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Influence of weather types on concentrations of metallic components in airborne PM_{10} in Zagreb, Croatia

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This study investigates the influence of weather types found over the continental part of Croatia on daily PM_{10} concentrations and concentrations of metallic compounds in PM_{10} (namely manganese, lead and cadmium) in air during 2000–2002 period. Pollutant concentrations were measured at the northern, residential part of Zagreb, far from major pollution sources. Weather types were determined from synoptic charts. In the employed categorization six different patterns were recognized: radiation weather type, high pressure ridge, precipitation weather type, southeastern advection, northeastern advection and wind weather type. The most frequently, elevated concentrations were related to radiation weather type and southeastern advection, while the lowest concentrations were recorded during the wind weather type. Obtained results generally suggest a major role of the local pollution sources, and particularly of the Zagreb industrial zone, in suspended particles/metallic compounds pollution. Since cadmium exhibits somewhat different behavior, the role of the long-range transport in cadmium pollution needs to be further investigated. Synoptic conditions favorable for elevated concentrations occurred in about 37% of investigated days. Typically, these conditions are characterized by 1) the weak winds and turbulence, and consequently, inefficient pollutant concentration dilution (the nighttime and wintertime radiative conditions); or 2) southeastern airflow (southeastern advection and daytime radiative conditions), which transports pollutants from industrial zone of Zagreb towards the measuring site. Due to the latter, particulate pollutants emitted in the industrial zone pass rather frequently above the eastern part of Zagreb.

Keywords: PM_{10}, lead, manganese, cadmium, local sources, long-range transport

1. Introduction

Processes governing the fate of airborne pollutants, and consequent pollution levels, can be divided into 1) production, 2) transport and dispersion, and, 3) removal processes. On the timescales from hours to days all of above processes can strongly depend on atmospheric conditions. Thus, for example, dispersion varies with turbulence intensity; wet removal depends on the precipitation intensity and duration, transport is governed by the wind field etc. Additionally, the interaction between meteorology and air quality is characterized by a large number of meteorological variables acting both simultaneously and at different time scales. Thus, it is complex and non-linear. Therefore, the use of individual weather variables in evaluation of meteorological impact on air pollution is inadequate.

A number of studies investigate the relationship between pollutant concentrations and atmospheric conditions. Some authors utilize principal component analysis (PCA) and cluster analysis in order to identify main synoptic patterns and to inspect their influence on the airborne pollutant concentrations or aerosol optical depth (e.g. McGregor and Bamzelis, 1995; Shahgedanova et al., 1998; Cheng and Lam, 2000; Shahgedanova and Lamakin, 2005; Khan et al., 2007).

Table 1. Weather types employed in the present study (Lončar and Bajić, 1994; Lončar and Vučetić, 2003)

Weather type	Description
Radiation weather type (Rad)	High pressure and stable weather conditions with weak wind and variable wind direction. Maximal influence of local factors (topography and the land-use) and the season (i.e. intensive turbulence during the summer, and, statically stable inversions during the winter).
High pressure ridge (g)	Although related to the high pressure field, it is not always characterized by the radiation weather conditions. Thus, it is treated separately from other high pressure types. It is accompanied by a weak wind.
Precipitation weather type (Pre)	Advection of warm and humid air, predominately related to the frontal systems and accompanied with a cyclonal curvature of isobars; windy, cloudy and rainy weather.
SE advection (SE)	Southeastern advection; during the wintertime cold advection with persistent inversions and weak wind.
NW advection (NW)	Northwestern advection; during the summertime cold advection with convective clouds and convective precipitation.
Wind weather type (Wind)	Cold advections with strong wind and intense horizontal and vertical air mixing.

Buchanan (2002) employs an objective classification of weather types based on daily grid-point mean sea-level pressure data which are analyzed by an automated procedure. For Edinburgh area, he obtains elevated black smoke and PM_{10} concentrations for weather types with anticyclonic vorticity. The same is in accordance with a number of PCA results for different pollutants (McGregor and Bamzelis, 1995; Shahgedanova et al., 1998; Khan et al., 2007). However, some pollutants, such as NO_2 during extremely low wintertime temperatures (Shahgedanova et al., 1998) and O_3 (Khan et al., 2007) exhibit different behavior.

