

SEASONAL CHANGES OF AEROSOL POLLUTANTS OVER BULGARIA

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Abstract: *In this work we present an investigation of seasonal behavior of atmospheric pollutant amounts over Bulgarian region. We use monthly averaged satellite and ground station data for the period of 2005 till 2018 and 2013 till 2018 respectively.*

СЕЗОННО ИЗМЕНЕНИЕ НА АТМОСФЕРНИТЕ ЗАМЪРСИТЕЛИ НАД БЪЛГАРИЯ

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Ключови думи: *дистанционни изследвания, атмосферни замърсявания*

Резюме: *В настоящата работа е представено изследване на сезонното поведение на атмосферните замърсители над територията на България. Използвани са средно месечни стойности на спътникови и наземни данни за периодите 2005–2018 и 2013–2018 съответно.*

Introduction

Broadly speaking, on the base of the spatial scales of the atmospheric processes, the atmospheric pollution can be studied on different scales. Here we will use the terms global for processes at continental scale, regional – for country level, and local - for city level.

There are two main methods for observation of the atmospheric composition – remote sensing and direct (in-situ) measurements near the surface. Each method has advantages and limitations, but their combination provides more complete information.

Example of global events reaching Bulgaria is the transport of Saharan dust over the Mediterranean and Aegean Sea. The source of pollution in this case (uplifting of mineral dust) is natural, located at the earth's surface, and the atmospheric circulation transports the particles far away. For studying such kind of events, it is better to use remote sensing methods, such as satellite data information. The larger the scale of the event is, the lower the needed spatial resolution of the satellite data for its registration is. Still, in order to relate satellite and in-situ observations high resolution satellite data are needed.

The regional and local pollution events originate from a number of different sources, such as soil, industrial facilities, ship transport, urban traffic, agricultural activities, etc. The sources are on or at a stack-height near the surface (point and area sources covering limited spatial scales), so it is better to investigate those kind of pollution events using near-surface measurement stations.

Some local sources may have impact on regional scale, as big industrial facilities and coal fired thermal power plants. In this case the pollutants can rise in the atmosphere and travel with the

winds, thus polluting large distant areas. Such kind of events has to be investigated combining in-situ and remote sensing methods.

In this project the activities foresee combined use of satellite data and data from near-surface measurement stations. This is a way to reach a better understanding of the whole pollution picture.

For investigation on the seasonal pollution behavior we choose to use different data sources as follows:

Optical images from Terra and Aqua satellites for the period of 2004–2018 [1].

Monthly values for AAI and NO₂ and SO₂ from MetOp A, B and C satellites for the period from 2007 to March 2019 [2]

Monthly images of NO₂ from Sentinel 5P satellite for first three months of 2019.

Daily data from selected near-surface stations for the period 2013-2018 [3].

Seasonal changes of the global atmospheric pollution

We can consider the sand storms from Africa as the main global dust pollution source over the Balkans. In Fig.1 we show the monthly variation of the number of days with sand storms directed towards the Balkan region for the period 2005–2018. The events are registered on the base of optical satellite data from *Terra* and *Aqua* satellites.

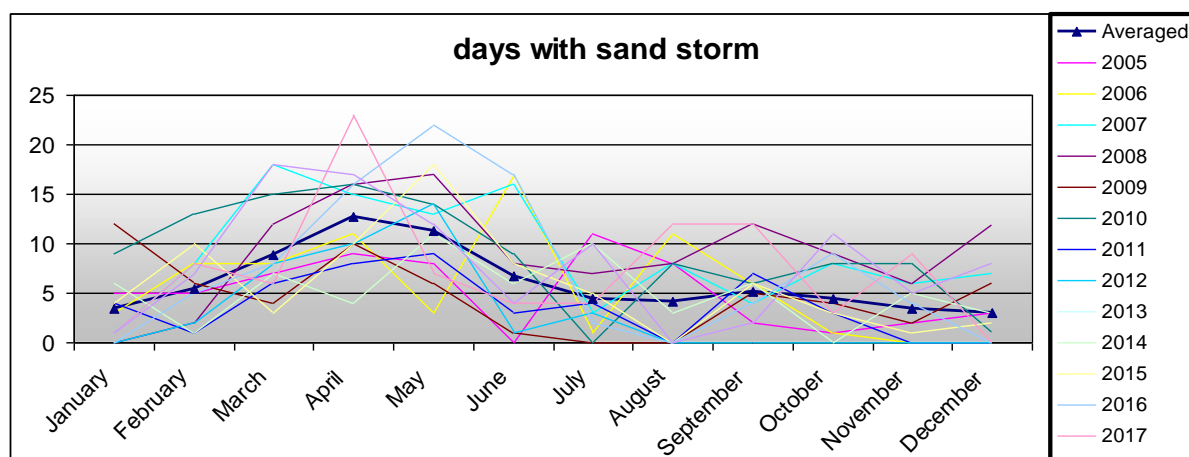


Fig. 1. Number of days with sand storms from Africa, directed to the Balkans

The number of days has monthly and yearly variability. The maximum number of days with sand storms, averaged over the 13-year period, is for April, while in specific years it occurs from March to May. Almost the same number of sand storm events is registered on the base of AAI from *MetOp A* and *MetOp B* satellites for the period 2007–2018.

For the investigation of the seasonal behavior of NO₂ vertical column density (VCD) over the Balkans, monthly values of NO₂ VCD from *MetOp A* satellite for the period from 2007 to 2018, and from *MetOp B* satellite for the period from 2013 to 2018 were used. For comparison, *Sentinel 5 P* data for the period from February 2018 until February 2019 and from *MetOp C* for the first three months of 2019 were used. All mentioned data show similar seasonal changes – a small increase in January, November and December. In Fig. 2 examples for monthly values of NO₂ VCD distribution for different months of 2017 are shown.

