### AIR POLLUTION MONITORING SYSTEM IN SKOPJE

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Abstract. During 1998 a Monitoring system with four automated air monitoring stations was established in Skopje. The location of the monitoring stations was evaluated on the basis of the meteorology and topographic conditions of emission sources and preliminary investigation of distribution pattern of ambient air concentrations in Skopje. Each station contains instruments for continuously measuring the concentration of CO, SO<sub>2</sub>, NO, NO<sub>2</sub>, SPM, O<sub>3</sub> in the air and at the same time, instruments for measurement of some meteorological parameters (temperature, humidity, wind speed, wind direction and solar radiation). All stations are connected to a central station for transferring data and for data processing. The analysis of automatic continuous monitoring results for meteorological, as well as ambient air quality data, carried out for the period of April 1998 to March 1999, is given. Results are compared with those obtained for stationary (factories, heating facilities etc.) and mobile (mainly traffic) emission sources. 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.

Key words: Air pollution, monitoring, meteorology, emission

#### 1. Introduction

The Japan International Cooperation Agency conducted the Study on Air Pollution Monitoring System in the Republic of Macedonia during the period of 1997-1999. The present state of ambient environment was studied with a focus on the model city, Skopje, such as related facilities including emission sources facilities, monitoring facilities and laboratories of related organizations and land utilization [1-3]. On the basis of the meteorology and topographic conditions of emission sources, and preliminary investigation of distribution pattern of ambient air concentrations, the locations for four automated air monitoring stations was evaluated and installed:

Station 1 (Gazi Baba): Background concentration located on the top of the hill

Station 2 (Center): City area location in the central area of Skopje City

Station 3 (Karpo (): Measuring point for measuring emission gases from automobiles located at the

cross-junction of trunk road

Station 4 (Lisi~e): Measuring point located between industrial area and the new towns.

Each station contains instruments for continuous measuring of the concentration of CO, SO<sub>2</sub>, NO, NO<sub>2</sub>, SPM. O<sub>3</sub> in the air, and at the same time, instruments for the measurement of some meteorological parameters (temperature, humidity, wind speed, wind direction and solar radiation). All stations are connected with the central station for transferring data and for data processing. The analysis of automatic continuous monitoring (AAM) results for meteorological as well as ambient air quality data 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. Those results are compared with those obtained for stationary (factories, heating facilities etc.) and mobile (mainly traffic) emission sources.

#### 2. Results and Discussion

#### 2.1. Meteorological Conditions

#### 2.1.1. Surface Meteorology

Taking into account the meteorological conditions due to the unique topographical conditions of Skopje City, it was decided to use the ultrasonic type of sensor is used for the measurement of surface wind direction and velocity. Station No. 4 (Lisice) 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 a N wind. In addition, the wind velocity is weak and this is a characteristic of Skopje city. The average wind velocity was observed based on time, the wind is strong from 16:00 to 18:00 in the non-heating season. In the heating season, the wind tends to be strongest from 12:00 to 15:00 though its velocity is weak. As for the variation of the monthly average wind velocity, there is a decreasing trend until June. The wind velocity does not change greatly after that, but it gets slightly stronger in December.

b) Temperature and Humidity

The fluctuation 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 minus 10 degrees 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. The atmospheric stability class is divided into following classes according to wind velocity, the amount of solar radiation and depends of the amount of clouds at night time:

Class A	Strong instability .	Class D	Neutral	
Class A-B	Strong instability- Instability	Class E	Moderately stability	
Class B	Instability	Class F	Stability	
Class B-C	Instability-Moderately instability	Class G	Strong stability	
Class C	Moderately instability			

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 found the high concentration. Contrarily, 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 (class A, B and D). 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 nighttime when there is no solar radiation, the atmospheric stability class is either neutral (D) or shifting towards the stable side (G), 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 obtained from this investigation is reasonable.

#### 2.1.2. 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.

#### 2.2. Ambient Air Quality

The parameters being monitored and the monitoring instruments used are: SO<sub>2</sub> (ultraviolet fluorescent method), NO<sub>3</sub> (chemiluminiscence method), CO (non-dispersive infrared analyzer method), O<sub>3</sub> (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.

