SOURCE IDENTIFICATION OF REGIONAL PM10 CONCENTRATIONS DURING EPISODES OF AIR POLLUTION IN EASTERN EUROPEAN AREA

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Abstract: Air pollution due to particulate matter with an aerodynamic diameter < 10 μ/m^3 was investigated in Eastern European Area. Available PM10 and PM2.5 measurements were analysed for urban and rural background Croatian stations in the period from 2006 to 2011 in order to assess the current level of air pollution with particles according to the defined limit values (CAFÉ Directive; EC, 2008). Significant difference between coastal and continental PM10 levels was found with high PM10 concentrations in urban and rural continental areas while at coastal stations concentrations were considerably lower and under the limit values. Rural background PM levels differed substantially for rural background stations going from annual average concentration ~ 30 µg/m3 at Kopački rit to 5 µg/m3 at Hum located on island of Vis. Sources of PM10 pollution events were identified during two episodes in November 2011 at urban and rural stations in Croatia, Hungary and Serbia using monitored air quality and meteorological data, backward air mass trajectories and results of a NWP WRF model. According to the calculated back trajectories all analysed stations were under the influence of the same air masses, eastern for the first episode and north eastern for the second episode. This is supported with the results of ARW WRF model. Elevated PM10 concentrations in the second episode are the consequence of local anthropogenic sources, regional transport as well as of the resuspension over the bare natural and agricultural surfaces in Hungary, Serbia and Croatia driven by the strong wind.

Key words: *Key words should be written in **9 pt** Times New Roman Italic**.***

INTRODUCTION

It has been shown that elevated atmospheric particulate matter concentrations have significant adverse health effects (Samet et al., 2000; Peters et al., 2001; Pope et al., 2002) and affects ecosystems and visibility. Particulate matter (PM) consists of complex mixture of solid and liquid particles of organic matter, core elements, secondary inorganic aerosols and trace metals and come from variety of natural and anthropogenic sources that might by directly emitted in the atmosphere or formed secondarily in atmospheric chemical reactions (Juda-Rezler et al. 2011).

Two episodes with considerably increased daily PM10 concentrations (>100 μ/m^3), were identified that were timely harmonized and relatively uniform in magnitude at almost all analysed continental stations in Croatia, Hungary and Serbia in the period starting from 2 to 20 of November 2011. First peak in PM10 daily concentrations occurred at almost all analysed stations on 2 November. The second peak comprised stations located on the eastern part of Hungary on 13 November and reached the stations on the west of Hungary on 17 November, higher PM10 concentrations occurred on Serbian stations on 15 November and on Croatian on 16 November. Increase in PM10 concentrations that occurred on 2 November 2011 was followed by a sudden decrease in PM10 concentrations at almost all analysed stations caused by a dilution with strong northeast wind. Second air pollution episode started after the decrease of north-east wind when stable stagnant atmospheric conditions were prevailing under the influence of high-pressure system over the Central and Eastern Europe.

DATA AND METHODS

Hourly PM10 and NO_2 measurements were used in the analyses from the available monitoring sites in Croatia, Hungary and Serbia during 2011. The location of all stations is shown in Fig.1.

Croatian stations Zagreb-1, Rijeka-1, Rijeka-2, Osijek, Sisak, Kutina and Slavonski Brod are urban. Stations Zagreb-1 and Rijeka-2 are under the influence of traffic while Sisak, Slavonski Brod and Rijeka-2 are under the influence of industry particularly oil refinery and petrochemical industry in Kutina. Rijeka is situated at the Adriatic coast while all other urban stations are continental. Observed hourly PM10 and PM2.5 concentrations were available from 8 rural background stations in Croatia that are distributed over the Croatian territory to capture the spatial and time concentration variations of different pollutants. One urban station Pecs-S, four urban background stations: Debrecen, Szeged-2, Veszprem, Ajka and one rural background station Sarrod were chosen for the analyses of PM10 levels in Hungary during November

2011. The measurements of PM10 concentrations were used from 6 stations in Serbia: four urban background and two urban traffic stations.

Figure 1. Measuring PM10 stations in Croatia, Hungary and Serbia during the year 2011. Urban traffic and/or industrial stations are marked with red colour, urban background stations are marked with orange colour and rural background stations are marked with green colour.

RESULTS

The analyses of the PM10 and PM2.5 concentrations in Croatia

Available PM10 and PM2.5 measurements were analysed for all Croatian stations in the period from 2006 to 2011 in order to assess the current level of air pollution with particles according to the defined limit values (CAFÉ Directive). Furthermore, the annual, inter-annual and daily variations of PM concentrations as well as their spatial distribution and specific local geographical differences were investigated. Measurements from rural background stations were used to quantify the contribution of regional and long-range transport to the locally measured concentrations.

