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The influence of meteorological conditions and weak to moderate traffic density on PM1 levels in a residential area of Zagreb, Croatia

Zvjezdana Bencetić Klaić [1], Kristijan Ružman [1], Ivan Smiljanić [1], Miran Stojnić [1] and Philipp Erbe [2]

[1] University of Zagreb, Faculty of Science, Department of Geophysics, Andrija Mohorovičić Geophysical Institute, Zagreb, Croatia

[2] Ilmenau University of Technology, Faculty of Mathematics and natural sciences, Institute of Physics, Ilmenau, Germany

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The levels of 1-min mean ambient particles with an aerodynamic diameter up to 1 μm (PM1) recorded at two nearby measuring sites in a residential area of Zagreb, Croatia, were analyzed with respect to the meteorological conditions (air pressure, horizontal and vertical wind speed, wind direction, global radiation, air temperature and relative humidity) and local traffic. PM1 mass concentrations were measured using a Model 8520 DUSTTRAK™ Aerosol Monitor (TSI Inc., Shoreview, MN, USA). One measuring site was placed next to a road with weak to moderate traffic, and the other was on a roof terrace approximately 100 m east of that road; the heights of the aerosol monitor inlet were at 1.7 m (road) and 15.8 m (roof) above the ground level (AGL). Both road and the roof experiments were performed by the same instrument, and thus, they correspond to different time intervals. The roof results show that nearby roads do not affect the PM1 concentrations at the site 100 m from the road, while, according to preliminary road results, the effects of the local traffic appear only if more than 22 vehicles per minute pass by the instrument. However, PM1 levels seem to be strongly affected by traffic occurring in the more distant areas of Zagreb. Additionally, the results for certain wind directions also exhibit the influence of the regional and/or long-range transport of pollution on PM1 levels. Finally, it is shown that 1-min mean PM1 mass concentrations depend on the 1-min mean meteorological variables investigated.

Keywords: advection of particles, ambient concentrations, DUSTTRAK™ Aerosol Monitor, traffic density, 1-min mean

1. Introduction

Particular matter (PM) is a complex, space- and time-varying (e.g., Querol et al., 2009) mixture of liquid droplets and particles suspended in the atmosphere

and comprises dust, dirt, soot and smoke. PM is a significant atmospheric constituent because it affects cloud formation, Earth's radiative balance and, consequently, climate (e.g., Buseck and Adachi, 2008; Monks et al., 2009; Uherek et al., 2010), human health (e.g., Bernstein et al., 2008; Ranft et al., 2009; Tach et al., 2010) and visibility (e.g., Kim et al., 2006). In many urban areas, PM has become a principal ingredient of polluted air (e.g., Yao et al., 2009).

The effects of PM on humans depend on the particle's chemical composition, concentrations, size, the length of human exposure and individual health conditions (e.g., Chalupa et al., 2004; de Kok et al., 2006; Nikasinovic et al., 2006; Pope and Dockery, 2006; Reich et al., 2009; Linares and Díaz, 2010; Tainio et al., 2010). Particles smaller than 10 μm in diameter (i.e., inhalable thoracic particles, PM10) enter the respiratory system. Those between 2.5 and 10 μm predominantly deposit in the upper respiratory tract, while particles smaller than 2.5 μm (PM2.5) can be inhaled more deeply into the lungs (Pope and Dockery, 2006). Ultrafine particles (PM0.1, aerodynamic diameter smaller than 0.1 μm) may even reach the bloodstream (e.g., Nemmar et al., 2005; Nemmar and Inuwa, 2008), thereby transporting harmful substances all over the body.

Because atmospheric particles are produced by a myriad of natural and anthropogenic processes, their sizes and other physical and chemical characteristics are diverse. Nevertheless, PM generally comprises metals, organic compounds, materials of biologic origin, ions, reactive gases and the particles' carbon cores (e.g., John, 2001; Engelbrecht et al., 2002; Watson et al., 2002; Kampa and Castanas, 2008; Khan et al., 2010). Particle dimensions range from the size of molecular clusters (a few nanometers) up to approximately 100 μm in diameter (particles larger than 100 μm are generally too heavy to be suspended in the air for a considerable time). Whitby divides airborne PM into three size modes (Whitby, 1978; John, 2001): nuclei (between 0.005 and 0.1 μm), accumulation (from 0.1 to 2 μm) and coarse (size above 2 μm), where each mode is log-normally distributed. The largest mode, coarse fraction, is produced by mechanical processes, and it contains crustal material (i.e., silicon compounds, iron, aluminum), sea salt and plant particles (e.g., John, 2001; Cheung et al., 2011).

The ultrafine nuclei mode, which is the smallest in both size and mass concentration, contains the largest number of particles. It is formed through combustion and photochemical reactions on gases in the atmosphere and is primarily composed of sulfates, nitrates and organic compounds (Buseck and Adachi, 2008). The mode diameter rapidly increases over time (John, 2001). Therefore, the nuclei mode is transient and is therefore noticeable in only the immediate vicinity of sources in the urban atmosphere (e.g., Fushimi et al., 2008). Nevertheless, it is very important for the formation and growth of larger particles (Buseck and Adachi, 2008). In contrast to the urban atmosphere, elevated concentrations of nuclei-mode particles have been observed in the free troposphere, far from the sources of the particles' precursors in the case of volcanic eruptions (e.g., Petäjä et al., 2012) and related to the flow regimes of elevated mountains (e.g., Rodríguez et al., 2009).

The accumulation mode is mainly anthropogenic. It is produced through the complex reactions of gases emitted into the atmosphere during fossil fuel combustion (i.e., gases containing sulfur, nitrogen and organic compounds). Both the nuclei and accumulation modes are known as fine particles. Fine particles comprise sulfates, organic ammonium, nitrates, carbon, lead, and some trace constituents (e.g., Čačković et al., 2001; Querol et al., 2001; Wilson et al., 2002; Funasaka et al., 2003; Rodríguez et al., 2004), and they predominately originate from traffic in many urban areas (e.g., Gertler et al., 2000; Abu-Allabam et al., 2003; Kristensson et al., 2004).

Most of the fine particle mass in the ambient atmosphere is in the accumulation mode. In accordance with a health-based standard and because the ambient particle size distribution has a minimum near 2.5 μm, a number of authors employ a threshold of 2.5 μm as a distinction between fine and coarse particles instead of Whitby's 2.0 μm (e.g., Pope and Dockery, 2006; Linares and Díaz, 2010). However, Wilson et al., (2002) recommend a threshold of 1.0 μm. These authors suggest that this threshold might better separate fine particles, which generally originate from combustion, from soil and other coarse particles.

More recent investigations (John, 2001) posit two different modes instead of Whitby's single accumulation mode: condensation (mean aerodynamic diameter of 0.2 μm) and droplet modes (mean aerodynamic diameter of 0.7 μm). The condensation mode directly or indirectly forms and grows with the condensation of gases through the coagulation of nuclei mode particles, and the rate of growth decreases with increasing condensation mode particle size. Particles in droplet mode are formed in aqueous phase reactions involving sulfur, and reaction rates decrease with increasing particle size.