On the other hand, in the studies of $PM_{2.5}$ and/or PM_{10} pollution Bešlić et al. (2003) and Kuo et al. (2007) use synoptic weather patterns determined subjectively from surface synoptic charts. Although such approach depends on the consistency of an experienced analyst, some investigations generally suggest close agreement between the objective and subjective schemes (e.g. Jones et al., 1993; Giaiotti and Stel, 2001).

In this study, we also investigate the relationship between weather types and suspended particle concentrations in Zagreb with an aim to detect synoptic conditions favorable for the occurrence of elevated pollution levels. Unlike the previous study (Bešlić et al., 2003), where chemical composition of the particles was not taken into account, here we focus on metallic compounds in PM_{10}, namely, manganese (Mn), lead (Pb) and cadmium (Cd).

2. Methods and data

Weather types provide a simple categorization of a variety of weather conditions which comprise a number of different meteorological elements such as temperature, pressure, precipitation, wind, and humidity (Lončar and Bajić, 1994). Apart from their influence on pollution levels, they also affect human health (e.g. Pleško, 1985) and behavior (e.g. Klaić, 2001).

Poje (1965) established the first weather type classification for Croatia. It comprises 29 different weather types based on surface pressure field over Europe. Pressure field is determined once per day (0, 6 or 12 UTC) depending on available synoptic data. Weather types are thereafter sorted into four main groups: high-pressure, low-pressure, transitory, and types with weak pressure gradients. Thus, weather types of a certain group do not necessarily characterize similar weather conditions, (for example, both the northern and the southern side of the cyclone are attributed to the same low-pressure group, although weather conditions in these two can be completely different), while some weather types with similar conditions are attributed to different groups.

In order to group weather types with similar meteorological conditions, a new assembling of types proposed by Poje was suggested by Lončar and Bajić (1994) and Lončar and Vučetić (2003). This new categorization comprises six »new« weather types. The same is employed in the present study (Table 1).

Figure 1. Position of the sampling site. (Source: Google Maps).

Here, we assume that weather types which are routinely determined on a daily basis for continental part of Croatia are representative for the greater Zagreb area.

The sampling site is located in the northern, residential part of Zagreb (45° 50' 09.4" N, 15° 58' 58.6" E, 147 m ASL), at the Institute for Medical Research and Occupational Health, Zagreb. It is about 20 m far from a road with moderate traffic density. The site is far from major pollution sources and it is approximately 8 km northwestward from the Zagreb industrial zone (Figure 1).

PM_{10} monitoring was carried out during the period 2000–2002. Sampling was compliant with the requirements of European Standard EN12341 (1999). Coarser particle fraction was removed by inertial impactor. Samples were collected daily on 90 mm cellulose membrane filters from 100 m^3 of ambient air. Mass concentrations of metallic components were determined using a Pye Unicam SP 9 flame atomic absorption spectrometer. Chemical analysis was performed as described in Čačković et al. (2006).

Concentration datasets were submitted to standard statistical procedures as follows (for full details please refer to any of a number of statistical text-box). Firstly, datasets were log-transformed, and afterwards the Kolmogorov-Smirnov test (KS) was applied. Since KS confirmed the normal distribution of the log-transformed datasets, we used the analysis of variance (ANOVA) in order to investigate the differences between pollutant concentrations categorized by weather types. If ANOVA showed significant difference at $p = 0.05$ level, multiple comparison t-tests were applied with an aim to identify which weather types resulted in significantly different pollutant concentrations. Number of tests is given by $n = (m\ (m - 1))\ /\ 2$, where m is the number of weather types. The tests were performed at three Type I error levels somewhat different than in Buchanan et al. (2002), i.e. they were defined as follows:

(a) No correction (ignores the issue of multiple testing in order not to increase Type II errors), $p = 0.05$;

(b) False discovery rate (FDR) correction (in order to avoid the increased Type I error due to multiple testing; less conservative compared to Sidak's or Bonferroni adjustment), $p = 0.05\ ((n+1)\ /\ 2n)$, where n is the number of categories compared;

(c) Sidak's adjustment (again, in order to avoid the increased Type I error due to multiple testing), $p = 1 - (1 - 0.05)^{1/n}$, where n is the number of categories compared.

