USEPA, daily maximum 8 h concentrations is also an important parameter from the aspect of health. Thus, two dataset (1 h and 8 h ozone concentrations) will be considered.

(2) In the past, the so called “single-site models” were used. This approach models the relationship between ozone and meteorological variables measured at the same site (Gardner et al., 2000; Thompson et al., 2001). The formation of ozone in the troposphere is a complex process, involving regional transport of ozone and its precursors. Hence, regional normalization models may be superior to single-site models. Thus, in this study we clustered of sites having similar O3 concentrations into same group by a hierarchical divisive clustering procedure. Six groups were considered.

## 2. METHODS

### 2.1 Linear regression model

A simple linear regression model was used in this study. It can be expressed as:

$$O_3 = \alpha + \beta Y + a\sin(kt) + b\cos(kt) + \sum_{i=1}^{n} c_i M_i \qquad (1)$$

where $\alpha$, $\beta$, $a$, $b$ and $c_i$ are coefficients to be determined by the regression procedure. The input variables include: Y a real number to represent year interval, $k = 2\pi/365$ the wave number, t the time in days starting from beginning of each year, M the meteorological parameters. Various combinations of meteorological variables were tested to determine the most appropriate form of the model. The meteorological parameters actually used in this study are shown in Table 1.

Eq. 1 consists three components: the long-term trend caused by anthropogenic emission change ( $\beta Y$ ), the seasonal term (*asin(kt)+bcos(kt)*), and meteorological effects. The coefficient $\beta$ represents the slope of the long-term trend. As mentioned before, six regions were considered and two $O_3$ concentration targets were tested for each region. Hence, a total of 12 different ozone dataset were considered.

Table 1 input meteorological parameters

| |
|---|
| Ave. of surface wind speed at 11:00 and 14:00 LST |
| Ave. of u-components at 11:00 and 14:00 LST |
| Ave. of v-components at 11:00 and 14:00 LST |
| Ave. of surface temperatures at 11:00 and 14:00 LST |
| Ave. of surface relative humidity at 11:00 and 14:00 LST |
| Ave. of cloudiness at 11:00 and 14:00 LST |
| Change of 850mb height between 02:00 and 08:00LST |
| Change of temperature at 850mb between 02:00 and 08:00LST |

Two regression models were run for each data set. The first model discards the meteorological parameters and obtains trends that are unadjusted. The second model considers all terms in the above equation and adjusted trend can be obtained. By comparing these two results, we can reveal the meteorological impacts on ozone trends.

The model was fitted using the robust MM regression technique in S-Plus (Insightful Co., 2001). The robust regression fit is minimally influenced by outliers in the dependent variables as well as dependent variable. This method is adopted because it has smaller RMSE than traditional linear least square method.

**2.2. MLP methodology**

For the MLP methodology, it is assumed that a time series of ozone $O(t)$ can be expressed as the sum of a long-term trend $T(t)$, a seasonal $S(t)$, meteorological $M(t)$ component, and error, or

$$O(t) = T(t) + S(t) + M(t) + E(t) \quad (2)$$

The seasonal component corresponds to the annual cycle cause by solar radiation, whilst the short-term component is associated with the variations of meteorological variables. The MLP ANN was used to estimate $S(t) + M(t)$ based on daily meteorological and seasonal predictors. The residual R(t) is defined as $O(t)$ minus $S(t)+M(t)$, which is the sum of the long-trend term and random errors, or

$$R(t) = O(t) - [\, S(t) + M(t)] = T(t) + E(t) \quad (3)$$

The long-term trend represents trends in ozone due to precursor emission changes and variations in the background concentrations of some related tropospheric trace gases.

We assume that:

$$R(t) = \alpha + \beta Y + E \quad (4)$$

The values of coefficients $\alpha$ and $\beta$ were determined by a simple least square method.

Three steps are needed to determine the meteorologically-adjust ozone trend in this approach: (1) Using MLP and input meteorological data to estimate $S(t) + M(t)$, (2) compute the residual $R(t)$ by Eq. 3, (3) estimate adjusted-trend $\beta$ by Eq. 4.

The feed-forward back-propagation MLP was used in this study; this model category will be abbreviated simply as MLP in the following. The MLP model was trained by using the trainbr algorithm in the MATLAB Neural network toolbox (Demuth and Beale, 2004). Training involves finding the set of MLP network weights, which enable the MLP model to represent the underlying patterns in the training data. As suggested by Gardner and Dorling (2001), MLP models with two hidden layers were used. There is no a standard way to decide the number of hidden neurons. A trial-and-error approach was used in this study. By changing the neurons numbers in first and second layers, the RMSE surface can be computed. Since the RMSE do not change significantly if the numbers of neurons larger than 6. Hence, six neurons in hidden layer 1 and four neurons in hidden layer 2 were adopted (see Fig. 1). The transfer function in the two hidden layer nodes was the log-sigmoid and tangent sigmoid functions, respectively, while for the output layer nodes the unbounded linear function was used.

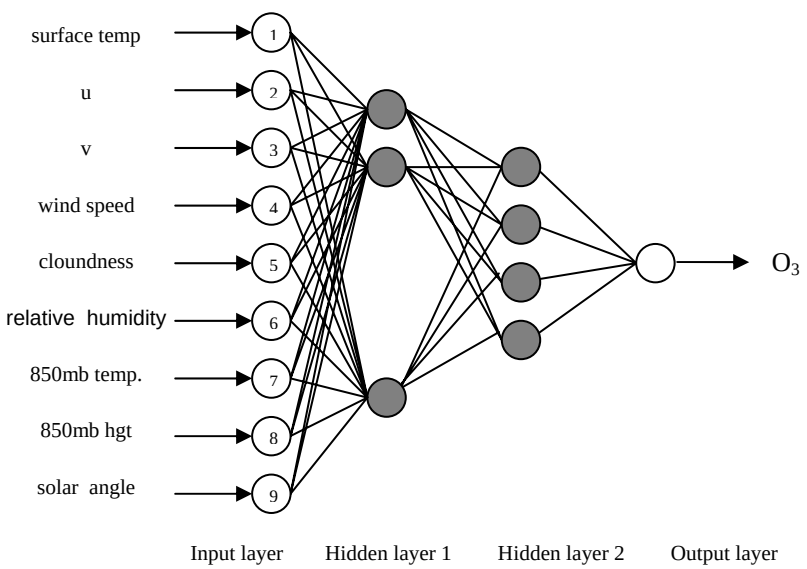


Fig 1 The architecture of a multilayer neural network

The available data were randomly divided into two subsets, training data and validation data. The training data is about half size of the dataset considered, the remaining data are used for validation. The MLP models were then trained to learn the relationship between the predictors and daily maximum ozone concentrations

using training data. Validation data set is to check the performance of the MLP network to determine the epoch at which training should be stopped to avoid over-training. Typically the global minimum is not reached and a good local minimum is treated as an acceptable solution. We train MLP models 50 times and selecting the model with the best generalization performance in order to reduce the likelihood of local minima causing problems.

Following training, the model residuals were calculated and interpreted as the meteorologically adjusted long-term trends and random errors. The trends were estimated from eq. 4 by a general least square method.

## 3. DATA

The original air quality data utilized in this study are obtained from EPA, Taiwan. The data consist of hourly averaged concentrations of ozone and other relevant pollutants collected from 16 stations in southern Taiwan over the four years from 2000 to 2003. Meteorological data were taken from weather stations close to air quality monitoring sites. These weather stations were operated by Central Weather Bureau. The locations of air quality monitoring stations are shown in Figure 2.

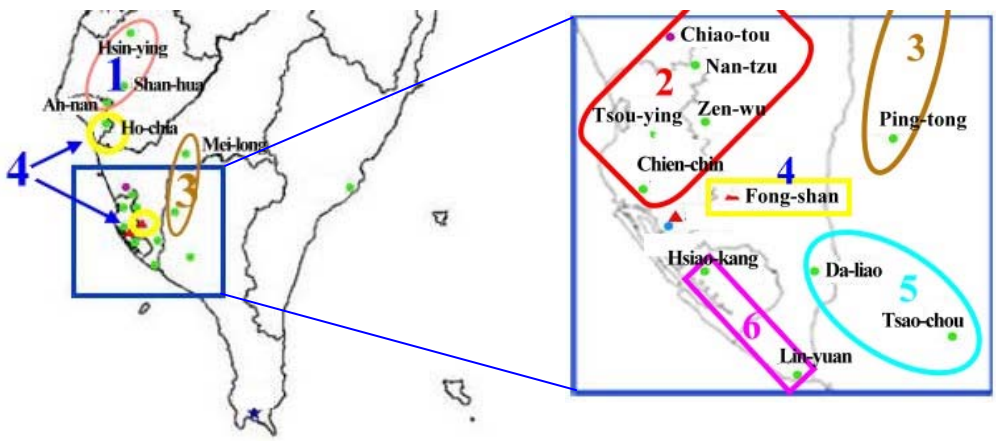


Fig 2 Locations of 16 air quality monitoring stations in southern Taiwan

The ozone response variable and meteorological predictor variables are calculated on a daily basis. If more than 6-h of data are missing on 1 day for any weather variable or for ozone, then the entire day is omitted from the analysis. The missing values typically reduce the available data by 5-10%. Most of the missing data correspond to extended down times caused by maintenance or instrument malfunction. There is no evidence that down times are related to ozone levels, so the missing values are omitted.

The hierarchical divisive clustering procedure was used to aggregate sites into several groups based on site-specific $O_3$ concentration data. Hierarchical divisive methods start with all observations in a single group and proceed until each observation is in a separate group. As shown in Fig. 3, air quality stations were

classified into six groups by cluster analysis. Since monitoring stations in each group are similar, averaged concentrations over each region were used for analysis.

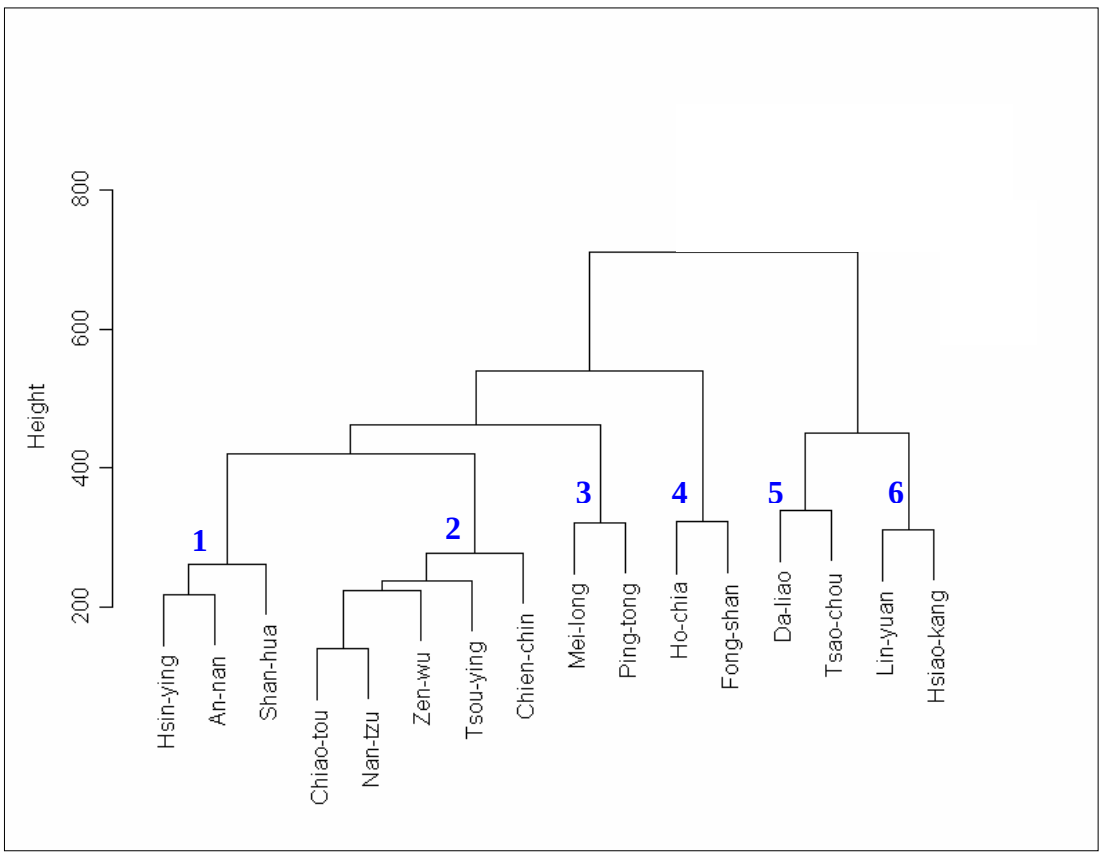


Fig 3 Air quality stations were classified into six groups by ierarchical divisive method

## 4. RESULTS AND DISCUSSION

(1) Trends of daily maximum 1 h ozone concentrations

Table 1 shows the meteorologically adjusted and unadjusted long-term components of the daily peak 1 h ozone time series at each region. Results including correlation coefficients, root mean square error (RMSE) and trend are listed in Table 1.

As shown in Table 1, $\beta$ values for trend only analysis are all positive (except for region 2), which indicates that the unadjusted ozone trends are increasing. Having taken the meteorological effects into account, the meteorologically adjusted trends (or demeteorological trends) are most likely associated with precursor emissions. It is noted that the demeteorological trends are smaller than unadjusted trends. The implication is that if one seeks ozone trend without considering the meteorological impact, one would end up with a trend that could misrepresent the effects of the emission reduction.

Table 1 also shows that the addition of meteorological variables to a model with only seasonality and trend will reduce the prediction error (RMSE) and the standard error of the trend estimate. The values of correlation coefficients will increase if the meteorological effects are considered. The $R^2$ values ranged from 0.4 to 0.6. It is vary reasonable when compared with the results of other studies.

In terms of RMSE or $R^2$, as shown in Table 1, the linear regression model and MLP model exhibit similar levels of performance. The absolute values of the demeteorological trends obtained by MLP methods are smaller then that obtained by liner regression method.

Table 1 meteorologically adjusted and unadjusted long-term statistics of the daily peak 1 h ozone time series at each regions

| Region | Trend only Linear regression | | | Demeteorological trend Linear regression | | | Demeteorological trend MLP | | |
|---|---|---|---|---|---|---|---|---|---|
| | $R^2$ | RMSE | $\beta$ | $R^2$ | RMSE | $\beta$ | $R^2$ | RMSE | $\beta$ |
| 1 | 0.20 | 22.61 | 1.14±1.09 | 0.57 | 16.62 | 0.73±0.66 | 0.55 | 16.29 | 0.4951 |
| 2 | 0.39 | 24.48 | -0.14±0.70 | 0.41 | 20.75 | -1.86±0.58 | 0.53 | 19.97 | -1.4675 |
| 3 | 0.16 | 25.63 | 5.95±0.66 | 0.44 | 18.30 | 4.38±0.52 | 0.59 | 18.03 | 3.5523 |
| 4 | 0.36 | 24.54 | 4.46±1.02 | 0.59 | 19.57 | 5.72±0.68 | 0.53 | 18.73 | 2.999 |
| 5 | 0.31 | 28.76 | 0.08±1.09 | 0.57 | 22.81 | -0.77±1.02 | 0.58 | 21.14 | -0.3475 |
| 6 | 0.43 | 26.27 | 1.24±0.89 | 0.43 | 22.08 | -0.53±0.64 | 0.56 | 20.82 | 0.0251 |

We can use two models developed in this study to 'predict' the daily peak ozone concentration for six regions. The results are shown in Fig. 4. This figure indicates that the differences between observed and predicted values are significant. It implies that some important factors, which have significant influence on the ozone concentration, were not well explained by the current models. These factors may include the short-term variations of emission conditions.

(2) Trends of daily maximum 8 h ozone concentrations

Since health effects research now shows that ozone affects public health over long periods of time, not just during a few 1 h peak events. A new standard based on daily maximum 8 h ozone concentration was promulgated in USA. Now, we will exam the results of daily maximum 8 h ozone concentration.

Table 2 is similar to Table1 except that daily maximum 8 h ozone concentrations were used. Since the values of maximum 8 h ozone concentrations are less than the values of daily peak 1-h ozone concentration, the values of RMSE and $\beta$ appear in Table 2 are less than their corresponding values in Table 1. The correlation coefficients in Table 2 are larger. The $\beta$ values would be reduced if meteorological conditions were considered.

Fig.5 shows the time series plots of the observed and predicted daily peak 8 h ozone concentrations of 2002 at six regions. The predictions of two models agree with the observations.

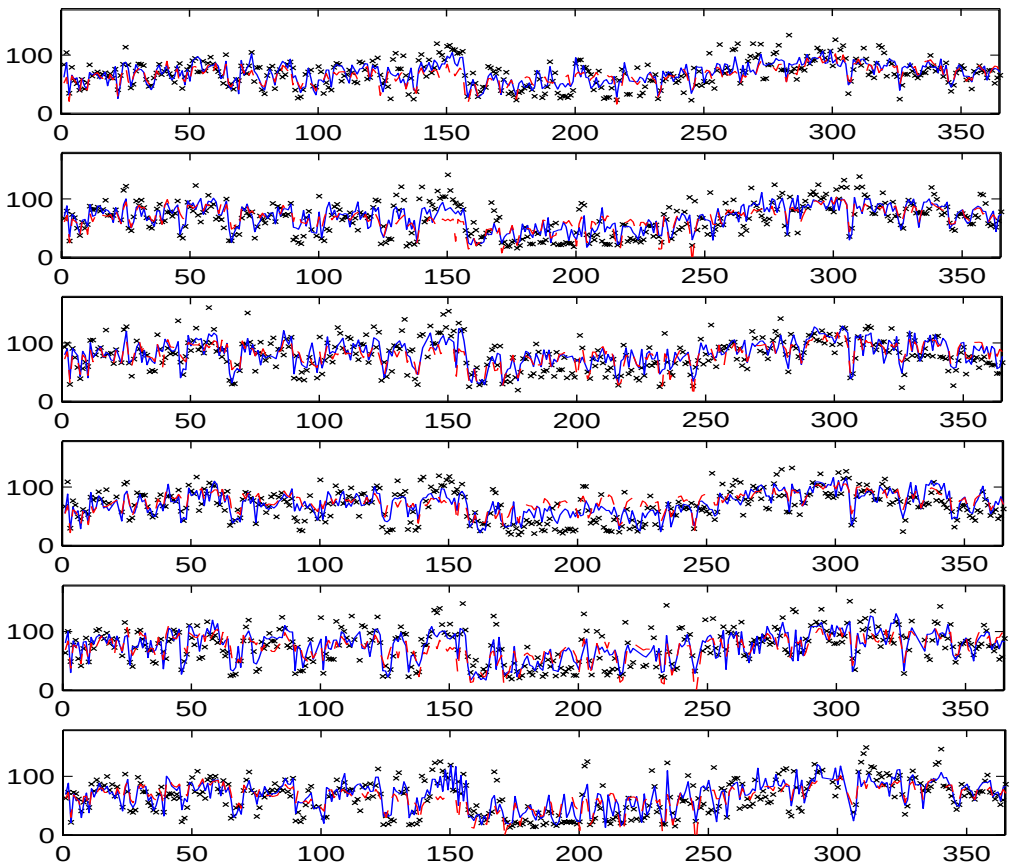


Fig. 4 The observed and predicted daily peak 1 h ozone concentrations of 2003 at six regions (blue line: least square method, red line: MLP method, x : observations)

Table2 meteorologically adjusted and unadjusted long-term statistics of the daily maximum 8 h ozone time series at each region

| Region | Trend only Linear regression | | | Demeteorological trend Linear regression | | | Demeteorological trend RBFNN | | |
|---|---|---|---|---|---|---|---|---|---|
| | $R^2$ | RMSE | $\beta$ | $R^2$ | RMSE | $\beta$ | $R^2$ | RMSE | $\beta$ |
| 1 | 0.28 | 18.00 | 1.20±0.62 | 0.61 | 13.25 | 0.64±0.59 | 0.61 | 12.35 | 0.4477 |
| 2 | 0.46 | 18.97 | 0.43±0.63 | 0.63 | 15.79 | -0.92±0.65 | 0.59 | 14.64 | -0.5549 |
| 3 | 0.34 | 18.59 | 4.30±0.76 | 0.64 | 13.68 | 3.66±0.60 | 0.63 | 12.97 | 3.1609 |
| 4 | 0.40 | 19.22 | 3.91±0.74 | 0.43 | 15.68 | 3.56±0.41 | 0.58 | 14.15 | 2.6529 |
| 5 | 0.44 | 19.50 | 0.11±0.71 | 0.64 | 15.69 | -1.79±0.57 | 0.62 | 14.55 | -0.5677 |
| 6 | 0.50 | 19.52 | 1.96±0.75 | 0.63 | 16.84 | 0.16±0.72 | 0.61 | 14.78 | 0.3757 |

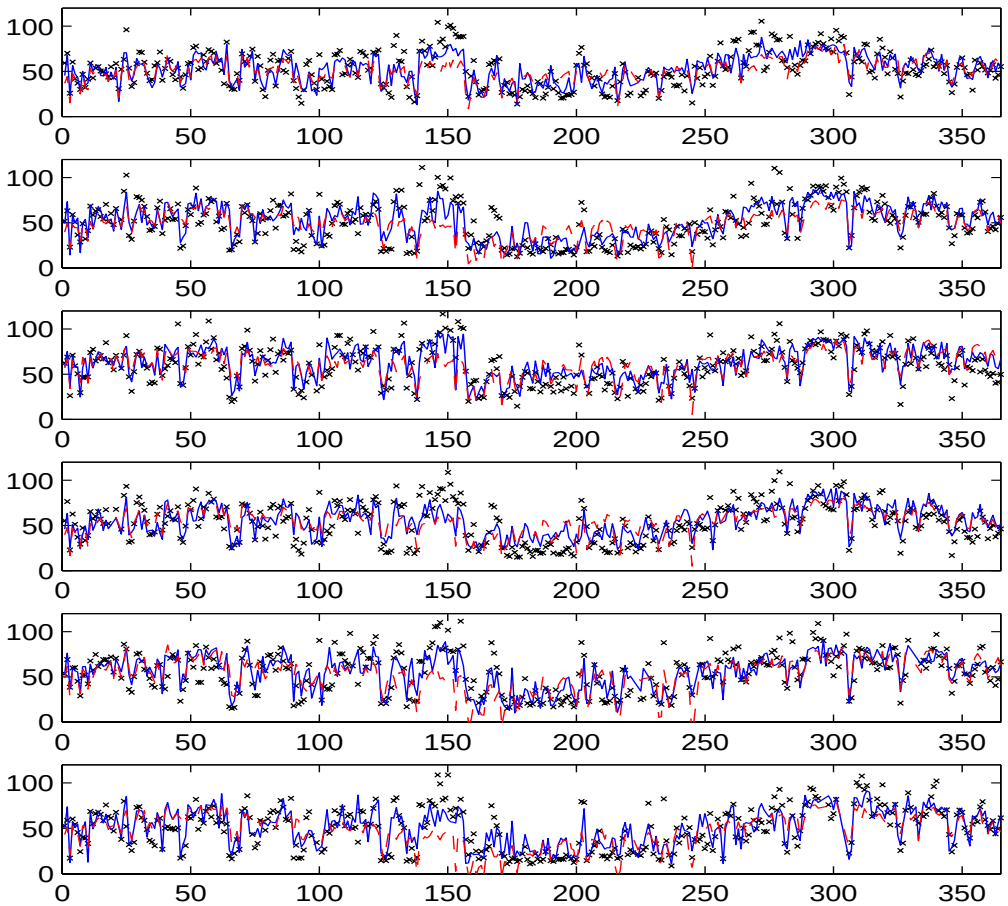


Fig. 5 The observed and predicted daily peak 8 h ozone concentrations of 2003 at six regions (blue line: least square method, red line: MLP method, + : observations)

As shown in Table 1 and 2, the meteorologically adjusted trends in region 2 and 5 are decreased. This may attribute to the effects of emission control. However, in other regions the meteorologically adjusted trends still exhibit increasing tendency. Since a new freeway was completed and operated in 2000, we believe the patterns of pollutant emission will be changed due to urban sprawl. It is well known that concentration distributions of air pollutant change while emission patterns change.

## 5. CONCLUSION

A robust linear regression method and a MLP neural network method were used to separate the effect of meteorological conditions on the ozone concentrations. After eliminating the meteorological factor in 2000-2003, the long-term trends ( $\beta$ ) obtained by linear regression and MLP method are decreased. In recent years, the meteorological conditions in Taiwan tend to increase ambient ozone concentrations.

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# SEVERE PARTICULATE POLLUTION IN LANZHOU CHINA

**Peter C. Chu, Yuchun Chen*, Shihua Lu*, Zhenchao Li* and Yaqiong Lu****

Naval Ocean-Atmospheric Prediction Laboratory, Department of Oceanography, Naval Postgraduate School, Monterey, California, USA

pcchu@nps.edu, http//www.oc.nps.navy.mil/~chu

*Cold and Arid Regions Environmental and Engineering Research Institute, Chinese Academy of Sciences, Lanzhou, China

**Department of Atmospheric Science, Chengdu University Of Information Technology, Chengdu, China

## ABSTRACT

Spatial and temporal variability of $SO_2$, $NO_2$, $NO_x$, $PM_{10}$ and total suspended particles (TSP) concentrations in Lanzhou, China is analyzed using the observational data collected from October 1999 to April 2001. The concentrations of $SO_2$, $NO_2$, $NO_x$ are within the national second-standard level of air quality in spring, summer and fall, but much worse in winter, reaching low to mid alert level of air pollution. Since 1999, the concentrations of $SO_2$, $NO_2$, and NOx have been decreasing. However, the concentrations of $PM_{10}$ and TSP have been increasing, and become major pollutants.

The mean concentration of $PM_{10}$ is 2.56 mg $m^{-3}$. Even in summer the air pollution due to $PM_{10}$ is at low alert level. The rate of polluted-day occurrence is 71% in a year, 89% in winter, and 79% in spring. Starting from November, the air pollution due to $PM_{10}$ intensifies, and reaches mid to high alert level of air pollution, and continues until April next year. The mean concentration of TSP is 5.92 mg $m^{-3}$, which is higher than the air quality standard. In winter and spring, the TSP concentration is 2-10 times higher than the third-standard level of air quality. Intrinsic factors and exterior preconditions for increase of $PM_{10}$ and TSP are discussed such as the propagation of dust storms.

**Key words:** Lanzhou, pollutants, total suspended pollutants, observational data analysis, dust-storm,

## 1. INTRODUCTION

Lanzhou is located at a narrow (2-8 km width), long (40-km), NW-SE oriented valley basin (elevation: 1,500-m to 1,600-m) with the Tibetan plateau in the west, Baita mountain (above 1,700-m elevation) in the north, and the Gaolan mountain in the south (Fig. 1a). The topographic characteristics make Lanzhou vulnerable to the invasion of dust storms (Fig. 1b). The aspect ratio of the valley (depth versus width) is around 0.07, which blocks the air streams due to the large frictional forces and causes weak winds and stable stratification (even inversion) to inhibit turbulent

diffusion. The meteorological conditions (low winds, stable stratification especially inversion) cause the pollutants difficult to disperse. These conditions make Lanzhou one of the most polluted cities in China (Fig. 1c).