This paper reports on ambient measurements of 1-min mean mass concentrations of submicron particles in residential area of Zagreb, Croatia. PM1 refers to fine particles with an aerodynamic diameter smaller than 1.0 μm, as defined by Wilson et al. (2002). This fraction, which is mainly anthropogenic and generally originates from traffic, incorporates entire nuclei-mode and major portion of accumulation-mode particles in terms of volume and mass (see, for example, Figs. 1 and 2 in Wilson et al., 2002). To our knowledge, apart from the study of Klaić (2012), other studies addressing the ambient PM levels in Croatia focused on larger particle fractions (i.e., PM10 and/or PM2.5) and longer sampling intervals (e.g., Čačković et al., 2001; Bešlić et al., 2005; 2007, 2008; Šišović et al., 2008; Čačković et al., 2008, 2009; Hrust et al., 2009; Jelić and Klaić, 2010). In this study, we investigate a larger dataset than a previous study (Klaić, 2012). We also report on the diurnal variations of the PM1 levels in residential area, and, we show preliminary results regarding the role of road traffic on near-road PM1 levels, which has not been investigated previously.

Since it is well known that meteorological conditions generally affect the fate of airborne pollutants (e.g., Finlayson-Pitts and Pitts, 1986), we investigate possible relationships between PM1 levels and available meteorological data. Hus-

sein et al (2006) also inspected the effects of meteorological conditions on the accumulation mode fraction (particles with an aerodynamic diameter between 100 nm and 2.5 μm) and ultrafine particles (smaller than 100 nm in diameter) in an urban environment. However, they analyzed the number concentrations of other small fractions, not PM1, and their results were derived from hourly data, while here we focus on mass concentrations and on a time scale of 1 minute.

2. Measuring sites and measurements

Meteorological and PM1 mass concentration measurements were performed in the northern residential part of Zagreb (Bubble A in Fig. 1a), approximately 1.5 km north-northeast of the city center (*Donji Grad* in Fig. 1a) and approximately 8 to 9 km northwest of the industrial area of Zagreb (*Peščenica Žitnjak* in Fig. 1a). This area is moderately hilly and is located at a south-facing slope of Mount Medvednica at an approximate height of 180 m above sea level. The mountain is approximately 1 km high and stretches from west-southwest towards the east-northeast.

Figure 1. (a) Satellite view of the town of Zagreb (source: Google maps). Positions of two nearby measuring sites are indicated with a single bubble denoted by A. The center of the town (*Donji Grad*) is approximately 1.5 km south-southwest of the measuring site, while the industrial area of Zagreb (*Pešćenica Žitnjak*) is approximately 8–9 km southeast of the measuring site. (b) A closer view of the measuring sites. Bubble A shows the position of the road (*Bijenička cesta*) PM1 measurements, while Bubble B shows the location of meteorological and roof PM1 measurements performed at the premises of the Department of Geophysics (DG). Point B is approximately 100 m east of Point A. (c) The view of the PM1 road measurement locations (Bubble A in Fig. b) from the west.

c)

Figure 1. Continued.

2.1. Meteorological data

Meteorological data (air pressure, air temperature, horizontal and vertical wind, global radiation and relative humidity) have been routinely measured by the Department of Geophysics (DG), Faculty of Science, University of Zagreb, at the DG premises. The data used in this study were recorded by automatic meteorological station META 2000 (AMES, Brezovica, Slovenia). Sensors measuring horizontal and vertical wind components and global radiation are placed at masts mounted on the roof terrace. The terrace is positioned on the western side of the approximately 50 m long three-story DG building (bubble B in Fig. 1b). The heights of 4.5 m, 4.3 m and 1.8 m above the terrace (17.3 m, 17.1 m and 14.6 m above ground) correspond to the positions of the horizontal wind, vertical wind and global radiation sensors, respectively. Air temperature and relative humid-

ity sensors are in a standard meteorological shelter (2 m above the ground) located in a grassy area north of DG building. An air pressure sensor is inside the building at a height of 7.5 m above the ground (187.55 m above the mean sea level). All meteorological data are measured every second, but 1-min averages are stored.

There are several three- to five-story university campus buildings west and northwest of the meteorological measurement sites. All of them have an east-west orientation and are approximately 50–70 m long (Fig. 1b). Other buildings in the area, one- to three-story family houses and urban villas with slanted roofs, are smaller. There are patches of grass and irregularly arranged trees with an average height of approximately 10 m between the buildings.

2.2. PM1 mass concentration measurements

PM1 mass concentrations were measured by the DUSTTRAK™ Aerosol Monitor, Model 8520 (TSI, Inc., Shoreview, MN, USA), previously calibrated by the factory. To collect PM1 data that correspond to the meteorological data, the instrument, which measures mass concentration every second, was set to store 1-min means. Measurements were performed at two adjacent locations (hereafter, road and roof measurements, respectively).

Road measurements were performed at *Bijenička* road on the western edge of the campus (Bubble A in Fig. 1b and Fig. 1c), approximately 160 m north of the bus stop. *Bijenička* road stretches in a north-south direction and is inclined southward. It has two lanes of opposite directions, separated by only a centerline. Generally, the traffic is low speed because the speed limit is 50 km / h, and the traffic intensity is low to moderate. *Bijenička* road connects the center of the town with the main cemetery. Thus, a substantial portion of traffic is related to the cemetery's daytime hours of operation. Due to the various heights of nearby roughness elements (up to approximately 10), the site could not be considered as representative for a well-defined urban street canyon. There is also a road in the vicinity that perpendicularly intersects *Bijenička* 70 m south of the measuring site (Fig. 1b). This road, which is roughly WSW-ENE oriented and is approximately 50 m south of the DG building, has very weak traffic.

The aerosol monitor was fixed on a 1.67 m high tower placed at the eastern pavement a half-meter away from the road edge and 12.5 m west of the campus building (Fig. 1c). The instrument inlet was 1.7 m above ground level (AGL) and was perpendicular to the roadway. The inlet height was selected to roughly correspond to the height at which pedestrians breathe. (According to the EU directive 1999/30/EC (EC, 1999), the breathing zone corresponds to a height of 1.5 m above ground. Further, according to the same directive, the inlet sampling point should be between 1.5 and 4 m above ground.) Road measurements were performed during ten separate, 45-min time intervals in February 2009 (Tab. 1), and 439 1-min mean PM1 mass concentration values were collected. Since the

traffic at the *Bijenička* road is generally weak out of working hours (meaning weekends, as well) we selected time intervals during working hours, when we expected denser traffic. Several wintertime working days in the row were selected due to similar weather conditions.

Table 1. List of the ten road experiments in February 2009. ST corresponds to the starting time (local standard time, LST) of the experiment.