3. Results and discussion

Among 1096 consecutive days investigated in this study, 838 days (i.e. 76.5% of days) could be attributed to one of the six weather types listed in Table 1. The remaining 23.5% of unclassified days are related to the centre of cyclone or anticyclone found over the continental part of Croatia, and they are not further analyzed.

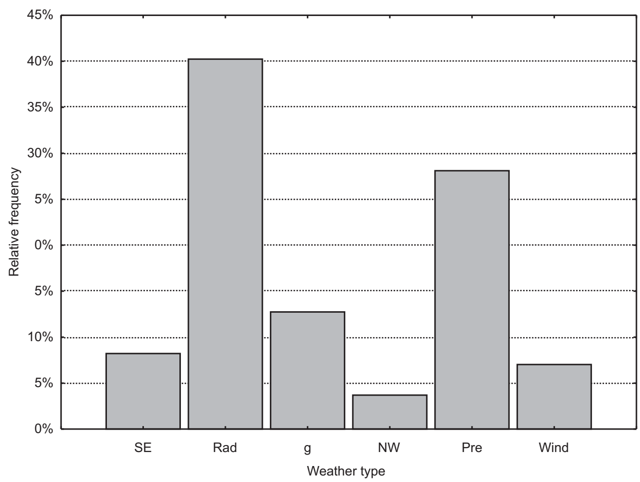

Figure 2. Relative frequencies of weather types occurring in the continental part of Croatia during the period 2000–2002. (Note: days with the centre of cyclone and centre of anticyclone found over the continental part of Croatia are not taken into account since they can not be attributed to any of above six types). Weather types are listed in Table 1.

Figure 2 shows relative frequencies of weather types during the period 2000–2002. Radiation and precipitation weather types were more frequent

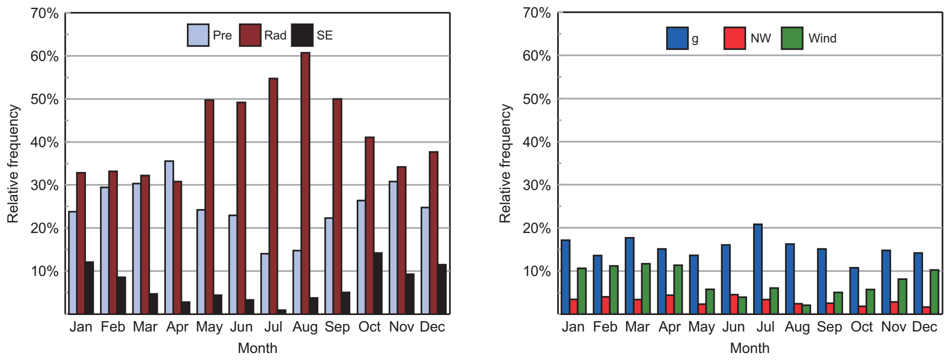

Figure 3. Annual variations of relative frequencies of weather types in the continental part of Croatia during the period 2000–2002. (Note: days with centre of cyclone and centre of anticyclone found over the continental part of Croatia are not taken into account since they can not be attributed to any of above six types).