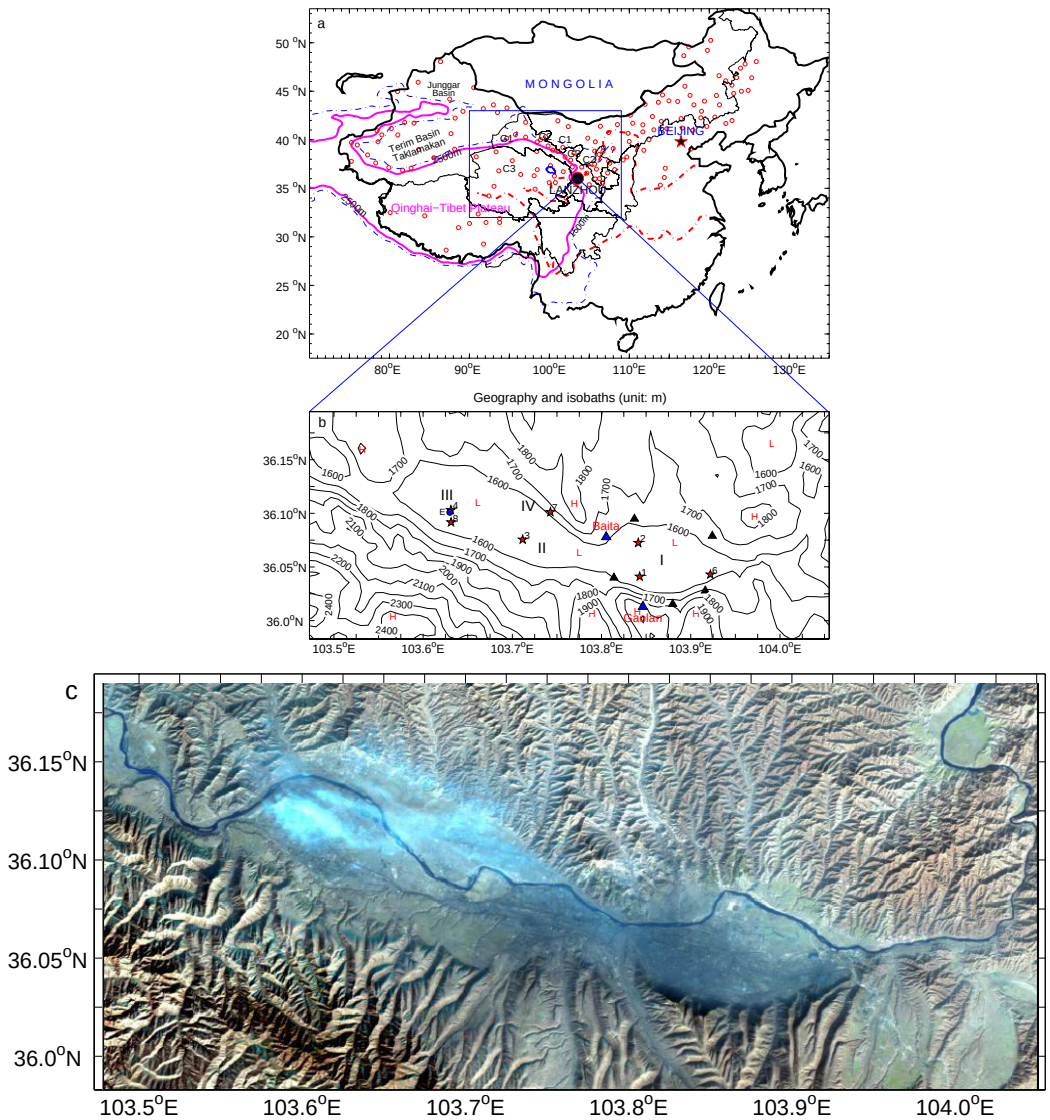


Figure 1: Lanzhou: (a) geography, (b) topography, and (c) LANDSAT-TM imagery representing air pollution on 3 January 2001.

Since mid 1990s, the local Lanzhou government has conducted afforestation on the mountain slope and shut down several factories that emitted large amount of pollutants. The gaseous pollutants such as $SO_2$ and $NO_x$ concentrations have been reduced. However, the particulate pollutants such as TSP and $PM_{10}$ still keep high concentrations [Chu et al. 2004]. Fig. 2 shows the evolution of annual mean concentration for the three major pollutants ($SO_2$, $NO_x$, TSP) measured at the local environmental protection agency (EPA) station (103.631°E, 36.103°N), which is

marked as the solid circle in Fig. 1b. The annual mean $SO_2$ has a maximum near 0.12 mg $m^{-3}$ (above the third level standard: 0.10 mg $m^{-3}$) in 1994, and decreases monotonically to 0.055 mg $m^{-3}$ (below the second level standard: 0.06 mg $m^{-3}$) in 2000 (Fig. 2a). The annual mean $NO_x$ has two maxima (above the third level standard: 0.10 mg $m^{-3}$) in 1990 and 1995, and decreases monotonically to 0.05 mg $m^{-3}$ (close to the second level standard: 0.05 mg $m^{-3}$) in 2000 (Fig. 2b). The annual mean TSP concentration is always above the third level standard: 0.3 mg $m^{-3}$ (Fig. 2c). What is the reason to cause high concentration of the particulate pollutants?

An air-quality monitoring system has been established in Lanzhou with multiple sampling and sufficient numbers of stations. This is the part of the project entitled Air Pollution and Control in Lanzhou (APCL), supported jointly by Gansu Province and the Chinese Academy of Sciences and carried out from 1999 to 2001.

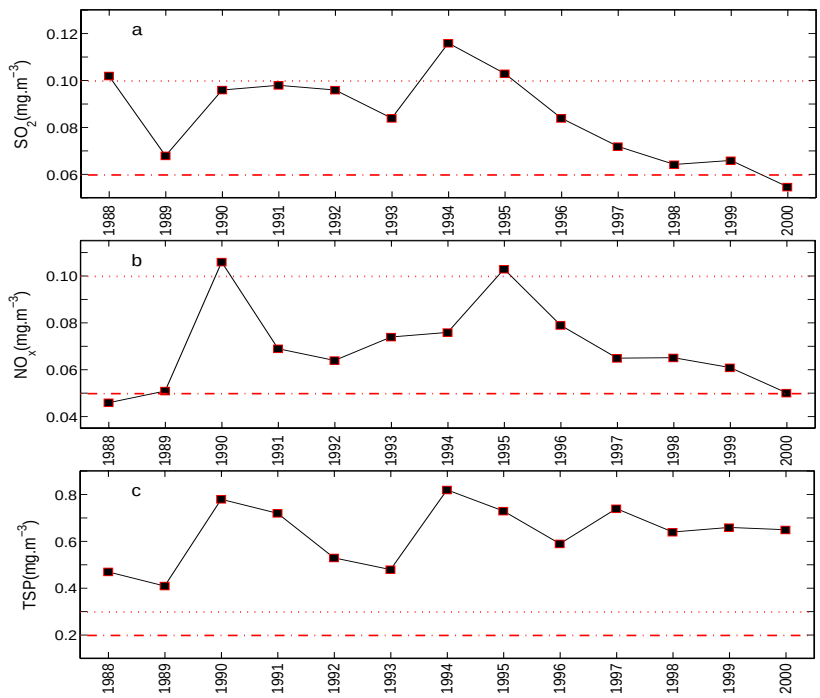


Figure 2. Annual mean concentration (mg $m^{-3}$): (a) $SO_2$, (b) $NO_x$, and (c) TSP measured at the local EPA station (103.631°E, 36.103°N), which is marked as the solid circle in Fig. 1b. The second-level standard is represented by the horizontal dash-dotted line and the third-level standard is represented by the horizontal dotted line.

In this study, detail analyses are conducted on the concentration data collected by the air-quality monitoring system as well as the associated meteorological conditions. The objectives are to detect temporal and spatial variability of various pollutants, to evaluate the air-quality objectively and quantitatively, to analyze the pollutant sources, and to find the favorable meteorological conditions for the pollutant dispersion.

## 2. DATA

The data are from several sources: the APCL project, routine air-quality observations (Table 1), and routine meteorological observations. From the APCL project, the air quality data were collected at observational stations (St-1 - St-5) from October 1999 to April 2001 and at observational stations (St-6 - St-8) from August 2000 to April 2001. Over these stations, daily concentration of $SO_2$, $NO_x$ and TSP is calculated. The data from routine air-quality observations include daily concentrations of $SO_2$, $NO_2$, $PM_{10}$ collected continuously by the local Environmental Protection Agency (EPA) in Lanzhou from June 2000 to May 2001 (station-E in Fig. 1b). Annual mean concentrations of $SO_2$, $NO_x$ and TSP are collected and computed from surface meteorological stations.

Table1. Location of observational stations and mean concentrations (mg $m^{-3}$) of major pollutants measured during the whole observational period. Note that the second-level annual mean standards of air quality are (0.06, 0.05, 0.20) mg $m^{-3}$ for ($SO_2$, $NO_x$, TSP).

| Site | Longitude E | Latitude N | Height Above Surface (m) | Region | $SO_2$ | $NO_x$ | TSP |
|---|---|---|---|---|---|---|---|
| St-1 | 103.84 | 36.04 | 25 | Chengguan (District-1) | 0.08 | 0.06 | 0.69 |
| St-2 | 103.84 | 36.07 | 11 | Chengguan (District-1) | 0.03 | 0.04 | 0.57 |
| St-3 | 103.71 | 36.08 | 15 | Qilihe (District-3) | 0.05 | 0.05 | 0.74 |
| St-4 | 103.63 | 36.10 | 22 | Xigu (District-2) | 0.08 | 0.06 | 0.68 |
| St-5 | 104.09 | 35.84 | 4 | Yuzhong County | 0.01 | 0.01 | 0.28 |
| St-6 | 103.92 | 36.04 | 19 | Chengguan (District-1) | 0.02 | 0.03 | 0.56 |
| St-7 | 103.74 | 36.10 | 15 | Anning (District-4) | 0.04 | 0.05 | 0.52 |
| St-8 | 103.63 | 36.09 | 4 | Xigu (District-2) | 0.06 | 0.05 | 0.54 |

## 3. TSP CONCENTRATION

The daily mean TSP concentration is larger than the second-level standard (0.3 mg $m^{-3}$) almost all the time and than the third-level standard (0.5 mg $m^{-3}$) sometimes (Fig. 3). The monthly mean TSP concentration exceeds the second-level daily mean TSP standard (0.3 mg $m^{-3}$) all the time at all the ACPL stations in the urban area of Lanzhou. Even in the background station (St-5) located in the countryside, the monthly mean TSP concentration often exceeds the second-level daily mean TSP

standard. The monthly maximum TSP concentration in the background station (St-5) always exceeds the second-level daily mean TSP standard and even exceeds the third-level daily mean TSP standard (1.0 mg $m^{-3}$) quite often. At the seven urban ACPL stations, the monthly maximum TSP concentration often exceeds the third-level daily mean TSP standard.

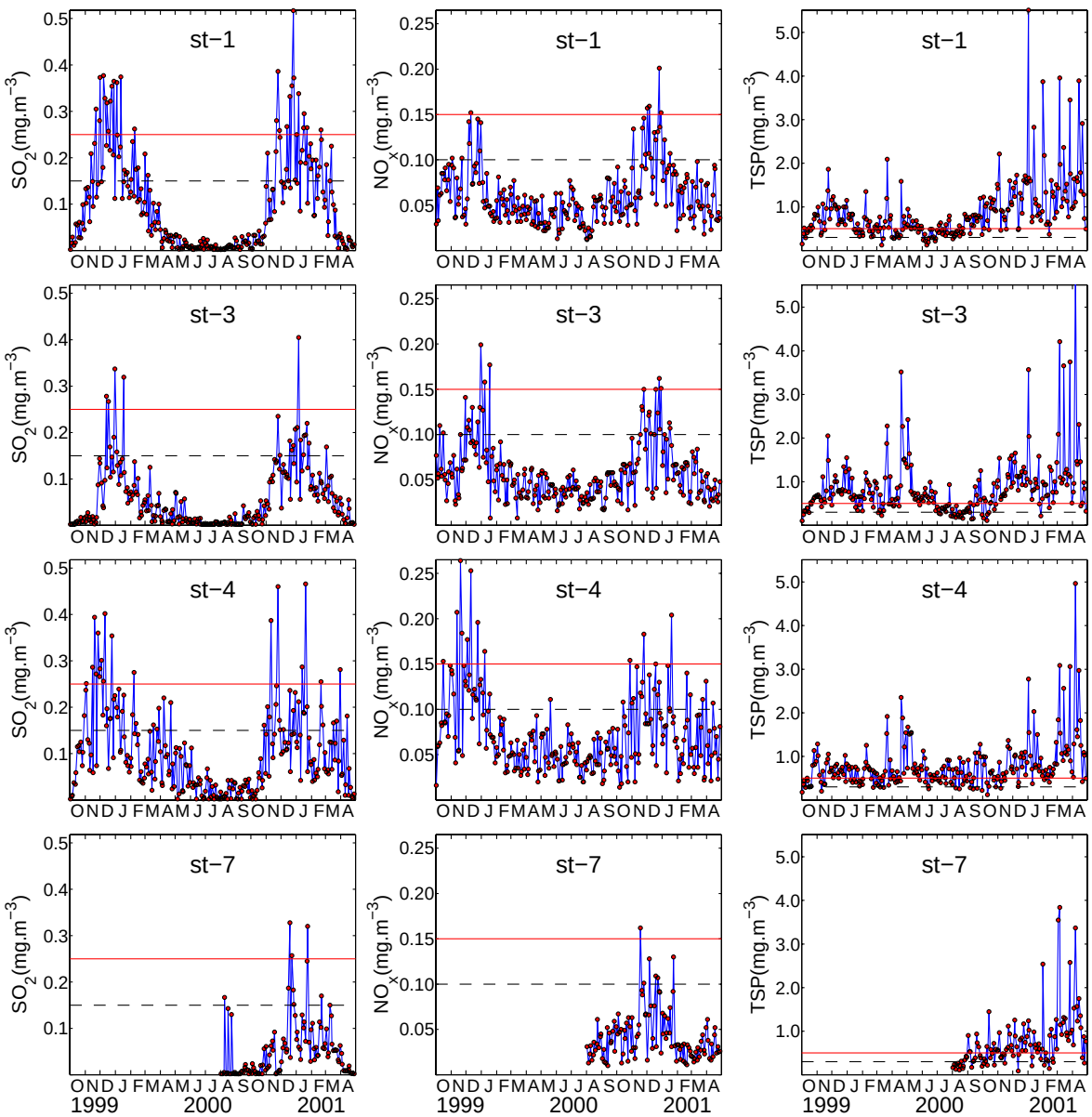


Figure 3. Daily mean $SO_2$, $NO_x$, and TSP concentrations (mg $m^{-3}$) at St-1, St-3, St-4 and St-7. Horizontal dashed line is daily mean second-level standard, while horizontal solid line is daily mean third-level standard. Note that the daily mean TSP concentration is usually above the second-level standard.

The TSP concentration has an increasing tendency (Fig. 4). Among the eight ACPL stations, five stations (St-1 to St-5) have two years' data. To filter out the seasonal effect, the monthly mean TSP concentrations (mg $m^{-3}$) of the same month between

period-1 (October 1999 - April 2000) and period-2 (October 2000 - April 2001) are compared. The monthly mean TSP concentration is more in period-2 than in period-1 all the time. The monthly maximum TSP concentration is more in period-2 than in period-1 almost all the time.

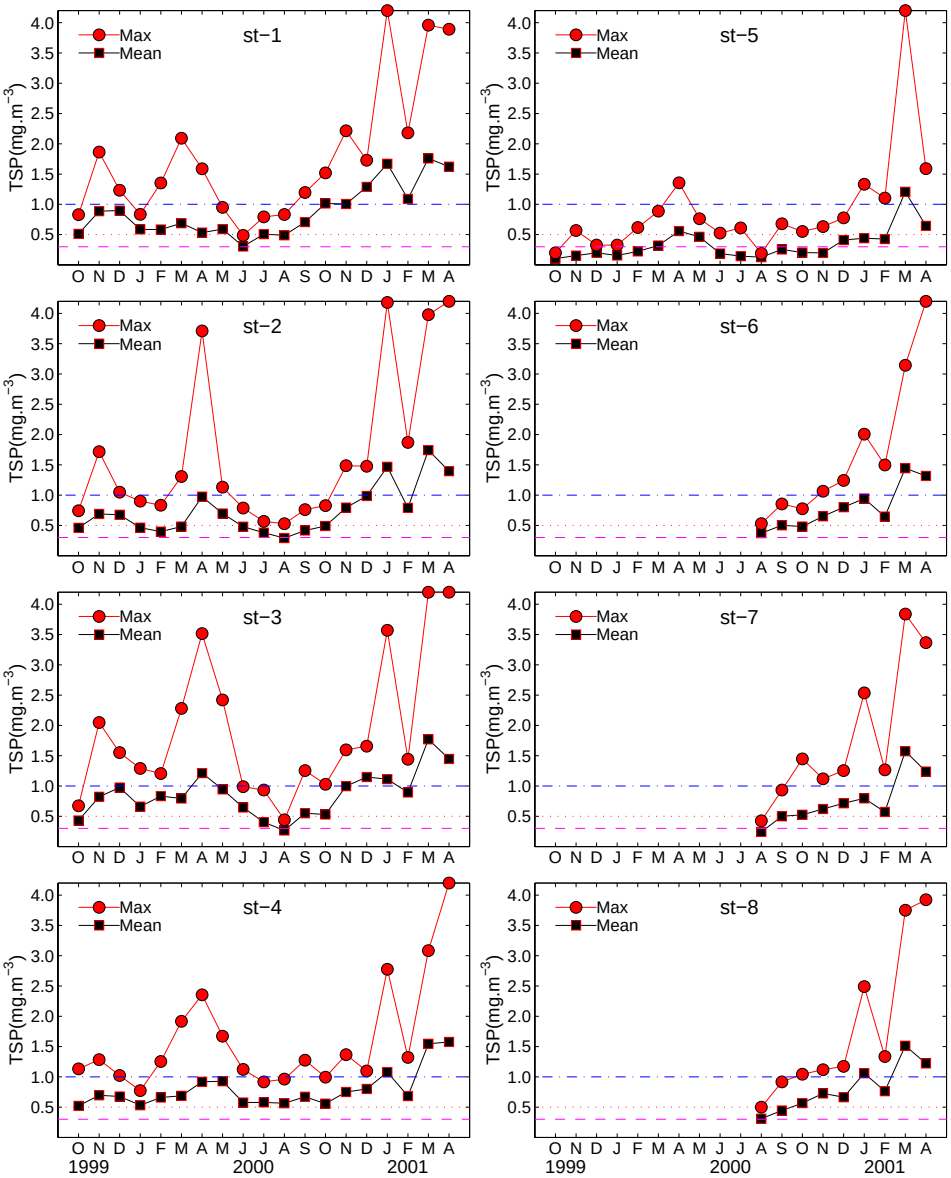


Figure 4. Monthly mean(■) and maximum(●) concentrations(mg m$^{-3}$) of TSP at St-1 to St-8.

Air pollution index (API) is a quantitative measure for uniformly reporting the air quality for different constituents and connects to the human health. CEPA classifies the air quality standards into 5 major categories due to API values (Table 2): I (clean), II (good), III (low-level pollution), IV (mid-level pollution), and V (high-level pollution). The categories III and IV have two sub-categories: ($III_1$, $III_2$) and ($IV_1$, $IV_2$).

Table 2. API and air quality management in China.

| Air Pollution Index | Air Quality Classification | | Air Quality Description and Management |
|---|---|---|---|
| API ≤ 50 | I | Clean | No action is needed. |
| 50 < API ≤ 100 | II | Good | No action is needed. |
| 100 < API ≤150 | $III_1$ | Low-level pollution | Persons should be careful in outdoor activities. |
| 150 < API ≤200 | $III_2$ | | |
| 200 < API≤ 250 | $IV_1$ | Mid-level pollution | Persons with existing heart or respiratory illnesses are advised to reduce physical exertion and outdoor activities. |
| 250 < API≤ 300 | $IV_2$ | | |
| API ≥300 | V | High-level pollution | Air pollution is severe; The general public is advised to reduce physical exertion and outdoor activities. |

Monthly mean API of $SO_2$, $NO_x$, and TSP for the eight observational stations shows the similar fact that the TSP pollution is much more series than the $SO_2$, and $NO_x$ pollutions and the $SO_2$ and $NO_x$ pollution has seasonal variation with larger value in winter and much smaller value in other seasons. At the background station (St-5), API for $SO_2$ and $NO_x$ is less than 50 all the time (Fig. 5).

Monthly mean API for TSP is large even in the background station (St-5) with the value larger than 200 during April-May 2000 and January-April 2001 (Fig. 5). It is always greater than 200 during the whole observational period at St-4 with a maximum value above 600 in March 2001. At the other stations, it is generally greater than 200 (but always larger than 100) during the period except in summer 2000. March 2001 is one of the severely polluted months of TSP. The maximum API for TSP at all eight stations is greater than 500. The monthly mean API of TSP is greater than 500 in all the city stations with 400 in the background station (St-5).

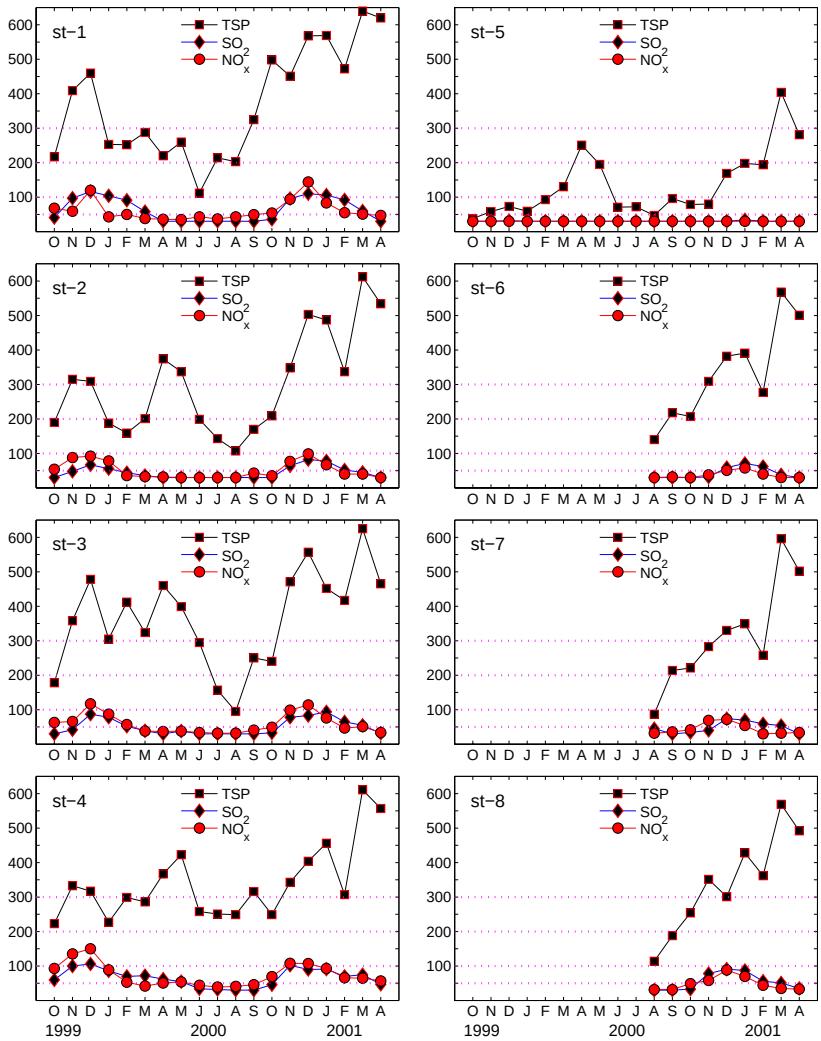


Figure 5. Monthly mean API at St-1 to St-8. Here, the symbol '♦' represents $SO_2$, '●' represents $NO_x$, and '■' represents TSP.

## 4. DUST-STORM AND TSP-$PM_{10}$ POLLUTIONS

As described in Section 5, TSP and $PM_{10}$ are the most series pollutant in Lanzhou. Occurrence of high TSP and $PM_{10}$ concentrations are associated with the propagation of dust storms. For example, Fig. 6 shows January 2001 daily TSP concentrations at the background station (St-5) and spatially averaged over seven (St-1 to St-4 and St-6 to St-8) city stations are illustrated in panel-a, and the daily $PM_{10}$ concentration at St-E is shown in panel-b. On the two panels the dust-storm is marked by the symbol '✍'. Dust-storms occur quite often in the vicinity of Lanzhou: 9 days in January 2001. For example, during the dust-storm from December 31, 2000 to January 1, 2001, dusts float in the sky for 7 days (January 1-5, 2001) [Ding et al., 2001], and causes high TSP and $PM_{10}$ concentrations on January 1, 2001 (TSP: 3.08 mg $m^{-3}$ and $PM_{10}$: 2.56 mg $m^{-3}$) and on January 2, 2001 (TSP: 1.75 mg $m^{-3}$ and $PM_{10}$: 2.01 mg $m^{-3}$).

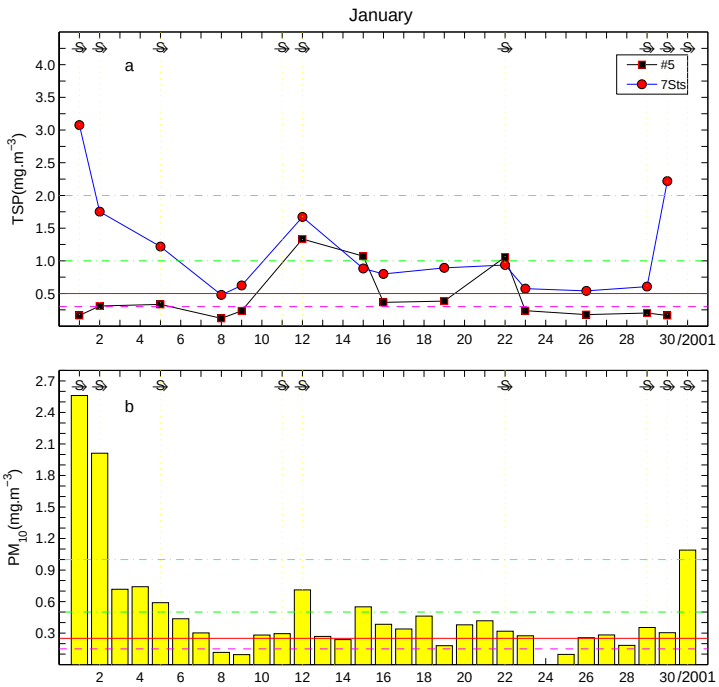


Figure 6. Daily mean (a) TSP, (b) $PM_{10}$ concentrations (dust storm represented by the symbol '🌬'), and (c) horizontal distribution of dust storms (solid dots) in January 2001.