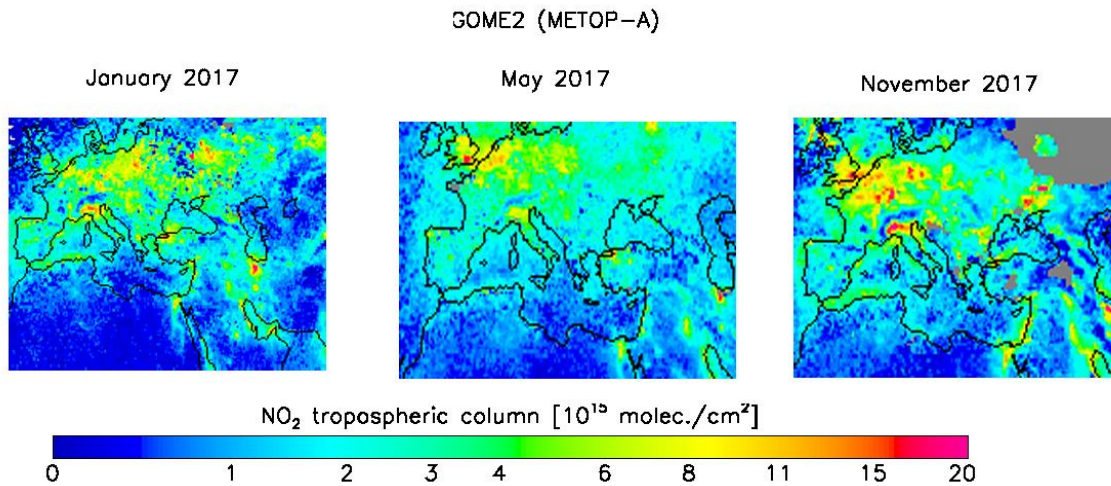


Fig. 2. Monthly averaged NO₂ distribution form MetOp A data

Seasonal changes of the local atmospheric pollution

The analysis here is presented for some selected points /urban and rural/ based on data from satellite and in situ measurements.

The region around the town of Stara Zagora is considered as one of the main industrial pollution sources in Bulgaria. The reason is the open coal mining and the complex of thermal power plants 'Maritza-Iztok'. In Fig. 3 the number of days with pollution for the period 2004–2011 registered on the basis of the optical satellite data from *Terra* and *Aqua* satellites is shown. The number of days with significant pollution in the region of Stara Zagora decreased strongly after 2012. This is confirmed also in [4] where the Aura OMI observations of regional SO₂ and NO₂ pollution changes from 2005 to 2015 over Eastern Europe and for the region of Stara Zagora are discussed. The vertical column density of SO₂ is compared for three periods 2005–2007, 2009–2011, 2013–2015, showing strong decreasing trend. Estimating the SO₂ emission retrievals and consistency of satellite and surface measurements with reported emissions in [5], these SO₂ emission reductions in the region of Stara Zagora after 2012 are also confirmed.

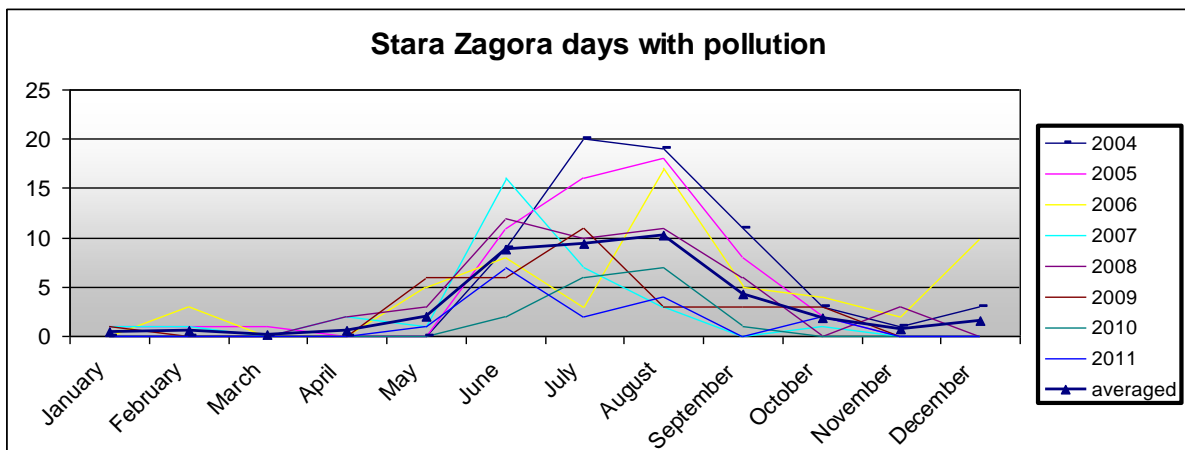


Fig. 3. Number of days with atmospheric pollution over the region of Stara Zagora

For the investigation of the local pollution, several near-surface stations from the monitoring network of the Executive Environmental Agency (ExEA) were chosen, as shown in Fig. 4. The abbreviation RIOSV in the Figures stands for the Regional Inspectorates of the ExEA.

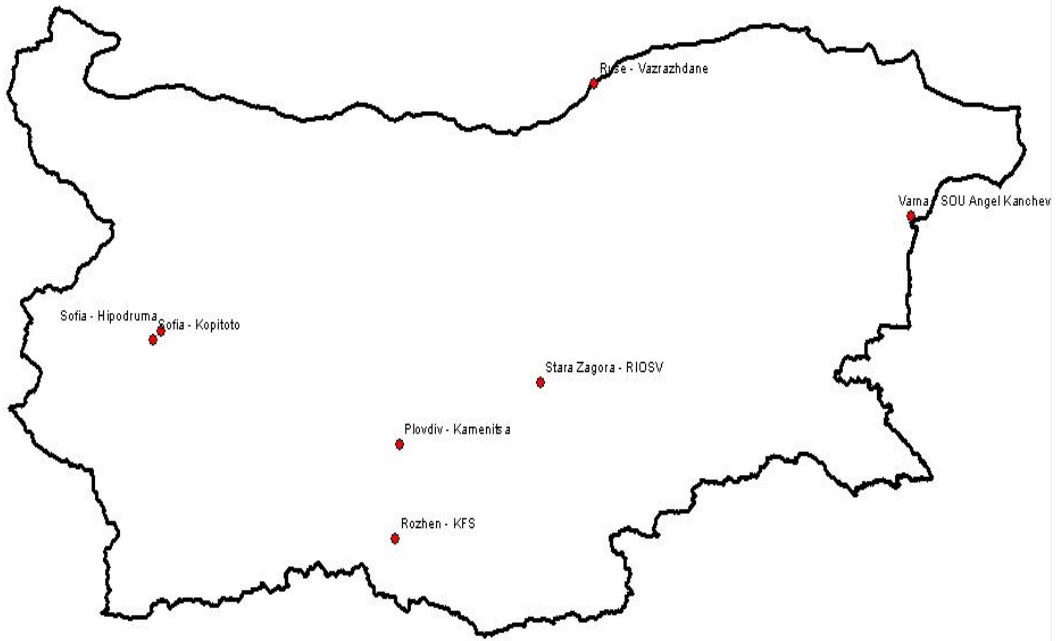


Fig. 4. Locations of the selected the near-surface stations for atmospheric pollution monitoring

The types of the selected stations are as follows:

- Big cities – Sofia, Varna, Plovdiv, and Ruse;
- Industrial region with well known pollution origins – Stara Zagora;
- Rural places far away from pollution sources (rural remote) – Rozhen and Kopitoto.

