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Air pollution monitoring

# AIR POLLUTION MONITORING SYSTEM IN THE REPUBLIC OF MACEDONIA

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Abstract. During 1998 a Monitoring System with four automated air monitoring stations was established in Skopje, FYR Macedonia. Each station contains instruments for continuously measuring the concentration of CO, SO<sub>2</sub>, NO, NO<sub>2</sub>, suspended particular matters (SPM), O<sub>3</sub> in the air and at the same time, instruments for measurement of different meteorological parameters (temperature, humidity, wind speed, wind direction and solar radiation). All stations are connected to the central station for transferring data and for data processing. It was found that the concentration of major pollutants (SO<sub>2</sub>, NO<sub>x</sub>, CO and SPM) increases remarkably during the heating season. High concentrations, especially in winter, are caused by additional pollution from heating facilities including home heaters, by geographical conditions peculiar for Skopje and by meteorological conditions which shows the tendency to form temperature inversion layers. On the basis of those data and data obtained from upper-layer meteorological data, atmospheric stability, which is an index expressing the relative difficulty of atmospheric dispersion, was determined.

Keywords: air pollution, monitoring, meteorology, emission, Skopje.

## AIMS AND BACKGROUND

Many of the cities in the FYR Macedonia, including the capital, Skopje, are located in the basins surrounded by mountains. The meteorological conditions unique to such basins are thus causing air pollution called "stagnation", due to gases emitted from factories, automobiles and households, often posing a serious problem<sup>1-3</sup>. Especially in some industrial cities, including Skopje, such air quality aggravation is serious especially in winter period when basin fogs generate. The Government of FYR Macedonia adopted various air pollution protection measures. However, due to lack of an automatic monitoring method, there still exist many problems such as being unable to respond immediately to the aggravation

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in the concentration of air pollution. Besides getting a better understanding of the present situation of air pollution, it is also important to make improvements in the monitoring system for the enactment of regulatory laws as well as assessments of improvements.

Under these circumstances, The Japan International Cooperation Agency conducted a Project during the period of 1997-1999 for constructing an air pollution monitoring system with four automatic monitoring stations in the city of Skopje. Each station contains instruments for continuous measuring of the concentration of CO, SO<sub>2</sub>, NO, NO<sub>2</sub>, suspended particular matters (SPM) and O<sub>3</sub> (for one station) in the air, and instruments for the measurement of some meteorological parameters (temperature, humidity, wind speed and wind direction, and solar radiation for one station). All stations are connected with the central station for transferring data and for data processing. Starting 2003 this automatic air pollution monitoring system will be extended with the stations in the cities of Kumanovo, Kocani, Kicevo and Bitola. In the city of Veles there are already 2 stations controlled by the "Zletovo" lead and zinc smelting plant.

In this paper the analysis of automatic continuous monitoring (AAM) results for meteorological as well as ambient air quality data which was carried out for the period of April 1998 to March 1999 (non-heating season from April to September 1998, and the heating season between October 1998 and March 1999) is given. These results are compared with those obtained for stationary (factories, heating facilities, etc.) and mobile (mainly traffic) emission sources. The results give many explanations about the air pollution in the region of the city of Skopje, distribution of the pollutants and the appearing of the "stagnation" phenomenon.

## **EXPERIMENTAL**

On the basis of the meteorological and topographic conditions of emission sources, and preliminary investigation of distribution pattern of ambient air concentrations, the locations for four automated air monitoring stations were evaluated and installed: Station 1 (Gazi Baba), which is background concentration located on the top of the hill; Station 2 (Center), city area location in the central area of Skopje; Station 3 (Karpos), measuring point for measuring emission gases from automobiles located at the cross-junction of trunk road; Station 4 (Lisice), measuring point located between industrial area and the new towns. The parameters being monitored and the monitoring instruments used are listed as below: SO<sub>2</sub> (ultraviolet fluorescent method), NO<sub>x</sub> (chemiluminescence method), CO (non-dispersive infrared analyser method). O<sub>3</sub> (ultraviolet absorption method) and SPM (gravity balance method). Meteorological parameters are: temperature, humidity, wind speed, wind direction and solar radiation.

The monitoring altitude is set at a range of 3-4 m, taking into consideration the influence due to rescattering of SPM. Auto-calibration method is used in the calibration of instruments. The monitored data is recorded in the data logger as well as the recorder and transmitted to the central station using the telemetric system. At the same time, this information is then sent out from the central station to the public information system set up in the town area. Each monitored station is well equipped with voltage regulators, UPS for data loggers, air-conditioners and security system for monitoring purposes.

Heavy metals determinations in the SPM samples taken from the high volume and low volume samplers are carried out by the atomic absorption spectrometry and by X-ray fluorescence spectrometry.

#### **RESULTS AND DISCUSSION**

#### METEOROLOGICAL CONDITIONS

Surface meteorology. Taking into account the meteorological conditions due to the unique topographical conditions of Skopje, it was decided to use the ultrasonic type of sensor used for the measurement of surface wind direction and velocity. Station No 4 was chosen for the measurement of solar radiation, as there were no obstacles nearby.