Deserts and barren lands are widespread in the northwest of China and they are expanding attributing to underdeveloped methods of producing and management, and irrational utilization of resources. Aridness and desertization cause frequent dust storms [Zhu, 1999; Dong et al., 1999; Wang and Cheng, 1999]. The dust storms that directly cause severe particulate pollution of Lanzhou are mainly generated in neighboring regions such as Hexi Corridor (Fig.1a marked by G1, G2, G3), Badanjilin desert (100°~103°E, 39°~42°N, centered at C1), south of Tenggeli desert (103°~106°E, 37°~39°N, centered at C2), and Caidam desert (about 92°~98°E, 36°~38°N, centered at C3). These regions are enclosed as a ladder-shaped region (Fig 1a). When the air is dry, strong winds blowing over the deserts cause the dust-storms and in turn brings high TSP and $PM_{10}$ concentrations in Lanzhou.

## 5. CONCLUSIONS

(1) For the past ten years, the gaseous pollutions such as $SO_2$, $NO_2$, $NO_x$ have been improved in Lanzhou. The annual mean $SO_2$, $NO_2$, $NO_x$ concentrations decrease steadily from 1995 to 2000. However, the pollutions of $PM_{10}$, TSP have been worsened, and become the major pollutants in Lanzhou.

(2) TSP pollution is very series in Lanzhou with 2~10 times greater than the third level standard in winter and spring. March 2001 is the most severely polluted month with API□500 all the time. The high-level TSP pollution (API > 300, category V)

occurs frequently with 68% in spring, 16% in summer, 45% in fall, and 63% in winter.
(3) Dust-storms occur quite often in the vicinity of Lanzhou (9 days in January 2001, 3 days in February 2001, 9 days in March 2001, and 12 days in April 2001), and cause high TSP and $PM_{10}$ concentrations in Lanzhou. The maximum TSP concentration was observed as 5.29 kg $m^{-3}$ (March 2001). Reduction of the TSP and $PM_{10}$ concentrations is an urgent issue for air quality control in Lanzhou and related to the reduction of dust storms in the vicinity. This may be achieved by the improvement of the land surface characteristics such as long term forestation and vegetation.

## 6. ACKNOWLEDGEMENTS

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# ATMOSPHERIC $PM_{10}$ AND $PM_{2.5}$ LEAD CONCENTRATIONS AND TEMPORAL VARIATIONS OVER A SUBURBAN SITE OF ISTANBUL (TURKEY)

**Ferhat Karaca[1], Omar Alagha[1], Ferruh Ertürk[2], Yusuf Ziya Yılmaz[3] and Türkan Özkara[3]**

[1] Fatih University, Department of Environmental Engineering, Büyükçekmece, Istanbul, Turkey
[2] Yıldız Technical University, Department of Environmental Engineering, Beşiktaş, Istanbul, Turkey
[3] Nuclear Research and Education Centre, P.K. 1, Atatürk Airport, Istanbul, Turkey

## ABSTRACT

In this study, 92 daily aerosol samples of fine and coarse particles were collected near the Buyukcekmece Lake (41º2'35"N, 28º35'25"E). The samples were collected in random days during July 2002 to July 2003. Thse annual mean $PM_{2.5}$ and $PM_{10}$ lead concentrations were found as 0.054 μg $m^{-3}$ and 0.129 μg $m^{-3}$, respectively. Seasonal behavior of $PM_{2.5}$ and $PM_{10}$ lead concentrations were investigated and their cyclic behaviors were defined, and the major sources of the lead emissions were investigated in study area. By comparing, the Pb levels measured in this study with those reported previously from various locations on Turkey and other parts of the world, the status of Pb pollution on a non-impacted suburban area of Istanbul metropolitan was evaluated. Emission sectors of lead in $PM_{10}$ and $PM_{2.5}$ samples from different sources have been quantified.

**Key Words:** Air Quality, Meteorology, Fine and Course Aerosols, Modeling

## 1. INTRODUCTION

The objective of the Third International Symposium on Air Quality Management at Urban, Regional and Global Scales is to bring scientists working in education universities, research organizations, government and industry together, working on management of air quality problems. One of the most important air quality criteria pollutants is the inhalable particulate matter. Recently the $PM_{2.5}$ part of the aerosols gained a big attention and new standards has been established by USA-EPA. In EU countries there are limited studies on $PM_{2.5}$ whereas in Turkey there are few studies (Samura, 2003). In Istanbul city, this is the first reported study regarding Pb in segregated aerosol samples. The current interest in atmospheric fine particulate matter ($PM_{2.5}$) is mainly due to its effect on human health (Christoph H. et al., 2005) Preliminary data indicate that large regions in the United States have ambient $PM_{2.5}$ concentrations that exceed the new $PM_{2.5}$ NAAQS resulting in unhealthy conditions for a significant fraction of the population in North America (Tony H., et al., 2005).
The aim of this study was to determine the Pb concentrations in segregated parts of aerosol and to determine their possible sources.

Concentrations of some pollutants in urban air may differ because of human activities. For this reason, a continuous air quality monitoring is important to understand the relationship between meteorological parameters and human related emissions. Atmospheric particulates, especially anthropogenic fine particles, typically enriched by a number of toxic metals (e.g., Pb, Sb, and Zn), are found to potentially influence human health (Samura, 2002; Karaca, et al. 2005).
On a global basis, anthropogenic inputs of Pb predominate over natural sources. Among several anthropogenic inputs, vehicular and roadside emissions of particles were often found to be the most important sources (Waleed, 1990). It was recorded that some 25% of vehicular emissions of Pb is coarse grained and deposited close to the road, while the remaining 75% is fine and may remain airborne (Hana and Al-Bassam, 1983).
During the study period, lead containing gasoline usage in cars decreased steadily and it was completely prohibited by the end of the February-2004 by the Turkish government. Because of this reason, this study has a particular importance to explain the outcomes of these actions. The goals of this study are:
1) To check and identify seasonal variations of Pb concentrations
2) To assess the influence of meteorological factors on temporal variations
3) To define the sources of Pb concentrations in airborne particles collected over a non-impacted suburban area of Istanbul metropolitan area
4) To compare the effects of limitations of lead containing fuels in cars

## 2. METHODS AND SAMPLING

### 2.1. Monitoring Site

Monitoring has been performed between July 2002 and July 2003. In this work, 92 daily aerosol samples of fine (< 2.5 μm) and coarse (2.5-10 μm) particles were collected. Samples were collected on daily basis randomly. Accordingly, at least six samples were collected monthly. The sampling site was chosen at a suburban area of Istanbul, located approximately 5 km north from the town of Büyükçekmece (41º2'35"N, 28º35'25"E). The Istanbul Water and Wastewater Administration (ISKI) compound, near the Büyükçekmece Lake, was chosen as the sampling station location because of easy access, availability of electrical power, and provision of enough protection for the sampling instrument. The location of the sampling station is shown in Figure 1.

The sampling site is located 10 m above the sea level and within the first zone of the watershed area, and sufficiently far away from residential areas, so that there is no significant stationary or mobile emission source within 5 km around the sampling site. In view of this fact, this area is suitable to monitor the regional and long-range effects on the characteristics of particulate matter (Karaca et al. 2005-a).

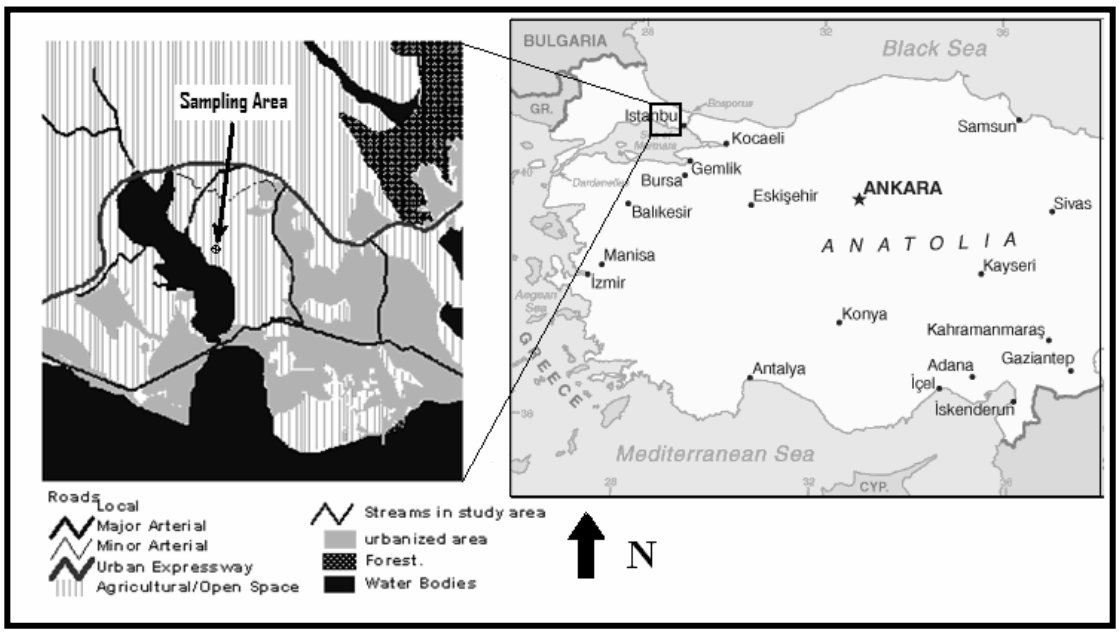


Figure 1. The location of the sampling site

## 2.2. Sampling Instrument

A size segregation dichotomous sampler (Anderson sampler) with automatic sample changer was used to collect aerosol samples. It has an aerosol inlet cut-off, which samples only particles smaller than 10 µm. In addition, the particles are being size-segregated by means of virtual impaction into fine particles smaller than 2.5 µm ($PM_{2.5}$) and coarse particles between 2.5 and 10 µm ($PM_{2.5\text{-}10}$). The particles were collected on 37 mm, 2 µm pore sized Teflon® membrane filters which are recommended for gravimetric and quantitative determination of trace metals (Karaca et al. 2005-a). The sampling flow rate was 1 $m^3$ $hour^{-1}$. The gravimetric determination technique is described by Karaca et al. (2005-a).

## 2.3. Chemical Analysis

The empty filters were pre-conditioned to stable weight at constant relative humidity (in silica gel desiccator) for at least 24 hours, and then the filters were transferred and weighted by means of a 4 digit sensitive balance. CEM MARS 5 microwave digestion system (CEM Corporation, Matthews, NC, USA ) was used for the dissolution of all the collected aerosol samples.

The optimized digestion mixture was found as: 5 ml of concentrated $HNO_3$ (Ultrex, J.T. Baker, Phillipsburg, NJ 08865, USA), 4 ml $H_2O_2$ (30-32%, semiconductor grade, Aldrich Chemical Company, Inc., Milwaukee, WI, USA), 0.5 ml HF (47-51%, environmental grade plus, Alfa AESAR, Ward Hill, MA. 01835, USA), and 5 ml of saturated $H_3BO_3$ solution 5%, (puratronic, 99.9995%, Alfa AESAR,. Ward Hill, MA 01835, USA ).

The digestion temperature was increased slowly to 210 °C by using maximum power (1200W) for 20 minutes digestion time. After cooling the digestion vessels, the

content was transferred to a 25 ml measuring flask and diluted to the mark with de-ionized pure water.
Graphite Furnace Atomic Absorption Spectrometer (GFAAS) technique was used to determine the Pb in aerosol samples, which is a suitable technique for the low concentrations found in aerosols matrix. Varian® (model) GFAAS instrument equipped with auto sampler and utilizing coated Varian® graphite tubes was used for Pb determination.
To obtain calibration curves aqueous standard solution method was used. The multi-standard solution (MERCK), which contains 23 different elements with concentration of 1000 mg/L, was used to prepare the calibration curve. $R^2$ value of the calibration curve was 0.99937 with curve-fit equation represented by the equation $y = 0.0038\ x$.
The accuracy of the instrument and method was tested using standard reference materials (SRM) IAEA-336 and IAEA Soil-5. The recovery of Pb from these SRM`s was 110.2% and 97.1%, respectively.

## 3. RESULTS AND DISCUSSION

The $PM_{10}$ aerosol mass annual arithmetic mean value of 47.1 μg $m^{-3}$ was found to be lower than the Turkish air quality standard of 60 μg $m^{-3}$. On the other hand, this value was found higher than the annual European Union air quality $PM_{10}$ standard of 40 μg $m^{-3}$. Furthermore, the annual $PM_{2.5}$ mean concentration of 20.8 μg $m^{-3}$ was found higher than the United States EPA standard of 15 μg $m^{-3}$. During wintertime, especially in November and December, there was a clear peak for both $PM_{10}$ and $PM_{2.5}$. This could be related to seasonal meteorological conditions and extensive use of fossil fuel for domestic heating (Karaca et al., 2005).

The annual mean Pb concentration of $PM_{2.5}$ and $PM_{10}$ was found as 0.054 μg $m^{-3}$ and 0.129 μg $m^{-3}$, respectively. The temporal variations of Pb concentrations for $PM_{10}$ and $PM_{2.5}$ fractions are given in Figure 2. The Pb concentration in both aerosol fractions followed nearly the same pattern with higher concentrations in the $PM_{10}$ fraction. Furthermore, there is a clear peak during summer time, especially in $PM_{10}$ fraction.

There is national neither standard nor international limits for Pb in fine and coarse aerosol fraction. Consequently, the best way to evaluate the air quality with respect to Pb levels is to compare our study with others. These measured aerosol Pb concentrations were compared with national and international studies and given in Table 1. As it can be seen from this comparison, the annual average Pb concentration over Buyukcekmece region is neither as high as polluted industrial areas in Milan-Italy (Vaccaro et al.., 2001) nor as low as in unpolluted remote areas. Result of this study could be classified among urban or sub-urban studies. This shows clearly the effect of urbanization and industry on suburban area, especially the one like Buyukcekmece watershed area.

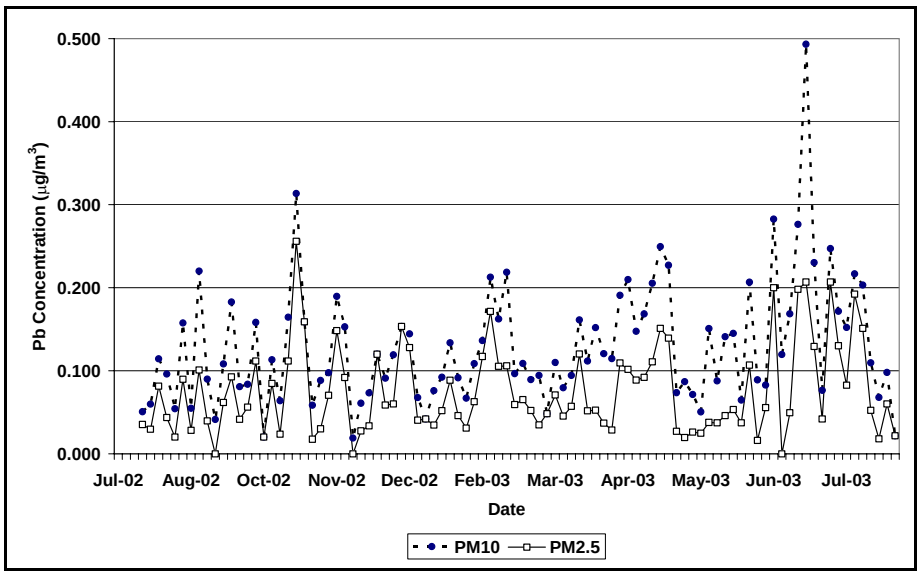


Figure 2. $PM_{10}$ and $PM_{2.5}$ Pb concentrations during study period

Table 1. Some measured Pb concentrations in similar studies

| **Region** | **Measured Concentration ($\mu g\ m^{-3}$)** | | |
|---|---|---|---|
| | $PM_{10}$ | $PM_{2.5}$ | $PM_{2.5-10}$ |
| Büyükçekmece (this study) | 0.129±0.074 | 0.075±0.054 | 0.054±0.038 |
| Geneva (Switzerland) (Chiaradia ve Cupelin, 2000) | 0.045±0.016 | | |
| Milan (Italy) (Vaccaro et al.., 2001) | 0.215±0.112 | 0.138±0.75 | |
| Shanghai (China) (Zheng et al., 2004) | 0.515 | | |
| Bursa (Türkiye) (Alusine et al., 2003) | 0.151 | 0.120 | 0.031 |

### 3.1. Seasonal Characteristics

In order to evaluate seasonal characteristics of the data set, monthly average Pb concentrations are calculated and illustrated in Figure 3. In general, lead concentrations of fine particles are higher than the coarse fraction, except in August 2002 and May 2003. This difference is much more significant during wintertime periods. One of the reasons of this seasonal behavior is the effect of the increase of anthropogenic activities such as fossil fuel combustion, and industries during heating season. We can assume that traffic is stable during whole sampling period; on the other hand there is significant increase in fossil fuel combustion to facilitate heating. The heating season of the sampling period corresponds to 10/2002-4/2003 interval. Another reason of this difference could be the effects of meteorological conditions. Exceptionally summer period of this study, an arid season was observed. There are

several studies, which mention that it is possible to observe less coarse particle concentrations during wet seasons than dry seasons (Helda et al. 2005, Hueglin, 2005).
Conversely, general trend of the data set shows that the atmospheric lead concentration is stable. This indicates that, principally, the effecting sources of lead over Büyükçekmece area are not conditional to seasons; expect the coarse fraction during wintertime. This outcome brings a necessity to the statistical investigation of the relationships of meteorological conditions and concentrations. This relationship is evaluated in the following sections.

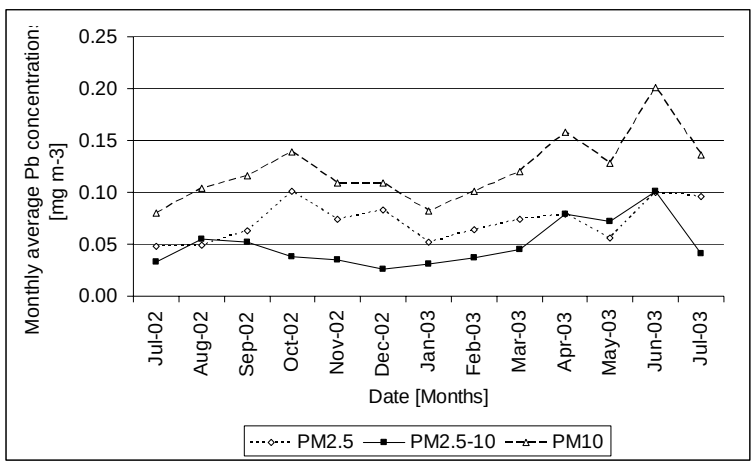


Figure 3. Monthly average atmospheric lead concentrations

The monthly average ratios of $PM_{2.5}/PM_{10}$ and $PM_{2.5–10}/PM_{10}$ could be a useful tool to figure the seasonal effects over lead containing particles (see Figure 4). As it seen from Figure 4, fine particles have more than 50% contributions to lead concentrations of inhalable particles during sampling period. This ratio changes only during two periods in August 2002 and May 2003. This result confirms the conclusions of previous assumptions.

Summer and winter time averages of lead concentrations of fine particles are calculated as $0.075 \pm 0.056$ μg $m^{-3}$, $0.076 \pm 0.052$ μg $m^{-3}$ respectively. The summer and winter values are nearly equal, which confirm that the emission of fine fraction lead concentration is fairly constant during seasons. Moreover, summer and winter lead concentration standard deviations are almost the same which confirm that their seasonal fluctuations have the similar characteristics. It is possible to speculate that atmospheric fine lead containing particles over Büyükçekmece area are emitted dominantly by traffic and industrial sources which are stable during seasons. Heating could not be the main source of atmospheric lead because otherwise it could not stable during all sampling period. A high standard deviation values indicates the receptor point is effected mainly by mobile sources which could fluctuate and unstable during weekly period.

On the other hand, if the standard deviation is considerably smaller, it means that the receptor site mainly effected by point and stable sources. Standard deviations of both

seasons are about 70% of average values, which could be accepted as noticeably high value. Because of this conclusion, one can say that, atmospheric lead over Büyükçekmece area is emitted generally by automobile sources as fine particles.

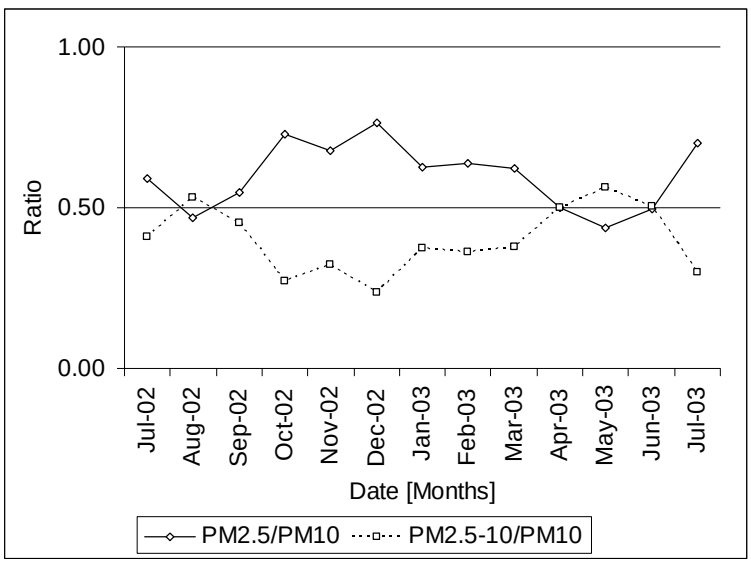


Figure 4. The monthly average ratios of $PM_{2.5}/PM_{10}$ and $PM_{2.5-10}/PM_{10}$

Summer and winter time averages of lead concentrations of coarse particles are calculated as $0.067 \pm 0.043$ μg $m^{-3}$, $0.036 \pm 0.022$ μg $m^{-3}$ respectively. Summer time average value is 1.84 times higher than winter time. It seems interesting due to lower concentrations, which are observed during winter times, but this acceptable if one consider the effects of meteorological conditions on the generation of atmospheric coarse particles.

Processes, especially, precipitation, dryness, and atmospheric wet deposition processes are the factors which governs the concentration of atmospheric particles (Karaca et al, 2005). Some rain samples were collected and analyzed in Büyükçekmece area by Başak and Alagha (2004), and they observed that precipitation samples collected during winter time have higher lead concentrations than summer times. This confirms that atmospheric lead particles are washed out by precipitation and atmospheric lead concentrations of wet seasons could be less than dry seasons. Generally, coarse particles are generated by natural effects like wind blowing, sea sprays, soil erosion etc. It was found that these coarse particles decreases in wet seasons and increase in dry seasons. This could be another reason of the decreased wintertime concentrations.

### 3.2. Relationships between Aerosol Mass and Lead Concentrations

Some useful information could be obtained by comparing measured lead and mass concentrations. By this way, it could be possible to figure that the cyclic behavior of enriched lead containing particles reaching the sampling site and the frequency of the episodes can be defined more properly. The time series of the ratios

$PM_{2.5}Pb/PM_{2.5}Mass$ [mg/g] and $PM_{2.5\text{-}10}Pb/PM_{2.5\text{-}10}Mass$ [mg/g] are given in Figure 5 and Figure 6 respectively. It is clear that the most observed particles peaks (episodes) are observed during summer times.

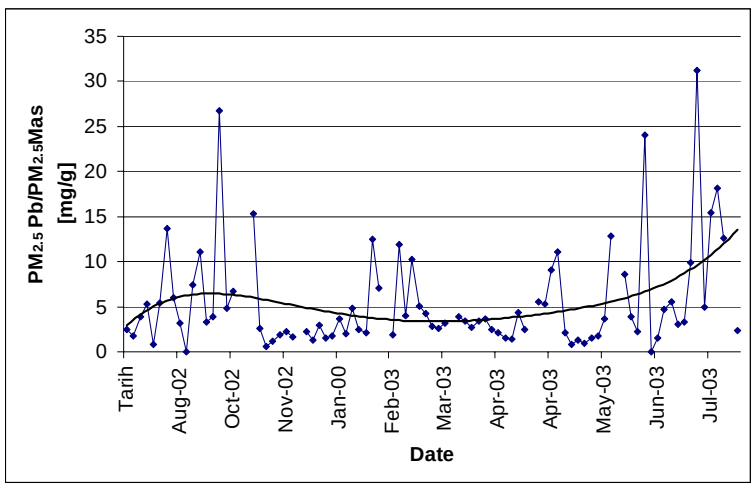


Figure 5. Seasonal behavior of $PM_{2.5}Pb/PM_{2.5}Mass$ [mg/g] ratio

According to these figures, the seasonal behaviors of both ratios are similar to each other. They have some sharp fluctuations during summer times, but more steady during winter times. We believe that this kind of unsteady behaviors can be explained by the effects of meteorological factors. Because of the similar trends in the cyclic behaviors of both ratios, one can think that the origins of fine and coarse lead containing particles are same.

Correlation coefficients ($R^2$) between particulate matter mass and lead concentrations for fine and coarse particles are calculated as 0.35 and 0.33, respectively. It shows that there is not any significant correlation between mass and lead concentrations. This means that the lead containing particles are not a constant share of particles and may not be emitted directly from any local sources but several regional sources. This figure is completely different for heating season concentrations. The correlation coefficients between mass and lead concentrations of fine and coarse particles are 0.62 ($P<0.01$) and -0.03 during heating season, respectively. The significant correlation (0.62) indicates that there is a valid relationship between mass and lead concentrations of fine fraction.

On the other hand no correlation (-0.03) is found for coarse particles. This result indicates that lead concentrations of fine particles are mostly generated by local and steady sources which are related by heating activities like some lead containing fossil fuels.

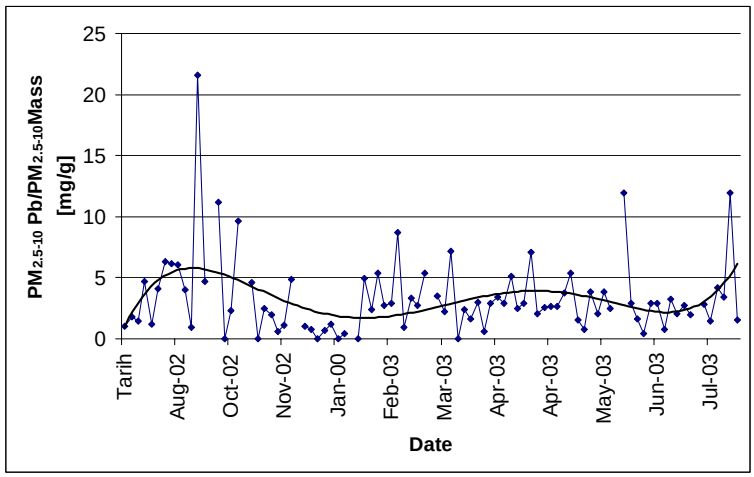


Figure 6. Seasonal behavior of $PM_{2.5-10}Pb/PM_{2.5-10}Mass$ [mg/g] ratio

Last of all, the relationships between lead concentrations of fine and coarse particles are calculated and found as 0.35. Contrary to the previous conclusion assumption, it can be speculated that lead concentrations of fine and coarse particles may be not generated by common sources or similar processes. Fine and coarse particles containing lead are generated mainly by traffic during summer time from several regional sources. However, maybe fine particles containing lead are mainly generated by seasonal heating during wintertime from local sources.