Data for PM_{2.5}, PM₁₀, SO₂, NO and NO₂ in the period 2013–2018 were analysed for all of the stations. As Plovdiv is the focus of interest in this project, the monthly averaged values for PM_{2.5}, PM₁₀, NO₂ and SO₂ concentrations at station Kamenitza are shown in Fig. 5 to Fig. 8, respectively.

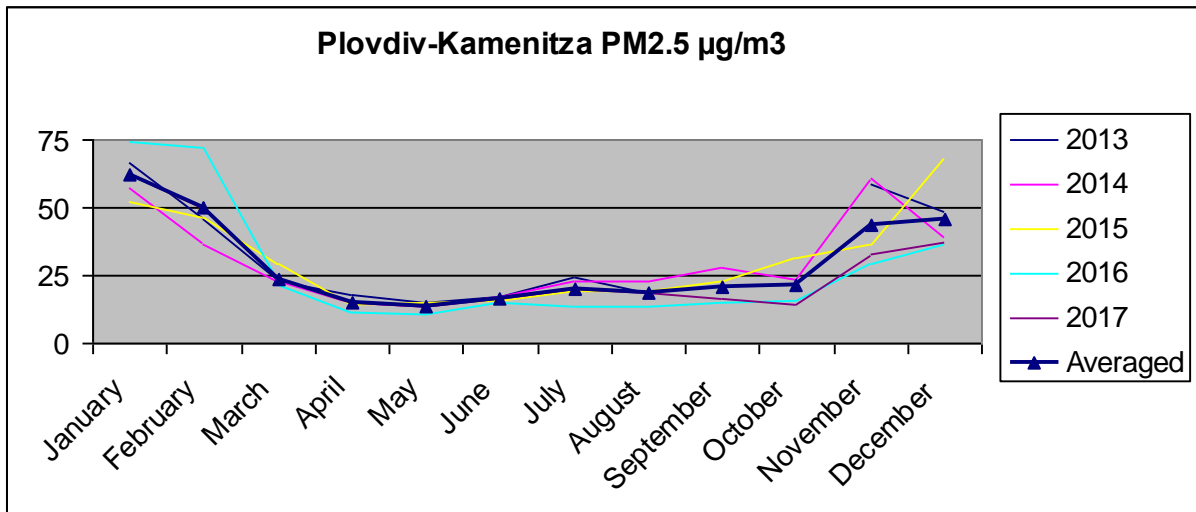


Fig. 5. PM_{2.5} monthly values (µg/m³) at Kamenitza- Plovdiv station

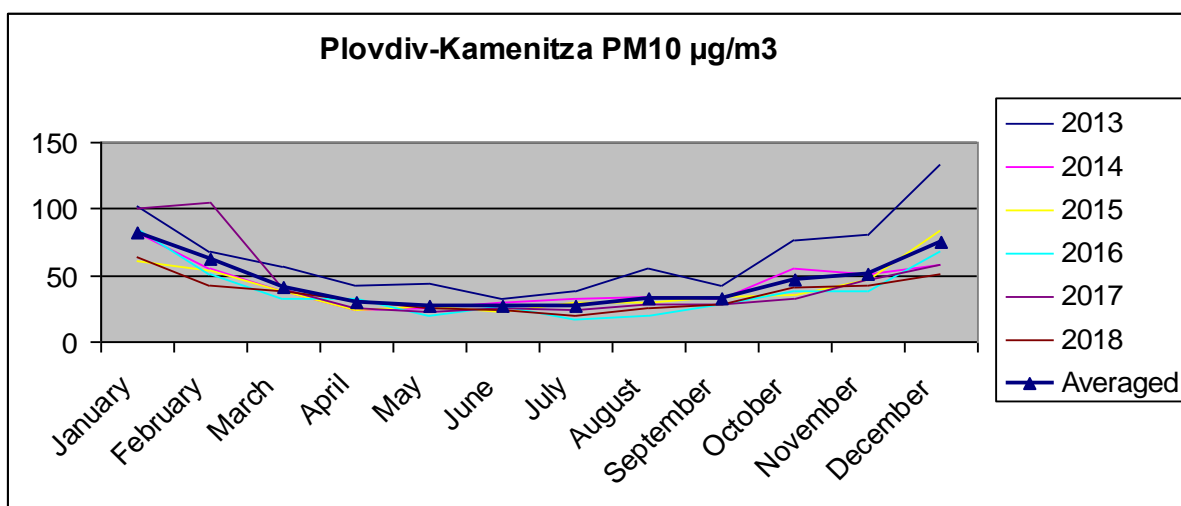


Fig. 6. PM10 monthly values ($\mu\text{g}/\text{m}^3$) at Kamenitza - Plovdiv station

As seen in Fig. 5 and Fig. 6, PM2.5 and PM10 concentrations show similar seasonal behavior with higher values in winter and lower in summer.

The SO₂ concentrations (Fig. 7) show low averaged values with no significant seasonal changes. In Fig. 8 and Fig. 9 the monthly variation of NO and NO₂ concentrations in Plovdiv are presented. The graphs show increased NO and NO₂ values during the winter months, similar to those registered with MetOp. The concentrations of NO₂ are higher than those of NO and show stronger seasonal variations.

For providing more information across the country in Fig. 10 and Fig. 11 the PM2.5 concentrations at the urban stations Hipodruma (in Sofia) and Vazrajdana (in Ruse) are shown.

As seen in Fig. 5, Fig. 10 and Fig. 11, the monthly variation for PM2.5 is similar in Sofia, Ruse and Plovdiv. It has a maximum during the winter months – January, February and December. The highest values are registered in Sofia, followed by Plovdiv and Ruse.

The averaged values for PM10 in the chosen big cities in Bulgaria show the same behavior as PM2.5, only with higher values.

For the investigation of the seasonal distribution of the pollutants over non-urban areas, two mountain stations not directly affected by pollution sources are chosen. In Fig. 12 and Fig. 13 the monthly variation of PM2.5 is shown, respectively at Rozhen (1760 m a.s.l) and Kopitoto (1350 m a.s.l).