Calculated annual average PM10 concentrations are considerably below the defined annual limit value of 40 $\mu g/m^3$ at all stations except for PM10 concentrations determined with the reference gravimetric method at Sisak-1 station during the years 2010 and 2011 (not shown). Decreasing trend is detected at all stations in the period from 2006 to 2010, while for 2011 an increase in average annual PM10 concentrations is present which can be contributed to different factors: local sources, transboundary and regional transport as well as to unfavourable meteorological conditions that were prevailing over the eastern Europe. The level of PM10 concentrations between the continental and coastal urban stations differs considerably with lower concentrations on stations under the influence of sea (i.e. Rijeka-1 and Rijeka-2). Annual average PM10 and PM2.5 concentrations are calculated for rural background stations in Croatia. The highest annual PM10 ~ 32 $\mu g/m^3$ and PM2.5 ~ 28 $\mu g/m^3$ values are found at Kopački rit station that is located on the eastern part of Croatia near the border with Hungary and Serbia. Values of PM10 concentrations at other continental and north Adriatic stations are around 15 $\mu g/m^3$ and PM2.5 ~ 14 $\mu g/ m^3$. The lowest values are found at south coastal station Žarkovica with PM10 ~ 10 $\mu g/m^3$ and PM2.5 ~ 8 $\mu g/m^3$ and at distant station Hum situated at the island of Vis where PM10 and PM2.5 are around 5 $\mu g/m^3$. According to the Directive daily average PM10 concentrations should not exceed 50 $\mu g/m^3$ more than 35 times in one calendar year. Intercomparison with limit values showed that all continental stations exceeded the allowed number of exceeding days for almost all years (exceptions are Sisak-1 during 2009 and 2010, and Zagreb-2 for 2009), while at Rijeka station on the Adriatic Sea the number of exceedance is significantly lower. The highest number of exceeding days is found for Sisak-1 station for PM10 concentrations determined with the gravimetric method. However it should be pointed that a considerable difference in the level of PM10 concentrations obtained with the gravimetric and the automatic measurements is found and further investigation and comprehensive analyses are required. Similarly as for trend in annual average PM10 values a decreasing trend in number of exceeding days is found for period from 2006 to 2010 and an increase in number of exceeding days is found for the year 2011.

Yearly course of daily average PM10 concentrations exhibits seasonal variations with the highest values of daily average PM10 concentrations during winter and late autumn at all urban stations. Significant deviations in the yearly course of PM10 concentrations among different years are not established while daily PM10 levels differ substantially among stations depending on the station location and the influence of local emission sources. The annual course of average daily PM10 concentrations is shown in Fig. 2 for 2011. The average daily concentrations ~ 80 μg/m^3 during the colder part of the year while they are ~ 30 μg/m^3 during the warmer part of the year. The highest daily PM10 concentrations are found at Sisak-1 station determined with the gravimetric method with daily values reaching up to 200 μg/m^3 while the lowest concentrations are measured at coastal stations in the Rijeka area.

While the daily average PM10 concentrations are well correlated at all stations, deviations and variability in average daily PM10 concentration levels between stations are pronounced. The yearly course of PM2.5 and PM10 ratios calculated from the observed hourly concentrations at rural background stations in Croatia during 2011 are shown in Fig.3. Stations mainly experience seasonal variations in PM25/PM10 ratios with higher values around 0.8 during winter and the lowest values ~ 0.6 in the summer time. Rural background station Hum is not showing significant seasonal variations and ratio is during the whole year > 0.9.

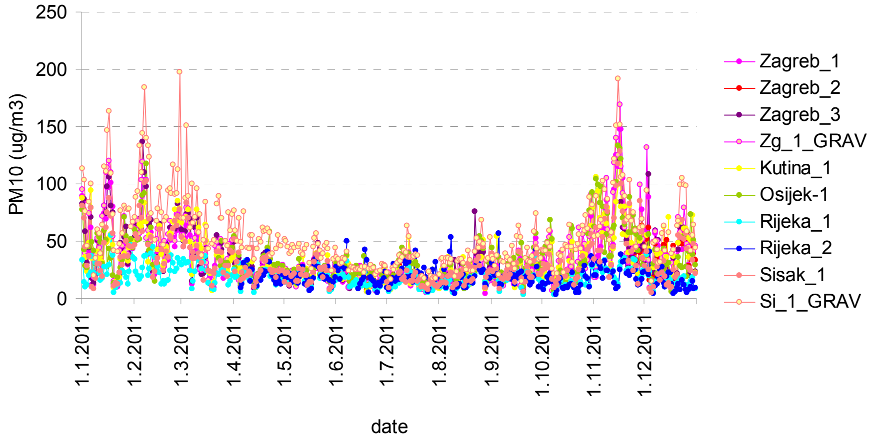

Figure 2. Annual course of daily PM10 concentrations at different urban stations in Croatia during 2011.