Experiment code	1	2	3	4	5	6	7	8	9	10
Date	13	13	16	16	16	17	17	17	19	19
ST (LST)	14:59	16:14	12:29	13:29	14:29	07:14	8:14	09:14	13:59	14:59
Experiment duration (min)	45	45	45	41	45	38	45	45	45	45

During the roof measurements, the aerosol monitor was mounted on the 3 m mast fixed on the same DG terrace as the meteorological sensors for global radiation and three-dimensional wind (Bubble B in Fig. 1b). The instrument inlet was 15.8 m AGL. The instrument was also equipped with a waterproof environmental enclosure designed for outdoor measurements. Measurements were performed during two time intervals, one from 17 March to 12 May 2010 and the other from 14 September to 26 September 2011. We note that the first of the two measurement intervals provided us with the near-ground PM1 mass concentration data at times following the eruption of the Eyjafjallajökull volcano in Iceland, which lasted from 15 to 21 April 2010 (e.g., Folch et al., 2012). As in the road measurements, 1-min means were calculated from the values measured every second. In total, 96738 1-min mean PM1 mass concentration roof values were collected.

3. Traffic intensity observations and calculation of weighted numbers of vehicles

To relate the road measurement data to traffic density, movies recording the passage of vehicles through *Bijenička* road were shot by digital camera (Fig. 1c) during each of the ten road experiments listed in Tab. 1. During these 10 experiments, up to 30 vehicles per minute passed in both directions. Based on recorded movies, vehicles passing in each minute in either direction were thereafter grouped into five different categories: cars, terrain-cars, vans, trucks and buses.

The mechanism of the fine particle emissions and their gaseous precursor emissions through road traffic is complex (e.g., Gertler et al., 2000; Abu-Allabam et al., 2003; Kristensson et al., 2004) because emissions depend on driving speed,

vehicle maintenance, vehicle type and fuel quality. Therefore, pollutant emissions differ from vehicle to vehicle, and for each vehicle dependent on the aforementioned variables. Because such detailed data could not be obtained, we assumed a simple relationship between the type of the vehicle and fuel consumption under speed-limited urban driving conditions based on the suggestions given by Vehicle Center of Croatia (2009). Assumed fuel consumption enabled us to introduce simple weighting coefficients that correspond to vehicle types (Tab. 2). A weighted total number of vehicles (*WNV*) per minute was thereafter calculated from

$$WNV = n_C \cdot 1.000 + n_{TC} \cdot 1.500 + n_V \cdot 2.000 + n_T \cdot 3.125 + n_B \cdot 3.750 \qquad (1),$$

where n_C, n_{TC}, n_V, n_T and n_B correspond to the number of cars, terrain-cars, vans, trucks and buses passing in both directions nearby the measuring site in the 1-min time interval concerned. The mean *WNW* for each of the ten road experiments are listed in Tab. 3.

Table 2. Assumed fuel consumption under urban driving conditions[1] and corresponding weighting coefficient.

Type of vehicle	Assumed fuel consumption (l/100 km)	Weighting coefficient
Car	8	1.000
Terrain-car	12	1.500
Van	16	2.000
Truck	25	3.125
Bus	30	3.750

4. Results and discussion

The contribution of background pollution (i.e., pollution from other urban and distant sources) to the total PM1 concentration at the measuring sites generally depends on the wind direction. Thus, we expect the northern flows to be accompanied by an advection of clean air from Mount Medvednica with consequently low background pollution, while airflows of southern directions transport polluted air from the town toward the measuring site (Fig. 1a) and thus, should contribute more to the total PM1 concentration.

Because the road measurements comprised only 439 1-min mean concentration values, the relationships between pollution levels and meteorological condi-

[1] Personal communication with Mr. Zoran Kalauz of Vehicle Center of Croatia, Zagreb, March 2009.

tions, except for concentration rose shown in Fig. 2a, were not analyzed with respect to each of the wind directions separately. However, the roof measurements provided a very large dataset of 1-min mean concentrations (96 738 values) and corresponding meteorological data. Therefore, they were analyzed separately for each of sixteen wind directions (N, NNE, NE, ENE, E, ESE, SE, SSE, S, SSW, SW, WSW, W, WNW, NW and NNW). Each of the attributed directions corresponded to an angle of 22.5 degrees. For instance, wind directions between 11.25 and 33.75 degrees were attributed to north-northeastern flows, and directions between 33.75 and 56.25 degrees, to northeastern flows, and so on.

Apart from distinguishing different pollution sources regions, separate analysis of PM1 concentrations for each wind direction also enabled a certain, rough grouping of concentrations with respect to the typical atmospheric conditions. Namely, different wind directions are generally caused by different synoptic setups, and consequently, accompanying by different weather conditions. Thus, for example, southerly and westerly winds mainly carry humid air towards Croatia; southerly winds carry warmer air, while northerly winds above Zagreb are on the average the strongest (not shown here), and they are generally cold and dry.

4.1. Road measurements results

Tab. 3 summarizes the basic statistics of the ten road experiments and the corresponding meteorological conditions. It shows that the mean 1-min PM1

Table 3. List of the ten road experiments in February 2009. MWNV is the average WNV (see Eq. 1) for the corresponding experiment. The mean values of meteorological variables are denoted by T (temperature), RH (relative humidity), GR (global radiation), P (air pressure) and V_h (horizontal wind speed).

Experiment code	1	2	3	4	5	6	7	8	9	10
MWNV (min^{-1})	15.1	15.0	14.0	13.7	12.3	17.6	16.4	13.0	13.4	14.1
PM1 ($\mu g\ m^{-3}$)										
Mean	15	16	40	44	25	48	63	69	39	29
Min	7	4	23	14	12	40	52	55	22	22
Max	180	170	256	413	391	82	180	361	277	180
T (°C)	4.0	3.9	1.9	2.8	3.0	2.2	2.6	3.4	1.9	1.7
RH (%)	32.8	33.5	46.4	41.9	41.1	51.6	54.8	53.0	42.5	42.5
GR ($J\ cm^{-2}$)	143.9	30.5	336.4	257.4	125.9	0.0	22.5	50.0	444.5	305.8
P (hPa)	989.1	989.1	999.3	998.3	997.7	990.4	989.8	989.4	999.5	999.6
V_h ($m\ s^{-1}$)	3.7	2.7	2.4	2.5	1.3	1.9	1.7	1.7	3.2	3.5

concentrations for individual experiments varied from 15 to 69 μg m^{-3}, in which the lowest levels (experiments 1 and 2, i.e., concentration values of 15 and 16 μg m^{-3}) are comparable to the annual mean regional background PM1 concentrations within the Mediterranean Basin (10–11 μg m^{-3}) (Querol et al., 2009).