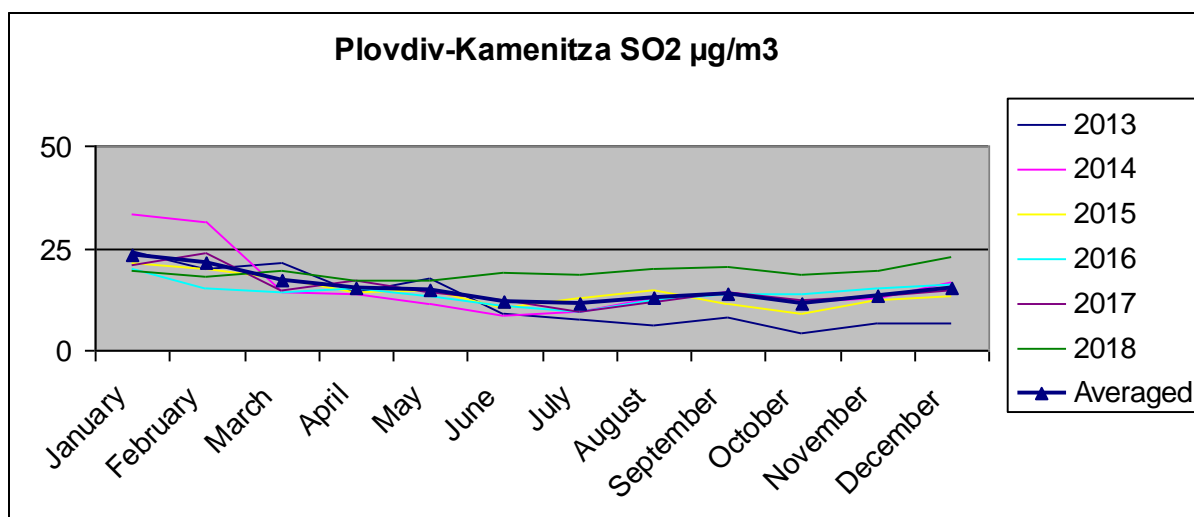


Fig. 7. SO₂ monthly values ($\mu\text{g}/\text{m}^3$) at Kamenitza - Plovdiv station

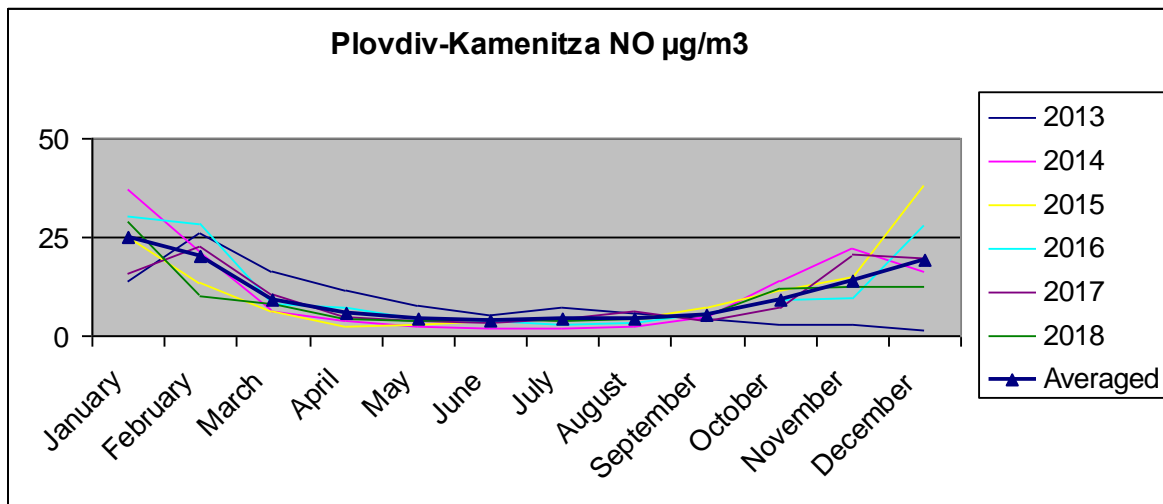


Fig. 8. NO monthly values ($\mu\text{g}/\text{m}^3$) at Kamenitza - Plovdiv station

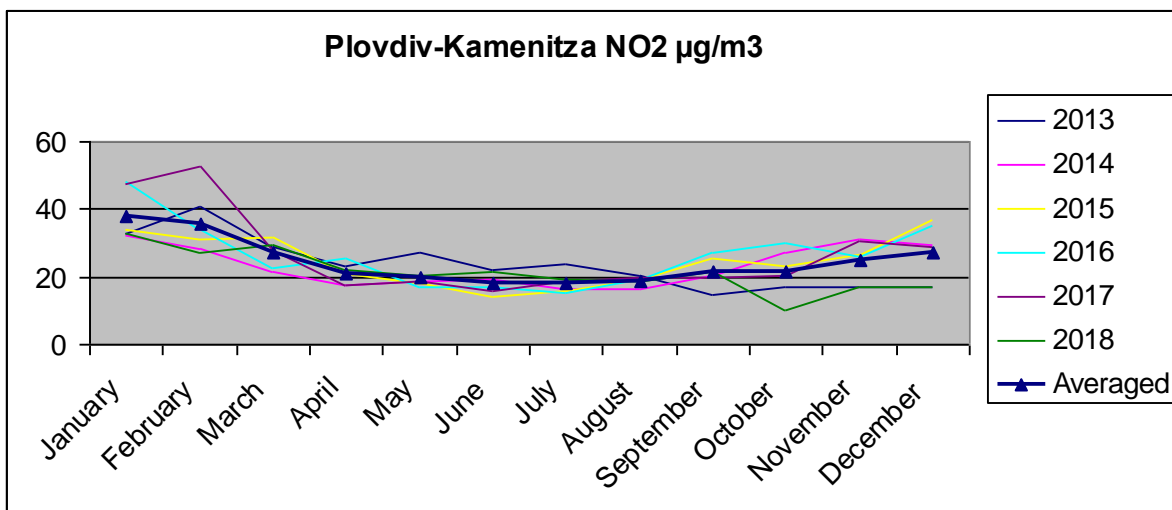


Fig. 9. NO₂ monthly values ($\mu\text{g}/\text{m}^3$) at Kamenitza- Plovdiv station

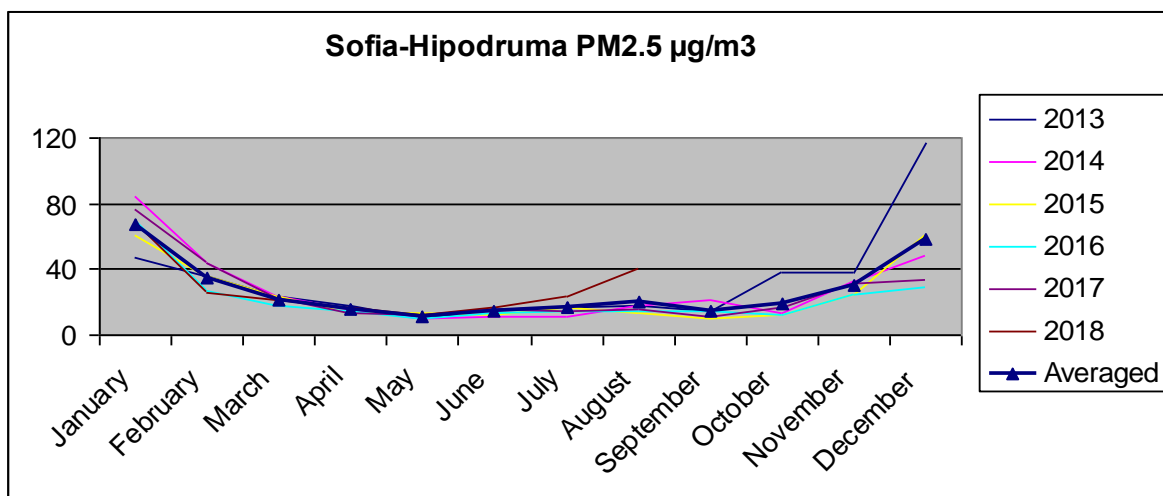


Fig. 10. PM_{2.5} monthly values ($\mu\text{g}/\text{m}^3$) at Hipodruma in Sofia