### 3.3. Definition of Possible Sectors of Episodes

Lead bearing episodes are of particular concern. Episode values are more critical than regular trends; possibly, they are enriched over a particular area or affected by some point sources. The highest 30% lead level samples of the total data set were defined as episodes. In addition to that, the region of the study is divided into geographically 16 sectors and these sectors are given in Table 2. Occurrences of the observed wind directions according to these sectors are counted and the repetition numbers are calculated. This is very useful method to define the sectors, which are the responsible of episodes. After that, it could be possible to focus on particular sectors to search possible sources of episodes within these sectors. These sectors could be collected into four main sectors: North-East (NE), South-East (SE), South-West (SW), and North-West (NW).

According to this sectoring, the major part of the episodes, about 54%, is generated from the North-East sector. South-West sector is found as the second major sector with the value of 23% (see Figure 7). Two main residential areas, Hadımköy and Büyükçekmece are located in the northeastern and southwestern parts of study area, respectively. During sampling period, natural gas distribution system was not connected to these residential areas. Coal and fuel oil had been used extensively for residential heating. The episode values seem related with residential activities, like city inside traffic, residential heating.

Secondly, in the northeastern part of the study area, industrial activities are standing out. It is clear that industrial activities of this sector are the one of the responsible

sources of atmospheric lead over Buyukcekmece lake basin. In addition to that, there is a cement factory located on the southwestern part of sampling area. However, in the literature, generating of atmospheric lead from cement factories is not clear, one should note that, as a conclusion of our personal observations; they possibly use several materials (car tires, coal, etc) as fuel.

Table 2. Wind directions and episodes occurrence

| Wind Direction | Repetition | Wind Direction | Repetition |
|---|---|---|---|
| E | 2 | S | 0 |
| ENE | 3 | SE | 1 |
| N | 4 | SSE | 0 |
| NE | 4 | SSE | 1 |
| NNE | 5 | SW | 2 |
| NNW | 2 | SWS | 2 |
| NW | 2 | W | 2 |
| NWW | 0 | WSW | 0 |

Finally, TEM and E-5 motorways can be pointed as atmospheric lead sources. E-5 and TEM are located in South-East, South-West and North-East, North-West locations, respectively. Probably they have significant contributions to atmospheric lead episodes.

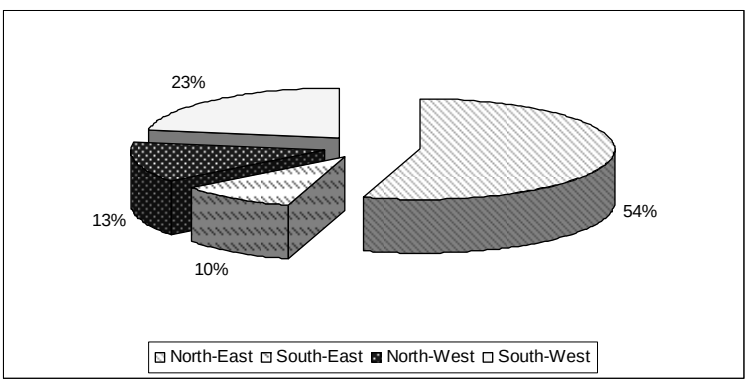


**Figure 7. Sectors of atmospheric lead episodes**

## 4. CONCLUSION

In this study, the annual mean Pb concentration of $PM_{2.5}$ and $PM_{10}$ was found as 0.054 μg $m^{-3}$ and 0.129 μg $m^{-3}$, respectively. These values are moderate according to literature values of remote and urban sites. Generally, fine particles lead levels are higher in summer that in wintertime, which is related to traffic and industrial activity. There was no significant correlation between particulate matter mass and lead concentrations for fine and coarse particle. This may be explained by the fact that, the lead containing particles are not a constant share of particles and may not be

emitted directly from any local sources but several regional sources. On the other hand, a significant correlation (0.62) between mass and lead concentrations of fine and coarse particles indicates that there is a valid relationship between mass and lead concentrations of fine fraction. Episodic sectoring resulted in a conclusion that the major part of the episodes, about 54%, is generated from the NE sector while SW sector is found as the second major sector with the value of 23% for the observed episode values.

## 5. ACKNOWLEDGEMENTS

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# EFFECTS OF METEOROLOGY ON ANKARA AIR QUALITY

Deniz Derya Genç[1], Canan Yeşilyurt[2], Banu Bayar[2] and Gürdal Tuncel[1]

[1]Middle East Technical University,Department of Environmental Engineering,Inonu Bulvari 06531 Ankara,Turkey,dgenc@metu.edu.tr
[2]Refik Saydam Hygiene Center, Air Pollution Laboratory, Ankara, Turkey

## ABSTRACT

The daily air pollution index (API) highly correlated with meteorological parameters over the whole urban area of Ankara and assimilative capacity of Ankara atmosphere are investigated in this study.$SO_2$ and $PM_{10}$ concentrations measured at eleven monitoring stations all over the Ankara are used in API. Two meteorological variables namely, maximum daily wind speed and thermic excursion, and the previous day's air pollution index are found to be highly correlated (r=0.72) with daily air pollution index by the multiple linear regression analyses. API forecasting is applied for the whole urban area of Ankara. Product of average wind speed and mixing height are used for the determination of assimilative capacity of Ankara atmosphere. The air pollution dispersion index (ventilation index) which is proposed by the State of Colorado Department of Health in Denver is used. The assimilative capacity of Ankara is found to be highest in summer and lowest in winter, while the highest values of ventilation coefficient are observed during afternoon hours in both seasons.

**Key Words:** API; API Forecast; Ventilation Coefficient; Mixing Height; Ankara

## 1. INTRODUCTION

Air pollution problem in Ankara has been known since 1926 and Tincer et.al., (1975) underlined the possible future air quality problems and suggested that Ankara was not a suitable urbanization region. Air pollution problem in Ankara was mainly associated with fossil fuel combustion and up to now some precautions were taken. Combustion of high sulfur content of coal was banned and usage of low sulfur content of coal was encouraged by a Governmental Decision in 1985, however the effect of this effort on air quality was not observed because of the illegal entrance of poor quality coal to the Ankara. The sharp decrease in fossil fuel combustion based pollutants in Ankara was observed after 1990s, as a result of the switching from coal to natural gas usage for heating. The decrease in fossil fuel combustion based pollutants were the most dramatic between 1990 and 1995, their concentrations in the city are still decreasing but at a slower pace.

Ankara, which is the capital of Turkey, has been urbanized faster due to migration from countryside to the city results in greater emissions into atmosphere, mainly produced by the increase of traffic, as well as from the use of old and badly maintained vehicles. Therefore present day urban air pollution in Ankara is thought

to be mainly associated with still domestic heating and traffic emissions which is accepted as the main source of the urban air pollution problem in most of the world cities (Mage et al., 1996; Mayer, 1999).

The occurrence of high concentrations of lower atmospheric pollution across Ankara is critically dependent upon the hilly topography of the region which lessens lateral transmission of air masses. Besides physical setting of Ankara, meteorology also plays an important role for dispersion and dilution of emitted air pollutants since meteorological factors such as wind speed, precipitation and mixing height all play important roles in determining the pollutant levels for a given rate of pollutant emission (Tayanc, 2000). Therefore, the aim of this study is to investigate the effect of meteorology on Ankara air quality.

## 2. MATERIALS AND METHODS

This study is based on hourly $SO_2$ and $PM_{10}$ data generated at three curbside stations which were placed at the most crowded street corners of Ankara, namely Iskitler, Kavaklıdere and Kızılay, and eight non-curbside stations which were not under direct influence of traffic activities, by the Ministry of Health, Refik Saydam Hygiene Center, between October 1999 and August 2000. A fully automatic mobile laboratory was used for measurements at curbside stations therefore measurements at curbsides were not continuous, but performed for approximately one week in each month. However, measurements at non-curbsides were continuous as these stations were regular monitoring stations of Ankara Air Quality Monitoring Network.

$SO_2$ was measured by conductometry (DKK Model GRH-72M) and $PM_{10}$ was measured by beta ray absorption (Model DUB-12).

There are two meteorological stations in Ankara operated by the Turkish State Meteorological Service, one located at Incirli, which is within the downtown area, and the other one at Etimesgut, which is at the outskirts of the city. As the measurements in this study performed at urban area, the meteorological data from the Incirli station was used.

## 3. RESULTS AND DISCUSSION

In this study, air pollution index (API) is used for the investigation of the effect of meteorology and used for forecasting the air pollution over the whole urban area of Ankara by the method developed by Cogliani (2001). The hourly highest concentrations in the day of $SO_2$ and $PM_{10}$ are used for the evaluation of the daily air pollution index from October 1999 to March 2000. Unfortunately, there is no daily threshold values of major air pollutants for the determination of daily air pollution index that are fixed by the Turkish Ministry of Environment and Forestry. Furthermore, some deficiencies of air pollution index API proposed by EPA and a need for different API is reported in the literature (Murena, 2004). Therefore in this study the new API which is proposed at CITEAIR Workshop (2005) was used. Three air quality levels are derived from the threshold values given in Table 1, as: good,

acceptable and poor. For a single station, a score is given depending upon the hourly highest concentration of $SO_2$ and $PM_{10}$. If the air quality level is good the score is 1, if the air quality level is acceptable the score is 3, if the air quality level is poor the score is 7. When the concentration is missing, the score is 0. The daily scores of $SO_2$ and $PM_{10}$ are summed, the total score is from 1 to 2, from 3 to 6, more than or equal to 7. When total is 0 not valuable (NV) is attributed. The new score is given depending upon the summed scores. When total is 2 the new score is 0 indicating the good air quality level. When the quality of the air is acceptable the value varies from 4 to 6 and the new score is 1. For the poor air quality level, 2 is attributed as the new score if the values are from 8 to 14. When concentrations are missing the total score is 0 and new score 2 (poor air quality level) is attributed to missing data. This reference is made to $SO_2$ and $PM_{10}$ concentrations measured at all available stations. Then, the sum of the new scores of the all available stations gives the daily air pollution index, I, of the whole city.

Table 1. Proposed Air Quality Index of $SO_2$ and $PM_{10}$ (CITEAIR ,2005)

| **Air Quality Level** | **Index** | **Class** | **$PM_{10}$[a]** ($\mu gm^{-3}$) | **$SO_2$[a]** ($\mu gm^{-3}$) |
|---|---|---|---|---|
| GOOD | Very Low | 0<br>25 | 0<br>25 | 0<br>50 |
| | Low | 25<br>50 | 25<br>50 | 50<br>100 |
| ACCEPTABLE | Medium | 50<br>75 | 50<br>75 | 100<br>300 |
| POOR | High | 75<br>100 | 75<br>100 | 300<br>500 |
| | Very High | >100 | >100 | >500 |

[a] corresponds to the maximum hourly value on a day

Air pollution levels over the Ankara calculated from the proposed API during the October 1999-March 2000 period is given in Table 2.Poor air quality dominates during this period. In fact, although overestimation is done in API calculation (the new score 2 (poor air quality level) is attributed if the total score is 0), the results are reasonable as the application period is in winter. Low air quality in winter is frequent in Ankara atmosphere as a result of the lack of ventilation (will be discussed in proceeding section) and high emissions.

Meteorological data are examined in the preliminary analysis to extract the meteorological variables that is strongly correlated to daily air pollution index. Therefore one by one correlation between the daily air pollution index and the meteorological variable (daily highest temperature, daily lowest temperature, daily average temperature, daily highest wind speed, daily average wind speed, daily average barometric pressure, daily highest mixing height, daily average mixing height and daily average barometric pressure) is investigated.

Table 2. Percentage of Occurrence of Pollution Levels during October 1999-March 2000 period

| **Pollution Level** | **%** occurunce |
|---|---|
| GOOD | 1 |
| ACCEPTABLE | 14 |
| POOR | 85 |

Daily highest wind speed and daily thermic excursion, which is calculated from the hourly temperature data, show a high correlation with daily air pollution index ($R^2$=43 % and $R^2$=14 % respectively); therefore other meteorological variables that show a weak correlation with the daily air pollution index are discarded.

Daily vehicular traffic is known as the major air pollution source in most urban areas (Mage et al., 1996; Mayer, 1999). Daily traffic data is missing in this study; therefore previous day's air pollution index value $I_{d-1}$, which explains 29 % of the variability in daily air pollution index, is chosen instead of the daily traffic data as another correlation variable to the daily air pollution index.

The calculated air pollution index $I_c$ is obtained by the linear multiple partial correlation among the three independent variables:

- daily highest wind speed;
- thermic excursion ;and
- previous day's air pollution index.

Statistical summary of the linear multiple correlation analysis is given in Table 3. The calculated air pollution index ($I_c$) equation for Ankara during the October 1999-March 2000 is:

$$I_c = 14.3101 - 0.503\,V + 0.178\,I_{d-1} + 0.145\,\Delta T \qquad (1)$$

Table 3. Statistical summary of the linear multiple partial correlation analysis

| Model | $R^2$ | r | Adjusted $R^2$ | Std. Error of Estimate |
|---|---|---|---|---|
| (Constants), maximum wind speed, thermic excursion, $I_{d-1}$ | 51.23 | 0.72 | 49.72 | 2.03 |

Correlation between the calculated air pollution index $I_c$ and the air pollution index I is 0.72. The good agreement between I and $I_c$ values during the measurement days is seen in Figure 1.

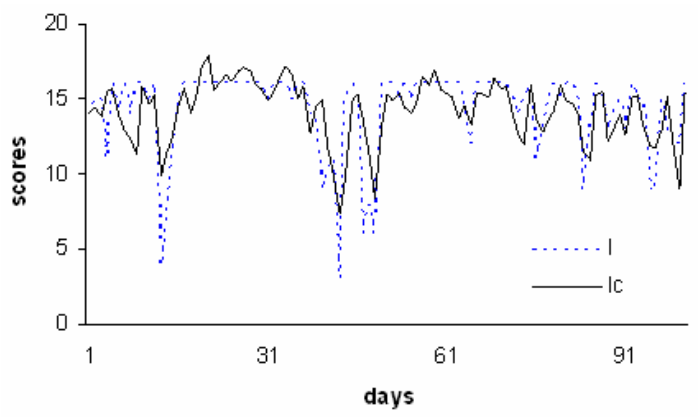


Figure 1. I and $I_c$ trends versus observation days during the period October 1999-March 2000

Verification of the $I_c$ obtained from the examination of a specific period is performed by using the three independent variables that is measured outside that period. Independent variables measured between January-March 2003 are used and the correlation is found as 0.45 while the standard error is 1.95. I and $I_c$ trends versus observation days during the period January-March 2003 is given in Figure 2. Although the correlation decreases from 0.72 to 0.45, this could be accepted as a good forecast. Because lower correlations are seen most of the air pollution index forecast models (Jiang et al., 2004).

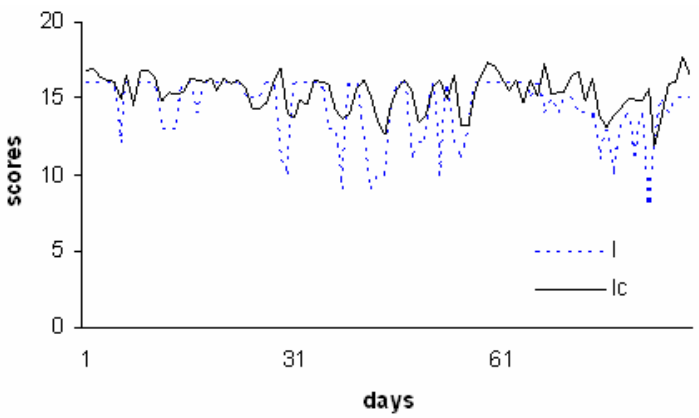


Figure 2. I and $I_c$ trends versus observation days during the period January-March 2003

In this study assimilative capacity of the atmosphere which is defined as the maximum pollutant load that can be discharged into the atmosphere without violating the best-designed use of air resources in the planning region (Manju et al., 2002) is also determined in terms of ventilation coefficient. Ventilation coefficient is product of mixing height and average wind speed through the mixing layer.

The air pollution dispersion index (ventilation index) which is proposed by the State of Colorado Department of Health in Denver is used to determine the ventilation coefficients for Ankara during the measurement period. In this study, surface wind speeds which are obtained from Incirli Meteorology Station and mixing heights which are obtained from EPA's PCRAMMET program are used. The categories used for air pollution dispersion are POOR: 0-2000, FAIR: 2001-4000, GOOD: 4001-6000 and EXCELLENT: ≥6001. The percent occurrence of each ventilation category during the sampling period is given in Figure 3.a. As can be seen from the Figure 3.a percentage of the poor dispersion conditions are dominant during the measurement period.

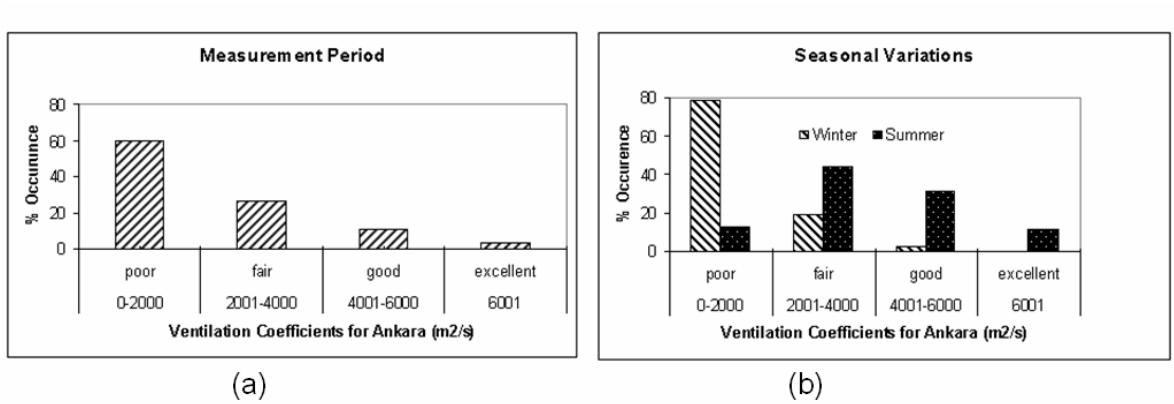


Figure 3. Variations of Ventilation Coefficients for Ankara (a) sampling period (b) seasonal

Seasonal variations of ventilation coefficients (dispersion conditions) are also investigated and depicted in Figure 3.b Seasonal variations in the ventilation coefficients show that poor dispersion conditions dominate over Ankara during the winter season while the increase in ventilation is obviously observed for summer season. Therefore, poor air pollution dispersion conditions besides the high source strength may explain the higher pollution levels over Ankara in winter season to a considerable extent.

Hourly computed ventilation coefficients during the winter and summer seasons are given in Figure 4. Lowest ventilation coefficients during the night and early morning indicate high pollution potential during this period. The highest values of ventilation coefficient are observed during afternoon hours due to increase in solar insolation. Viswanadham and Anil Kumar (1989) stated that when the incoming solar insolation increases as the day progresses the ventilation coefficient also increases during afternoon hours; further during evening hours when the incoming solar radiation ceases the ventilation coefficient also gradually decreases. Therefore, the assimilative capacity of the Ankara is at its best during noon. The lowest ventilation coefficients for both winter and summer are nearly same however the highest ventilation coefficient is about a factor of 4 higher in summer than in winter.

Diurnal variations of the mixing height also shows similar pattern in two seasons. The highest value observed is 2038 m in summer and 850 m in winter. The very low

values of mixing height that are reflected in the ventilation coefficient during the late nights and early morning hours could be due to the occurrence of ground based inversions that hamper dispersion (Padmanabhamurty and Mandal,1979). The wind speed in summer is only slightly higher than the winter wind speeds. However, there is a significant difference between the summer and the winter ventilation coefficients. This could be due to the fact that the contribution by wind speed to ventilation coefficient is less in comparison to mixing height. Investigation of the wind speeds during the measurement period showed that almost 60 % of winds blew at a speed between 2 and 4 m/s and less than 8 % had a speed higher or equal than 5 m/s. Thus, the low wind speed surpassed over Ankara during the measurement period. Not only because of the physical setting but also lower wind speeds restrict the horizontal dispersion, therefore the main dispersion mechanism over Ankara is vertical dispersion.

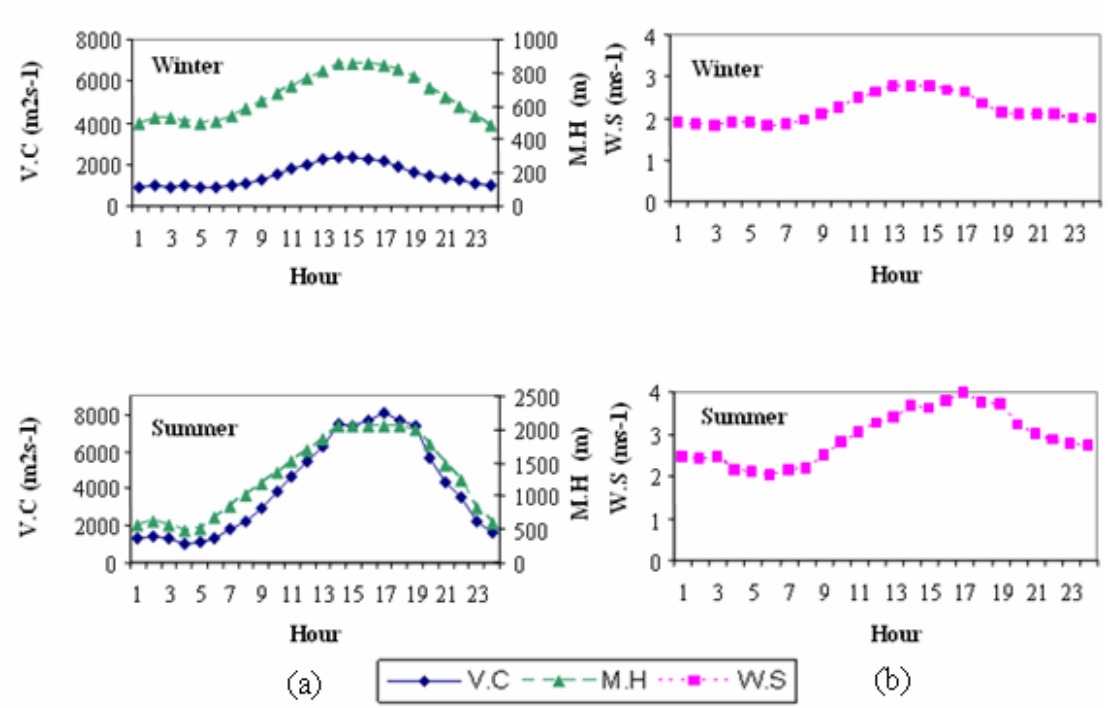


Figure 4. Seasonal diurnal variations of (a) ventilation coefficient (V.C) and mixing height (M.H) (b) wind speed (W.S)

## 4. CONCLUSION

Application of API forecast method in Ankara showed that meteorological parameters (maximum daily wind speed and thermic excursion) play an important role in forecasting the air pollution over Ankara.

The assimilative capacity of Ankara is found to be highest in summer and lowest in winter. The highest values of ventilation coefficient are observed during afternoon hours in both summer and winter. The contribution of mixing height to ventilation coefficient is higher in comparison to wind speed. Lower wind speeds surpass over Ankara, therefore the main dispersion mechanism over Ankara is vertical dispersion.

Reporting air pollution data to the public become a requirement or desire in most of the world cities (Brimblecombe, 2005). Although forecasting the next day air pollution is not a requirement yet, most of the cities report the next day's air pollution to the public. Unfortunately, there is no attempt to report neither the previous day's air quality nor the next day's air quality in Turkey. Therefore, this study may aid governmental officials in their efforts to regulate and improve air quality in Turkey.

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# GENOTOXICITY EVALUATION OF ATMOSPHERIC ENVIRONMENTS OF SUBWAY STATIONS IN SEOUL USING TRADESCANTIA MICRONUCLEUS ASSAY

**Hae Shik Shin, Jin Kyu Kim[1], Jeong-Joo Lee[3] and Jin-Hong Lee[2]**

[1]Korea Atomic Energy Research Institute, Jeongeup, 580-185, Korea,
shs9645@hanmail.net, jkkim@kaeri.re.kr
[2]Dept. of Environmental Engineering, Chungnam Nat'l University,
Daejeon, 305-764, Korea
[3]Dept. of Environmental Health, Yong-In University, Yongin, 449-714, Korea

## ABSTRACT

Airborne pollutants in the subway facilities can be potentially harmful to the health of passengers. This study was designed to check whether the suspended particulates have mutagenic or carcinogenic effects on the plant cell systems. Total suspended particulates were collected with a high volume air sampler, in the entrance, the waiting room, and the platform of each subway station. The biological end-points in this experiment were the micronuclei in the pollen mother cells of Tradescantia. The exudates were prepared by shaking the filter paper from the sampler in distilled water for 30 minutes. The differences in genotoxicity between air samples from the subway station and the control value was statistically significant. Almost every plant cutting exposed to the exudates resulted in a positively clastogenic response. The chemical analysis showed that the aqueous extracts from the air samples contained several heavy metals. Relationship between the induced genotoxicity and heavy metal concentration could not be clearly defined. The meiotic pollen mother cells of Tradescantia clone 4430 are particularly sensitive to genotoxic pollutants. Thus the Tradescantia micronucleus (Trad-MCN) assay was selected as a biological test system to evaluate airborne particulates collected in the subway facilities. A significant correlation was observed between the intensity of the pollution and the frequency of micronuclei appearing at the tetrad stage of meiosis.

**Key Words:** Tradescantia micronucleus assay; Air particulates, Genotoxicity, Subway station

## 1. INTRODUCTION

Airborne pollutants in the subway facilities can be potentially harmful to the health of passengers. Experimental studies have shown that extracts of urban air particulates can induce cancer in animals and are mutagenic in bacteria and mammalian cells (Sasaki et al., 1987, Crebelli et al., 1995, Pagano et al., 1996). Several epidemiological studies have revealed possible health risks related to air pollution and have suggested an association with an increase in lung cancer and respiratory diseases for the urban population (Schwartz, 1991, Pershagen and Simonato, 1993).

The quality of the indoor air we breathe and the attendant consequences for human health are influenced by a variety of factors. These include hazardous material discharges indoor and outdoors, meteorological and ventilation conditions, and pollutant decay and removal processes. Over 80% of our time is spent in indoor environment so that concentration of indoor air pollutants, exposure and ventilation are important consideration for evaluation of human risks (Dockery and Spengler, 1981). It is important to identify the level of genotoxic activity in the environment and to relate it to biomarkers of cancer risk in humans.

The Tradescantia-micronucleus test has been used in studies on air pollution because this plant is particularly sensitive to chemical mutagens and can be used for in situ monitoring programs (Ma, 1983, Ma et al., 1994).

This study was designed to check whether the suspended particulates have mutagenic or carcinogenic effects on the plant cell systems. Total suspended particulates were collected with a high volume air sampler, in the entrance, the waiting room, and the platform of three subway stations in Seoul.