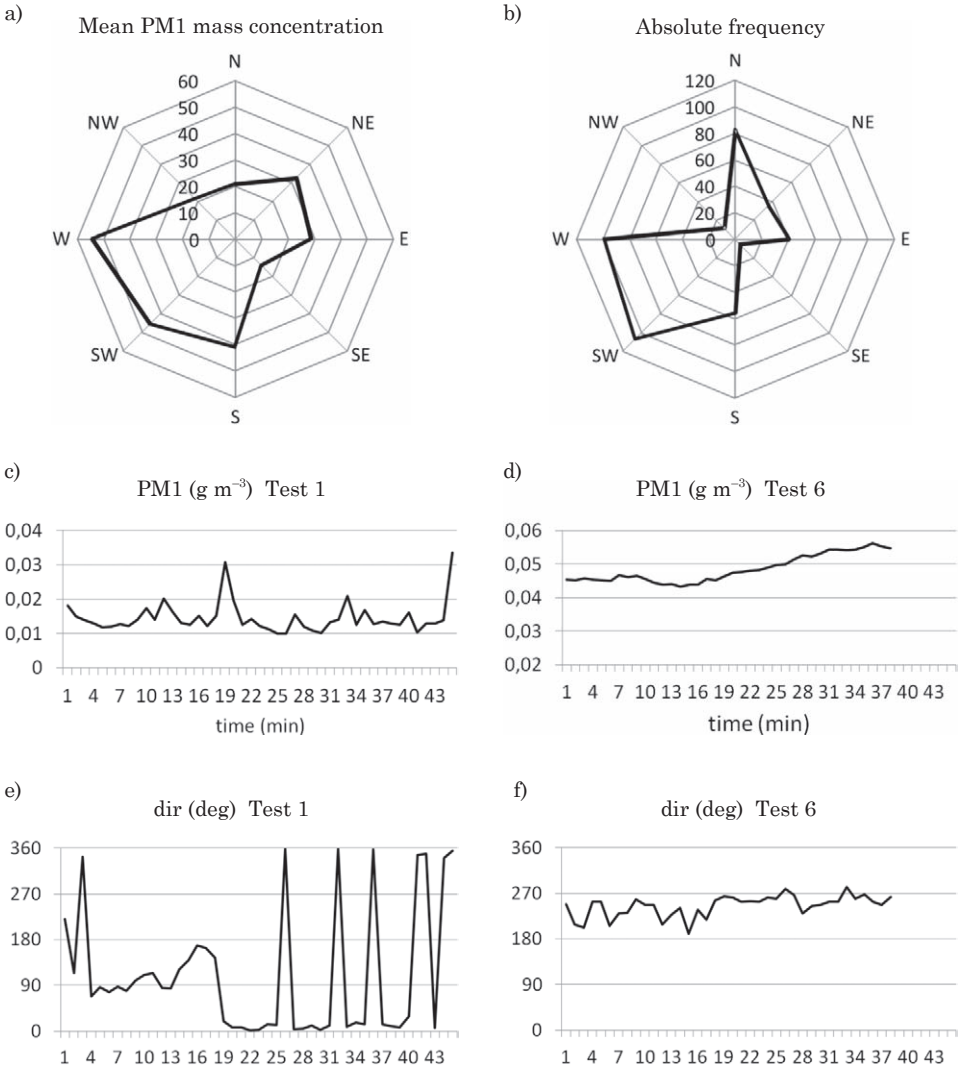


Figure 2. Mean PM1 concentrations (μg m^{-3}) at *Bijenička* road *vs.* wind directions (a) and the absolute frequency of wind directions (b). The instrument inlet height was at 1.7 m AGL. Panels c and d show time series of 1-min mean PM1 concentrations for the 1st and the 6th experiment, respectively (see Tabs. 1 and 3), while Panels e and f show corresponding 1-min mean wind directions.

Further, 1-min values of the entire data set varied for two orders of magnitude, that is, from 4 μg m^{-3} (minimum that occurred during experiment 2) to 413 μg m^{-3} (maximum recorded during the experiment 4). The average PM1 concentration for all ten experiments was 38.7 μg m^{-3}. We note that this value is similar to the estimate of the exposure of a cyclist to PM1 particles (37.4 μg m^{-3}), which is obtained in the Mol (Belgium), based on seven different sampling experiments (Berghmans et al., 2009).

Fig. 2 shows the dependence of the mean PM1 mass concentrations on wind directions. Because the data set was relatively small, road concentrations were inspected against only 8 main wind directions, where each of the directions corresponds to an angle of 45 degrees. As seen from the figure, the PM1 road concentrations were affected by the advection of pollutants from other urban and distant sources. Thus, N and NW winds, which generally transport clean air from the nearby Mount Medvednica, were related to low PM1 concentrations, while southern (S and SW) flows, which transport the air from the town of Zagreb (Fig. 1a), were accompanied by relatively high concentrations. However, the highest 1-min average PM1 concentrations were obtained for western flows, perhaps due to the superimposed effects of several factors: 1) the inlet exposure towards west; 2) local traffic (instrument was placed at the eastern pavement); 3) urban sources within the western portion of Zagreb; and 4) regional and/or long-range transport of pollutants. However, we cannot quantitatively distinguish between these possible effects.

Nevertheless, in order to better understand local versus the long-range transport effects, time variations of 1-min data for individual experiments were inspected. Here we show two time series of PM1 concentrations (Panels c and d in Fig. 2) and corresponding wind directions (Panels e and f). Panels c and e correspond to Experiment 1, while Panels d and f correspond to Experiment 6 (see Tabs. 1 and 3). During the Experiment 1, which was accompanied with variable wind directions during first 19 minutes, followed by more or less northerly winds at latter times (Fig. 2e), concentrations show more rapid temporal variations compared to experiment 6 (compare Figs. 2c and d), suggesting thus, the effects of local sources. On the contrary, during the Experiment 6, which is at the beginning accompanied with roughly southwestern winds (although they slowly vary between southern and western), and, as of 17^{th} minute with predominately western flows (Fig. 2f), concentrations during the last two thirds of experiment slowly increase in time (Fig. 2d), exhibiting thus the effects of distant sources. Similar, slow increase of concentration was also observed for Experiments 7 and 8 (not shown here). Again, in both experiments slow concentration increase went along with predominately western flows. Above findings corroborate our hypothesis that western flows support regional and/or long-range transport of particles.

At first glance, the concentration related to SE flows is surprising (Fig. 2a); it is the lowest concentration because southeastern winds generally transport the air from the polluted industrial zone towards the measuring site (Fig. 1a).

Previous studies of airborne PM2.5 and PM10 pollution levels in the northern part of Zagreb (Bešlić et al., 2007; 2008), which were executed on a different temporal scale (daily), reported the highest average concentrations for southeastern flows. However, SE flows over the Zagreb area were rare during the experiments; thus, among a total of 439 1-min time intervals, only 5 (i.e., ≈ 1.1% of all intervals) were southeastern (Fig. 2b). Similarly, several studies of multiyear periods on the daily scale also show that southeastern advection over the greater Zagreb is rare, with a relative frequency of only up to 8% (Bešlić et al., 2007; 2008; Čanić et al., 2009). Thus, we believe that the concentration minimum associated with SE flow is a consequence of a small sample size and is not a reflection of reality.