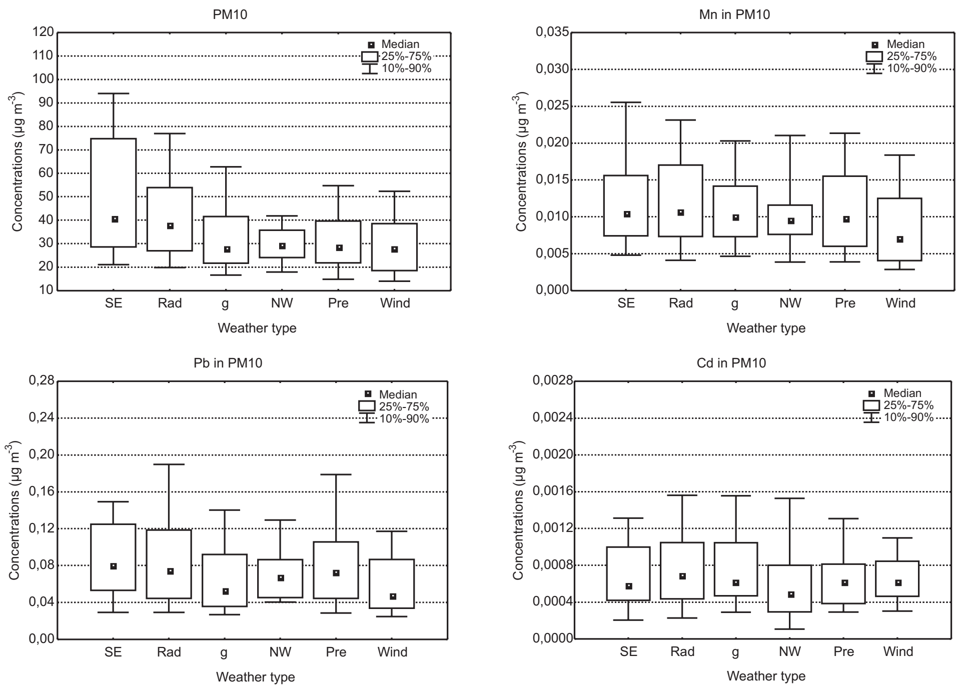

Figure 4. PM_{10} and metallic compounds concentrations in Zagreb vs. weather groups for all data during the period 2000–2002. Medians, 10^{th}, 25^{th}, 75^{th} and 90^{th} percentiles are also shown.

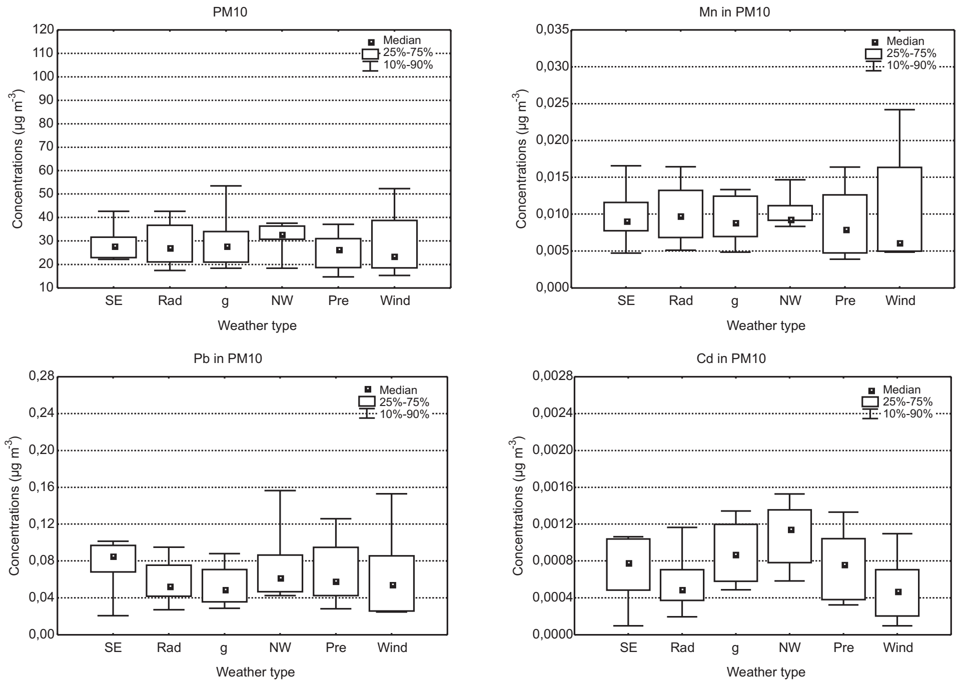

Figure 5. Same as in Figure 4, except for the spring.

compared to other types. Similar results were also obtained for previously investigated 30-year period (Lončar and Vučetić, 2003), where these two types were both, most frequent and of longest duration. Radiation type prevailed during the warm part of a year (Figure 3). Precipitation type was the most frequent during a spring and fall, while for July and August it was the rarest. Similarly, southeastern advection and wind type were also rarest during the warm part of the year. On the other hand, northwestern advection occurred infrequently during the period of investigation and it did not exhibit any prominent annual variation.

Summary statistics of pollutant concentrations versus weather types is shown in Tables 2–4 and Figures 4–8. As listed in the Table 2, pollutant concentrations for whole datasets, as well as for fall data, differ significantly for PM_{10}, Mn and Pb for different weather types, while no significant difference is found for Cd. On the other hand, a significant dependence of pollutant concentration on the weather type is found for both PM_{10} and Cd for a winter, while for a spring and summer only Cd concentrations depended on a weather type.