## 2. MAIN TEXT

Collection of subway particulate sample

Suspended particles were collected on fiber glass filter (8" x 10", Whatman, PM2000) by high-volume air sampler (Sierra Anderson, Model 305, USA) operated at a flow rate of 1.2 m3/min for 20h. The air volume pulled through filter was 1221.7 – 1682.4 m3. Air samples were collected with a high volume air sampler, in the entrance, the waiting room, and the platform of each subway station. The

biological end-points in this experiment were the micronuclei in the pollen mother cells of Tradescantia. The exudates were prepared by shaking the filter paper from the sampler in distilled water for 30 minutes. The aqueous extracts for heavy metals from these glass filters were extracted by sonicated for 30 min using Branson 5210 sonicator. The aqueous extracts were tested for Trad-MCN assay after extraction.

**Tradescanita-micronucleus test**

The aqueous filter extracts for heavy metals were tested according to the procedure used by Ma et al. (1994). Briefly, 10 - 15 plant cuttings bearing young inflorescence of Tradescantia clone 4430 were exposed to 100 ml aqueous filter extracts in Erlenmeyer flasks with various sampling location samples. The duration of young inflorescence treatment was 24-h, followed by a 24-h recovery in Hoagland solutions. Next young inflorescences were fixed for 24-h in prepared 1:3 aceto-ethanol solutions and then transferred to 70% ethanol for storage to reuse.

For scoring of Trad-MCN, five slides per treatment were prepared using the aceto-carmine technique. The genetic damage was expressed as Trad-MCN /100 tetrads. Negative controls were performed in Erlenmeyer flasks with deionized water.

The concentration of the total suspended air particulates and genotoxicity induced in the Trad-MCN assay are given in Table 1. The micronucleus frequencies in the Tradescantia inflorescence exposed to the extracts from the total suspended air particulates were from the lowest of 5.27±0.27 (waiting room of City hall subway station) to the highest of 10.13±0.37 (platform of Shindorim station).

Almost every plant cutting exposed to the exudates resulted in a positive response. The differences in the genotoxicity between the air samples from the subway station and the control value (2.41±0.32) were statistically significant. In case of Shindorim station, the Trad-MCN frequencies were 7.27±0.43 MCN/100 tetrads ($p < 0.001$) for the entrance, 8.53±0.47 MCN/100 tetrads ($p< 0.001$) for the waiting room, and 10.13±0.37 ($p < 0.001$) for the platform of the subway station.

Table 1. The frequency of micronuclei in Tradescantia inflorescences exposed to the extracts from the total suspended airborne particulates

| Sampling sites | | TSP Conc. (ug/m3) | Exposure (h) | MCN / 100 tetrads | Significance |
|---|---|---|---|---|---|
| Sindorim | A | 219.4 | 24 | 7.27 ± 0.43 | *** |
| | B | 278.8 | 24 | 8.53 ± 0.47 | *** |
| | C | 269.5 | 24 | 10.13 ± 0.37 | *** |
| City Hall | A | 141.5 | 24 | 9.80 ± 0.65 | *** |
| | B | 188.2 | 24 | 5.27 ± 0.27 | *** |
| | C | 238.6 | 24 | 8.07 ± 0.36 | *** |
| Shincho n | A | 138.7 | 24 | 8.27 ± 0.55 | *** |
| | B | 282 | 24 | 7.87 ± 0.40 | *** |
| | C | 252.5 | 24 | 9.87 ± 0.62 | *** |
| N.C. | D | | 24 | 2.41 ± 0.32 | |

A) Entrance, B) Waiting room, C) Platform, D) D.W., N.C. : Negative Control, * : Duncan' s t-test

## 3. CONCLUSION

The chemical analysis showed that the aqueous extracts from the air samples contained heavy metals such as copper, iron, antimony, barium, lead, manganese, etc. However their concentrations were relatively lower than those of any other environmental samples such as sediments, soils and river waters. And thus relationship between the induced genotoxicity and heavy metal concentration could not be clearly defined in this study.

It has been reported in several studies that the micronucleus assay proved more reliable and sensitive to the test than the stamen hair assay (Monarca et al., 1999, Kim et al., 2003). The results indicate that the air particulates can give an adverse effect on the health of subway passengers. The Trad-MCN assay was selected as a biological test system to evaluate airborne particulates collected in the subway facilities since the meiotic pollen mother cells of Tradescantia clone 4430 are particularly sensitive to genotoxic pollutants. A significant correlation is observed between the intensity of the pollution and the frequency of micronuclei appearing at the tetrad stage of meiosis. It was reported that the Tradescantia-MCN assay was

sensitive, reproducible and well standardized (Ma et al, 1994) and our results also support that the assay is easy and inexpensive to use. As found in the previous researches on air pollution and indoor pollution monitoring (Monarca et al., 1999, Kim et al., 2003), the Trad-MCN assay is sensitive to environmental genotoxins and a promising test tool for monitoring airborne genotixins in the environments.

## 4. ACKNOWLEDGEMENTS

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# STUDIES ON WIND ENVIRONMENT AROUND HIGH BUILDINGS IN URBAN AREA

**Liu Huizhi[1], Jiang yujun[2], Liang Bin[3], Zhu Fengrong[3] and Sang Jianguo[2]**

[1]LAPC, Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing 100029 huizhil@mail.iap.ac.cn
[2]Department of Atmospheric Sciences, School of Physics, Peking University, Beijing 1000871, jyj@pku.edu.cn
[3]State Key Laboratory of Turbulence and Complex System, Peking University, Beijing 1000871, sjg@pku.deu.cn

## ABSTRACT

In this studies, hot-wire wind measurement, wind scouring in wind tunnel and numerical computation were carried out to evaluate the wind environment of a tall buildings in the prevailing flow conditions in Beijing area. The results obtained by three techniques were compared and mutually verified. The conclusions of the buildings drawn from three approaches are agreement with each others. The studies analyze the advantages and limitations of each method, and point out that the combination of different techniques may produce better assessment of wind environment around high buildings.

**Keywords:** high buildings in urban area, wind environment, wind tunnel, numerical simulation.

## 1. INTRODUCTION

Along with the social and economic development, the scale of urban area as well as the quantity and height of tall buildings increase rapidly. Those structures change outstandingly the wind environment of the urban area. The tall and dense architectural complex reduces the ability of urban ventilation and self-purification, and intensifies urban air pollution and heat island effects in weak wind conditions. On the other hand, in strong wind conditions the tall building may induce local gusts in its vicinity and affect the pedestrian comfort and safety. Then the pedestrian level wind environment problem arises (Isyumov, 1986; Bottema, 2000).

In the lower flow fields of the urban surface layer with the intrusion of the tall building as a bluff body, the strong wind may appear around the buildings with the effects of the secondary circulations induced by the buildings such as channel flow,

corner flow, passing through flow as well as blocking, weak flow and vortex, etc. The appearance of the strong wind flow phenomena may bring uncomfortable for the pedestrian passed throng this area, and the damage of the building's gates and windows, the decorations etc. Those problems induce by the buildings have been paid more attentions by the government and the persons from the world. So in many developed countries there has legislation on the evaluation of the wind environment around the high buildings during the design phase of this building. The designer, proprietor, architect and building developer also hope that there has a comfortable and safety environment in the buildings. Thus studies on the pedestrian wind environment problems caused by tall building have a great development in recent tens years (Capelaoet.al,2003, Chang and Meroney, 2001, Ferreira et. al, 2002, He and Song,1999, William and Soligo, 1991, Hunt. et.al, 1976, Penwarden and Wise, 1975, Cochran and Cermak, 1991).

Since the assessment must be carried out before the construction of the new building, it can only do according to the design scheme. The main methods of the wind evaluation are fluid experiment (wind tunnel and water tank experiment) and numerical simulation. The PIV Particle Image Velometry can give a quantitative results about the flow around the buildings, and it has been used on the basic research of wind environment (Liu et. al, 2003, Baik and Park, 2000), but until now it has not been used in the applicant assessment. To obtain a quantitative flow field around the buildings in wind tunnel is the main method in wind assessment in the present. However it needs to spend lot of time to measure the wind field in an area. Some other methods including the scouring technique to replace point measuring method are also beginning to use in some wind environment evaluation, so that the whole flow field could be obtained in an area at the same time during one experiment. Numerical simulation now is used more on the basic research and applicant study in wind assessment, because of the simple method to obtain three-dimensional flow, temperature and pressure field in the space. The basic research includes comparing the model results with the field experiment and observation data, and improving the numerical model (He and Song, 1999, Sang et. al, 2000). Jones et. al. found that the wind tunnel and numerical simulation results could predict the strong wind location around the high buildings which the wind amplification factor is near 3.0, but the exact location and the range of the strong wind area are not the same by comparing the two results. In order to improve the numerical models, more studies on the comparing and improvements of the different method on wind environment are needed.

In the present paper by using wind tunnel tests, numerical computation and meteorological data analyses, the wind environment of S Building in Chaoyang District, Beijing has been studied. In the tunnel tests hot wire anemometer measurement and scouring technique were conducted. Those two approaches companied with numerical methods compensated each others and verified mutually to

provide a convenient and reliable way for wind environmental assessment of tall building or architectural complex.

In the design scheme of S building, the main part of S Building is a 30 storied tower with height of 123m and a 5 storied wing building. Around S Building there have already been some buildings, for instance the Forum Hall 16 meters west to S Building. The main objective of the assessment is to study the characteristics of flow fields in the vicinity of S Building in the strong approach wind, especially the strong northwesterly in winter and spring of Beijing area. According to the original design the Forum Hall should be pulled down. So another objective of the assessment is to evaluate the differences of the wind fields with and without the Forum Hall. So that reasonable suggestions may be made to modify the design so as to provide an acceptable wind field around the buildings.

In the studies the results of the three evaluation methods were mutually compared and the reliabilities as well as the advantages and limitations of them were analyzed. On the basis of these studies an appropriate, multi-discipline approach for the assessment of wind environment around tall buildings can reasonably be proposed.

## 1. FLOW VELOCITY MEASUREMENT IN WIND TUNNEL

### 1.1 Measurement method and facilities

The measurement was carried out in Peking University atmospheric boundary layer wind tunnel with scale of 32m(length)×3m(width) ×2m(height). The models of S Building and the surrounding structures were made acrylic plastic with ratio of 1:150 as shown in Figure 1.

Figure 2 shows the distribution of the measurement points at the height of 4m in full scale. The key areas of measurement were the leeside and the windward side of S Tower in northwesterly, where the strong wind might occur. In order to study the

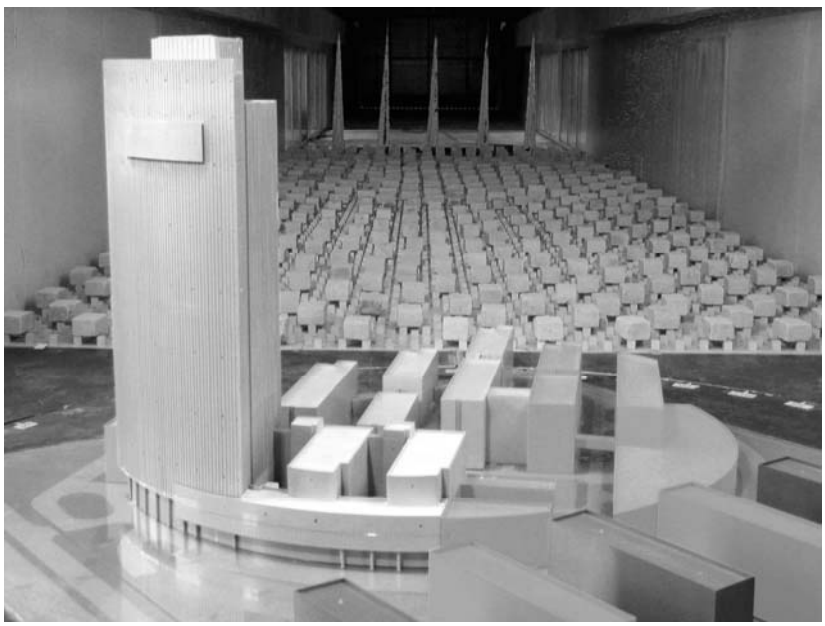

Figure 1. Model of S building in wind tunnel (scale 1:150). The viewpoint is from west to east while the high buildings in the left of the model is S building, and the roughness elements constructed from cubes is located in the far side direction.

variation of the wind field affected by the Forum Hall, the measurements were conducted again in the same flow conditions after the model of Forum Hall was moved away. In addition, the vertical profile of the flow velocity at the northeast corner of the Tower was also measured to study the three dimensional structure of the strong wind fields.

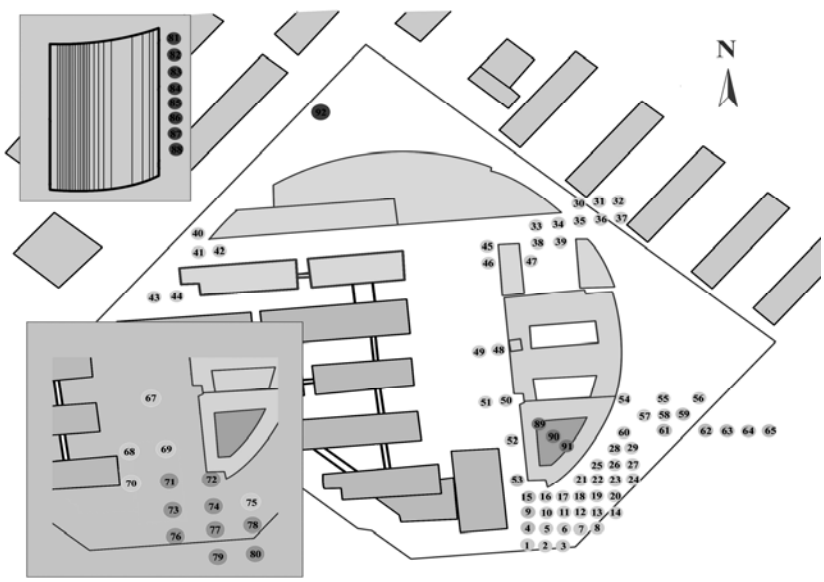


Figure 2 The positions of the measurement points in wind tunnel. In the upper-left the measurement points in the vertical at the point 54 at the corner of the tower, in the lower-left that with the Forum building pulled down.

### 1.2 Results of the measurement

In the following we focus our attention on the strong wind area in the south side of the Tower. Figure3 shows the distribution of the wind speed at the measured points. The maximum wind speeds appear at the southwest corner. The strong wind was composed of the corner flow caused by the south end of the Tower and the channel flow between the Tower and the Forum Hall. Parameter *R* is taken as the wind speed amplification factor,

$$R(x, y) = U(x, y) / U_0 , \qquad (2)$$

where $U_0$=5.4m/s is the speed of the approach flow( at 4m above the land surface), $U(x, y)$ is the speed measured at point $(x, y)$. The maximum *R* with value of 2.1 appears in the narrow channel between the Tower and the Forum Hall. The *R* value of 2.1 means that the speed in the strong wind area is two times of the approach one. Figure 4 shows the wind speed distribution after the model of the Forum Hall moved away. Without the Forum Hall the channel effect was removed. However, without the blockade of the Forum Hall the corner flow area extended. The strong wind with *R* values over 2 covered larger areas than that in Figure 3 at the south end of the Tower.

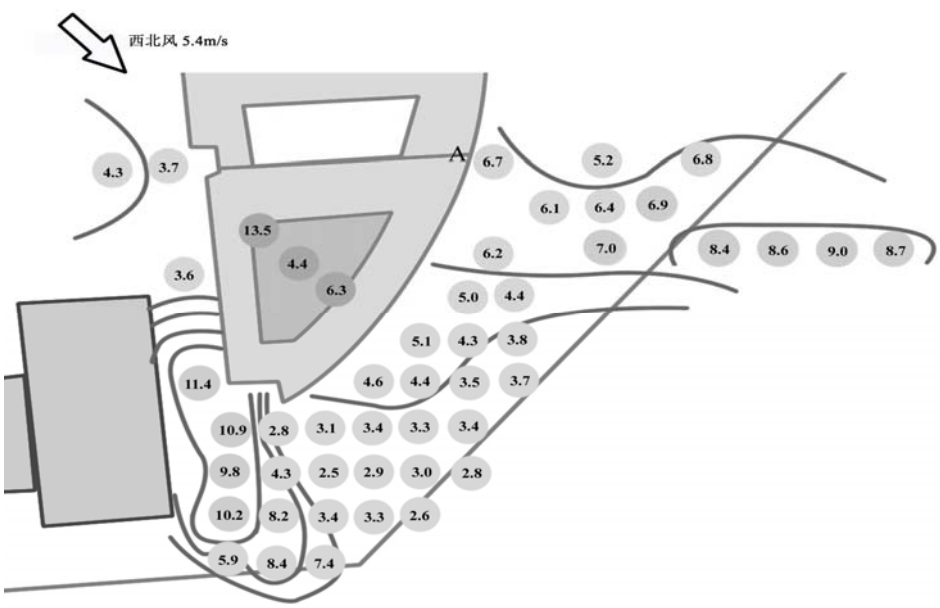


Figure 3 The distribution of speed measured by the hot wire in wind tunnel m/s .The measured height is at 2.67cm, while is at 4 m in the real atmosphere. The in flow is 5.4m/s.

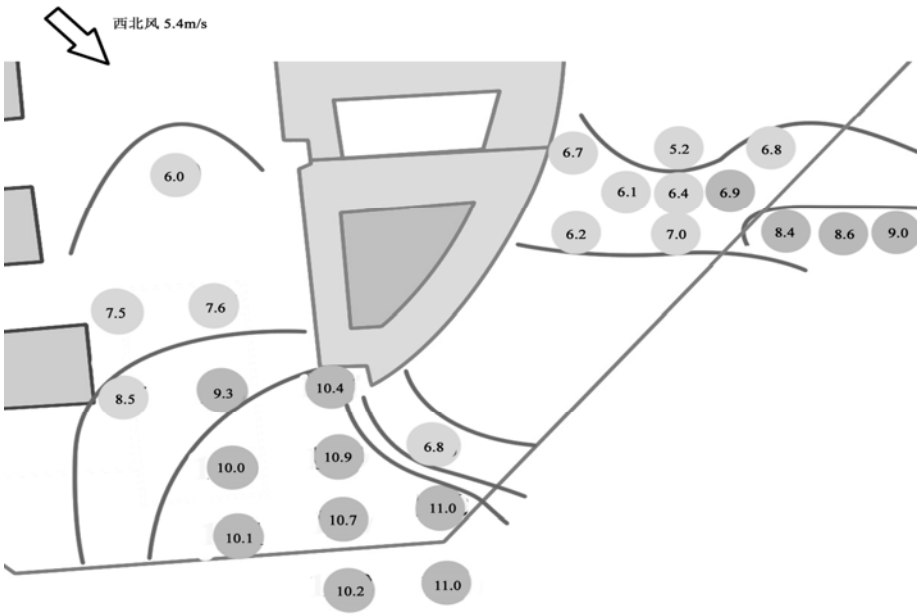


Figure 4. The same with figure 3, but without Forum building.

## 1. Application of scouring technique

The pedestrian wind field measurement techniques in wind tunnel can be divided into two categories. One is the instrumental measurement, for instance the hot wire measurement described in last section. The instruments for the measurement also include hot film, anemobiagraph, omni directional anemometer, etc. Enough quantity and density of measurement points are needed in the method in order to obtain a full view of wind fields. Another way of the pedestrian wind field measurement is to provide an overall, spatially continuous description of wind conditions at pedestrian level. Scouring technique is one of these methods ( Livesey et.al ,1990). A thin layer of particles was spread uniformly over the ground surface of the wind tunnel around the models of the building. By increasing the wind speed of the wind tunnel the particles in strong wind area were blew away and the scour patterns were formed.

In the study the flaky bran with size of 0.25-0.3 mm was used as the material of particles. The brawns put in oven to remove the moisture, resulting in cohesionless particles. As wind speed was gradually increased to a threshold velocity $V_t$, the particles started to move. By repeating the test the mean threshold velocity was obtained $\overline{V_t}$ =2.7m/s. In the study four grades of wind speed were set, that is, $V_r / V_t$ =0.55, 0.69, 0.76 and 0.85, respectively.

At the first grade the approach wind speed should be $V_r = 0.55 V_t$ =1.5m/s. As the

speed was kept constant in 2-3 minutes, a scouring pattern appeared in the vicinity of the models. This means that the wind speed enclosed in the sour area has exceeded the threshold velocity (2.7m/s). The ratio of the wind speed within the enclosed area to that of the approach flow is $R = V_t / V_r$ =1.82. By analyzing the photograph of the scouring area the envelope of strong wind area of R=1.82 was obtained. By increasing the test speed successively and repeating the procedure described above, the envelops of the strong wind areas for different grades of *R*=1.82,1.54,1.32 and 1.18 were obtained, respectively. Figure 5 shows the envelopes of strong wind areas for different grades in northwesterly. Figure 6 is the same as Figure 5, but without the Forum Hall. Comparing those two figures we may find that the area of strong wind will extend and the wind speed will strengthen if the Forum Hall is pulled down.

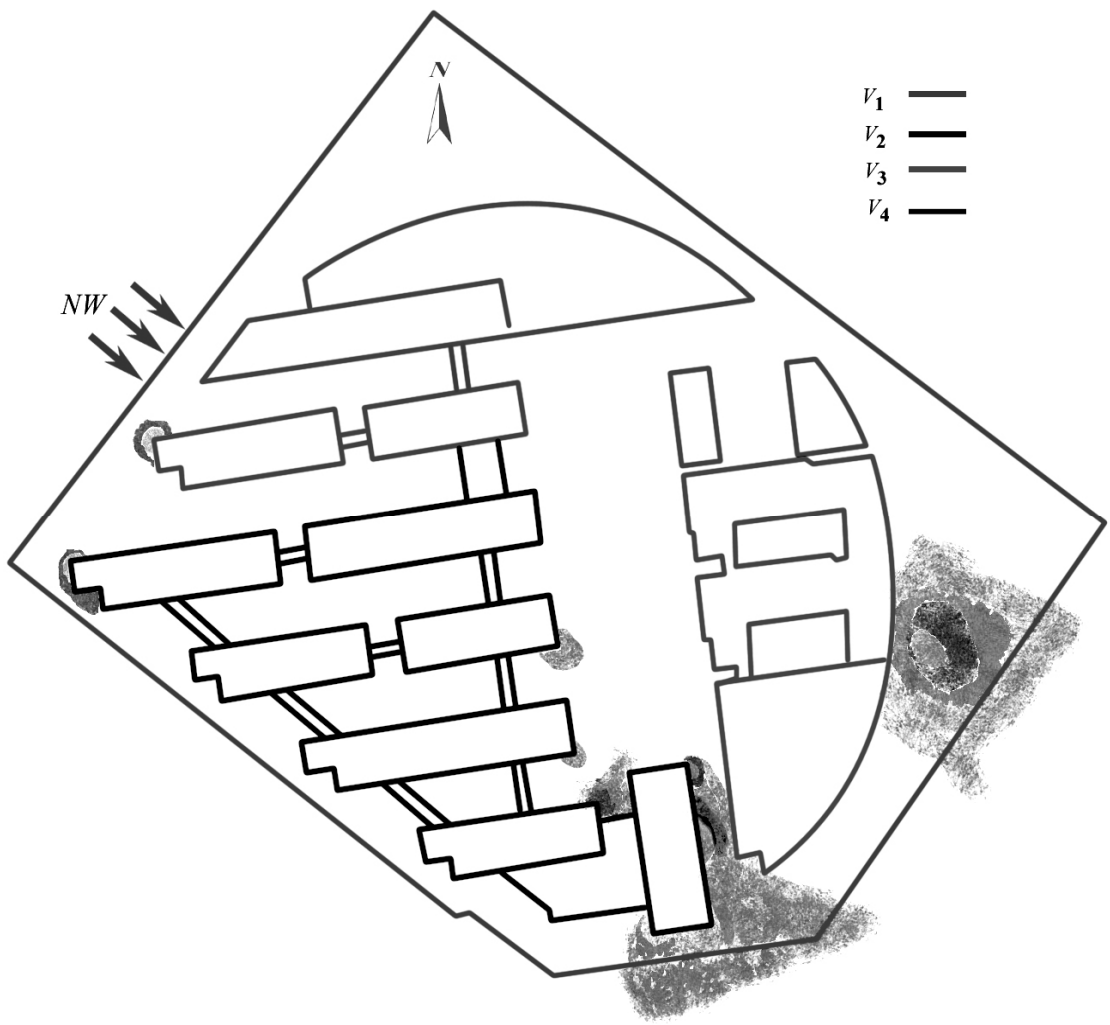


Figure 5 Contour of wind speedup ratios for the NW wind. V1 to V4 representing $R = 1.82$ 1.45 1.32 and 1.18 respectively, while $V_1 = 0.55V_t$ $V_2 = 0.69V_t$ $V_3 = 0.76V_t$

$V_4 = 0.85V_t$ ; and the threshold wind speed $V_t = 2.7m/s$.

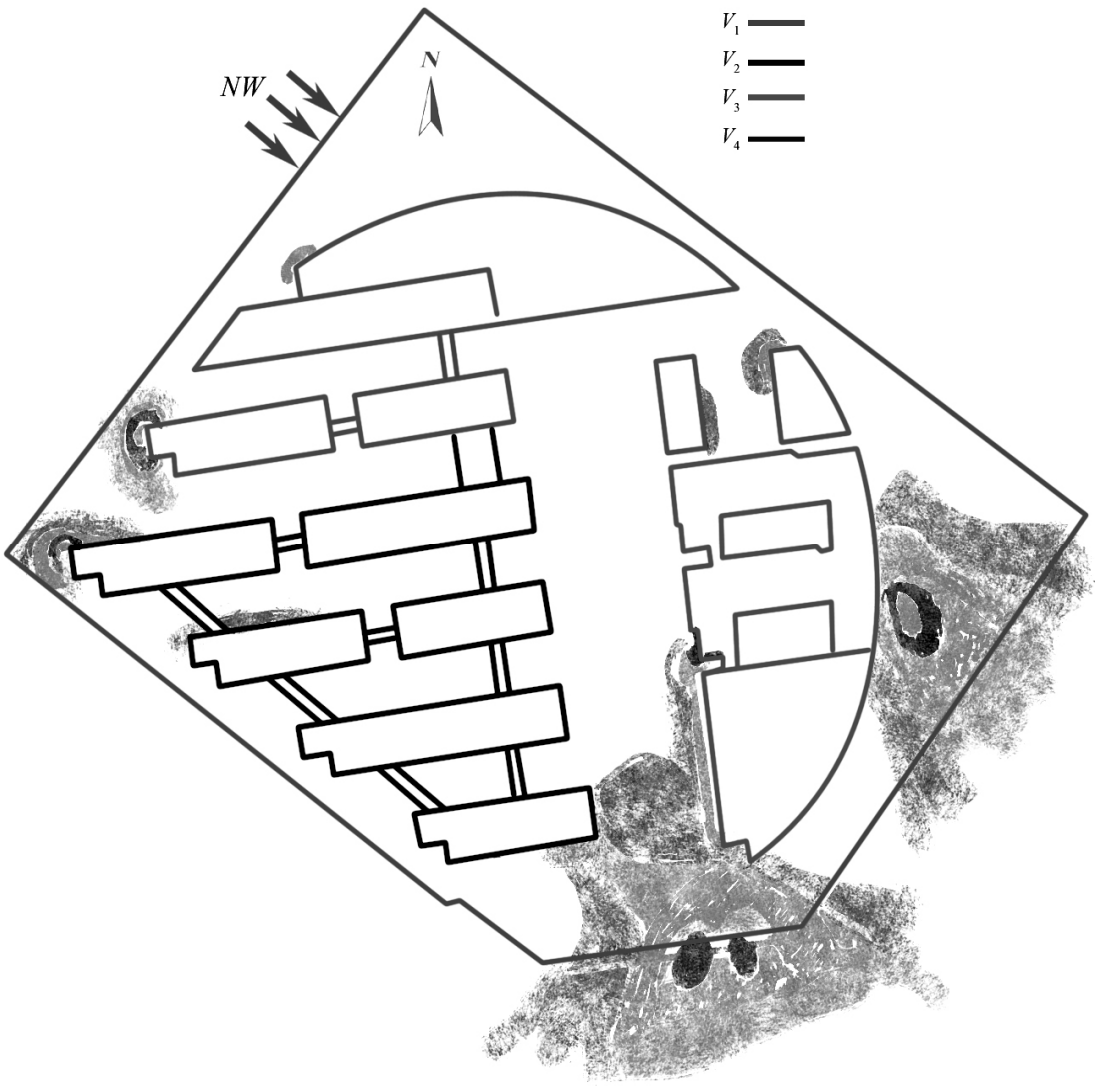


Figure 6 Same as figure 6. Contour of wind speedup ratios for the NW wind. The forum building pulled down, V1 to V4 representing $R = 1.82$ 1.45 1.32 and 1.18 respectively.