Fig. 3 shows temporal variations of the mean, maximum and minimum concentrations and the temporal variation of the mean *WNV* (see Eq. 1) based on the ten road experiments listed in Tab. 1. The highest mean and the highest minimum mass concentrations (69 and 55 μg m^{-3}, respectively) were obtained during the morning hours, namely for experiment 8, which lasted from 09:14 LST to 09:58 LST. The highest maximum concentration (413 μg m^{-3}) was recorded during the time interval from 13:29 to 14:09 LST (experiment 4). We note that the mean, maximum and minimum values do not follow the pattern of temporal variation of the local traffic density (i.e., the traffic on *Bijenička* road). This finding suggests that the advection of particles produced in the other parts of town (and maybe in more distant areas) dominated the particles produced by the traffic on *Bijenička* road. Theoretically, the discrepancies between the local traffic density and observed concentrations could also arise due to the boundary layer depth influences. We note however, that the lowest maximum concentration value, and moderate mean concentration were observed in the morning, around 7:30 LST, when we generally expect the boundary layer to be shallower than in latter times, and in spite of the fact that the traffic count was the highest. On the other hand, the highest among the ten recorded maximum concentrations was observed around 13:40 LST, when the traffic was moderate, and when the boundary layer should generally be deeper compared to morning hours. Therefore, we conclude that the effects of boundary layer depth were dominated by the effects of advection. The more so, our experiments correspond to wintertime period when, due to the low values of global radiation (Tab. 3), we expect generally shallow boundary layer without prominent temporal variations of its depth.

Fig. 4 depicts box-plots of the PM1 1-min mean mass concentrations vs. the traffic intensity and vs. the measured meteorological variables. As seen from the Panel a, the effects of local traffic on PM1 levels are observed only if at least 23 vehicles per minute pass along the road in both directions. (Note that values shown at the intersection correspond to the centers of the classes. Thus, the abscise value of 25 actually corresponds to the values from 23 to 27.) However, we note that more plausible picture of the local traffic influence on the PM1 levels would be obtained if the road measurements were accompanied with the

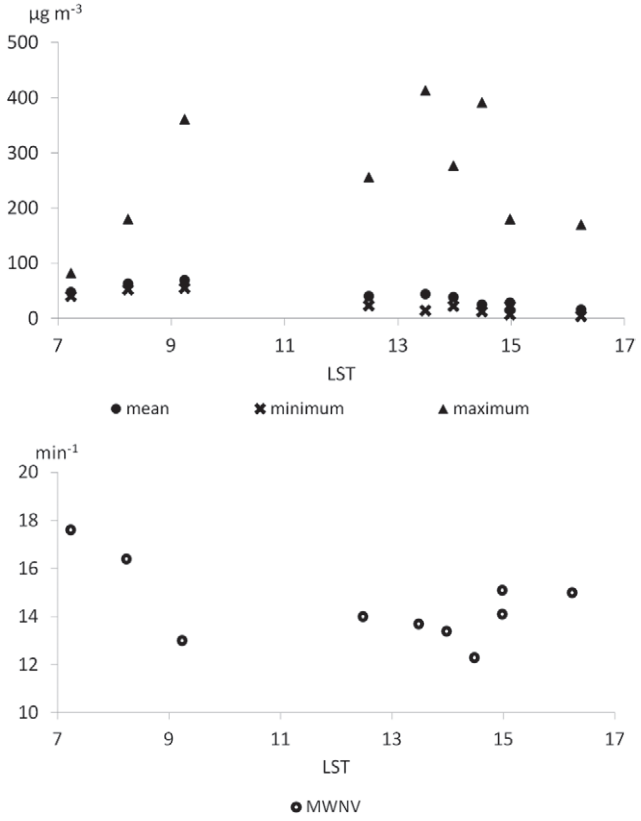


Figure 3. Mean, maximum and minimum PM1 mass concentrations (μg m^{-3}, *top*) and the mean weighted number of vehicles per minute (*MWNW*, *bottom*) at *Bijenička* road for each of the ten road experiments listed in Tab. 1. At the abscissa, the center of the time interval (local standard time, LST) corresponding to each experiment is shown.

simultaneous measurements at the site far away from the road (which would require another instrument), and if the dataset was longer. Thus, results shown in Panel (a) should be considered as preliminary.

Panel (b) does not reveal any relationship between the 1-min mean PM1 mass concentration and the air pressure, which can be attributed to the fact that atmospheric pressure is generally a slowly varying variable, while the PM1 concentrations were found to vary more quickly in time. Thus, the ten experiments captured a relatively small portion of the possible surface pressure values (for all ten experiments, the pressure values ranged from only 989.0 to 999.7 hPa, while during a single experiment, the variations were up to 0.5 hPa). Simultaneously, the PM1 values varied much more (for all ten experiments together, these concentrations ranged from 4 to 413 μg m^{-3}). The relationships between the PM1 values and global radiation and temperature (Panels c and d in Fig. 4) for the