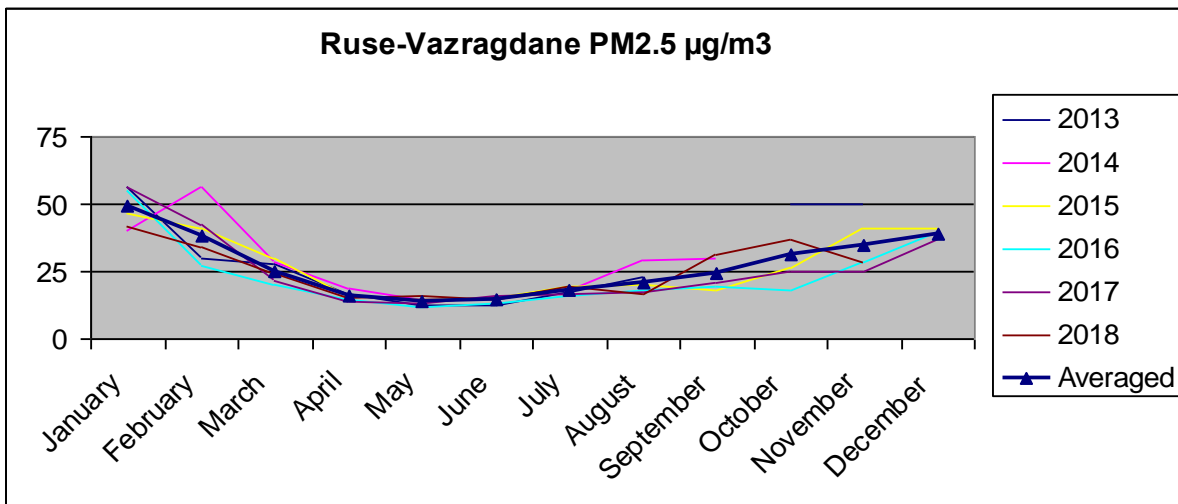


Fig. 11. PM2.5 monthly values (µg/m³) in Ruse

As seen in Fig. 12 and Fig. 13, the seasonal behavior of PM2.5 concentrations at the mountain sites is quite different from that in the big cities. The maximal values are registered in spring and summer months. Main reason for this is the seasonal variation of the height of the atmospheric boundary layer (ABL), which is low in winter and keeps pollution below the mountain tops. In summer, the ABL height reaches above the mountain tops and brings the urban pollution up. The high values at Kopitoto in spring may be influenced also by Saharan sand storms (reference Fig. 1). The PM2.5 concentrations at mountain stations are much lower than in urban areas.

As mentioned above, one well known polluted industrial region in Bulgaria is around Stara Zagora. In Fig. 14 the monthly variation of PM2.5 concentrations at a monitoring station in the city of Stara Zagora is shown. The PM2.5 values are low and their seasonal changes are negligible. Historically, the region was a “hot spot” for PM pollution before 2012. Then measures were taken and the emissions were reduced significantly. Previous investigations in the region showed that even when PM pollution was registered from satellite observations, no high values were measured at the ExEA monitoring stations near the ground [6, 7]. Similarly, decrease of SO₂ concentrations was observed in the region after 2012 [8].

