ABSTRACT

Comparisons of Private Stations (PS) measurements with Automatic Monitoring Stations (AMS) included in the National Environmental Monitoring System of the Ministry of Environment and Waters in Bulgaria has been made. The chosen PS (a total of four) are located close to AMS. In order to check whether the PS data are reliable and could be used for air quality monitoring, a statistical analysis of the data from the PS and the AMS was performed. The concentrations measured by two methods are quite different, but a good correlation between them has been established. In order to improve the PS measurement accuracy models for correction of the PM$_{10}$ and PM$_{2.5}$ concentrations measured using sensors were derived. They are characterized with good accuracy but are only valid for the warm part of the year. It is necessary to derive another set of coefficients for the cold half of the year. Without correction, the PS measurement error is large and their records may be misleading.

Keywords: air quality, PM measurement methods, Sensor Stations Data Analyses.

INTRODUCTION

Particulate matter pollution consists of very small liquid and solid particles flying in the air. Nowadays, the particulate matter air pollution, especially with small size particles PM$_{10}$ and PM$_{2.5}$ is associated with a number of adverse health effects. That is why it is important to measure and control their concentrations in the ambient air.

The Bulgarian government has taken commitment to achieve certain levels of fine particulate matter in the air since the country has joined the European Union. Over the years, air pollution has been decreasing, but in terms of fine particles sufficient air quality has not been achieved yet. Two methods for measuring of the concentration of fine particles in ambient air are officially recognized in Bulgaria - gravimetric (reference) method and beta absorption. At 71 % of monitoring stations in Bulgaria PM$_{10}$ concentration is measured by beta absorption. At the remaining stations (29 %) the gravimetric method is applied. For PM$_{2.5}$ the corresponding values are 40 and 60 %. In Bulgaria, PM$_{10}$ concentration is measured at 45 monitoring stations while only 10 stations are able to perform measurements of PM$_{2.5}$. The latter means that no real data for air quality assessment with respect to PM$_{2.5}$ are available for a large part of Bulgaria.

The so called “Private stations” (PS) are able to measure particulate matter by means of sensors. Since they are quite cheaper than the automatic monitoring stations (AMS) it should not be a serious problem to install and use a relatively large number of personal stations located in the area of interest. This means that the PS are an additional option to provide data on the PM$_{2.5}$ concentration in the air. The main purpose of this research is to check whether the PS data are reliable and could be used for air quality monitoring.

AIR POLLUTION MONITORING IN SOFIA MUNICIPALITY

PM air pollution is a long-standing problem for Sofia and its surroundings. The air quality monitoring is carried out by 6 AMS and one manually sampling sta-
tion (MS Gara Yana). These stations are included in the National Environmental Monitoring System managed by The Ministry of Environment and Waters. Data from four AMS are used for the purposes of this study, namely: AMS Mladost (traffic oriented station); AMS Drujba and AMS Hipodruma (background urban stations) and AMS Pavlovo (background urban/traffic monitoring station). The four stations mentioned above provide data about PM$_{10}$ concentration but only one of them - Hipodruma is able to measure the PM$_{2.5}$ concentration.

Monitoring stations Mladost, Drujba, Hipodruma and Pavlovo use automated continuous measurement systems based on the use of $\beta$-ray attenuation [1, 2]. They produce 1-hour average concentration values. The volume of the air passing through the sampling line is measured considering the actual weather conditions. The Air Quality Directive 2008/50/EC allows the use of such systems if their data are equivalent to those obtained by the reference method, i.e., after demonstration that these systems meet the Data Quality Objectives for continuous measurements [3 - 6].

According to the European and Bulgarian legislation, it is allowed the daily limit value for PM$_{10}$ concentration (50 μg m$^{-3}$) to be exceeded up to 35 times per year. However the four AMS have registered many more than 35 exceedances for each of the last 10 years.

Although a certain reduction of air pollution with PM$_{10}$ can be noted during the last years in Sofia (may be due to the implementation of measures included in the Programme for reduction of emissions and acquisition of the limit values for harmful substances on the territory of Sofia Municipality), the air quality still does not meet the air quality standards for human health protection.

PM analyzers based on beta absorption are expensive. They cannot be applied in many places and with high frequency of measuring. This means that the cheap personal stations would be a good alternative provided, if their accuracy is satisfactory. Situating many personal stations which cover a large part of the city’s territory can help to identify sources of PM pollution that have been unknown before.