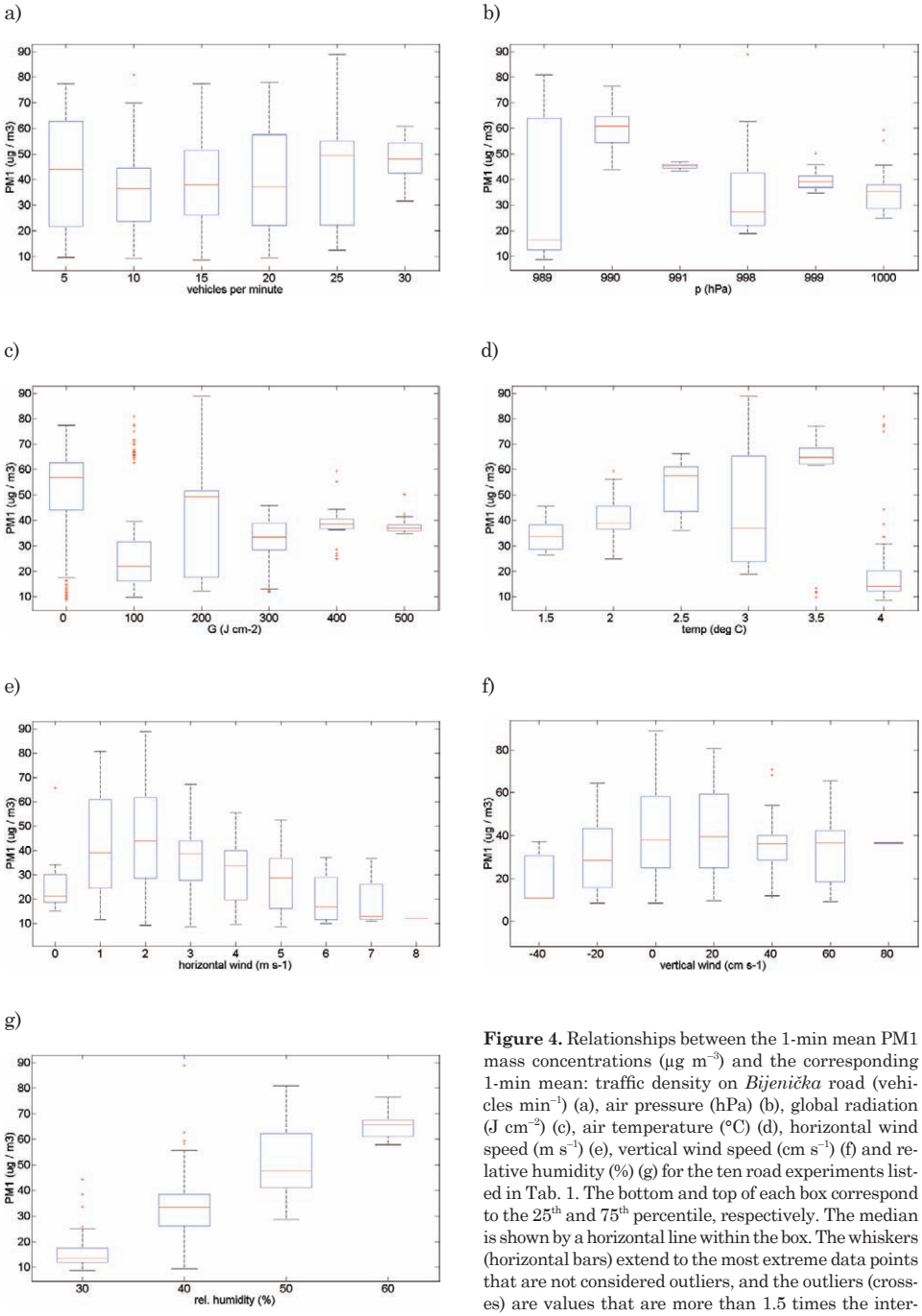


Figure 4. Relationships between the 1-min mean PM1 mass concentrations (μg m^{-3}) and the corresponding 1-min mean: traffic density on *Bijenička* road (vehicles min^{-1}) (a), air pressure (hPa) (b), global radiation (J cm^{-2}) (c), air temperature (°C) (d), horizontal wind speed (m s^{-1}) (e), vertical wind speed (cm s^{-1}) (f) and relative humidity (%) (g) for the ten road experiments listed in Tab. 1. The bottom and top of each box correspond to the 25th and 75th percentile, respectively. The median is shown by a horizontal line within the box. The whiskers (horizontal bars) extend to the most extreme data points that are not considered outliers, and the outliers (crosses) are values that are more than 1.5 times the interquartile range away from the top or bottom of the box.

road experiments are also unclear. We note that the road experiments were performed during the winter when the weather was predominately cloudy and cold. Considering the data from all ten experiments, the global radiation varied from 0 to 528 J cm^{-2}, while the temperature was only between 1.4 and 4.2 °C.

In contrast to global radiation and temperature, the effects of the wind and relative humidity on road PM1 levels are evident (Panels e, f and g). Panel e suggests the importance of advection (that is, the importance of urban sources other than the local traffic) for the weak horizontal wind speeds. Namely, for horizontal winds weaker than 2.5 m s^{-1}, PM1 concentrations increase alongside an increase in wind speed. This trend suggests that for weak winds, the advection of pollutants from other areas dominates over local ventilation. Because winds are weak, they obviously transport pollution from shorter distances (i.e., from other parts of the town). Conversely, for horizontal winds stronger than 2.5 m s^{-1}, PM1 concentrations decrease with an increase in wind speed, thus exhibiting the effects of ventilation and the consequent pollutant dilution.

Considering the vertical wind component (Fig. 4f), PM1 mass concentrations decrease as subsidence strengthens. We believe that this decrease is due to a suppression of particles at heights lower than those the instrument inlet (1.7 m AGL). The highest concentrations were obtained for weak convection (i.e., for vertical wind speeds between 10 and 30 cm s^{-1}). As expected, PM1 concentrations decreased with convection strengthening. This trend can be attributed to pollution dilution, which is caused by the efficient transport of particles in greater heights. We expect that the pattern of the dependence of PM1 concentration on vertical velocity depends on the height of the inlet. Thus, for example, if the inlet was next to the ground, the maximum concentration should be expected for the strongest subsidence.

In contrast to the results obtained by Hussein et al. (2006), in which relative humidity was not the controlling factor for both the hourly number concentrations of ultrafine particles (smaller than 100 nm) and the accumulation mode fraction (particles between 100 nm and 2.5 μm), our results show a clear increase in the 1-min mean PM1 mass concentrations with an increase in the 1-min mean relative humidity (Fig. 4g). We note that above results were obtained from the small dataset (i.e., from 439 1-min data). Nevertheless, as it will be shown in the following subsection, the dependence of the PM1 mass concentrations on the relative humidity was also obtained for the larger dataset of roof measurements.

4.2. Roof measurement results

The mean PM1 concentration for the entire roof dataset was 26.5 μg m^{-3}, while the standard deviation was 20.6 μg m^{-3}. The minimum and maximum 1-min mean measured values were 0 and 462 μg m^{-3}, respectively. The mean value is substantially lower than the road mean (38.7 μg m^{-3}), which can be attributed to several factors. First, the road measurements correspond to the win-

ter, when emissions are generally higher due to heating (heating in Zagreb is predominately based on gas) and denser road traffic (a major source of submicron particles in urban environments), while roof measurements mainly correspond to the warm season. During the warm season, the atmosphere is generally less stable than in the winter, and pollution dilution is enhanced due to the higher boundary layer depth. Consequently, concentrations should generally be lower. Second, the road measurements were performed next to the major urban particle source (road traffic), while the roof measurements were 50 m far from the closest road with very weak traffic, and approximately 100 m from the road with weak to moderate traffic (*Bijenička*). Finally, the road measurements were performed closer to the heights of major urban sources (i.e., the vehicles' tailpipes).

According to Folch et al. (2012), volcanic ash cloud due to the April 2010 eruption of Eyjafjallajökull was found above Croatia in the period from 20 to 21 April 2010. Similarly, the calculations performed at the Croatian Meteorological and Hydrological Service suggests the presence of a volcanic ash cloud above Croatia from 17 to 20 April 2010 (Pavić, 2011). However, the roof measurements did not exhibit any unusual increase in PM1 concentrations during this period (not shown here). Similarly, surface measurements of PM2.5 and PM10 at several other Croatian urban sites during the same period also do not show elevated particle concentrations (Pavić, 2011). We conclude that the more detailed inspection of possible influences of the volcanic eruption on surface particle concentrations in Croatia would require an analysis of the background concentrations, which is beyond the scope of this study.