#### 2.2.1. Sulfur Dioxide (SO<sub>2</sub>)

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 maximum set by environmental standards (maximal mean daily concentration of 150  $\mu$ g/m³) 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 particular the winter season, very often the environmental standards are being exceeded and in some cases, extremely severe air pollution occurred continuously. Taking into the 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> exceed the limit for alarm in the First Stage.

#### 2.2.2. Nitrogen oxides (NO, NO<sub>2</sub>, NO<sub>x</sub>)

During the heating season and in particular the winter season, the limit 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 value for  $NO_2$  and  $NO_3$  concentration sometimes is 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 ratio of  $NO_2$  to  $NO_x$  ( $NO_2/NO_x$ ) in the non-heating season and the heating season are shown in Table I.

Table I. Ratio of NO<sub>7</sub> to NO<sub>7</sub> (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  $NO_x$  formed by combustion normally takes the form of NO. Thus by analyzing the ration  $NO_2/NO_x$ , the emission source condition and oxidation mechanism can be clarified. The NO is oxidizing in the air and changes into  $NO_2$  while influenced by the combined effect of solar radiation and  $O_3$ . The high value of Gazi Baba Station was thought to be the active  $NO_2$  conversion-taking place because there is no  $NO_2$  sources in the vicinity. As for Lisice Station, however, the influence of mobile sources on  $NO_2$  conversion was thought significant. It was thought that solar radiation in the winter period was week and less influential on ratio of  $NO_2$  to  $NO_x$  together with the heating plant effect.

#### 2.2.3. 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 Western standards. Among the four Monitoring Stations, like in the case for NO<sub>x</sub>, 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 Station 2 and 4 being influence of such automobile emission that Station 1.

#### 2.2.4. 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 µg/m³ 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 increases. 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/m³) and like the case of SO<sub>2</sub> causes severe air pollution.

#### 2.2.5. Ozone $(O_3)$

The ozone concentration was monitored at Karpos and Lisice stations. The result of both stations resulted 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.

# 2.2.6. 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 season such making it obvious that the air pollution in Skopje City is concentrated in the heating season. The Figures of frequency distribution on each item reflect the characteristics of each point:

- As for NO<sub>x</sub> in the non-heating season, 90 % of the frequency is within the range of 30 ppb concentration at Station 1. However, those at Station 2 and Station 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<sub>x</sub> in the heating season, 90 % of the frequency is within the range of 100 ppb at Station 1, and 230 ppb at Station 4. Concerning the other points, then 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<sub>x</sub> in the non-heating season appears higher at Station 2 and Station 4 and that of SO<sub>2</sub> at Station 1 and Station 2, and that of SPM at Station 2 and Station 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.

## 2.2.7. Correlation Analysis of Air Pollutant Concentration

#### a) Correlation Analysis of Pollutant

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<sub>x</sub>, NO, NO<sub>2</sub>, and CO show high correlation coefficients. However, the correlation between NO and NO<sub>2</sub> is not high at Gazi Baba Station, which probably resulted from the conversion rate from NO to NO<sub>2</sub>.
- 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 are also not high.
- There are some cases where the correlation between  $O_3$  and the other monitoring parameters are minus.

In the heating season there is high correlation between SPM and CO,  $NO_2$  and  $SO_2$  at Gazi Baba and the Center; high correlation between  $NO_2$  and  $SO_2$  at each station. However, the correlation of  $NO_2$  and  $NO_x$  decreases at each station; the correlation of SPM and the other monitoring parameters tend to increase.

#### b) Correlation Analysis of Monitoring Stations

- 1) In the non-heating season, the correlation of SO<sub>2</sub> is high between Center and Lisice, where the coefficient is 0.626. In the heating season, however, it is high between the Center and Gazi Baba, where the coefficient is 0.804.
- 2) The correlation of NO, NO<sub>2</sub>, NO<sub>x</sub> is high between Center, Karpos and Lisice. The correlation coefficient of NO<sub>2</sub> is 0.816 between Center and Karpos in the heating season. It seems that three points are more commonly influenced by automobiles.
- 3) The correlation of O<sub>3</sub> among points in the non-heating season is high, as the coefficient is 0.827. However, the high correlation could not be seen during the heating season. It shows that both the process of creation and that of consumption differ between the heating season and the non-heating season.
- 4) The correlation of SPM in the heating season is quite high among each point and SPM seems to be a widely spread pollutant.

#### References

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