In order to identify weather types for which pollutant concentrations differ significantly, we applied multiple comparison *t*-tests, where six different

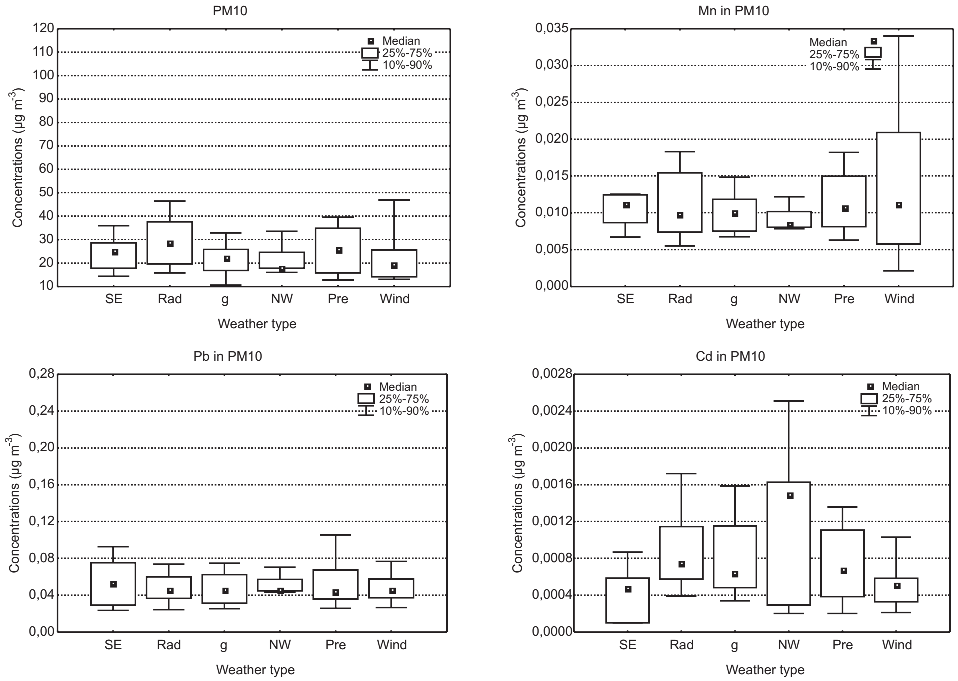

Figure 6. Same as in Figure 4, except for the summer.

weather types called for 15 pair-wise tests. The investigated Type I errors were determined at three levels as listed at the end of section 2, where a), b) and c) adjustments here correspond to $p < 0.05$, p < 0.027 and $p < 0.00344$, respectively. Results for entire investigated period are shown it Tables 3–5. (Note that results for Cd did not show statistically significant difference for different weather types (Table 2, all data). Thus, *t*-test pair wise comparison for Cd was not performed.)

It is obvious that, concerning PM_{10} concentrations, weather types are grouped in two subgroups (Table 3 and Figure 4). The SE advection and radiation regime form a separate subgroup characterized by elevated concentrations, while concentrations for other weather types show no difference. In the case of Mn (Table 4 and Figure 4), weather types are again grouped in two subgroups. Wind type forms a separate subgroup characterized by lower concentrations levels, while concentration levels for other weather types show no difference. With regard to the Pb concentrations (Table 5 and Figure 4), the radiation type, SE advection and precipitation type are related to elevated concentrations, while the wind type and a high pressure ridge are accompanied by lower concentration levels. The NW advection could be attributed to both subgroups.

Further inspection of results for which the relationship with weather conditions is statistically significant (namely, PM_{10}, Mn and Pb for whole datasets and for fall data; Cd for spring and summer; and PM_{10} and Cd for winter in Figures 4–8) shows that elevated concentrations are mainly related to the most frequent (Figures 2 and 3), radiation type and to southeastern advection. The former points out to the importance of statically stable conditions accompanied by the weak wind and weak turbulence (and consequently, the inefficient pollutant dilution). These generally prevail during a nighttime and wintertime. Further, radiation type is favorable for the establishment of the daytime upslope, northwestward and the nighttime downslope, southeastward local circulation due to Medvednica mountain (e.g. Klaić and Nitis, 2001–2002; Klaić et al., 2002; Klaić et al., 2003). Such circulation results in the daytime transport of pollutants from the Zagreb industrial zone towards the measuring site. The latter can also be attributed to the relative, northwestern position of the measuring site with respect to the Zagreb industrial zone.