Fig. 5 shows diurnal variations of PM1 mass concentrations and meteorological variables based on the 1-min mean roof measurements. As seen from Panel a, the PM1 concentrations follow the general pattern of traffic intensity expected in a residential area, wherein traffic increases when residents drive to work or return home. In addition, the pattern of the diurnal variation of PM1 agrees with the pattern obtained for the road measurements (Fig. 3, top). Roof measurements revealed two PM1 concentration maximums of similar magnitude. One (33.6 $\mu g\ m^{-3}$) corresponds to the morning (08 local time, hereafter LT), while the other (30.7 $\mu g\ m^{-3}$) occurs in the early evening (19 LT). However, the morning interval of elevated concentrations is longer than the evening interval. The PM1 concentration is minimal in the afternoon hours (22.5 $\mu g\ m^{-3}$, at 16 LT). The afternoon minimum is somewhat lower than the nighttime values (for example, mean concentration for 02 LT was 24.7 $\mu g\ m^{-3}$), when we expect the lowest traffic-related emissions of PM1 and its precursors. This difference might be due to nighttime conditions, which are generally characterized by a stronger static stability (i.e., weaker turbulence), which results in a lower boundary layer and, consequently, weaker pollutant dilution (that is, higher near ground concentrations). Panels b–f, which illustrate the mean diurnal variations of meteorological variables determined from two roof measurement intervals, exhibit

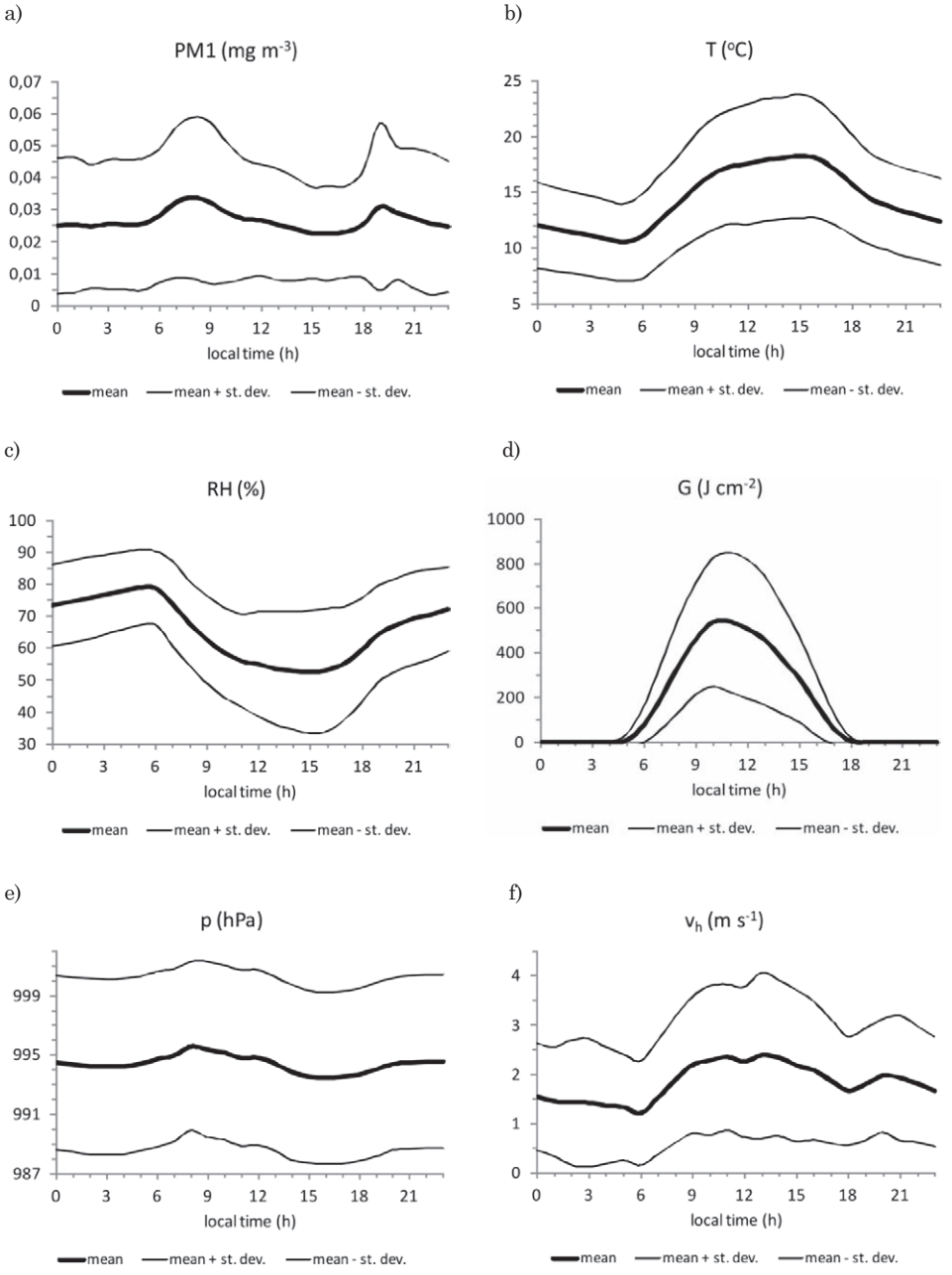


Figure 5. Diurnal variations of the roof PM1 mass concentrations (a), temperature (b), relative humidity (c), global radiation (d), air pressure (e) and horizontal wind speed (f), based on two measurement intervals (from 17 March to 12 May 2010 and from 14 September to 26 September 2011).

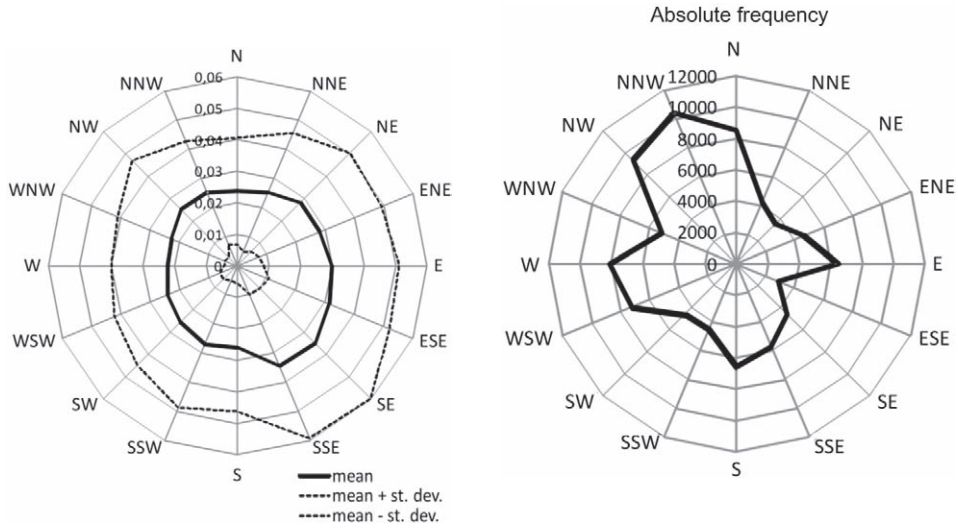


Figure 6. Mean PM1 roof concentrations *vs.* wind directions (mg m^{-3}) (*left*) and absolute frequency of wind directions (*right*) based on two measurement intervals (from 17 March to 12 May 2010 and from 14 September to 26 September 2011). Aerosol monitor inlet height was at 15.8 m AGL.

expected behaviors. However, we present them here to report on the average meteorological conditions under which PM1 concentrations were measured.