- a) Wind direction and speed. In observing the general trend, it was found that W and E winds have high frequencies while there are hardly any signs of an N wind. In addition, the wind velocity is weak which characteristic for the city of Skopje. As for the variation of the monthly average wind velocity, there is a decreasing trend until June and gets slightly stronger in December.
- b) Temperature and humidity. The fluctuations in the daily average temperature and humidity are the same for every station. In addition, together with the temperature increase from May to August, humidity also decreased. In August, the highest daily temperature is about 30 °C while the humidity is about 40%. The highest hourly value is about 40 °C. However, in the last half of December, approximately -10°C was recorded even in the daytime.
- c) Solar radiation. According to the data from Lisice station, there is a significant increase in the solar radiation in summer. This is based on the monthly average temperature.
- d) Appearance frequency of atmospheric stability classes. The atmospheric stability is an index for expressing the relative difficulty of atmospheric dispersion. When the atmosphere is unstable, dispersion of emitted pollutants in the air is accelerated. The pollutants emitted from a low stack are easily dispersed and the concentration level rapidly decreases. On the other hand, pollutants released by a tall stack are easily carried a long distance by advection, and a high concentration of pollutants occurs on the ground. When the atmosphere is stable,

it is difficult to disperse pollutants. Pollutants emitted from a low stack are with high concentration. On the contrary, pollutants released from a tall stack do not easily fall to the ground and the concentration of pollutants on the ground is low.

The investigated heating season, in particular the winter season, shows a tendency to be stable as compared to the non-heating season. In Skopje, the wind is often weak and there are many fair days in the summer season. The atmosphere is very unstable in this season when the solar radiation is high. The air pollutants are mixed and dispersed and hence unlike the winter season, the concentration does not get very high. On the other hand, during night when there is no solar radiation, the atmospheric stability class is either neutral or shifting towards the stable side, and thus unlike the daytime, the mixing and dispersing of the atmosphere does not occur. As a result, the concentration of the air pollutants is often found higher at night than in the day, despite little traffic. In the case whereby the meteorological conditions in Skopje are being considered, the air pollutant concentration level and concentration variation obtained from this investigation are reasonable.

Upper-layer meteorology. Using radiosondes and measuring temperature, wind direction and the vertical distribution of the wind velocity, additional survey of the upper layer meteorology was conducted. During the autumn season temperature inversion was found in the region of 400-1000 m above the ground. It was also found that the lower layers are unstable and the upper layers are stable. During the winter season (December–January) the inversion layer phenomenon occurs very frequently from about few hundred meters to 1000 m is 4°C/100 m). One of the reasons for this could be due to the geographical conditions whereby there are flat plains in the valley area. Under such meteorological conditions, emitted pollutants accumulate near the ground level without spreading to a wider region and severe air pollution occurred.

## AMBIENT AIR QUALITY

The parameters being monitored and the monitoring instruments used are:  $SO_2$  (ultraviolet fluorescent method),  $NO_x$  (chemiluminescence method), CO (non-dispersive infrared analyser method),  $O_3$  (ultraviolet absorption method), SPM (gravity balance method). The monitoring altitude is set at a range of 3-4 m, taking into consideration the influence due rescattering of SPM. The auto-calibration method is used in the calibration of instruments. The monitored data is recorded in the data logger as well as the recorder and transmitted to the central station using the telemetric system. At the same time, this information is then sent out from the central station to the public information system.

Sulphur dioxide  $(SO_2)$ . In the non-heating season,  $SO_2$  concentration level in the air neither exceeds the daily average value and hourly 98% value as well as maxi-

mum set by environmental standards (maximal mean daily concentration of 150  $\mu g/m^3$ ) for any of monitoring stations. When comparing the concentration level for each monitoring station, non-heating season shows low concentration and there is not any significant difference. In the winter season very often the environmental standards are being exceeded and in some cases extremely severe air pollution occurred continuously. Taking into consideration the cases for combined pollution with SPM, from the period of the end of December to the start of January, the concentration of SO<sub>2</sub> exceeds the limit for alarm in the First Stage.

Nitrogen oxides (NO, NO<sub>2</sub>, NO<sub>x</sub>). During the heating season and in particular the winter season, the limits for the environmental standards are frequently being exceeded and like the case of  $SO_2$ , there are cases when the environmental standards are being continuously exceeded. When comparing the  $NO_2$  concentration level for each monitoring station, Station 2 clearly shows the highest value. On the other hand, Station 1 tends to show low values as compared to the other monitoring stations. In addition, the highest 24-hour and hourly values for  $NO_2$  and  $NO_x$  concentrations sometimes are high in the case of Lisice. During the non-heating season, most of the time, the causes of  $NO_x$  can be considered to be due to automobile emission. While it is plausible to conclude that this is reflective of the surrounding areas near each monitoring stations. The ratios of  $NO_2$  to  $NO_x$  ( $NO_2/NO_x$ ) in the non-heating season and the heating season are shown in Table 1.