On the other hand, the lowest concentrations are the most frequently found for wind type. This is not surprising, since windy conditions are favorable for both pollutant outflow from the area of interest due to advection (i.e. ventilation), and, mixing with adjacent air, which results in pollutant concentration dilution.

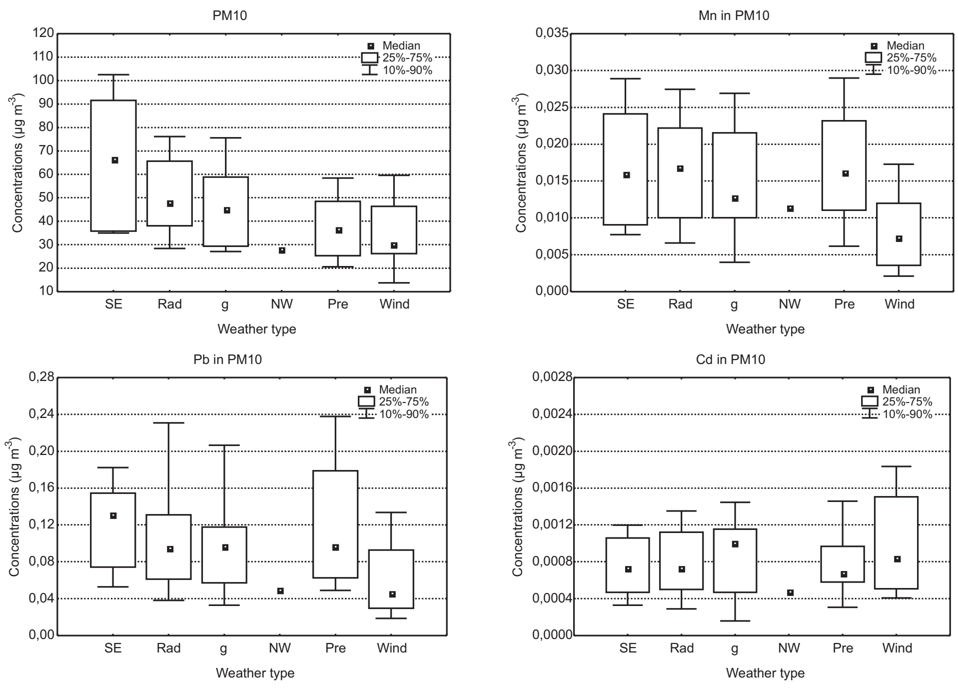

Figure 7. Same as in Figure 4, except for the fall.