**LUFTDATEN.INFO PROJECT AND PM SENSORS WORLD APPLICATION**

In 2015 the citizens of Stuttgart, Germany also faced the problem connected to the high levels of air pollution with PM$_{10}$ and PM$_{2.5}$. The idea to organise a large scale civil control over the air quality, by deployment of many private stations, was born. For 2 years, a network consisting of 2349 stations was built covering the major cities of Germany under the Luftdaten.info project.

Luftdaten.info project uses Optical Particle Sizer sensors as SDS011 and temperature, humidity and atmospheric pressure sensor as BME 280. Technical Parameters of sensor SDS011 are given in Table 1. The sensors count particles of a certain size in a given volume of air. By use of an appropriate model the number of particles is converted into μg m$^{-3}$ PM$_{10}$ or PM$_{2.5}$. Worldwide, a large number of experiments have been carried out to check and prove the effectiveness and reliability of sensors similar to those used in Germany [7 - 9].

Adaptive coefficients are used to improve the model accuracy in conversion the number of particles into mass

<table>
<thead>
<tr>
<th>Item</th>
<th>Parameter</th>
</tr>
</thead>
<tbody>
<tr>
<td>Measurement parameters</td>
<td>PM$<em>{2.5}$, PM$</em>{10}$</td>
</tr>
<tr>
<td>Range</td>
<td>0.0 - 999.9 μg/m$^3$</td>
</tr>
<tr>
<td>Temperature range</td>
<td>Storage environment: -20 - 60ºC</td>
</tr>
<tr>
<td></td>
<td>Work environment: -10 - 50ºC</td>
</tr>
<tr>
<td>Humidity range</td>
<td>Storage environment: Max 90 %</td>
</tr>
<tr>
<td></td>
<td>Work environment: Max 70 %</td>
</tr>
<tr>
<td>Air pressure</td>
<td>86 KPa - 110KPa</td>
</tr>
<tr>
<td>Corresponding time</td>
<td>1 s</td>
</tr>
<tr>
<td>Minimum resolution of particle</td>
<td>0.3 μm</td>
</tr>
<tr>
<td>Counting yield</td>
<td>70 %@0.3μm</td>
</tr>
<tr>
<td></td>
<td>98 %@0.5μm</td>
</tr>
<tr>
<td>Relative error</td>
<td>Maximum of ± 15 % and ±10 μg/m$^3$</td>
</tr>
</tbody>
</table>
concentration. These adaptive coefficients take into account the influence of the air temperature, humidity and pressure and what is very important, the nature and the properties of the counted particles, especially their density. The results obtained are quite various. The correlation coefficient between the data measured by use of the reference method and the sensors vary between 0.36 and 0.8 - 0.9 [10, 11].

Many different factors can strongly influence the PS measurements, including the PS location, the distance between the PS and the corresponding AMS, the PM characteristics, even the wind direction. Currently, the sensors are unsuitable for indicative monitoring purpose in EU since they generally cannot meet the data quality objectives as specified in the 2008/50/EC directive [12]. Nevertheless, the use of sensors private stations for registry of PM in the air, is still interesting although serious error are possible in converting the number of particles into mass concentration. The results of their application show that PS provide coarse information about air quality, but they are able to categorise the air quality as good, moderate or heavy polluted.

LUFTDATEN.INFO PROJECT IN BULGARIA
The Luftdaten.info project in Bulgaria was launched in 2016 with the deployment of the first stations in Sofia. There are 267 stations in Bulgaria and this number is increasing continuously. This ranks Bulgaria in the second place after Germany. The stations measure air quality in Sofia, Varna, Plovdiv and other large and small Bulgarian towns. The data are presented in real time on https://airtube.info. In Sofia, have been installed and are in use 166 private stations. They provide data about the PM$_{10}$ and PM$_{2.5}$ concentration, the pressure, the temperature and the humidity. PS provide records for periods of 240 seconds. Those data are used to calculate the daily averaged values of PM$_{10}$ and PM$_{2.5}$ concentrations.

It was mentioned already, the personal stations could be adapted and used together with the AMS included in the National Monitoring System even more when only one AMS provides data about PM$_{2.5}$ concentration. The ideal case is the PS to be located very close to the AMS and to work at the same height from the ground level. At the moment, this requirement can be satisfied and the closest PS was chosen for the purpose of this study. The parameters of the four couples AMS and PS are given in Table 2. Their location is presented on Fig. 1.