## 2. Numerical computation

The numerical simulation is one of the important methodologies in science research. It has developed rapidly in the latest decade that the method of calculating flow around blunt body in computational fluid dynamics applied to simulation of flow around buildings in the atmospheric boundary layer. The computational fluid dynamics and the computer capability have been developed so fast in recent years that numerical simulation can now be made more economically and yield more complete and equally as accurate results, and its results have been verified by the wind tunnel and observation data (Jones and Launder, 1972,Sang et. al, 2002, Wang et. al, 2003, Wang et.al., 2004).

In numerical computation two wind directions, the northwesterly, the prevailing wind in winter and the southwesterly in summer are chosen as the initial and the inflow boundary conditions. In the following only the northwest wind is discussed. Two kinds of computational domain are used. Domain A is 360m×320m with grid interval of $\Delta x = \Delta y = \Delta z$ =4m, including the planning S Building and the surrounding buildings, while domain B is 150m×210m with $\Delta x = \Delta y = \Delta z$ =2m, including only the S Building and the Forum Hall.

Figure 7 shows the horizontal flow fields of domain A at level of 4m in northwesterly. It can be seen that there are two strong wind areas in the vicinity of S Tower. One at the east side of the Tower is caused by the corner flow of the northeast corner of the Tower. The east side is planned to be green spot and the driveway, where is no sidewalk for pedestrian. Therefore the strong wind there will not affect much the pedestrians. Another strong wind at the south side of the Tower is composed of the corner flow by the south corner of the Tower and the channel flow between the Tower and the Forum Hall. Since there is the main entrance of the Building at the south end of the Tower, the strong wind there may make the pedestrians uncomfortable. Therefore the mechanism and structure of this strong wind area must be carefully studied.

Figure 8 shows the results in domain B, in which the resolution is higher, so that the structure of the wind field can be investigated in detail. The speedup ratio parameter R is taken as $R(x, y) = U(x, y, h)/U_0$, where $U(x, y, h)$ is the wind speed at h=4m in grid point of $(x, y)$, $U = (u^2 + v^2)^{1/2}$. $U_0$ is the approach flow speed at z=4m.

In the northwesterly the huge pressure difference between the windward side and the leeside of the Tower induces strong pressure gradient force to make the flow speedup. As the Forum Hall exists its blockade effect reduces the intensity of the flow on the windward side of the Tower, and then decreases pressure difference and the pressure gradient force, diminishes the corner flow. If the Forum Hall is pulled down the blockade effect is removed and thus the corner flow strengthens. Then the strong wind area increases and extends southward.

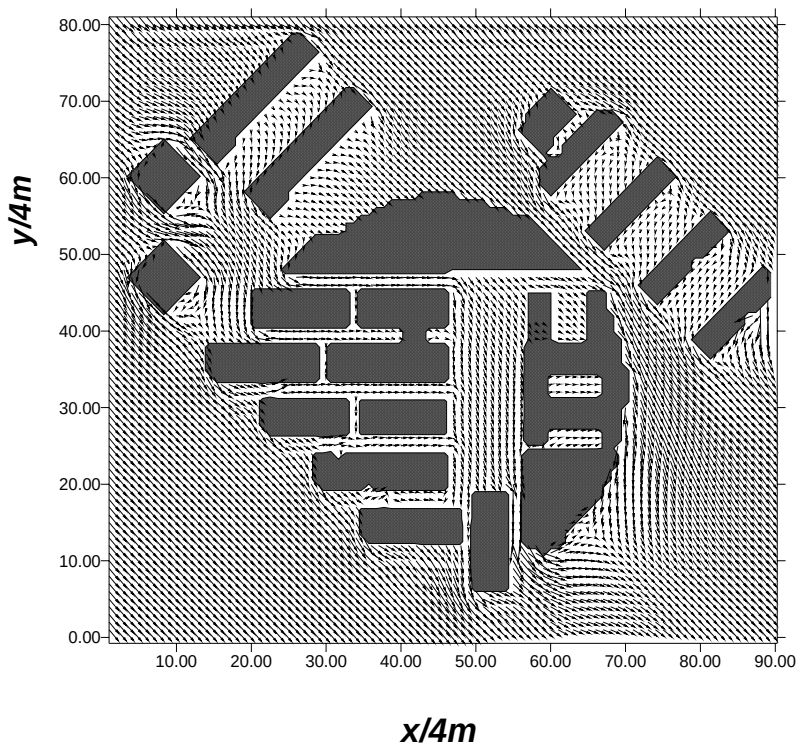


Figure 7 The computed wind fields of the northwester at 4m above the land surface in domain A. The coordinates are the numbers of grids. The grid interval is 4m.

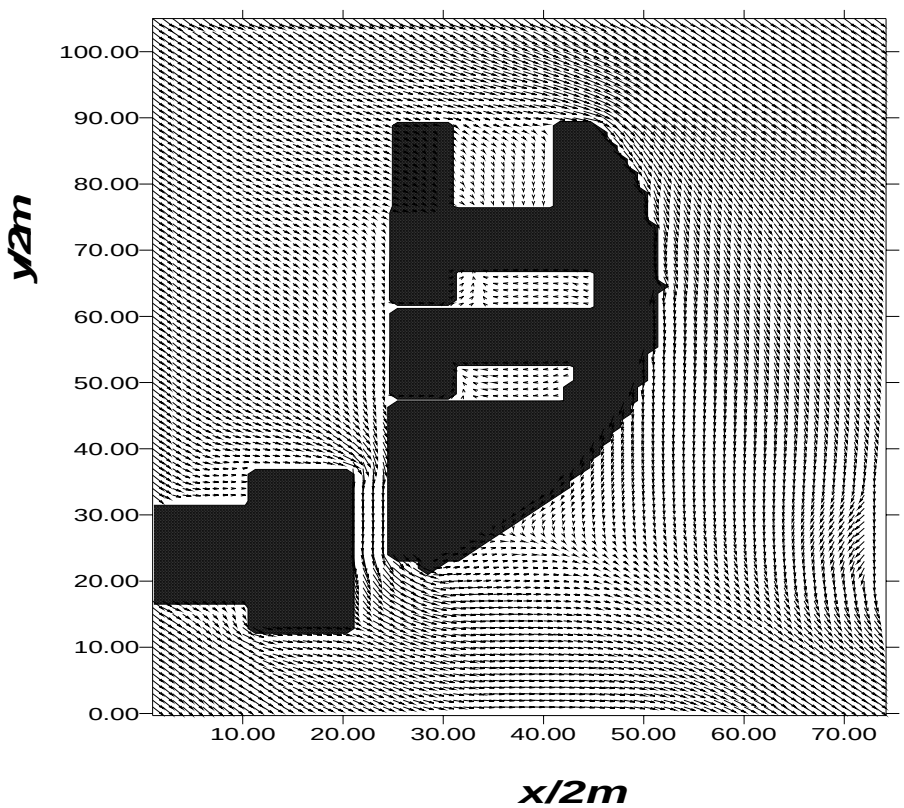


Figure 8 The computed wind fields of the northwester in domain B. The grid interval is 2m.

## 3. Discussion and summary

In the present studies numerical computation, hot wire measurement and scouring technique in wind tunnel were carried out simultaneously to study the wind fields around a designed building to seek for a better way for pedestrian wind field assessment. The studies show that the conclusions by the three approaches are in agreement with each others and each approach has its own advantages.

The souring technique can provide an overall view for a large area wind environment. It can be used for the wind environment assessment of a single tall building, and it demonstrates special advantages for the assessment of a large scale architectural complex.

Using the hot wire measurement we can obtain the wind fields of a whole evaluated area, we can also carry out a detailed survey of the concerned areas, for example, the open space of busy pedestrian activities, or a possible strong wind area, etc. In addition, we can make three dimensional measurements to study the structure and the cause of formation of the strong wind.

Numerical computation is a more flexible method. Using different grid interval we may have the ground surface or spatial wind fields within different scale computational domain. Furthermore, by using nested grid or varied grid the assessment may vary from a large scope to a small one, and from coarse resolution to fine one.

As described above, a correct assessment can be made independently by each approach.. However, by using two or more approaches, the assessment would be more accurate and more reliable, and the procedure would be more effective. The appropriate methods provided in the studies guarantee the scientific basis to improve the wind environment in the vicinity of a tall building and provide a comfortable open space for the pedestrians.

In the fast development of urbanization how to improve the living quality of the urban residents is of great concern. The wind environment of urban tall buildings will attract more and more attentions. The methods of assessment described above will be further tested and perfected.

## 5. ACKNOWLEDGEMENTS

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# BLOOD-LEAD MONITORING EXPOSURE TO LEADED-GASOLINE AMONG SCHOOL CHILDREN IN JAKARTA, INDONESIA 2005

**Budi Haryanto**

Center for Health Research, University of Indonesia, Dept. of Environmental Health, School of Public Health, Jakarta – Indonesia bharyant@cbn.net.id

## ABSTRACT

In Indonesia, except Jakarta, lead (Pb) is still being used in many products especially as octane booster for gasoline. It means that the negative effects of lead, such as: decreasing IQ, decreasing hearing, decreasing growth, and decreasing hemoglobin content are still occurring and will continue in the future. Meanwhile, the longer lead exposure occurs in the environment more dangerous the health effects especially to the children. The Jakarta blood-lead study in 2001 showed that 35% elementary school children have blood lead level (BLL) more than 10 μg/dl, and 2.4% had BLL more than 20 μg/dl (CDC-USAEP 2001).

The objective of this study is to assess the blood lead level patterns of elementary school children in Jakarta urban area in 2005, with the aim to evaluate mean of blood lead levels of 3$^{rd}$ and 4$^{th}$ grade elementary school children in Jakarta urban area. A cross-sectional survey was implemented.

The study was conducted on January to February 2005. The equipment of portable LeadCare and Hemocue were used for analyzing blood-lead and blood hemoglobin (Hb) respectively from children finger blood on site. All of the 20 selected elementary schools agreed to participate the test. A total of 203 students allowed by their parents participate the test. About 5% of the blood samples could not be analyzed. The overall average for Hb-blood is 12.6 mmHg and for Pb-blood is 4.2 μg/dl. Percentage of those children with Pb-blood equal and more than 10 μg/dl is 1.3%.

The declining Pb-blood prevalence from 2001 study is obviously shown the success of Leaded-gasoline phase-out program in Jakarta. It means that the program is success in preventing people exposed to leaded-gasoline in Jakarta, especially to the children from the risk of lead toxic impacts. Thus, the program of phasing-out of leaded-gasoline must be expanded and implemented to all of Indonesia provinces and cities in order to protect lead exposure to people.

**Key Words**: Air Quality, Blood-lead levels, Health Effects

# 1. INTRODUCTION

Rapid urbanization and industrialization in Indonesia have created severe air pollution problems, particularly in its major cities. The United Nations Environmental Program ranks Jakarta as the world's third most polluted mega-city, after Mexico and Bangkok. Vehicular traffic emissions are obvious and major sources of air pollution. An ADB-funded regional Technical Asisstance[1] estimated that vehicle emissions in Jakarta amount to approximately 71% of the oxides of nitrogen ($NO_x$), 15% of sulfur dioxide ($SO_2$), and 70% of particulate matter ($PM_{10}$)[2] in the total emission loads, and the annual economic cost of the associated health problems will be about $450 million by 2015. The World Bank study[3] estimated health damage due to all sources of air pollution in Jakarta at approximately $300 million per year in 1990, and this was projected to increase tenfold by 2010 unless serious control efforts were implemented. Total population in DKI Jakarta is expected to grow from approximately 9.5 million in 1998 to 11 million in 2005, and to 13 million in 2015.

In Indonesia, except Jakarta-the capitol city, lead (Pb) is still being used in many products especially as octane booster for gasoline. Means that the negative effects of lead, such as: decreasing IQ, decreasing hearing, decreasing growth, and decreasing hemoglobin content are still occur and will continue in the future. The efforts to eliminate effects of environment lead exposure from gasoline had been starting in Jakarta City by replacing leaded gasoline to unleaded gasoline since July 1$^{st}$, 2001 and will continue to all of provinces in Java Island (the most populated island with about 60% of Indonesia population) in the year of 2002, and finally to all of Indonesia provinces by the year of 2003 for the first scenario, and by the year 2005 for the last scenario. However, the plan seems very difficult to be implemented on time due to political issues and lack of government willingness rather than the economic problems. Meanwhile, the longer lead exposure occurs in the environment more dangerous the health effects especially to the children will be.

The human health impacts of exposure to environmental lead have been extensively researched overseas for many years as well as the respiratory diseases caused by air pollution. Progressive studies have shown these impacts to occur at progressively lower levels of exposure, such that it is now considered by the overwhelming majority of health scientists that there is no threshold level below which these effects do not occur. It is regarded to be one of the most serious health problems facing populations, particularly children. Common symptoms include IQ loss, reading and learning difficulties, hearing loss, difficulties in concentration, adverse on kidney function effects, blood chemistry, and the cardiovascular system as well as adverse reproductive effects for women. The negative impacts of lead pollution on human health are well documented. The most usual indicator of human exposure to environmental lead is the amount that can be measured in blood. The World Health Organization has set a standard for the maximum acceptable blood lead level at 20 micrograms per deciliter and the Center for Diseases Control and Prevention USA suggested children blood lead level is not exceed of 10 micrograms per deciliter.

However, more recent studies have shown measurable and chronic health effects at much lower levels.

Exposure is primarily caused by airborne lead. In congested urban areas, exhaust fumes from vehicles using leaded gasoline typically account for some 90 percent of airborne lead pollution.

- In pregnant women, blood lead concentrations of 10 μg/dl have been associated with potentially adverse development of fetuses and newborn infants.
- In children, blood lead concentrations of 10 μg/dl can cause significant IQ decrement and lower intellectual achievement; can slow down growth rate; and can affect hearing.
- In adults, blood lead concentrations of 10 μg/dl can increase hypertension and heart disease.
- Other health problems include: damage of haemopoietic system, damage of renal system, disturbance of cardiovascular system, neurobehavioral effects, abortion, still-birth, neonatal death, disturbance of immunity mechanism, carcinogen, and so on.

Several studies related to blood-lead level conducted in Jakarta found that: the average of Jakarta public bus drivers' blood-lead was 24.6 μg/dl in 1978 (Achmadi et al, 1978). A 1991 study of blood lead concentrations in Jakarta residents showed such levels to be typical ranging to over than 30 μg/dl in some sections of the population. In particular, 74% of 66 slum dwellers, who live and work in heavily trafficked areas, had blood lead level >30 μg/dl. The 2001 blood –lead study among elementary school children showed 35% had blood lead level (BLL) more than 10 μg/dl, and 2.4% had BLL more than 20 μg/dl (CDC-USEPA 2001). Other studies in Bandung city show similar trends. They are: 30-46% of policemen and public transportation drivers having blood-lead above 40 μg/dl (Djuangsih 1984), and 50% of street vendors having blood-lead above 40 μg/dl (Haryanto, 1993). It must be concluded, therefore, that significant chronic health impacts are caused in many sections of the population in Jakarta, by chronic exposure to environmental lead, and that the major source is lead in gasoline.

The lack of evidences from epidemiological studies, concerning lead and dust air pollution to people's health effects in Jakarta, is believed as a potential reason that may affects to the lack of awareness and willingness of public policy decision makers to develop appropriate strategy for preventing Jakarta population from the hazards of air pollution. Therefore, the study providing information in epidemiological evidences of children health effects caused by air pollution in Jakarta is needed and very important to be conducted in the near future.

## 2. OBJECTIVES

To assess the blood lead level patterns of elementary school children in Jakarta urban area 2004-2005;

## 3. METHODS

**Study Design**
A ***time series cross-sectional survey*** implemented for assessing the blood lead level patterns of school children in Jakarta city and determining changes between 2001 and 2005.

**Study Population**
The study population will be 3rd and 4th grade children who attend school in Jakarta. Children will be studied because they are the age group that is most vulnerable to health damage from lead and air pollution dust. It is also not feasible to examine all primary school grades because of time and logistic considerations.

**Location**
For blood-lead study, 4 elementary schools will be selected randomly at surroundings of every single 5 monitoring stations near the roads in Jakarta urban area. The 5 stations are located in: Jasa Marga Office (East Jakarta), EMC-Puspiptek Serpong (Banten), Taman Anggrek Lebak Bulus (South Jakarta), Hotel Indonesia (Central Jakarta), and University of Trisakti (West Jakarta).

**Sampling design**
Given the size of Jakarta in terms of both population (~12 million) and geography, a simple random sample at the household level was not considered to be logistically feasible. Therefore, a cluster survey design was used for the cross-sectional study. Clusters were defined as elementary schools in five selected air monitoring stations in Jakarta. Four elementary schools nearest every single of the monitoring station are assigned as clusters. Thus, 20 elementary schools will be the source of samples (children). An equal probability random sample of 10, 3rd- and 4th-grade children in each of the 20 schools is selected for inclusion in the study. The number of sample will be 200 elementary school children.

**Cross-sectional sampling calculation**
Sample sizes were calculated to provide a large enough sample so that the margin of error around the mean (95% CI) was ±10% accounting for clustering. Sample sizes were derived using the following equation:

$$n = \frac{(z)^2 (SD)^2 (1 - IC)}{(\%ME * E\bar{x})^2 \left( (1 - (IC)(z)^2 (SD)^2) / (\%ME * E\bar{x})^2 (\#cluster) \right)}$$

$$= \frac{(2.8)^2 (2.8)^2 (1\text{-}0.3)}{(0.1 \times 8.3)^2 \times (1\text{-}(0.3)(2.8)^2 (2.8)^2 / (0.1 \times 8.3)^2 (20)} = 189$$

z : z-score from previous study
SD : standard deviation from previous study

IC : intracluster correlation (No data were available from the Jakarta study to determine the intracluster correlation. Therefore, a conservative estimate that assumes relatively high clustering was used for intracluster correlation)
ME : margin of error
Ex : expected mean

From calculation above, the number of sample is 189. In order to enhance more validity for the data, we decide to increase the number of sample up to 200.

**Data collection**
There were 5 study teams of 6 people each (30 people total): Each team includes the following: 1 phlebotomist, 3 interviewers, 1 person to handle all paperwork (consent forms, results sheets, labeling of blood sample), and 1 CHR-UI (Center for Health Research-University of Indonesia) supervisor.

Consent forms and questionnaires distributed to school principals in the selected schools prior to the beginning of the study. The principals asked to send the forms home with the children so that the parents can fill them out and return them to the schools with their children before the CHR-UI team arrives.

The blood draws are performed by the phlebotomist on the study team who was one of the local fieldworkers. The procedure for collecting the blood sample for lead was the following:

- The child's hand is washed with soap and water, dried with a paper towel, wiped with an alcohol prep swab and dried with gauze.
- The finger is punctured with a sterile, non-reusable Tenderlett lancet.
- The first drop of blood is wiped away with gauze.
- 50 ml (about 4 drops) of blood is collected in a capillary tube for lead analysis.
- After the blood is taken, a band-aid is applied for further protection of the puncture site.

Letters are generated to inform parents of their children's blood-lead test results and these will be given to each school principal for distribution 1-3 days after the sample has been collected. The blood samples are tested for lead using the LeadCare portable analyzer (ESA Laboratories, Chelmsford, MA, USA).

The procedure involves placing the sample into a reagent tube and allowing it to rest for at least 1-2 minutes, followed by placing the sample onto the LeadCare electrode. The analyzed result is available in 3 minutes. Because the upper end of the operational range for the LeadCare analyzer is $95^{o}$F and ambient temperatures in Jakarta was higher, the CHR-UI team analyzed the bloods in air conditioned rooms in the evenings. Blood specimens do not stored after analyses are completed. Instead, all excess bloods and used materials (lancets, tubes, swabs, wipes, and gloves) are collected in biohazard bags and taken for biohazard disposal.

**Duration of the study**
The duration of the study is 24 months and started at January 2004 to December 2005, including study preparation, materials development, field preparation, data collection, data analyses, interim report writing, report improvement, and seminar of finding. In general, the study divided into two terms, year-1 of study and year-2 of the study. Comparison between the study years will also be generated.

**Organization**
The study executed by researcher team from the Center for Health Research – University of Indonesia in collaboration with the Department of Environmental Health - School of Public Health – University of Indonesia and the Environmental Monitoring Center – Sarpedal/Bapedal – the Ministry of Environment. The study is fully supported by JICA-DEMS (Decentralized Environmental Management System) and United States Asia Environmental Partnership (USAEP) – USAID.

## 4. RESULTS

This study was conducted in January to February 2005. All of the 20 selected elementary schools agreed to participate the test. Totally, a number of 203 students (third and fourth graders) are allowed by their parents participate the test. The participation rate is 100%. Unfortunately, about 5% of the blood samples (11 students) could not be analyzed. The results in detail are as the followings:

**Serpong, Tangerang site**

The overall average for Hb = 12.8 mmHg
The overall average for Pb = 9.0 μg/dl
**Percentage of those children with Pb-blood equal and more than 10 μg/dl is 26.8%.**

**DKI Jakarta site**

The average Pb-blood in Jakarta Selatan = 5.1 μg/dl
The average Pb-blood in Jakarta Timur = 4.4 μg/dl
The average Pb-blood in Jakarta Pusat = 4.6 μg/dl
The average Pb-blood in Jakarta Barat = 2.4 μg/dl

| Variable | Obs | Mean | Std. Dev. | Min | Max |
|---|---|---|---|---|---|
| hb | 161 | 12.56 | 1.10 | 8.8 | 15.6 |
| pb | 151 | 4.15 | 1.67 | 0.0 | 11.2 |

The overall average for Hb in DKI Jakarta = 12.6 mmHg
The overall average for Pb in DKI Jakarta = 4.2 μg/dl

**Percentage of those children with Pb-blood equal and more than 10 μg/dl is 1.3%.**

**Comparison with 2001 Pb-blood study:**
Albalak et.al. of CDC-USAEP Pb-blood study among 396 elementary school children, in DKI Jakarta in 2001, found that geometric means of Pb-blood children is 8.6 μg/dl and about 35% of the children having Pb-blood levels above 10 μg/dl. With almost same methodology and design study, our findings obtain Pb-blood means of 4.2 μg/dl and 1.3% children have Pb-blood levels above 10 μg/dl. This declining Pb-blood prevalence is obviously shown the success of Leaded-gasoline phase-out program in DKI Jakarta. It means that the program success in preventing people in Jakarta, especially children from the risk of the dangerous of leaded-gasoline. Thus, the program of phasing-out of leaded-gasoline must be expanded and implemented to all of Indonesia provinces and cities in order to protect lead exposure to people.

## 5. ACKNOWLEDGEMENTS

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# URBAN AIR QUALITY MANAGEMENT IN INDIA (WITH SPECIFIC EMPHASIS ON PARTICULATE MATTER)

**Sumit Sharma[1], T.S. Panwar[2]**

[1] Research Associate, Center of Environmental Studies, TERI, India Habitat Centre Lodi Road, Delhi, India
[2] Fellow, Center of Environmental Studies, TERI, India Habitat Centre Lodi Road, Delhi, India

## ABSTRACT

Urban air quality is an issue of major concern across many cities and towns in the developing countries, including India. In particular, high levels of particulate matter (both SPM and RSPM) are responsible for non-compliance against air quality standards. This paper suggests various air quality management options that can be applicable to major urban centers which are at present most affected by the deteriorating air quality. As an example, Pune city is chosen, which is a top-ten urban agglomerate in India (based on population). Analysis of ambient air quality is carried out and the causal factors for poor air quality are determined. In order to highlight the gravity of the problem, the health impacts due to exposure to particulate matter emissions in Pune city are estimated, and economic evaluation of health damage is done. Sectoral emission loads are estimated for transport, industrial, and domestic sectors, which provide an estimate of the major contributors to air pollution with specific reference to particulate matter, which is a major pollutant of concern. A detailed scenario analysis is carried out to estimate the changes in emissions that would take place due to various interventions. On the basis of the above exercise, an air quality management plan is developed that specifically accounts for factors contributing to deterioration of air quality in Pune. This overall assessment of the air quality issues for Pune can provide useful insights for the development of the air quality management plan for other cities as well.

**Key Words:** Urban Air Quality Management, Particulate matter, Emission Loads, Health impacts, Scenario analysis

## 1. INTRODUCTION

Environmental quality is rapidly becoming a major issue in urban India. Indian cities suffer from some of the worst air quality problems in the world, which continue to be a major public health issue. Urban air pollution is largely a result of combustion of fossil fuels for transportation, power generation, industrial and other economic activities. Major contributing factors for urban air pollution are: 1) Increased migration of population towards urban areas, 2) Industrial development & higher levels of energy consumption, 3) Increasing traffic & related issues such as poor inspection & maintenance, congestion and outdated vehicular technology, and 4) poor quality fuels.

**Highly Polluted Indian Cities**

Delhi, the capital of India, itself is one of the most polluted city in the country. The trends of air pollutants: sulphur dioxide ($SO_2$), nitrogen oxides ($NO_x$), suspended particulate matter (SPM), and respirable suspended particulate matter (RSPM) are shown in Figure 1 for the years 1995 to 2004. With the intervention of Honourable Supreme Court, various strategies have been implemented to abate air pollution levels in the city. Though, $SO_2$ concentrations have come down significantly, but no substantial decrease is observed in the particulate matter concentrations. On a positive note, the SPM and RSPM concentrations have not increased despite the increased activity levels in the city. While $SO_2$ and $NO_X$ are within the permissible limits, the SPM and RSPM concentrations have violated the ambient air quality standards in most of the years.