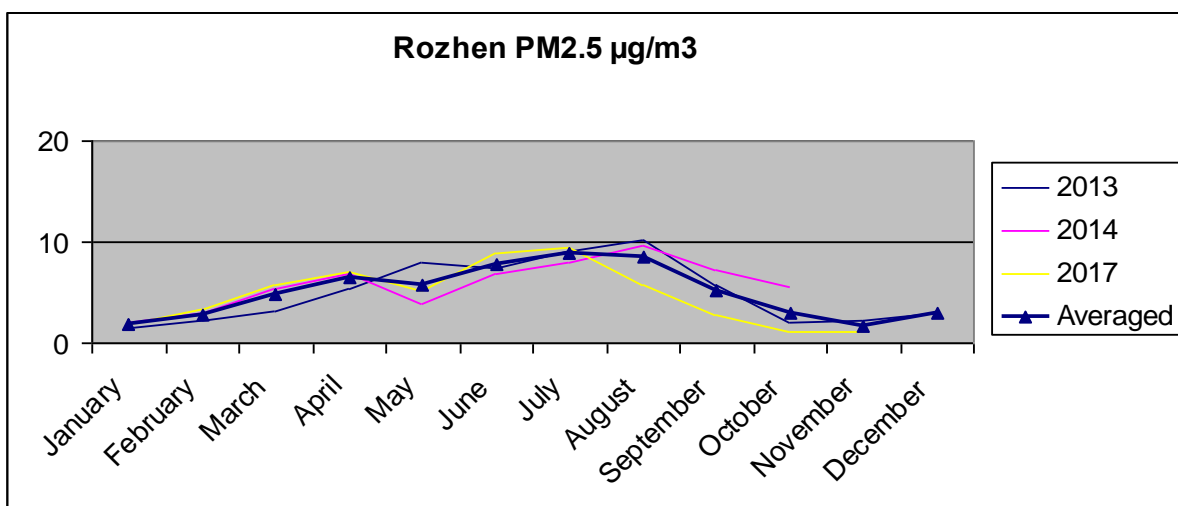


Fig. 12. PM2.5 monthly values (µg/m³) at Rozen

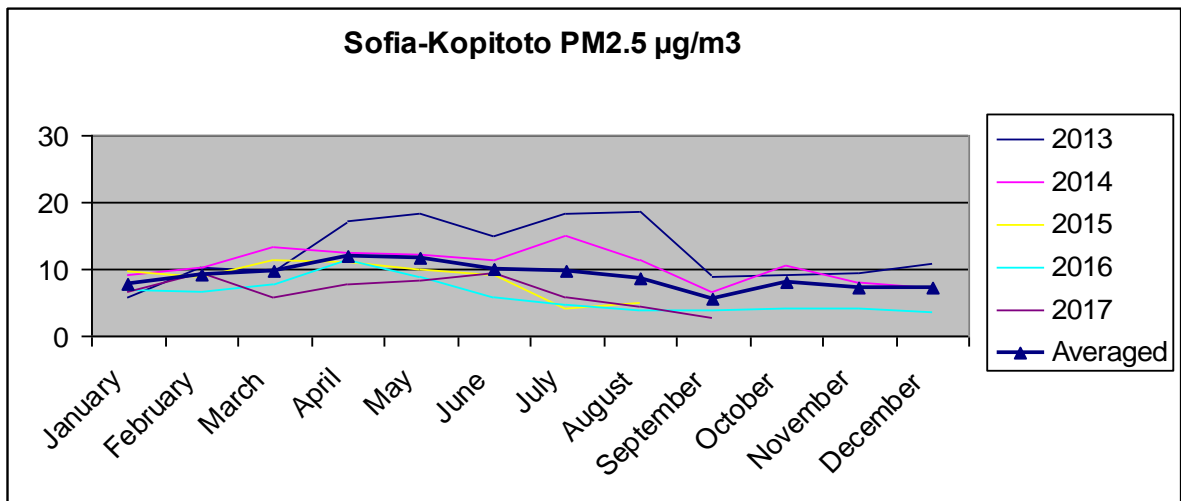


Fig. 13. PM2.5 monthly values (µg/m³) at Kopitoto

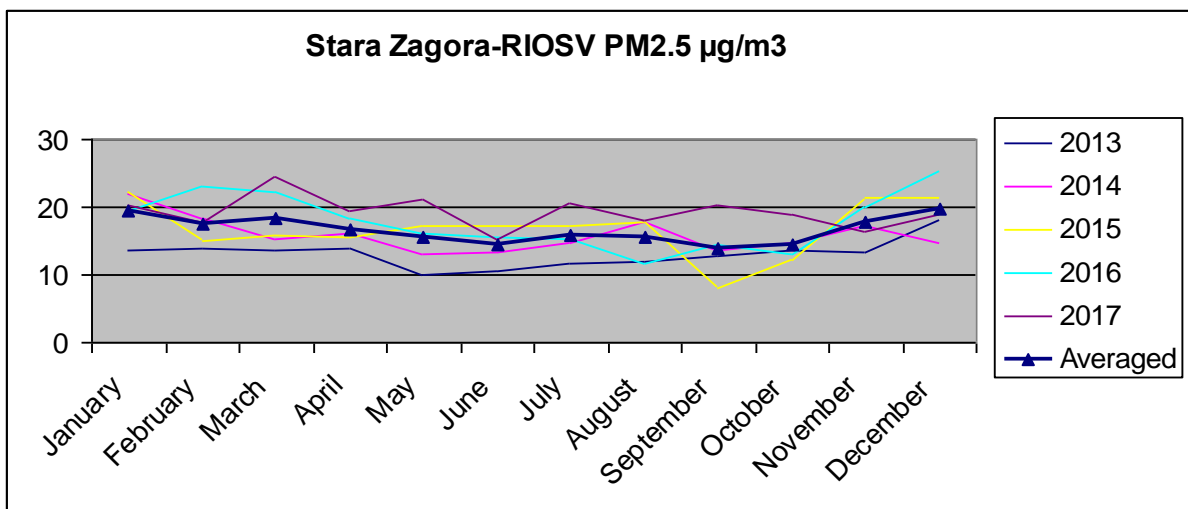


Fig. 14. PM2.5 monthly values (µg/m³) in Stara Zagora

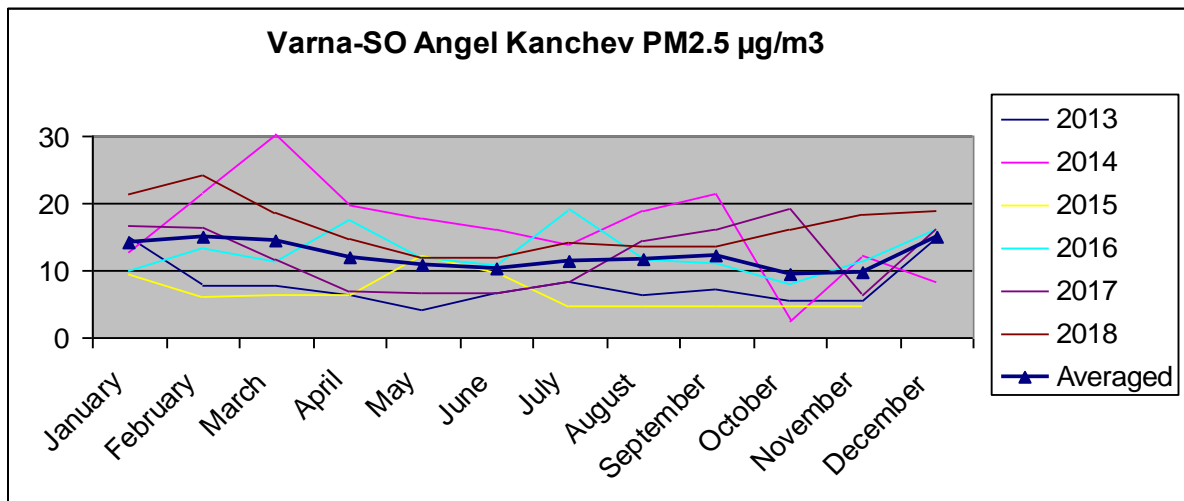


Fig. 15. PM2.5 monthly values (µg/m³) in Varna

At the end, in Fig. 15, the monthly variation of PM2.5 in Varna, located at the Black Sea coast is shown. Low values and small seasonal variations are observed.

Comparison of the seasonal pollution behavior registered on the base of different data sources
 For the comparison of dust pollution over Bulgaria registered from the satellites and near-surface stations, monthly values of AAI from the MetOp A and B satellites are used. The behavior of AAI from MetOp A for the period of 2007–2015 for Sofia, Plovdiv and Varna regions, respectively, is shown in Fig. 16, Fig. 17 and Fig. 18. The record stops in 2015, as after the middle of the 2015 AAI data from the MetOp A satellite decrease significantly.