The software used for converting the number of particles into mass concentration should not be applied before verification. Something more, such verification and validation should be done for each site of measuring. That is why the purpose of the present study is to check and evaluate the applicability and accuracy of these measurements on the territory of Sofia by statistical analysis of PS and AMS data series.

**Table 2. Characteristics of AMS and PS couples in Sofia municipality.**

<table>
<thead>
<tr>
<th>AMS</th>
<th>AMS coordinates</th>
<th>PS No</th>
<th>PS coordinates</th>
<th>Distance, $m$ AMS - PS</th>
<th>Period* from to</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hipodruma</td>
<td>42.680526° N 23.296797° E</td>
<td>3473</td>
<td>42.680000° N 23.293000° E</td>
<td>315</td>
<td>13.06.2017 - 12.09.2017</td>
</tr>
<tr>
<td>Pavlovo</td>
<td>42.669840° N 23.268299° E</td>
<td>5299</td>
<td>42.671000° N 23.258000° E</td>
<td>860</td>
<td>27.08.2017 - 12.09.2017</td>
</tr>
<tr>
<td>Drujba</td>
<td>42.666506° N 23.400075° E</td>
<td>3101</td>
<td>42.665221° N 23.391905° E</td>
<td>690</td>
<td>30.05.2017 - 12.09.2017</td>
</tr>
<tr>
<td>Mladost</td>
<td>42.655134° N 23.383278° E</td>
<td>1102</td>
<td>42.655120° N 23.375916° E</td>
<td>600</td>
<td>13.03.2017 - 12.09.2017</td>
</tr>
</tbody>
</table>

*operation period of the private station
STATISTICAL ANALYSIS

The daily averaged values of PM$_{10}$ concentration at the AMS and the corresponding PS are presented on Figs. 2 - 5. Fig. 6 represents a similar information about PM$_{2.5}$ concentration at AMS Hipodruma. It can be seen that changes in PM concentration measured with PS and AMS have a similar character with one exception - PM$_{10}$ concentration at Mladost AMS as shown on Fig. 4. Here quite different values have been measured by PS for the end of May and the beginning of June 2017. The big deviations could be caused by plant pollens, contamination of station chamber or unknown sensor issue. Taking into

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Hipodruma, N = 92</th>
<th>Hipodruma, N = 92</th>
<th>Drujba, N = 95</th>
<th>Mladost, N = 158</th>
<th>Pavlovo, N = 11</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>PM$_{2.5}$</td>
<td>PM$_{10}$</td>
<td>PM$_{10}$</td>
<td>PM$_{10}$</td>
<td>PM$_{10}$</td>
</tr>
<tr>
<td>$\sigma_o$</td>
<td>5.02</td>
<td>8.96</td>
<td>8.54</td>
<td>7.80</td>
<td>13.18</td>
</tr>
<tr>
<td>$\sigma_p$</td>
<td>3.81</td>
<td>4.68</td>
<td>4.41</td>
<td>6.20</td>
<td>6.09</td>
</tr>
<tr>
<td>RMSE</td>
<td>5.29</td>
<td>16.92</td>
<td>12.09</td>
<td>10.68</td>
<td>19.79</td>
</tr>
<tr>
<td>$r$</td>
<td>0.82</td>
<td>0.72</td>
<td>0.69</td>
<td>0.70</td>
<td>0.89</td>
</tr>
</tbody>
</table>

Table 3. Statistical parameters.

Fig. 2. PM$_{10}$ concentration at Hipodruma during the period 13.06 - 12.09.2017, µg m$^{-3}$.

Fig. 3. PM$_{10}$ concentration at Drujba during the period 30.05 - 12.09.2017, µg m$^{-3}$.

Fig. 4. PM$_{10}$ concentration at Mladost during the period 13.03 - 12.09.2017, µg m$^{-3}$.
account the altitude of the remaining data, the mentioned outliers were excluded (Fig. 7).