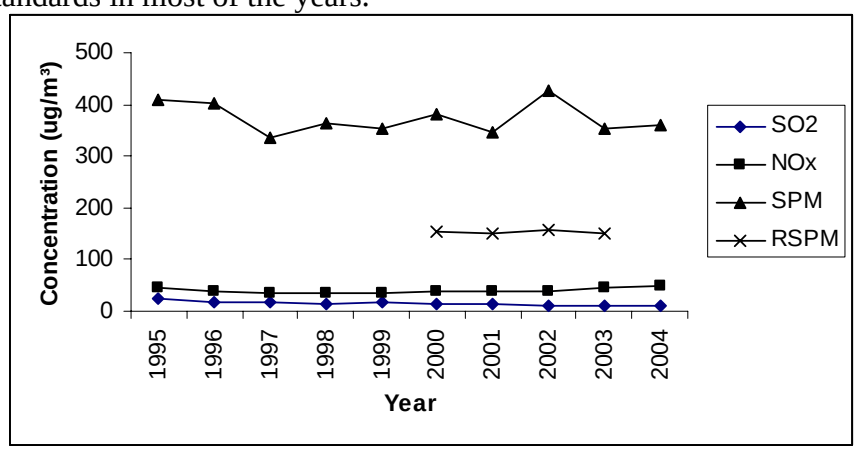


Figure 1: Trends of annual average $SO_2$, $NO_x$, SPM, and RSPM concentrations (average for all the stations) in Delhi for the years 1995 to 2004
Source : CPCB, 1997, 1998, 1999, 2000, 2001, 2002, 2004a, 2004b, 2005

The Honourable Supreme court of India ordered for preparation of scheme with regard to improvement of the air environment in 16 cities other than Delhi, which are equally, or more polluted. The cities are Agra, Ahmedabad, Kanpur, Solapur, Lucknow, Bangalore, Chennai, Hyderabad, Mumbai, Kolkata, Pune, Faridabad, Jharia, Jodhpur, Patna, and Varanasi. The brief status of ambient air quality in these cities for the period 1995 to 2003, is described in following sections.

**$SO_2$ :** $SO_2$ levels in all the selected cities are under permissible standards except in Jharia where $SO_2$ concentrations violated the standards in 1998/99. In 2003, the $SO_2$ concentrations in the cities were even less than 20 $\mu g/m^3$, which is one third of the conservative residential area standards. Among all the 16 cities, $SO_2$ levels were highest in Jharia, Faridabad, Kolkata, and Lucknow.

**$NO_x$ :** $NO_x$ levels in most the selected cities are under permissible standards. However, Kolkata, Pune, and Jharia have registered $NO_x$ levels exceeding the annual average standards. On a positive side, the $NO_x$ concentrations in all the other cities were less than the conservative residential area standards. Trends of $NO_x$ concentrations are increasing in Pune, Kolkata and Jharia.

**SPM :** SPM emerges out as the pollutant of concern for our urban centers. Almost all the cities violated the residential area standards and cities like Kanpur, Agra, Lucknow, Patna, Jharia, and Jodhpur have even violated the less stringent industrial

area standards many times. Chennai stands out with hardly violating even the residential area standards. On a positive note, trends are decreasing in Kolkata, Ahmedabad, Patna, Pune, and Jharia while they are increasing in Kanpur, Solapur and Jodhpur.

**Case- Pune City**

To simplify and to arrive at problem specific unique air quality management plan we chose Pune as the representative city among all the urban agglomerates. Pune, the second largest city in Maharashtra, has a population of 3.75 million people (PMC, 2003). Few important transformations of the post independence period in Pune are marked by: 1) Rapid growth of large industries in all peripheral areas, 2) Growth of new National level institutions, 3) Spilling over of residential development across the Municipal Corporation limits in all directions. Some of this spill bears a resemblance to planned development and a large part being in the form of unauthorized development.

- **Air Quality in Pune**

Annual average $SO_2$ concentrations across the Pune city have varied from 22-54 µg/m³ during 1995 to 2003, but remained under the prescribed residential area standards. While annual average NOx concentrations have varied from 23-78 µg/m³ and violated the residential area standard few times. Annual average SPM concentrations have violated the residential area standards in all the years and have shown increasing trend during the years 2000 to 2003. RSPM monitoring started in the year 2000 and the annual average RSPM concentrations have followed an increasing trend thereafter and also violated the industrial area standard during the years 2002-03. (Figure 2)

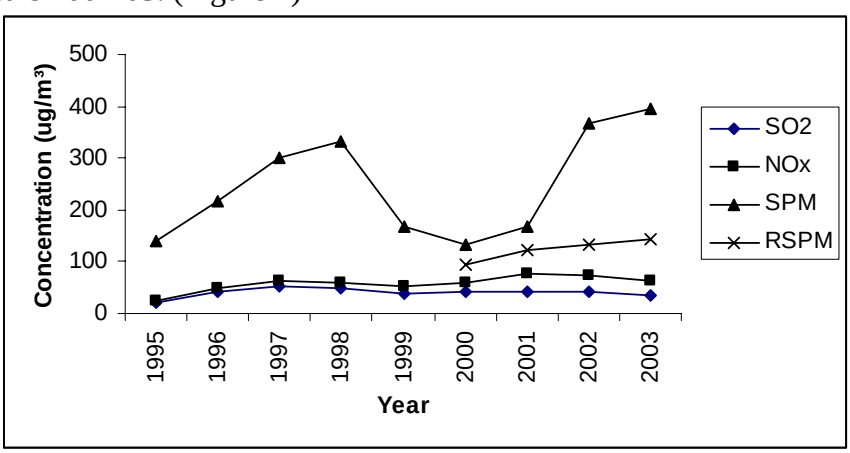


Figure 2: Trends of annual average $SO_2$, $NO_x$, SPM, and RSPM concentrations (average for all the stations) in Pune for the years 1995 to 2003
Source : CPCB, 1997, 1998, 1999, 2000, 2001, 2002, 2004a, 2004b, 2005

- **Health Impacts And Economic Damage**

To highlight the enormity of problem, the health impacts due to exposure to respirable particulate matter emissions in Pune are estimated. The estimation exercise started off with the estimation of the population exposed to the pollution. The figures for the population of Pune city are collected from Census India 2001 with categorizations of slum and non-slum, by gender and age groups. Health impact

coefficients (Ostro B.,1994) are used to arrive at health effects due to change in concentration of the particulate matter. These effects are then monetized using the monetary cost attached with unit health state (Brandon and Hommann (1995). The results are presented in Table 1, which reveals that due to the annual average RSPM concentration of 141 µg/m³ in the year 2003-04, which is well above the earlier suggested WHO guideline value of 40µg/m³, nearly 6633 mortalities are expected to occur which amounts to an economic damage of nearly 977 millions. Also, economic damage of nearly 966 millions is expected to occur due to restricted activity days.

Table 1: Health impacts and economic damage due to RSPM concentration in Pune in 2003-04

| **Outcome** | **Number** | **Economic damage (Million Rs)** |
|---|---|---|
| Mortality | 6633 | 977 |
| Hospital admissions | 5362 | 17 |
| Emergency room visits | 105186 | 23 |
| Restricted activity days | 14210532 | 966 |
| Lower respiratory illness in children | 337493 | 1 |
| Asthma Attacks | 1019688 | 14 |
| Symptoms of respiratory disorder | 81771618 | 164 |
| Chronic Bronchitis | 27347 | 7 |
| **Total (Millions)** | | 2169 |

- **Causal Factors**

*Population growth:* Pune city has grown at moderate rates in the post independence period with decadal growth rates ranging from 7 to 102 percent over the last eight decades. Pune urban population has grown from 17.35 lakhs to about 26.97 lakhs between 1991 and 2001 (Census of India, 2001). Continuously growing population exerts tremendous pressure on the limited resources and thus affects the environment significantly.

*Vehicular growth:* Due to increase in personal vehicles and less dependency on public transport, Pune is facing major problems of congestion and air pollution. The total numbers of on-road vehicles in 2002-03 were about 7 lakhs, which are projected to double by 2014-15, and the use of liquid fuels like petrol and diesel can only make the situation worse.

*Industrial growth:* There are about 30000 small-scale industries in Pune district. The major types of industries are paper and wood, foundry, metal, forging, automobile, engineering, textile, dairy, steel mills, refractory and ceramic, galvanizing units.

*Domestic & Commercial:* Though, cleaner fuel like LPG has taken over in urban areas, but people living in slums continue to use polluting fuels like firewood, biomass, etc. With ever-rising population, the emissions from domestic sectors are always increasing.

## EMISSION INVENTORISATION

The approach towards emission inventorisation used in this paper is based on the activity data/ fuel consumption estimates and use of appropriate emission factors. The base year for the calculations was assumed as 2004-05 and projections were

made for a decade i.e., up to 2014-15. Different scenarios, which are developed to study the impact of various interventions on the SPM emission loads, are:

a) *Business As Usual (BAU):* This scenario simulates the SPM emission loadings, if no interventions are taken in any of the sectors.

b) *CNG scenario:* This scenario simulates the SPM emission loadings,

- if CNG is replaced with traditional fuels like petrol and diesel with city specific switch over percentages in the vehicular sector, and
- if natural gas is replaced with traditional fuels like petrol, diesel, furnace oil, biomass etc with city specific switch over percentages in the industrial sector.

c) *PCV Scenario (Phase-out 10 yr. old commercial vehicles):* This scenario simulates the SPM emission loadings, if all the commercial vehicles (auto-rickshaw, taxi and buses) older than 10 years are phased out.

d) *PAV Scenario (Phase-out all 10 yr. old vehicles):* This scenario simulates the SPM emission loadings, if all the vehicles older than 10 years are phased out.

e) *IS Scenario (Industrial scenario):* This scenario simulates the SPM emission loadings, if 10 % of the emissions coming out of industrial sources can be avoided due to technology change, shifting, less fuel usage or modified production processes.

f) *PTS Scenario (Public transport system scenario):* This scenario estimates the emission loadings, if the existing private vehicles (2-wheeler & cars) fleet is replaced by an efficient public transport system (bus based). Estimations are also made for public transport system based on fuel usage (diesel as well CNG) (PTS-d & PTS-c). The emission estimates were made for three major sectors (transport, industrial, and domestic) and the same are dealt below in detail.

- **Transportation Sector**

For each year, estimates of breakup of different types of vehicles (old and new) and vehicle kilometer travelled were made along with the switchover percentage to CNG. The emission factors for different reference years are based on the type of vehicle and fuel used (TERI, 2002). Besides this, the emission factors for the period 2010-11 to 2014-15 for LCV and passenger cars were taken as per the norms specified for Euro IV vehicles under the auto fuel policy of the Government of India. However, in the case of diesel heavy-duty vehicles, the emission norms were adopted from a study that evaluated the alternative fuels and technologies for buses in Mumbai (TERI, 2001). Emission loads are calculated as:

Emissions Load/Day = No. of vehicle x Kilometers travelled per day (Km/d) x Emission factor (g/km)

The emission inventory of SPM for the transport sector for years 2004-05 and 2014-15 under different scenarios is shown in Figure 3.

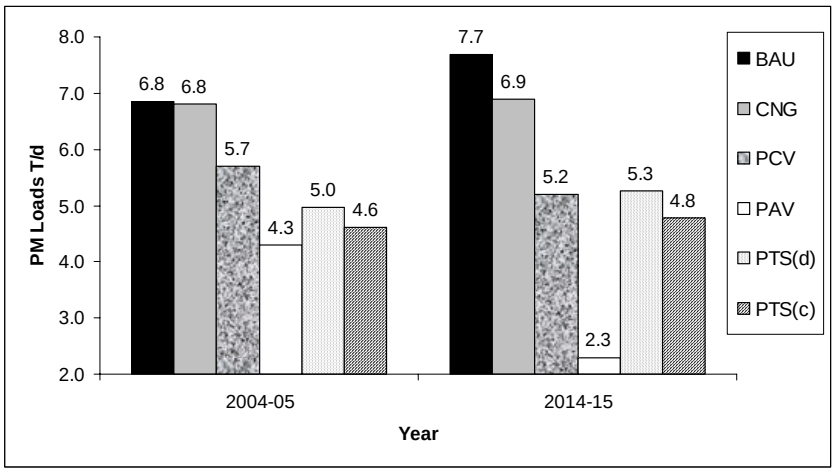


Figure 3: Vehicular SPM loads for Pune under different scenarios

Figure 3 clearly compares the vehicular emission loads of Pune under different scenarios and clearly exhibits the fact that the vehicular pollution load will increase to its maximum if activity levels go with the same pace as they are going presently.

- *BAU scenario:* Figure 3 reveals the increase in emission loads during 2004-05 to 2014-15 due to increase in number of vehicles. As per the present rate, the total SPM emission loads from vehicular sector will increase from 6.8 T/D in 2004-05 to 7.7 T/D in 2014-15.
- *CNG scenario:* Figure 3 reveals the decrease in emission loads due to intervention of CNG in the automobile sector. Therefore, it is estimated that SPM emission loads will be reduced by nearly 10% in 2014-15 when compared to BAU scenario.
- *PCV scenario:* Phasing out commercial vehicles older than 10 years can have a significant impact on the SPM emission loads generated by vehicular sector. Immediate phase out of 10 year older commercial vehicles results in a decrease of 16 % SPM emissions loads in Pune. However, continuation of this phasing out programme, leads to a decrease of 33% of SPM emissions in 2014-15, in comparison to BAU scenario, in Pune.
- *PAV scenario:* This emerges out to be the most effective scenario. Phasing out all vehicles older than 10 years can have a tremendous impact on the SPM emission loads generated by vehicular sector. Immediate phase out of 10 year older vehicles results in a decrease of 37 % SPM emissions loads, in Pune. However, continuation of this phasing out programme, leads to a decrease of 70% of SPM emissions in 2014-15, in comparison to BAU scenario, in Pune.
- *PTS scenario:* PTS(d) scenario replaces all the 2-w, cars and existing buses with an equivalent efficient public transport system of new diesel buses. Immediate action can result in reduction of 26% SPM emission loads when compared to BAU scenario and continuation of program till 2014-15 may result in a decrease of 31% SPM emission loads when compared to BAU scenario. PTS(c) scenario replaces all the 2-w, cars and existing buses with an equivalent efficient public transport system of new CNG buses. Immediate action can result in reduction of

32% SPM emission loads when compared to BAU scenario and continuation of program till 2014-15 may result in a decrease of 38% SPM emission loads when compared to BAU scenario.

- **Industrial Sources**

The consumption of various fuels (Petrol, Diesel, Furnace Oil, Coal, Firewood, LPG & Natural gas) in various categories of industries has been estimated. The emission factors (in Kg/Unit of fuel consumed) have been adopted from WHO(1993). Emission loads have been estimated for SPM for BAU, CNG, and IS scenarios.
Emissions Load/Day = Fuel Consumed x Emission Factor (Kg/Unit of fuel consumed)
The emission inventory of SPM for years 2004-05 and 2014-15 under BAU, CNG, and IS scenarios is shown in Figure 4.

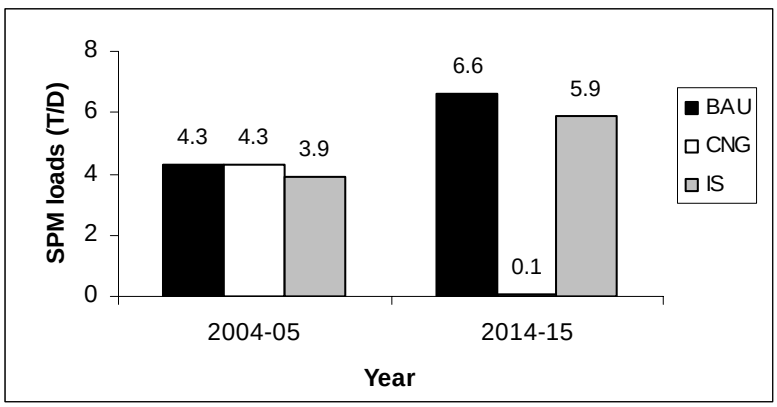


Figure 4: Industrial SPM loads for Pune under different scenarios

- *CNG scenario:* Figure 4 reveals the drastic decrease in emission loads due to intervention of natural gas in the industrial sector. This is with the assumption of very good percentage switch of industries towards natural gas due to economical or regulatory reasons.
- *IS scenario:* This scenario lowers down the emission levels by 10 % due to various factors like technology change, process modifications, energy conservation and cleaner production initiatives. Therefore, a marginal decrease can be observed under this scenario.

- **Domestic Sector**

The total fuel consumption has been estimated accounting for the population in Pune city and the per capita fuel consumption based on NSSO (2001). Emission factors for LPG, fuel-wood, and kerosene have been adopted from World Bank1997 and TERI 2003 in order to estimate emission loads.
Emissions Load/Day = Fuel Consumed x Emission Factor (Kg/Unit of fuel consumed)

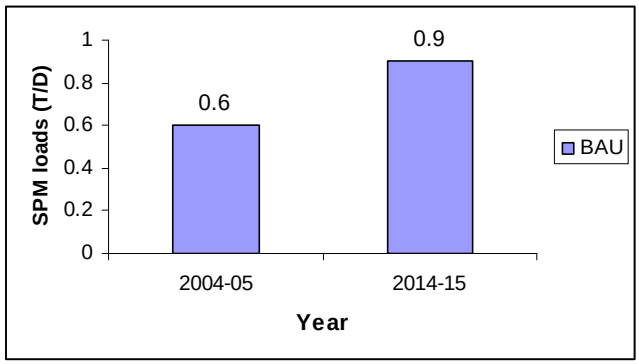


Figure 5: Domestic emission load in Pune in BAU scenario for 2004-05 and 2014-15

- **Aggregated SPM Emission Loads**

Aggregated SPM emission loads are calculated by adding the emission loads from all the three sectors. Analysing the results, CNG scenario emerges out to be the best, which targets all the sectors and expected to reduce the emissions by 48% by the end of 2014-15. PAV scenario reduces the total emissions by nearly 21% immediately and 36% after 10 years. However, if commercial vehicles are only phased out, then reduction is 9% immediate and 16% after 10 years. PTS scenarios (diesel & CNG) also show immediate reduction of 15-18% emission loads, and a reduction of 16-19% in the year 2014-15. (Figure 6)

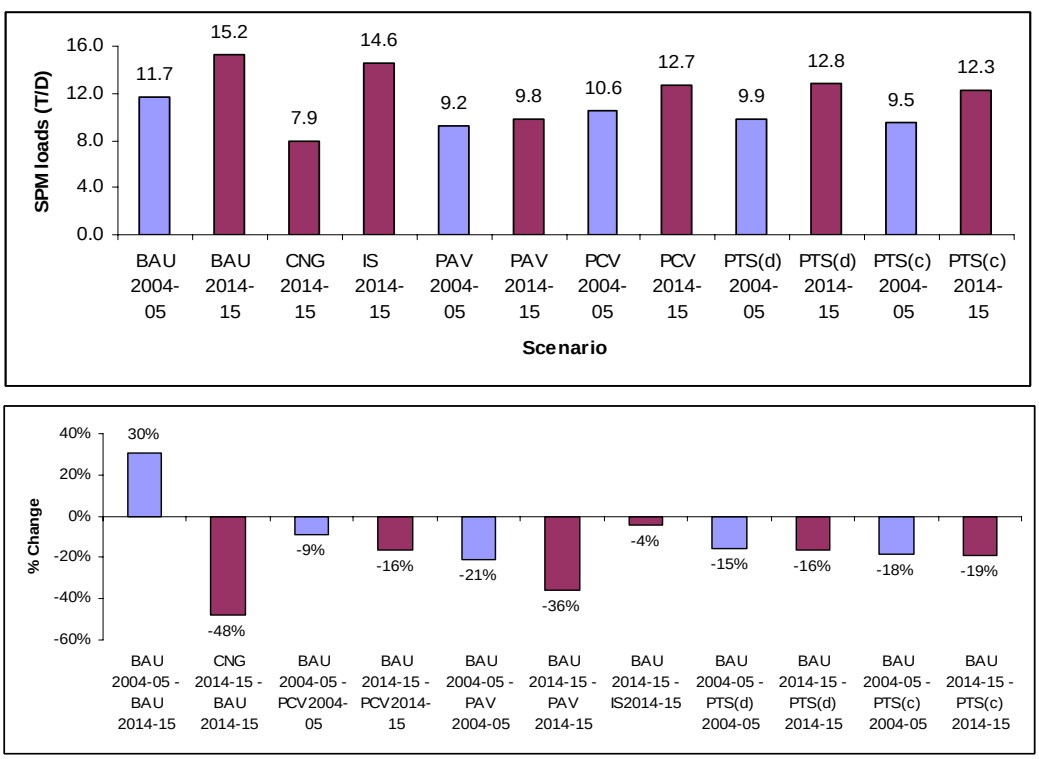


Figure 6: Total and percentage change in emission load in Pune under various scenario for 2004-05 and 2014-15

- **Percentage share of SPM loads from various sectors under different scenarios**

Figure 7 clearly explains the percentage share of SPM emission loads under different scenarios. CNG scenario targets the industrial sector and reduces its share drastically. However, PCV, PAV and PTS scenarios aim the transport sector and reduce their share from total SPM emission loads. In 2004-05, percentage share of vehicular emission load is maximum (58%) with other major contributors are the industries with 37% share. However, in the BAU scenario 2014-15, the percentage share of industries increases to 43%. Though, with the intervention of Natural gas in the CNG scenario, there has been a drastic decrease in industrial emissions and its share goes down to 1%. In PAV and PCV scenarios, percentage share of vehicular sector reduces to 24% and 41% respectively, in the year 2014-15, while under the PTS scenarios the share remains nearly 40% in the same year.

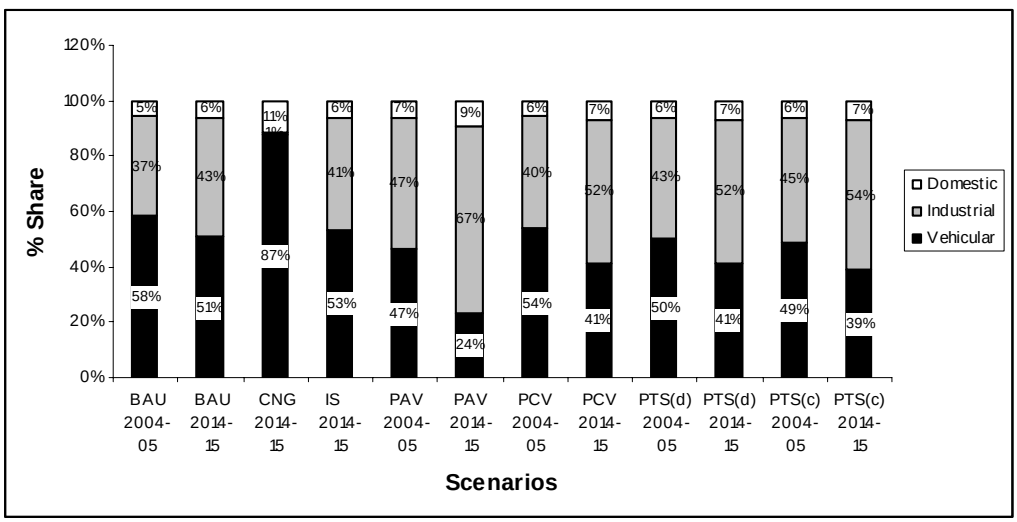


Figure : 7 Percentage share of each sector under different scenarios in Pune.

## AIR QUALITY MANAGEMENT PLAN

Pune is a city in which vehicles and industries both contribute almost equally. Therefore management strategies should focus upon both the sectors. Some of the city specific key strategies can be:-

- Introduction of CNG in vehicular and natural gas in industrial sector can reduce the SPM emissions substantially.
- Notification of vehicles norms like Bharat-II, III, IV.
- Phasing out of 10-year old commercial or all the vehicles (if possible).
- Shifting of industries to the outskirts of the city.
- Checking fuel adulteration.
- In non-CNG scenario, supply of better quality of diesel and petrol.
- Proper inventorisation of industrial emission loads.
- Round the clock vigilance of industries.
- Major augmentation of public transport system is required. Mass Rapid Transport system may be considered for the fast expanding and major urban areas in the country.

- Strengthening of inspection and maintenance system is required for regulating pollution from the large fleet of in-use vehicles

Also, few general strategies can also be adopted as: -

- Development of comprehensive air quality management plans based on scientific studies that involve information related to urban planning, ambient air quality, emission inventory, and air quality models.
- Systematically planned emission load mapping studies should be undertaken at regular intervals. Development of emission factors for Indian conditions to be expedited.
- Review of air quality standards based on health studies
- Strengthening of air quality monitoring network (both in terms of number of stations as well as parameters monitored, including air toxics)
- Development, maintenance and continuous updating of a credible database on air quality, noise levels, emission inventory, source apportionment, health effects, etc
- Proper siting of projects to minimize the adverse impact on people and environment
- Urban planning with focus on environmental issues
- Non-point sources of pollution (such as generator sets, waste burning, etc) also need to be controlled
- Promoting use of cleaner fuels like LPG and kerosene for domestic consumption for reducing indoor air pollution
- Incentives for environmentally benign substitutes, technologies and energy conservation
- Use of fiscal measures for pollution prevention and control. Economic instruments need to be in place to encourage a shift from curative to preventive measures, internalization of the cost of environmental degradation, and conservation of resources.
- Promotion of appropriate research and development (R & D) studies in areas such as source apportionment, environmental health, exposure assessment, environmental modeling etc.
- Thrust to information dissemination & public awareness activities
- Capacity building (both in terms of infrastructure and human resources) for various pollution control boards/Department of Environment

## 3. CONCLUSION

The scenario analysis approach provides various options for air quality management under different circumstances. Analyzing the options, a priority list of actions can be framed. In the case of Pune, vehicular and industrial sectors contribute almost equally to the total SPM emission loads, which directs us to frame the strategies targeting both the sectors. Analysing the results of scenario analysis, intervention of CNG in vehicular and industrial sectors appears to be the most appropriate option to reduce the SPM emission loads most effectively. Alternatively, phasing out all the 10-year-old vehicles can also help in reducing the SPM loads significantly.

This overall assessment of the air quality issues for Pune provides useful inputs for framing the strategies for air quality management in other cities as well.

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# LEVELS AND MAJOR SOURCES OF PM-2.5 AND PM-10 IN BANGKOK METROPOLITAN REGION

**Nares Chuersuwan[1], Subuntith Nimrat[2], Sukanda Lekphet[3] and Tida Kerdkumrai[4]**

[1]School of Environmental Health, Institute of Medicine, Suranaree University of Technology, Nakhon Ratchasima 30000 THAILAND (nares@sut.ac.th)

[2]Faculty of Sciences, Burapha University, Chonburi 20200 THAILAND (subunit@bucc4.buu.ac.th)

[3]Buraeu of Research-Development and Hydrology, Dept. of Water Resources, Bangkok 10300 THAILAND (s_lekphet@yahoo.com)

[4]Physics and Engineering Program, Department of Science Service, Bangkok 10400 (tida@dss.go.th)

## ABSTRACT

This research was the first attempt to concurrently measure and identify major sources of both PM-10 and PM-2.5 in Bangkok Metropolitan Region (BMR), Thailand. Daily averages of PM-10 were 108.1±35.5, 62.1±30.7, 61.1±25.2, and 37.9±18.9 μg $m^{-3}$ at traffic, two residential areas, and low impact sites, respectively. Average 24-hr of PM-2.5 at the traffic site was also higher than other locations while two residential sites were relatively similar. Seasonal difference of PM-10 and PM-2.5 concentrations was distinct between dry and wet seasons. Spatial correlations of PM-2.5 among sampling sites, in term of $R^2$, ranged from 0.74 to 0.81 comparing to 0.67 to 0.78 of PM-10. Evidence from PM-10 and PM-2.5 concentrations showed a significant role of urban-scale meteorology in spatial distributions of both PMs within BMR.