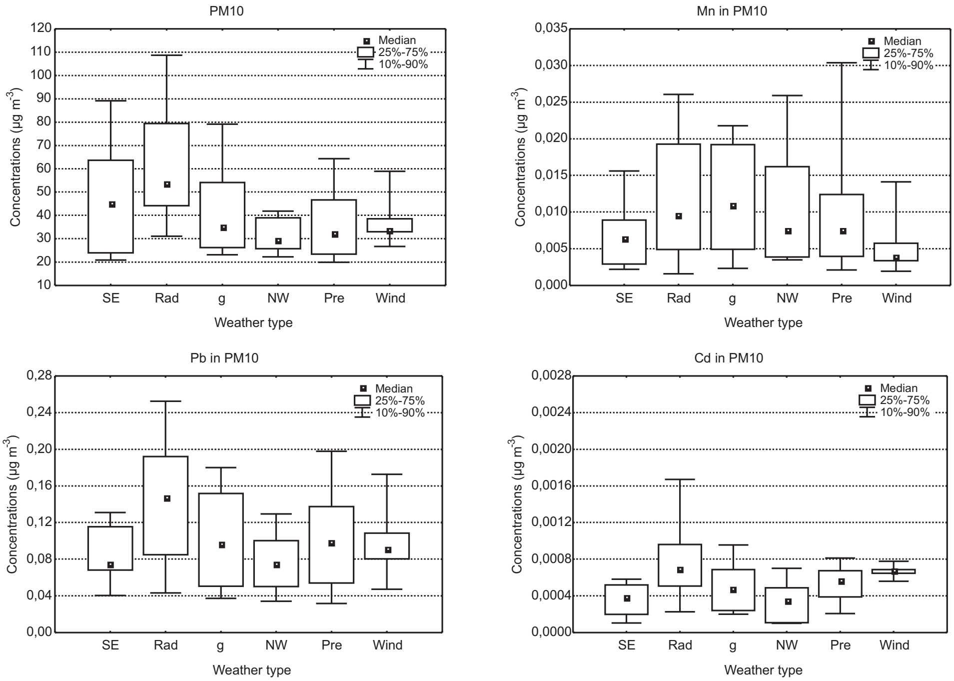

Figure 8. Same as in Figure 4, except for the winter.

Apart from cadmium, above results suggest generally low impact of the long-range transport of pollutants from Western Europe on suspended particles/metallic compounds concentrations in Zagreb. Yet, for Cd, the highest springtime and summertime concentrations are related to northwestern advection. Thus, it is very likely that Cd pollution is affected by distant western-European sources. The present study, however, cannot clearly distinguish between the effects of local and distant pollution sources. Hence, its primary aim is to investigate the relationship between the pollution levels and synoptic-scale weather conditions. In future work, however, it would be desirable to inspect the role of the long-range transport in cadmium pollution. Such an investigation should require a modeling approach, where a coupled meteorological and chemical model should be employed.

4. Conclusion

Analysis of the 3-year daily airborne concentrations of PM_{10}, manganese, lead and cadmium in PM_{10} measured in residential part of Zagreb revealed the dependence on synoptic conditions. Synoptic conditions were represented by six weather types defined for continental part of Croatia. The dependence was

Table 2. Analysis of variance of pollutant concentrations with respect to weather types for Zagreb data during the period 2000–2002. F is variance ratio and p is significance level. Results for p ≤ 0.05 are shown in bold.

Season	Pollutant	F	p
All data	PM_{10}	**16.09238**	**0.00000**
	Pb	**4.23426**	**0.00083**
	Mn	**4.04872**	**0.00123**
	Cd	1.64470	0.14584
Spring	PM_{10}	0,827254	0,532413
	Pb	0,595479	0,703445
	Mn	0,903036	0,481254
	Cd	**3,115805**	**0,011651**
Summer	PM_{10}	1,731265	0,131815
	Pb	0,270174	0,928718
	Mn	0,286611	0,919713
	Cd	**2,928613**	**0,015601**
Fall	PM_{10}	**6,247561**	**0,000026**
	Pb	**3,616238**	**0,003991**
	Mn	**7,365301**	**0,000003**
	Cd	0,430717	0,826597
Winter	PM_{10}	**7,805484**	**0,000001**
	Pb	2,104229	0,067568
	Mn	1,244343	0,290902
	Cd	**4,743327**	**0,000486**

statistically significant at the level $p = 0.05$ for PM_{10} (whole datasets, fall and winter data), Mn (whole datasets and fall data), Pb (whole datasets and fall data), and, finally for Cd (spring, summer and winter data).

The most frequently, elevated pollutant concentrations were related to the radiation weather type and southeastern advection, while lower concentrations where the most often found for wind weather type. The same is in accordance with previous results for PM_{10} and $PM_{2.5}$ (Bešlić et al., 2003).

Although the present study is not able to clearly distinguish between the effects of local and distant pollution sources, results suggest generally minor role of the long-range transport from Western Europe except for the springtime and summertime cadmium concentrations. Thus, PM_{10}, Mn and Pb exhibit different behavior compared with sulfur dioxide and precipitation acidity in Zagreb (Klaić, 1988; Klaić and Lisac, 1988; Klaić, 1991) where elevated con-

Table 3. t-test pair-wise comparisons of PM_{10} concentrations in Zagreb, classified by weather types for all data during the period 2000–2002. Sign '–' indicates that there is no statistically significant difference in concentrations for the two particular weather types. Letters a, b and c correspond to statistically significant differences at the levels $p<0.05$, $p<0.027$ and $p<0.00344$, respectively, i.e. they correspond to the tests (a)–(c) listed at the end of the section 2.