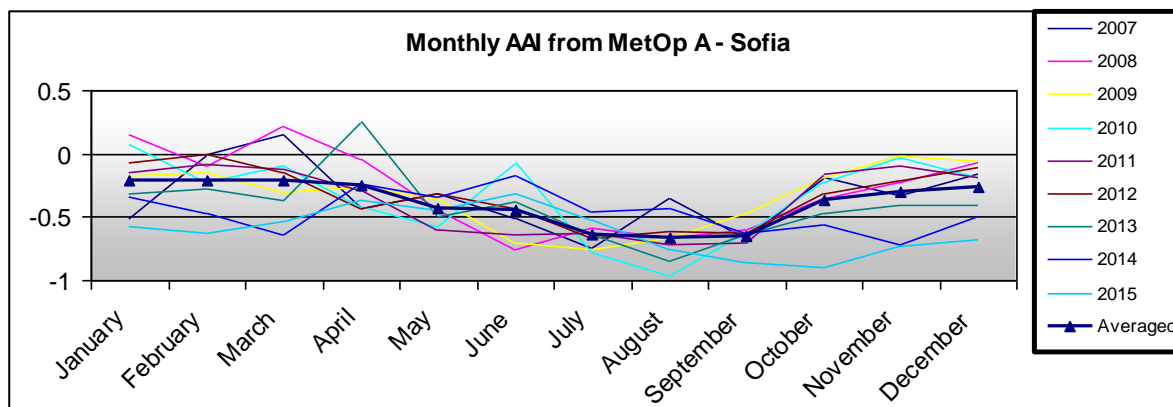


Fig. 16. of AAI in Sofia

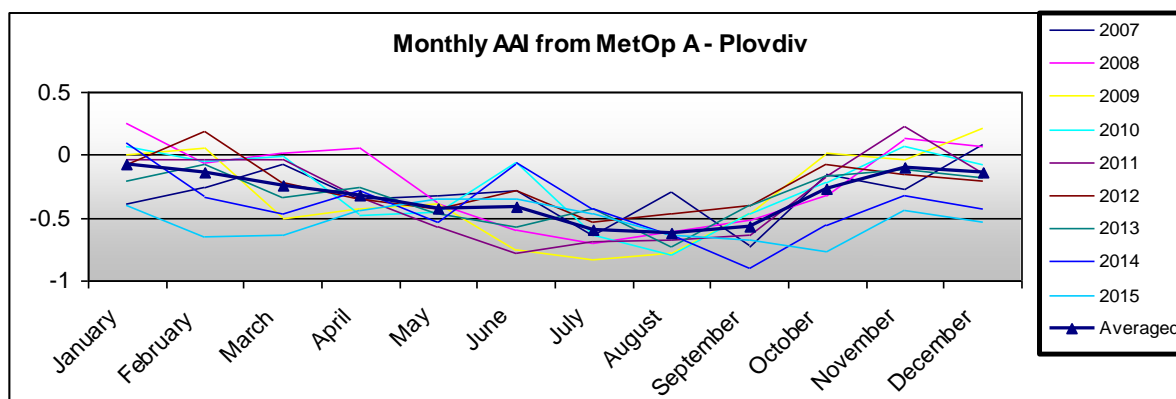


Fig. 17. Monthly variation of AAI in Plovdiv

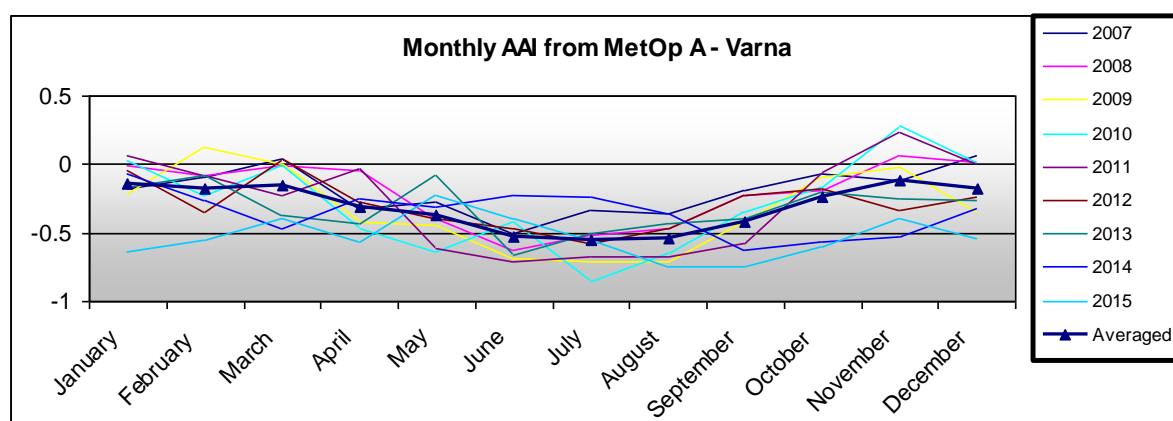


Fig. 18. Monthly variation of AAI in Varna

The monthly variation of AAI from MetOp B satellite for the period of 2012–2018 for Plovdiv is shown in Fig. 19 for comparison.

Almost all monthly AAI values in Figs. 15–18 are negative. A negative AAI value corresponds to presence of cloud coverage and scattering aerosols. Higher AAI values are registered in winter and spring.

To compare observations from the three MetOp satellites, monthly AAI data for March 2019 are used. The results are shown in Fig. 20.