Chemical mass balance analysis of PM-10 at the traffic site indicated that automobile emissions and biomass burning-related sources contributed approximately 33 percent each. Automobiles contributed approximately 39 and 22 percent of PM-10 mass at two residential sites while biomass burning contributed about 36 and 28 percent. PM-10 from re-suspended soil and cooking sources accounted for 10 to 15 percent at a residential site. Major sources of PM-2.5 at traffic site were automobile and biomass burning, contributing approximately 32 and 26 percent, respectively. Biomass burning was the major source of PM-2.5 mass concentrations at residential sites, accounting for 25 and 41 percent. Automobile contributed about 16 and 41 percent at residential sites, but meat cooking was significant, 19 percent, at a residential site located in the northeast of BMR. Meat cooking also accounted for 31 percent of PM-2.5 mass at a low impact site. Automobile, biomass burning, and road dust were less significant, contributed 10, 6, and 5 percent, respectively. However, approximately 25 to 33 percent of PM-10 could not be identified in this study because of the lack of some source signatures for BMR or Thailand. PM-2.5 mass had larger unexplained sources than PM-10, ranging from 27 to 47 percent partly due to inadequate source profiles of PM-2.5 in BMR.

**Key words**: PM-10, PM-2.5, Bangkok, Air quality, Source contribution

## 1. INTRODUCTION

Air quality in Bangkok Metropolitan Region (BMR), Thailand, was seriously deteriorated in the early 1990's regarding to airborne particles, carbon monoxide, and lead concentrations, especially at monitoring stations near major roadways (PCD, 2004). Gradual improvement of air quality in BMR has been observed after the Asian economic crisis partly due to the slowing down of economic expansion as well as stringent regulations. A report from the World Bank showed that air quality in BMR was better than several Asian cities, for example, Beijing, Jakarta, New Delhi and Manila (World Bank, 2003). However, problems still exist in some urban centers and near traffics, especially the problem on particulate matter (PM). Airborne fine particles in recent years have increased and started to exceed the national ambient air quality standards (120 $\mu g\ m^{-3}$ for 24-hr average of PM-10) in some monitoring stations in BMR and major cities (PCD, 2004). In 2002, the maximum 24-hr concentration of PM-10 was as high as 300 $\mu g\ m^{-3}$ (1.5 times higher than the standards).

The World Bank study on daily hospital admissions for cardiovascular and respiratory illnesses associated with air pollution levels at five hospitals in BMR. The results indicated that PM-10 (particulate matter with aerodynamic diameter less than 10 $\mu m$) concentrations in BMR associated with 4,000 to 5,500 premature deaths each year, based on the population of 10 millions. The deaths were attributed to short-term exposures to outdoor PM. Hospital admissions caused by respiratory and cardiovascular illness were higher when PM-10 concentrations were higher (Radian, 1998). An increase of 1 $\mu g\ m^{-3}$ in PM-10 was estimated to increase the mortality rate by 0.084%, chronic bronchitis cases to 3.06 per 100,000 and incidence of respiratory symptoms to 18,300 per 100,000. The study estimated that reduction of annual 10 $\mu g\ m^{-3}$ of PM-10 would reduce adverse health effect in Bangkok: 700-2,000 premature deaths, 3,000-9,300 new cases of chronic respiratory disease, 560-1,570 respiratory and cardiovascular hospital admissions. The World Bank report has found encouraging results in term of health and financial benefit. Recently study by Vichit-Vadakan (2004) examined the relationship between daily mortality and daily mortality from 1996 to 2001. The results indicated a 10 $\mu g\ m^{-3}$ change in daily PM-10 is associated with a 0.5 percent increase in total mortality. Concerns over possible adverse health and environmental effects have prompted responsible agencies to investigate and control fine PM. Unfortunately, PM-2.5 has not been in the routine measurement in BMR.

## 2. MAIN TEXT

Comprehensive sampling campaign for PM-10 and PM-2.5 measurements began every third day between February 2002 and January 2003 at four separated locations in BMR. The sampling locations were located along prevailing wind directions, northeast and southwest monsoon winds (Figure 1). Din Daeng (DD) station locates in the inner city next to a busy road representing a high impact (traffic) site. Two

residential sites are located on the northeast and southwest of BMR, known as Jan Krasem (JK) and Bann Somdej (BD). A low impact site, Bank Na (NA), locates in the outskirt of BMR. Each sites equipped with two low volume air samplers, MiniVol$^{TM}$ (AirMetrics, Oregon), for 24-hr measurement of PM-10 and PM-2.5 mass concentrations. Samplers ran simultaneously at all locations, sampling at 5 LPM from 12:00 – 12:00 AM local time. This design flow rate can achieve particle cut-point of 2.5 and 10 μm. A primary reference flow device (Gilian Instrument Corp., New Jersey) was used to calibrate the flow rate prior and after the sampling. It is important to note that the MiniVol$^{TM}$ is not a standard device designated by the U.S.EPA. The samplers have been tested and provided approximately 5 – 10 percent of comparable mass concentrations in the U.S. (Baldauf et al., 2001). Samples were alternated collected on either 47 mm stretch PTFE filters (Whatman, New Jersey) or Pallflex® quartz fiber filters (Pall Corp., Michigan) for gravimetric and chemical analyses. PTFE filters are known to have least effects from high humidity. Pre-heated of quartz fiber filters were performed to eliminate residues in the filters. Mass concentrations were determined by a single pan microbalance (Mettler Toledo, Switzerland) with 1 μg resolution. Temperature and relative humidity (RH) were controlled during filter equilibration and weighing to be within 23±5°C and 40±10%RH, respectively. Filters were kept below 4°C protected from light in individual filter holders prior to chemical analysis.

Fifteen elemental compositions of both PM-10 and PM-2.5 were determined using an Inductively Coupled Plasma – Mass Spectrometry (ICP-MS), Elan 6000 (Perkin Elmer Instrument, Connecticut). Quartz fiber filters were extracted using hot acid extraction according to Compendium Method IO-3.1 (U.S.EPA, 1999). Total carbon (TC) was analyzed using thermal technique described by Chen and Wang (1997) with PE 2400 (Perkin Elmer Instrument, Connecticut) and only PM-2.5 samples were analyzed for TC at present. Dionex DX-120 (Dionex Corp., California) was used to determine sulfate, nitrate, chloride, and ammonium concentrations in both PMs. Chemical Mass Balance (CMB8) was used in apportion major sources of the PMs (Watson et al., 2002).

Average 24-hr PM-10 and PM-2.5 at traffic site, DD, had higher concentrations than other stations, 108.1±35.5 and 69.0±28.8 μg $m^{-3}$, respectively. Despite of large distant (about 13 km) between JK and BD, two residential sites, they had relatively similar concentrations of average 24-hr PM-10, 61.1±25.2 and 62.1±30.7 μg $m^{-3}$, respectively. Similar results found for PM-2.5 concentrations at these residential sites while low impact site, NA, exhibited lower concentrations of both PMs than other stations (Table 1). Time-series data showed that both PM-10 and PM-2.5 concentrations had similar trend throughout the sampling stations within the metropolitan while daily fluctuations were observed partly due to approximately 66% of average PM-10 mass concentration accounted by particles less than 2.5 micron and influenced by meteorological conditions. PM-2.5/PM-10 ratios from all stations ranged from 0.64 to 0.67 lower than those found in the western U.S., approximately 0.75, but higher than the ratio found in the eastern U.S. about 0.52 (U.S. EPA, 2002) and Mexico City, less than 0.61 (Vega et al., 2002). However, the ratios found in BMR were in the range of those found in Spain, ranging from 60 to 90 percent (Hernádez et al., 2002; Querol et al., 2002) meaning that PM-2.5 is of

important for PM-10 mass concentrations since it constituted more than a haft of the mass in the area.

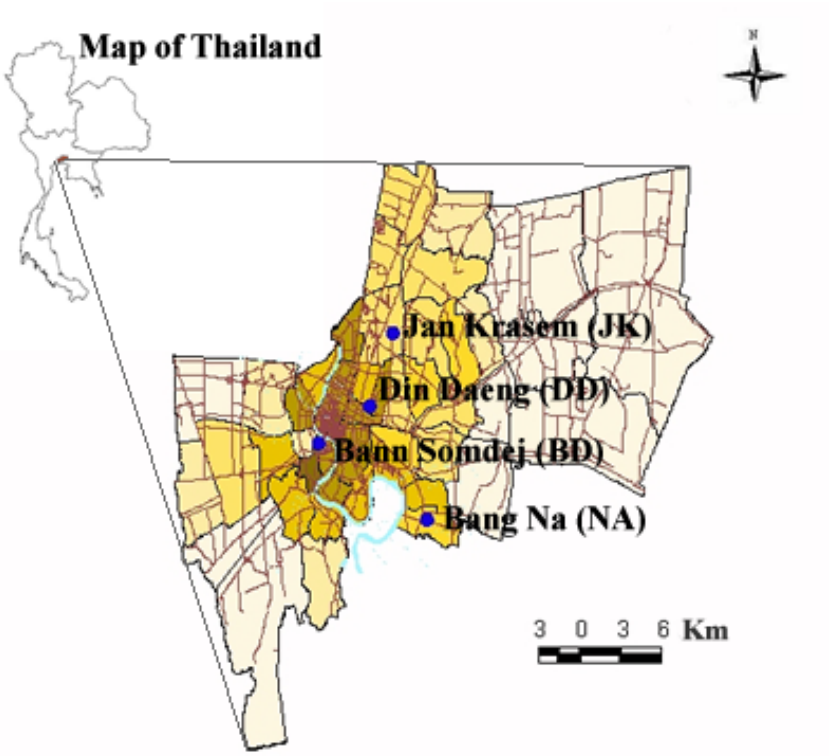



Figure 1. Map of Thailand and Bangkok Metropolitan Region (BMR) including sampling locations.

Table 1. Statistical summary of average 24-hr PM-10 and PM-2.5 in BMR during 2002 – 2003.

| Average 24-hr PM-10 and PM-2.5 concentrations (μg $m^{-3}$) | | | | | | PM2.5/ PM10 ratio |
|---|---|---|---|---|---|---|
| Stations | Type | N | Average ± σ | Min | Max | |
| • DinDaeng (DD) | • PM-10<br>• PM-2.5 | 107<br>99 | 108.1±35.5<br>69.0±28.8 | 37.7<br>12.3 | 205.7<br>150.3 | 0.64 ±0.11 |
| • Jan Krasem (JK) | • PM-10<br>• PM-2.5 | 106<br>103 | 61.1±25.2<br>40.9±21.4 | 21.3<br>9.2 | 130.0<br>109.8 | 0.67 ±0.13 |
| • Bann Somdej (BD) | • PM-10<br>• PM-2.5 | 100<br>93 | 62.1±30.7<br>41.5±24.6 | 20.9<br>11.3 | 176.6<br>144.4 | 0.67 ±0.27 |
| • Bang Na (NA) | • PM-10<br>• PM-2.5 | 92<br>88 | 57.6±23.9<br>37.9±18.9 | 20.7<br>8.0 | 145.1<br>103.9 | 0.66 ±0.14 |

Seasonal variations are divided into wet and dry seasons despite three seasons are recognized in Thailand (summer, rainy, and winter). Dry season (October – February) had higher PMs than wet season (March – September). Conditions during dry season in Asia play a major role in causing high particulate matter e.g., biomass burning and lack of rain scavenging (Ogunjobi et al., 2004). Relationships between

PM-10 and PM-2.5 mass concentrations at each station were analyzed in terms of linear regression and the results revealed that coefficients of determination ($r^2$) were relatively high, ranging from 0.75 to 0.83, shown in Figure 2. Equations (1) to (4) indicated that most of the slopes closed to unity meaning that factors influencing PM-10 concentrations were likely due to site-specific local conditions, e.g., local source strength, locations, meteorological conditions, etc. These local contributions could be observed from the intercepts. For instance, local contributions around DD accounted for approximately twice of the contributions in other areas, more likely from high traffics and surrounding activities.

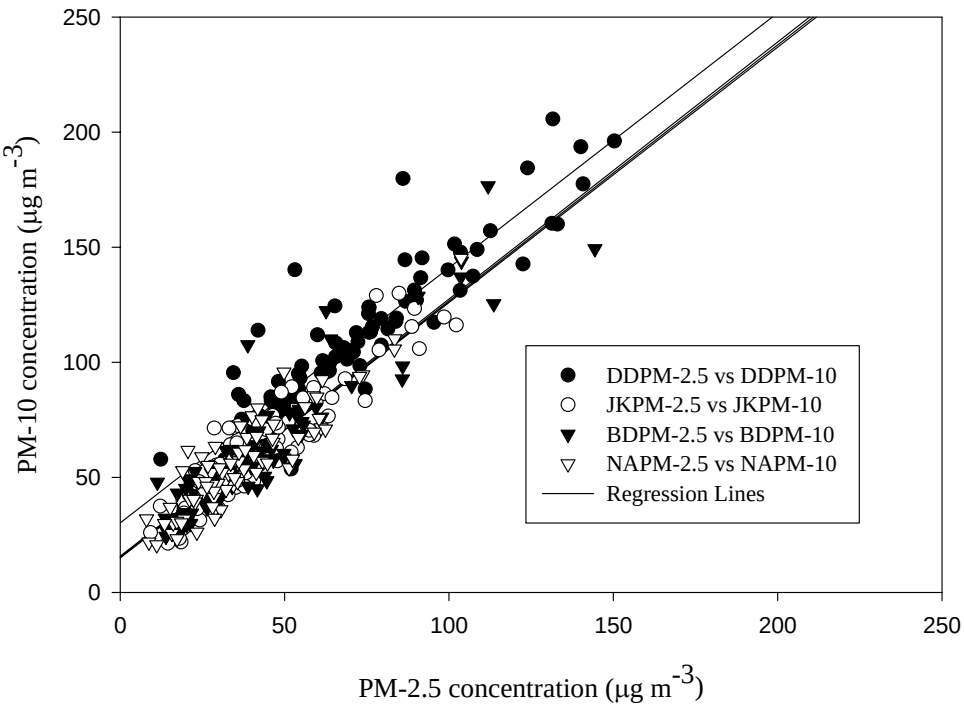



Figure 2. Correlations between PM-10 and PM-2.5 concentrations, 2002-2003.

$DD_{PM\text{-}10}$ (μg $m^{-3}$) = 1.08[$DD_{PM\text{-}2.5}$] + 32.3 (μg $m^{-3}$), $r^2$= 0.79 ----- (1)
$JK_{PM\text{-}10}$ (μg $m^{-3}$) = 1.03[$JK_{PM\text{-}2.5}$] + 18.02 (μg $m^{-3}$), $r^2$= 0.75 ----- (2)
$BD_{PM\text{-}10}$ (μg $m^{-3}$) = 1.08[$BD_{PM\text{-}2.5}$] + 15.49 (μg $m^{-3}$), $r^2$= 0.80 ----- (3)
$NA_{PM\text{-}10}$ (μg $m^{-3}$) = 1.11[$NA_{PM\text{-}2.5}$] + 15.33 (μg $m^{-3}$), $r^2$= 0.83 ----- (4)

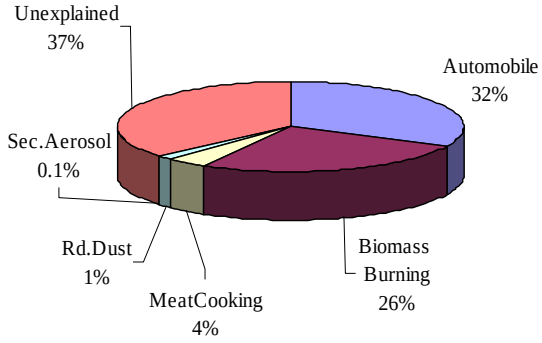
Spatial relationship between sites of PM-10 in term of coefficient of determination ($r^2$) ranged from 0.60 to 0.78 while PM-2.5 had better correlation, 0.74 to 0.81. Chemical analyses indicated that total carbon was a major composition in both PMs, accounting for about a half of the mass of PM-2.5 in traffic influenced station and residential stations while low impact site had total carbon less than a half of the mass concentrations. Sodium, aluminum, iron, sodium, potassium, sulfate, and nitrate were predominantly found in both PMs. Average concentrations of chemical species analyzed from PM-2.5 samples are presented in Table 2.

Table 2. Average concentrations of composition found in PM-2.5 samples.

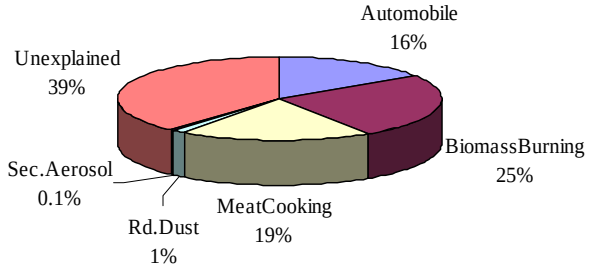
| Parameter | Unit | PM-2.5 | | | |
|---|---|---|---|---|---|
| | | DD | JK | BD | NA |
| Mass | $\mu g\ m^{-3}$ | 69.0±28.8 | 40.9±21.4 | 41.5±24.6 | 37.9±18.9 |
| TC | $\mu g\ m^{-3}$ | 38.48±19.32 | 21.72±12.75 | 21.92±13.33 | 17.57±11.01 |
| $NH_4^+$ | $\mu g\ m^{-3}$ | 0.49±0.20 | 0.72±0.24 | 0.52±0.21 | 0.85±0.52 |
| $Cl^-$ | $\mu g\ m^{-3}$ | 0.80±0.34 | 1.01±0.56 | 1.02±0.43 | 0.96±0.25 |
| $NO_3^-$ | $\mu g\ m^{-3}$ | 0.88±0.30 | 0.70±0.56 | 0.89±0.40 | 0.76±0.51 |
| $SO_4^{2+}$ | $\mu g\ m^{-3}$ | 1.84±0.55 | 1.33±0.59 | 1.66±0.49 | 1.96±0.57 |
| Cr | $\mu g\ m^{-3}$ | 0.13±0.06 | 0.15±0.15 | 0.12±0.07 | 0.13 ± 0.06 |
| Cu | $\mu g\ m^{-3}$ | 0.08±0.14 | 0.07±0.14 | 0.05±0.05 | 0.06±0.04 |
| Fe | $\mu g\ m^{-3}$ | 1.43±0.82 | 1.73±1.47 | 1.66 ± 1.59 | 2.20±2.18 |
| Mn | $\mu g\ m^{-3}$ | 0.05±0.02 | 0.06± 0.11 | 0.05±0.03 | 0.07±0.04 |
| Ni | $\mu g\ m^{-3}$ | 0.26± 0.31 | 0.47± 0.91 | 0.45±0.72 | 0.38± 0.37 |
| Pb | $\mu g\ m^{-3}$ | 0.18±0.18 | 0.28±1.02 | 0.15±0.13 | 0.22±0.17 |
| Zn | $\mu g\ m^{-3}$ | 0.78±0.74 | 0.74± 0.68 | 0.92±0.72 | 1.09±0.53 |
| V | $\mu g\ m^{-3}$ | 1.11±0.51 | 1.19±0.54 | 1.17± 0.51 | 1.09±0.53 |
| Na | $\mu g\ m^{-3}$ | 1.46±1.06 | 1.31± 0.91 | 1.62± 1.11 | 1.31± 0.66 |
| Mg | $\mu g\ m^{-3}$ | 0.47 ± 0.25 | 0.51± 0.54 | 0.46± 0.27 | 0.75±1.42 |
| K | $\mu g\ m^{-3}$ | 0.98±0.56 | 0.75± 0.66 | 1.10± 0.88 | 0.93±0.67 |
| Ca | $\mu g\ m^{-3}$ | 2.98±2.28 | 3.33 ±2.97 | 3.14 ±2.75 | 3.12± 2.25 |
| Al | $\mu g\ m^{-3}$ | 1.91±1.29 | 2.74±3.14 | 2.13± 1.58 | 2.95 ± 2.39 |
| Sn | $\mu g\ m^{-3}$ | 0.09±0.15 | 0.13 ± 0.28 | 0.06±0.12 | 0.097±0.16 |
| As | $\mu g\ m^{-3}$ | 0.31±0.13 | 0.34± 0.14 | 0.33±0.139 | 0.32±0.16 |

Chemical compositions were used in Chemical Mass Balance (CMB8) to identify major source contributions of the PMs (Watson et al., 2002). Only five major sources were able to identify in this study with a relatively large unexplained mass, up to one-third, due to limited source profiles. Automobile was accounted for approximately 32 percent of PM-2.5 mass at traffic site, followed by biomass burning, 26 percent while cooking and secondary aerosol were less significant. Although average mass concentrations of PM-2.5 at residential sites were relatively similar, source contributions were differed from one another at residential sites in the northeast (JK) and southwest (BD) of BMR. Biomass burning was a major contribution (25 percent) to PM-2.5 mass at JK, followed by cooking (19 percent), and automobile (16 percent). PM-2.5 mass concentrations at BD were influenced by biomass burning (41 percent) while cooking was not important. In contrast, cooking was predominant (31 percent) including automobile (10 percent) and biomass burning (6 percent) at low impact site (Figure 2). Unexplained source for PM-2.5 mass ranged from 28 to 47 percent. In the case of PM-10, automobile and biomass burning were equally significant at traffic site, accounting from 66 percent of the mass (Figure 3). Automobile and biomass burning were important sources of PM-10 at other stations as well, ranging from 36 to 28 percent, but cooking and road dust contributed approximately 32 and 26 percent, respectively. Unexplained sources, however, accounted for 23 to 33 percent of PM-10 mass concentrations.

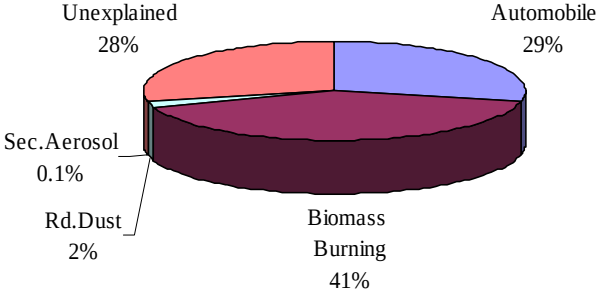
(a) Din Daeng PM-2.5




(b) Jan Krasem PM-2.5




(c) Bann Somdej PM-2.5




(d) Bang Na PM-2.5

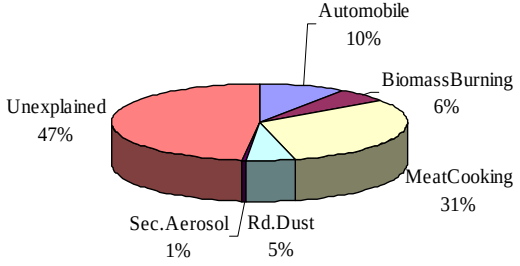
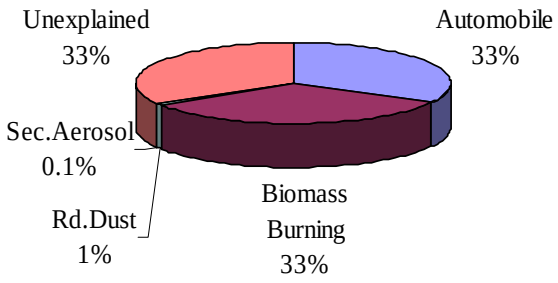
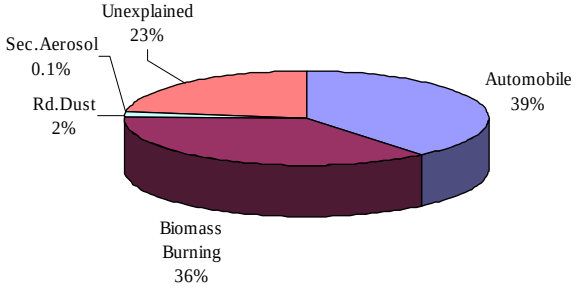



Figure 2. Estimated source contributions of PM-2.5 at each station.

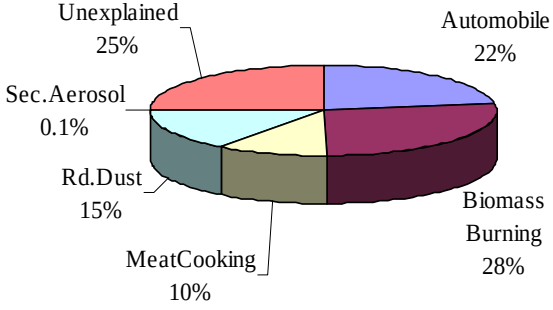
(a) Din Daeng PM-10




(b) Jan Krasem PM-10




(c) Bann Somdej PM-10




(d) Bang Na PM-10

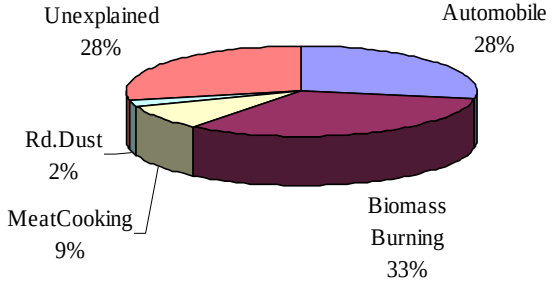



Figure 3. Estimated source contributions of PM-10 at each station.

## 3. CONCLUSION

Situation of PM-10 average concentrations were within ambient air quality standard of 120 μg $m^{-3}$ for 24-hr average. While Thailand has not yet promulgated PM-2.5 standard, concentrations of PM-2.5 at traffic site exceeded 65 μg $m^{-3}$ if a U.S. standard was considered but the concentrations were lower at residential and low impact site. PM-2.5 concentrations correlated well with each other within BMR and the correlations were better than PM-10. PM-2.5 accounted for approximately 66 percent of PM-10 mass concentrations found in BMR. Daily fluctuation of both PM-10 and PM-2.5 mass concentrations were in concert with one another across the sampling sites suggesting that urban-scale meteorology was partly influenced such phenomenon. Seasonal difference of PM-10 and PM-2.5 concentrations was distinct between dry (October – February) and wet seasons (March – September), which is typical in Asian countries with high biomass burning and lack of rain scavenging during dry season. Burning-related sources in residential sites contributed a significant portion of PM-2.5 mass, approximately 1.4-1.5 times more than automobile sources, reflecting the important sources within the areas. Major source contributions of PM-10 were automobiles and biomass burning at both traffic and residential sites, but automobile was more important. Differences in source contributions found in two residential sites when cooking and road dust were contributed approximately 10 to 15 percent, respectively, at a resident site in the northeast. Large unexplained mass was observed, especially for PM-2.5 suggesting that appropriate sources profiles were crucial for BMR to effectively identify source contribution of both PMs. Possible improvement can be made thru specific source profiles regarding to fine particulate matter, which in turn will assist an effective air quality management in the future.

## 4. ACKNOWLEDGEMENTS

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