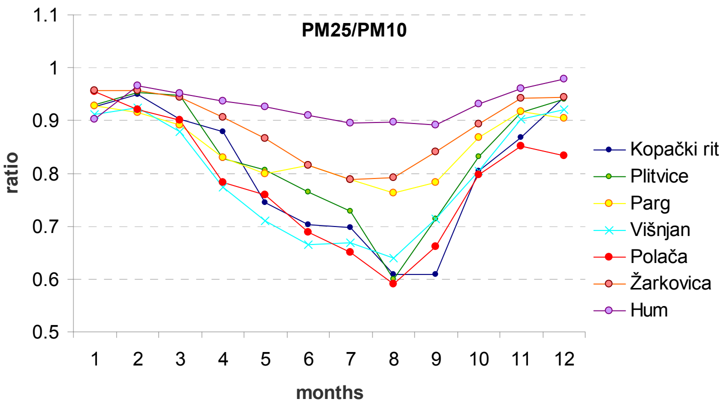

Figure 3. Monthly ratios of PM2.5 and PM10 averages calculated from the observed hourly concentrations at rural background stations in Croatia during 2011.

The analyses of air pollution episodes in Eastern European area

The time series of daily NO_2 and PM10 concentrations for selected stations in Hungary, Croatia nad Serbia are shown in Fig. 4 in the period from 15 October to 22 November 2011. Two peaks in daily PM10 concentrations with values over 100 µg/m^3 were identified at almost all stations. First peak started at almost all stations on 2 November and second comprised stations located on the eastern part of the Hungary on 13 November and reached the stations on the west on 17 November. The magnitude of daily PM10 concentrations was the strongest for the easterly stations in Hungary. Difference between the daily NO_2 and PM10 concentrations was high especially during the episodes. In Croatia two peaks in daily PM10 concentrations higher than 100 µg/m^3. An increase in daily PM10 concentrations that could be related to the episodes measured at continental stations was not observed at urban stations Rijeka-1 and Rijeka-2 located on the north part of the Adriatic Sea. In Serbia PM10 measurements from three urban background and one traffic urban stations in Belgrade were available as well as measurements from Novi Sad located approximately 30 km north from Belgrade and Niš located in the south east part of Serbia. Two peaks in daily PM10 concentrations first one observed on 1 and 2 of November and second observed on 14, 15 and 16 of November. However the magnitude varied across the stations.