Table 1. Ratios of NO, to NO, (April 1998-March 1999)

Season	Station 1	Station 2	Station 3	Station 4
Non-heating season	0.7597	0.4882	0.5610	0.4323
Heating season	0.4968	0.2837	0.3597	0.2664

The NO<sub>x</sub> formed by combustion normally takes the form of NO. Thus, by analysing the ratio of NO<sub>2</sub>/NO<sub>x</sub>, the emission source condition and oxidation mechanism can be clarified. The NO is oxidising in the air and changes into NO<sub>2</sub> while influenced by the combined effect of solar radiation and O<sub>3</sub>. The high value of Gazi Baba station was thought to be the active NO<sub>2</sub> conversion taking place because there is no NO<sub>2</sub> sources in the vicinity. As for Lisice station, however, the influence of mobile sources on NO<sub>2</sub> conversion was thought significant. It was thought that solar radiation in the winter period was week and less influential on ratio of NO<sub>2</sub> to NO<sub>x</sub> together with the heating plant effect.

Carbon monoxide (CO). In the non-heating season the concentration level of CO in the air exceeds that required by the environmental standards in almost every case except Station 1. In the heating season the environmental standards were exceeded in all the cases for all the monitoring stations. The reason for this

is that the standard values set are higher compared with that of the Western standards. Among the four monitoring stations, like in the case for  $NO_x$ , the center tends to show a slightly higher concentration level. As for the non-heating season, a high contribution was from automobiles and it was thought that high CO concentration of Stations 2 and 4 being influenced by the same automobile emissions that at Station 1.

Suspended particular matter (SPM). SPM concentration during the non-heating season exceeds the standard value in most cases in all monitoring stations. Aberrant hourly concentration such as  $209~\mu g/m^3$  in the 98 percentile is not merely resulted from stationary source and exhaust gas from vehicles. It can be resulted from suspended dust. It is known that in the cases of weak wind and dry air, like Skopje, the atmosphere tends to stay and SPM concentration level to increase. Moreover, the running car also effectuates the ground dust pick-up. During the heating season the environmental standards have been exceeded in all the cases at all the monitoring stations. In particular, the hourly value for Karpos station shows a high value (800  $\mu g/m^3$ ) and like the case of  $SO_2$  causes severe air pollution.

Ozone  $(O_3)$ . The ozone concentration was monitored at Karpos and Lisice stations. The result of both stations were almost identical but exceeded the standard from April to August. From September onwards and until mid-November, the concentration was observed to decrease, and after that, the level of concentration did not show much decrease, similar to that in the summer.

Appearance frequency distribution of air pollution concentration. The appearance frequency distribution of air pollutant concentration provides strong support for the presumption that the pollutant source affects the characteristics of the air pollution phenomena at each monitoring station. There are significant differences observed between the heating and non-heating seasons such making it obvious that the air pollution in the city of Skopje is concentrated in the heating season. The figures of frequency distribution on each item reflect the characteristics of each point:

- a) As for  $NO_x$  in the non-heating season, 90% of the frequency are within the range of 30 ppb concentration at Station 1. However, those at Stations 2 and 4 are about 90 ppb. The value of 50 % frequency is about 10 ppb at Station 1, while those at the other points are at concentrations a few times higher than that at Station 1, where the characteristics of the monitoring points could be seen. As for  $NO_x$  in the heating season, 90% of the frequency are within the range of 100 ppb at Station 1, and 230 ppb at Station 4. Concerning the other points, they are within that range.
- b) Comparing the monitored parameters, there are some differences in their pattern and it was noticed that their emission differs a little.

c) Frequency of  $NO_x$  in the non-heating season appears higher at Stations 2 and 4 and that of  $SO_2$  at Stations 1 and 2, and that of SPM at Stations 2 and 3 are high. Each parameter always shows higher values at Station 2. However, the distribution of concentration differs, for example SPM appears to be high at Station 4 in the heating season.

Correlation analysis of air pollutant concentration. By obtaining the correlation coefficient of the concentration of two pollutants it is possible to evaluate whether or not they are affected by the same emission source. It was found that in the non-heating season:

- the correlation of  $NO_x$ , NO,  $NO_2$  and CO shows high correlation coefficients. However, the correlation between NO and  $NO_2$  is not high at Gazi Baba station, which probably resulted from the conversion rate from NO to  $NO_2$ ;
- the correlation between SPM and the other monitored parameters are not high. There seems to be various emission sources of SPM, other than that by burning;
- the correlation between SO<sub>2</sub> and the other monitored parameters is also not high;
- there are some cases where the correlation between  $O_3$  and the other monitoring parameters is minus.

In the heating season there is high correlation between SPM and CO, NO<sub>2</sub> and SO<sub>2</sub> at Gazi Baba station and the Center; high correlation between NO<sub>2</sub> and SO<sub>2</sub> at each station. However, the correlation of NO<sub>2</sub> and NO<sub>x</sub> decreases at each station; the correlation of SPM and the other monitoring parameters tends to increase.

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