Some statistic characteristics of the data series shown on Figs. 2, 3, 5, 6 and 7, such as standard deviation of AMS measurements

\[
\sigma_O = \frac{1}{N} \sum_{i=1}^{N} (O_i - \bar{O})^2
\]  

\[
\sigma_P = \frac{1}{N} \sum_{i=1}^{N} (P_i - \bar{P})^2
\]  

root mean square error

\[
RMSE = \sqrt{\frac{1}{N} \sum_{i=1}^{N} (P_i - O_i)^2}
\]
and correlation coefficient

\[ r = \frac{1}{N-1} \sum_{i=1}^{N} (O_i - \bar{O})(P_i - \bar{P}) \]

\[(4)\]

are given in Table 3.

The data analysis shows comparatively strong positive correlation between the results for PM$_{10}$ from AMS and PS (after removal of outliers at station Mladost). In addition, the values of PM$_{10}$ and PM$_{2.5}$ concentrations, registered by the PS, are significantly lower than those measured at AMS.

To improve the PS measurements, correction coefficients are to be used so that AMS and PS values of PM$_{10}$ and PM$_{2.5}$ concentration to be as close as possible. The Least-squares method is applied for that purpose. The measured values are corrected according to the model

<table>
<thead>
<tr>
<th>PM Station</th>
<th>$b$</th>
<th>$R^2$</th>
<th>RMSE</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hipodruma, PM$_{2.5}$</td>
<td>1.37802</td>
<td>0.9617</td>
<td>3.11</td>
</tr>
<tr>
<td>Hipodruma, PM$_{10}$</td>
<td>2.05095</td>
<td>0.9494</td>
<td>6.95</td>
</tr>
<tr>
<td>Mladost, PM$_{10}$</td>
<td>1.50440</td>
<td>0.9242</td>
<td>6.94</td>
</tr>
<tr>
<td>Pavlovo, PM$_{10}$</td>
<td>2.18699</td>
<td>0.9728</td>
<td>5.82</td>
</tr>
<tr>
<td>Drujba, PM$_{10}$</td>
<td>1.80771</td>
<td>0.9236</td>
<td>6.54</td>
</tr>
</tbody>
</table>

Fig. 8. Model accuracy for measurement of PM$_{2.5}$ in station Hipodruma.

Fig. 9. Model accuracy for measurement of PM$_{10}$ in station Hipodruma.

Fig. 10. Model accuracy for measurement of PM$_{10}$ in station Mladost.

Fig. 11. Model accuracy for measurement of PM$_{10}$ in station Pavlovo.
where, $y$ is the calculated and $x$ - the measured PS value of PM$_{10}$ or PM$_{2.5}$ concentration. The values of coefficient $b$ for the sensor stations are given in Table 4, and the model accuracy is illustrated on Figs. 8 - 12. Models for all stations, except for Drujba, are characterized by relatively high accuracy. At Drujba the error of the model is relatively high, but only a few observations with deviation over 50% have been registered.

**DISCUSSION**

Table 4 shows that the correction coefficients for PM$_{10}$ varied from 1.5 to 2.2, which could be due to the different origin of the predominant particles at the different sampling points. For example, the model coefficient for Mladost is 1.5, probably due to particles of predominantly fuel origin from road traffic and those for Hipodruma and Pavlovo (with values above 2) are probably determined by predominantly high density mineral particles.

According to the world literature and the present study, it is important to note that the accuracy of PS measurements and whether the measured values will be higher or lower compared to the referent methods, depend on the meteorological conditions and the origin of the particles. The sensors measure lower PM concentration values during the warm and higher during the cold half of the year compared to the referent methods.

That is why the derived coefficients are valid for the warmer period of the year and it is necessary to derive another set of coefficients for the cold half of the year. Without correction, the PS measurement error is large and their records may be misleading.

**CONCLUSIONS**

It was found in this study that:
- During the warm period of the year, PS measured significantly lower levels of fine particulate matter in the ambient air compared to AMS;
- It is necessary to derive an appropriate set of correction coefficients for the cold period of the year;
- A good correlation between the AMS and PS measurements has been established;
- The AMS and PS data sets for PM$_{2.5}$ demonstrate a significantly higher correlation than the PM$_{10}$ measurements.

The PS measurements of PM$_{2.5}$ concentration are characterized with a higher accuracy. That is why a lower correction coefficient is derived compared to the PM$_{10}$ data sets.

If accurate correction coefficients are derived, the PS can be used successfully for particulate matter measurement.

Further analyzes have to be performed with PS located close to AMS. So it will be possible to assess the accuracy of PS data better.

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