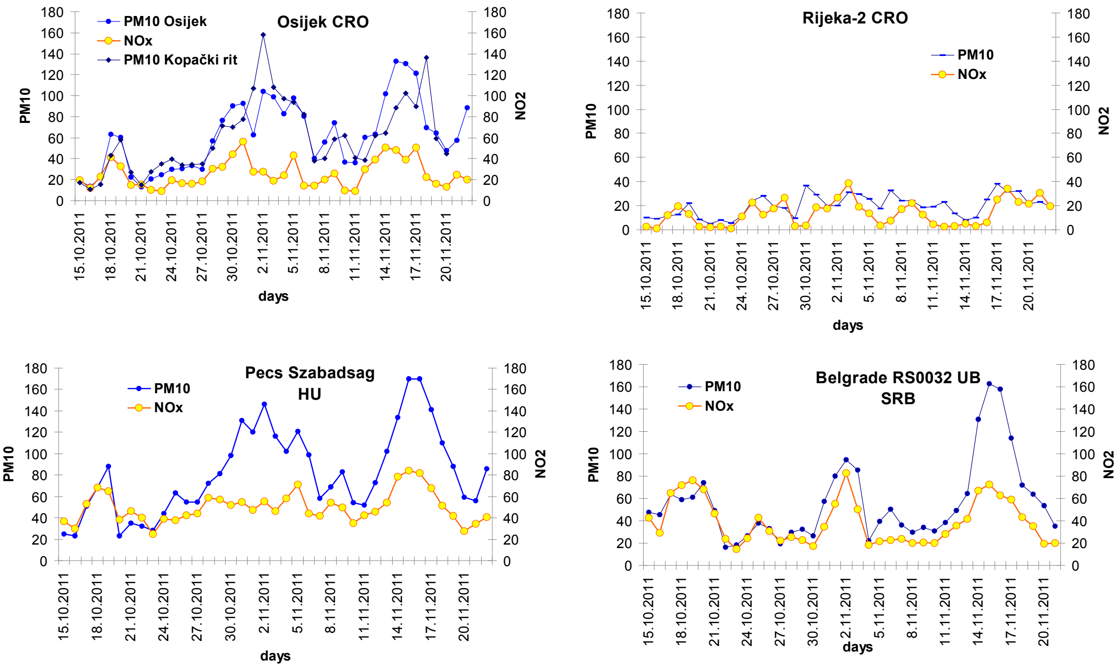

Figure 4. Time series of daily NO_2 and PM10 concentrations at different stations in Croatia, Hungary and Serbia during the episodes from 15 October to 22 November 2011.

Trajectory studies

In order to assess the influence of long range transport to the measured PM10 levels 6 days back trajectories for all analysed stations were calculated starting at 2 November and 4 day back surface trajectories starting at 16 November (Fig. 16). Obviously all analysed stations were under the influence of the same air masses, eastern for the first episode and north eastern for the second episode.

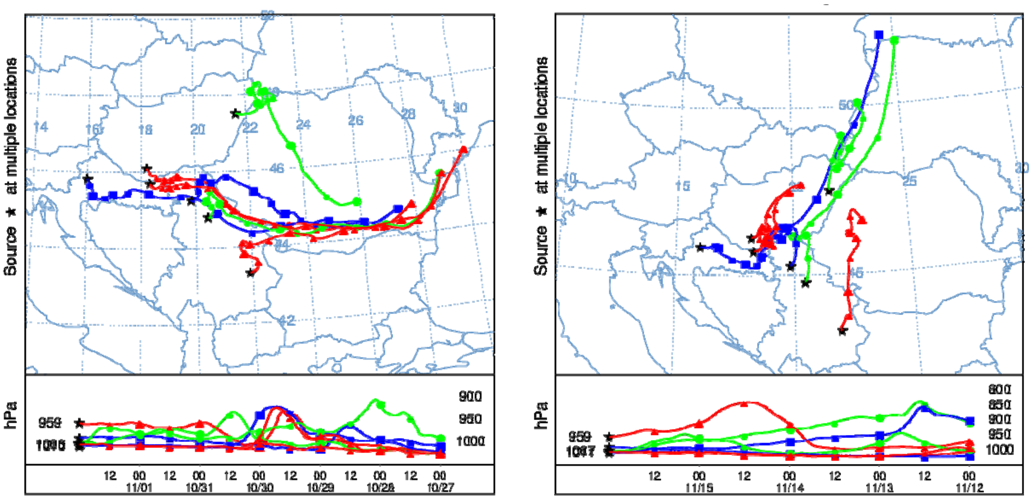

Figure 16. Backward surface trajectories from the analysed stations in Croatia, Hungary and Serbia starting at 2 November (left panel) and 16 November 2011 (right panel).

CONCLUSIONS

Two episodes with considerably increased daily PM10 concentrations (> 100 ug/m3), were identified that were timely harmonized and relatively uniform in magnitude at almost all analysed continental stations in Croatia, Hungary and Serbia in the period starting from 2 to 20 of November 2011. According to the calculated back trajectories all stations with peaks in PM10 concentrations were under the influence of the same air masses, eastern for the first episode and north eastern for the second episode. This is supported with the results of ARW WRF. Light wind of variable direction was present over the most of Croatian territory while over the eastern part of Croatia, Serbia and west and central Hungary moderate to strong south east wind was present during the first peak in measured PM10 on 2 November 2011.

Strong north eastern wind started to blow over Hungary and Croatia when a decrease in PM10 concentrations was measured at most of the stations, and at the same time Bora wind was present along the Adriatic coast. Stable conditions with light wind were present over the continent when an increase in PM10 concentrations was identified in the period from 12 to 17 November after which a south eastern and eastern moderate wind prevailed. Daily PM10 concentrations were generally higher during the second episode compared to the PM10 levels observed during the first episode. Elevated PM10 concentrations in the second episode are the consequence of local anthropogenic sources, regional transport as well as of the resuspension over the bare natural and agricultural surfaces in Hungary, Serbia and Croatia driven by the strong wind.

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