	NW	Pre	Rad	SE	Wind
g	– – –	– – –	a b c	a b c	– – –
NW		– – –	a b c	a b c	– – –
Pre			a b c	a b c	– – –
Rad				a – –	a b c
SE					a b c

a ($p < 0.05$) b ($p < 0.027$) c ($p < 0.00344$)

Table 4. Same as in Table 3. except for Mn.

	NW	Pre	Rad	SE	Wind
g	– – –	– – –	– – –	– – –	a b c
NW		– – –	– – –	– – –	a b c
Pre			– – –	– – –	a b c
Rad				– – –	a b c
SE					a b c

a ($p < 0.05$) b ($p < 0.027$) c ($p < 0.00344$)

centrations and higher precipitation acidities were related to northeastern advection, i.e. affected by western-European sources.

The major role of the local pollution sources in elevated pollutant concentrations is further supported by the fact that such events predominately occurred during the radiation type and southeastern advection. For the radiation type, two mechanisms can be responsible for elevated concentrations: 1) the nighttime and the wintertime weak wind and weak turbulence (and consequent, inefficient pollutant concentration dilution); and 2) radiation type supports the development of the daytime upslope, southeastern airflow, i.e. the transport of polluted air from the from the Zagreb industrial zone towards the measuring site. Similarly, elevated pollution levels during southeastern advection can also be attributed to the transport of pollutants from the industrial zone.

Above two weather types occurred on the average in about 37% of days (out of the total 1096 investigated days, 31% and 6% were related to the radiation and SE type, respectively), and they are both accompanied with the southeastern airflow. Accordingly, pollutants originating from the industrial zone rather frequently pass above the eastern part of the town. Thus, it can be concluded that the air quality in Zagreb, and particularly in its eastern part, may often be affected by its industrial zone.

Finally, the established relationships between the weather types and pollutant concentrations may serve as a tool for rough assessment of pollution levels which are expected based on synoptic conditions.

Table 5. Same as in Table 3. except for Pb

	NW	Pre	Rad	SE	Wind
g	– – –	a b –	a b c	a b c	– – –
NW		– – –	– – –	– – –	– – –
Pre			– – –	– – –	a b –
Rad				– – –	a b c
SE					a b c

a ($p < 0.05$) b ($p < 0.027$) c ($p < 0.00344$)

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SAŽETAK

Utjecaj tipova vremena na razine koncentracija metala sadržanih u frakciji lebdećih čestica PM_{10} u atmosferi iznad Zagreba

Ivan Bešlić, Krešimir Šega, Mirjana Čačković, Zvjezdana Bencetić Klaić i Višnja Vučetić

U radu je istražen utjecaj tipova vremena na dnevne koncentracije frakcije lebdećih čestica PM_{10} te olova, mangana i kadmija sadržanih u PM_{10} u razdoblju od 2000. do zaključno 2002. godine. Koncentracije su mjerene u sjevernom, rezidancijalnom dijelu Zagreba, podalje od većih izvora onečišćujućih tvari. Tipovi vremena (radijacijski, greben visokog tlaka, oborinski, vjetreni te jugoistočna i sjeverozapadna advekcija), određeni su na temelju sinoptičkih karata. Povišene koncentracije primijećene su tijekom radijacijskoga tipa i jugoistočne advekcije, a najniže tijekom vjetrenoga tipa vremena. Rezultati općenito ukazuju na važnu ulogu lokalnih izvora onečišćenja, naročito industrijske zone. Kako je slika za kadmij donekle drugačija, u daljnjem radu potrebno je ispitati ulogu daljinskog prijenosa u onečišćenju kadmijem. Uvjeti koji pogoduju povišenim razinama onečišćenja dogodili su se tijekom promatranog razdoblja u 37% ispitanih dana, a općenito ih karakterizira: 1) slab vjetar i slaba turbulencija, odnosno slabo razrjeđivanje onečišćujuće tvari (noću i zimi tijekom radijacijskog tipa vremena), ili 2) jugoistočno strujanje (pri jugoistočnoj advekciji te danju tijekom radijacijskog tipa) koje prenosi polutante od industrijske zone prema mjernom mjestu. Stoga onečišćujuće tvari koje su emitirane u industrijskoj zoni prilično često prolaze nad istočnim dijelom Zagreba.

Ključne riječi: PM_{10}, olovo, mangan, kadmij, lokalni izvori, daljinski transport

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