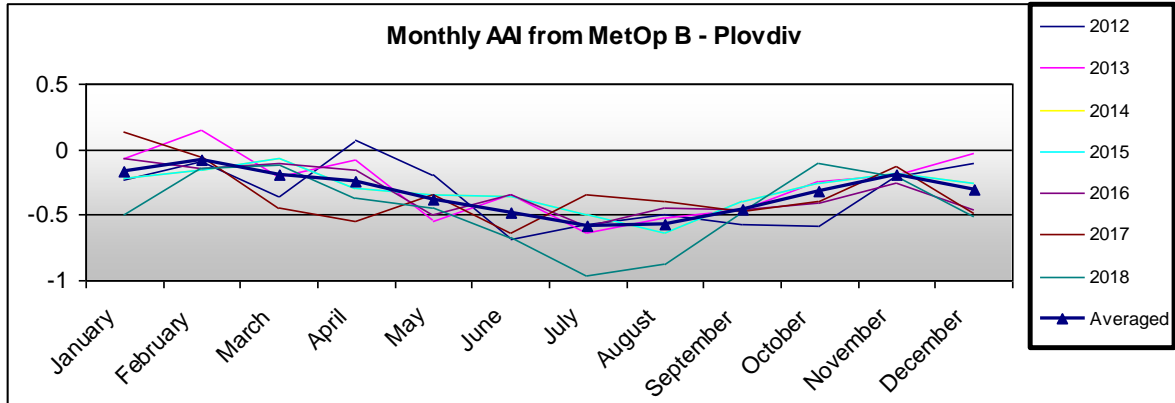


Fig. 19. Monthly variation of AAI in Plovdiv

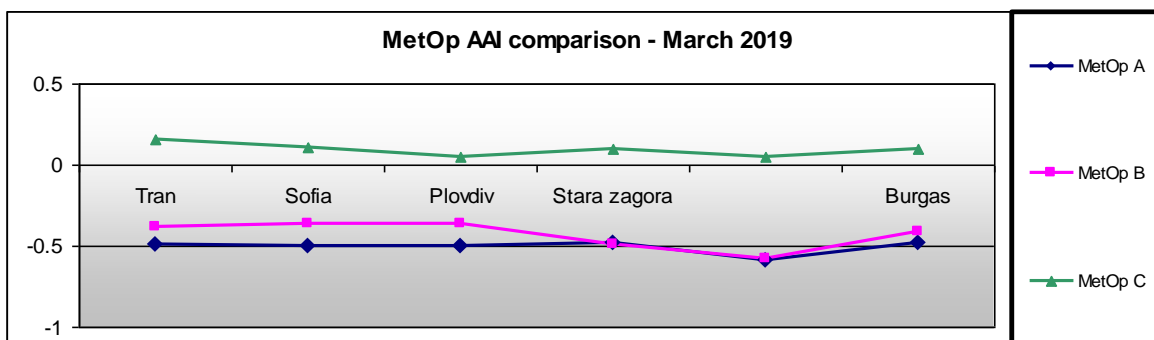


Fig. 20 Comparison of AAI data from three MetOp satellites at selected locations in March 2019

AAI values from MetOp-C satellite are higher than those from the two other satellites and the differences change with place. This means that, best is to use available AAI data from all satellites for a selected period and place, to compare and to study the reasons for the differences.

There are no available for us satellite monthly data for SO₂, but, on the basis of available daily data, we can say that SO₂ values are very small and show no significant seasonal changes. The same behavior is seen in Fig. 7 for near-surface measurements.

To study the seasonal behavior of NO₂ from satellite data Sofia, Plovdiv and Varna are chosen as big cities, and Stara Zagora as industrial region.

In Fig. 21 and Fig. 22 the monthly variation of NO₂ registered on the base of monthly data from the *MetOp A* satellite for the period 2007–2018 for the regions around Sofia and Plovdiv is shown, respectively. The values of NO₂ are given in relative units.

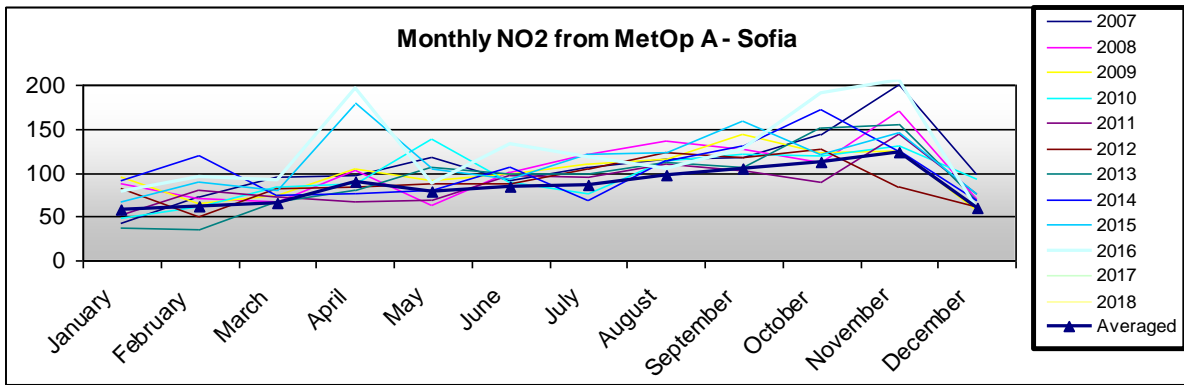


Fig. 21. Monthly variation of NO₂ around Sofia

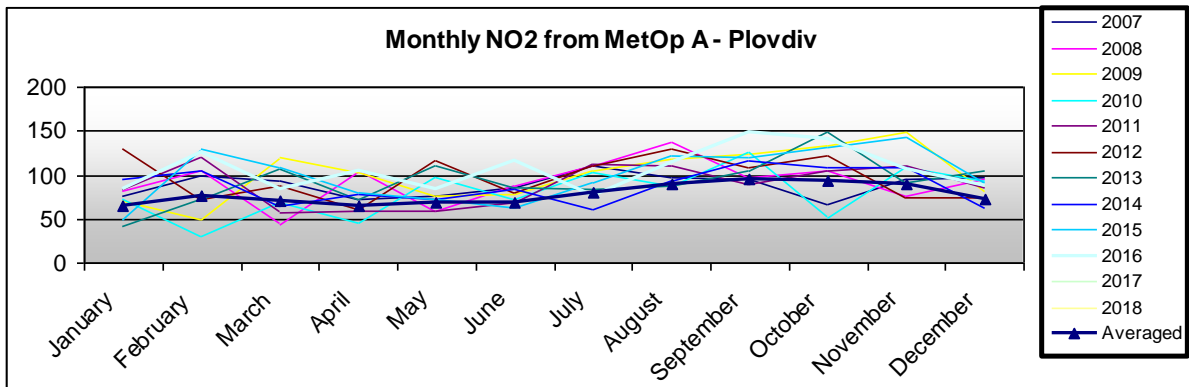


Fig. 22. Monthly variation of NO₂ around Plovdiv

As seen from the two Figures above, in both cases NO₂ shows a maximum during the autumn months. For Sofia region the well pronounced maximum of averaged over the years distribution is in November. In the region of Plovdiv the seasonal variation is smaller. In Stara Zagora region (not illustrated here) maximum values of NO₂ are registered during the summer months.

All the NO₂ data from the MetOp satellites show comparable values for the whole period of investigation. The data for NO₂ from Sentinel 5P, as presented in monthly images, is comparable with that from the MetOp satellites.

Conclusions

Comparing satellite data with the data from near-surface measurement stations, we must remember that:

A value for the total column content of pollutants over a region, is corresponding to the spatial resolution of the satellite's instruments, while at the near-surface stations the in-situ values (point values) for the concentrations are measured;

The satellite data contain both the influence of regional and global events;

For the dust pollution satellite data for AAI or AOD are available, while in-situ PM_{2.5} and PM₁₀ are registered separately. AAI and AOD include represent various types of existing aerosol;

In the presented analysis for the near-surface stations daily values of PM_{2.5} and PM₁₀ were used, while for other pollutants the temporal resolution was 1 hour.

The similar seasonal behavior seen in the data from different origins (satellite, in-situ) provides an opportunity to use them together, but only after a good understanding of their differences.

In a summary:

- Based on satellite data for 9 to 12 years, the monthly variation of AAI, NO₂ and SO₂ at selected big cities in Bulgaria is analysed.
- Satellite data show increased amount of dust and gaseous components in autumn and winter above big cities.
- The registered differences in the seasonal behavior of pollution ingredients at the chosen places could be explained with local sources of pollution, long lasting unfavorable meteorological conditions or pollutants transported from longer distance.

Acknowledgments

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