Fig. 6 illustrates the dependence of the mean PM1 concentrations on the wind directions and absolute frequencies of the recorded 1-min mean wind directions. Each direction corresponds to an angle of 22.5 degrees. Patterns of both concentrations rose (Fig. 6, left), and wind direction rose (Fig. 6, right), which are similar to the results obtained for a shorter measurement period (17 March–12 May 2010) in the previous study (Klaić, 2012). Unsurprisingly, the corresponding concentration values are slightly different. The highest mean PM1 mass concentration is again related to southeastern flows, which transport polluted air from the industrial zone of Zagreb, passing in addition along the rather long path above town (i.e., above a number of other urban sources) (Fig. 2.a). Compared to the result from that previous study (0.0398 mg m^{-3}), the maximum concentration for the larger dataset is somewhat lower (0.0350 mg m^{-3}) in our study. Elevated concentrations are also related to south-southeastern (0.0347 mg m^{-3}) and east-southeastern flows (0.0316 mg m^{-3}). As in the previous study, the lowest mean concentrations are related to western (0.0221 mg m^{-3}) and west-northwestern winds (0.0226 mg m^{-3}), which are followed by northern (0.0237 mg m^{-3}) flows. We note that these flows transport the air above the non-urbanized slopes of Mount Medvednica towards the measuring site (Fig. 2a).

During the two investigated periods, the most frequent flows were those from the north-northwest (10 417 minutes out of the total 96 738 minutes, which cor-

responds to 10.8% the entire data dataset) (Fig. 6, right). These were followed by northwestern flows (9323 minutes, i.e., 9.6% of the data) and northern flows (8533 minutes, i.e., 8.8% of the data). The most rare were east-southeastern flows (2940 cases, i.e., 3.0%) and northeastern flows (3578 minutes, i.e., 3.7%), while southeastern flows, which were accompanied by the highest mean PM1 concentrations, occurred relatively rarely (4611 cases, i.e., 4.8%).

We note that both concentration and wind direction roses for the roof measurements (Fig. 6) are completely different from the roses for the road measurements (Fig. 2). This difference can be attributed to several facts. First, road measurements comprise a relatively small dataset (439 concentration and corresponding wind direction data), while the roof measurements correspond to a large dataset (96738 pairs of concentration and wind data). Accordingly, the road data were analyzed with respect to only eight main wind directions, while the roof results corresponded to more detailed wind roses with sixteen wind directions. Finally, the road and roof data correspond to different seasons; thus, the difference in the airflow patterns is not surprising. The elongation of the wind direction rose for the roof data (Fig. 6, right) in the NNW–SSE direction, that is, in the direction perpendicular to the elongation of Mount Medvednica, suggesting the presence of the of up- and down-slope thermally induced winds, which are generally established over the south-facing slopes of Mount Medvednica under synoptically undisturbed atmospheric conditions (e.g., Klaić et al., 2002; 2003). Daytime up-slope winds should generally contribute to the increase of concentrations at the measuring site, since they transport the urban air toward the site, while down-slope north-northwestern winds should result in the decrease of PM1 concentrations. However, Fig. 5a does not suggest such pattern, since the lowest concentrations occur at times when up-slope winds are well established (15–16 LST). Obviously, the diurnal variation of PM1 concentrations exhibits the domination of pollutant dilution due to the deep afternoon boundary layer over pollutant advection by up-slope winds. Although not seen in the diurnal variation of PM1 concentrations (since they are dominated by other processes), the effects of advection by up- and down-slope winds are clearly seen in the mean concentration rose (Fig. 6, left). Namely, average PM1 concentrations are the lowest for down-slope winds (N, NNW and NW), while they are the highest for upslope winds (SSE and SE).

Fig. 7 illustrates the increase in the 1-min mean PM1 mass concentrations with the 1-min mean air pressure. Here, we show only the results for the entire roof dataset (Panel a) and for the western flows (b). For the western winds (Panel b), PM1 concentrations for the highest pressure values (over 1007.5 hPa) are lower than the concentrations for the air pressure between 1002.5 and 1007.5 hPa. However, this is a consequence of a generally small number of concentrations accompanied by the occurrence of the air pressure over 1007.5 hPa (Panel c), rather than a real air pressure effect. Similarly, the results for other wind directions also show an increase in PM1 mass concentrations with air pressure. These

results are in accord with the fact that the high atmospheric pressure is generally accompanied by a stable atmospheric stratification and consequent elevated pollutant concentrations. The above results differ from those of Hussein at al. (2006), in which a clear relationship was not found between the aerosol particle number concentrations and air pressure.

At first, the relationship between the 1-min mean PM1 mass concentrations and the 1-min mean global radiation does not seem to be straightforward (Fig. 8). While the results for the entire PM1 dataset suggest a decrease in PM1 concentrations with an increase in global radiation for global radiations over 900 J cm^{-2} and almost no variation in the concentration for the lower values of global radiation (Panel a), investigations of individual wind directions exhibit several patterns. The majority of directions (namely, S, SSW, SW, W, WNW and WSW) show similar patterns as the entire data set (see, for example, the results for southern winds in Panel c). Conversely, other wind flows (specifically, N, NE, NNE and NNW) show U-shaped variation (e.g., results for the northern winds in Panel b), while the south-southeastern flows suggest the opposite, ∩-shaped pattern (Panel e). Meanwhile, for the remaining flows (E, NW, SE, ENE and ESE), it seems that there is no relationship between global radiation and PM1 concentration (see, for example, the results for northwestern flows in Panel d). The U-shaped pattern, however, is related solely to the airflows, which on their way towards the measuring site, pass above the slopes of Mount Medvednica in which there are no major urban emission sources (Fig. 2a). In contrast, the pattern shown in Panel c is related to 1) flows passing above many urban sources (that is, S, SSW, SW and WSW flows) and/or to 2) flows with a prominent western component (namely WNW, W, WSW and SW), which can begin due to larger scale forcing (it is well known that westerlies generally prevail in the midlatitudes) and may therefore transport particles and/or their precursors from more distant regions. In both cases related to the pattern shown in Panel c, it is reasonable to assume that particles arriving at the measuring site are generally older than the particles whose concentration exhibits the U-shaped relationship shown in Panel b.

Similar to the road results (Fig. 4d), the roof results do not suggest any straightforward relationship between air temperature and PM1 mass concentration, although the roof measurements captured a wide range of 1-min mean temperatures (from 2.8 to 34.9 °C, see frequency distribution in Fig. 9b). For the majority of wind directions and the entire data set, the dependence of concentration on air temperature was wavelike and exhibited an increase in concentration for higher temperatures (as an example, we offer the results for southeastern winds, Fig. 9a). Hussein et al. (2006) show that the annual variation in the total number concentration of PM2.5 fine particles is inversely proportional to air temperature, which is not found in the present study of PM1 mass concentrations recorded during the warm season. However, Hussein et al. also showed that the number concentrations of accumulation-mode particles (particles between

100 nm and 2.5 μm) increase significantly alongside temperature for temperatures above 15 °C. Because the mass of fine particles primarily comprises the mass of the accumulation-mode particles (e.g., John, 2001) and because higher number concentrations imply higher mass concentrations, our results agree with the findings of Hussein et al. for accumulation-mode particles. In our case, however, an increase in mass concentrations with temperature was observed for temperatures higher than in the study of Hussein et al. Most frequently (for 5 out of 16 inspected wind directions, not shown here) such an increase in PM1

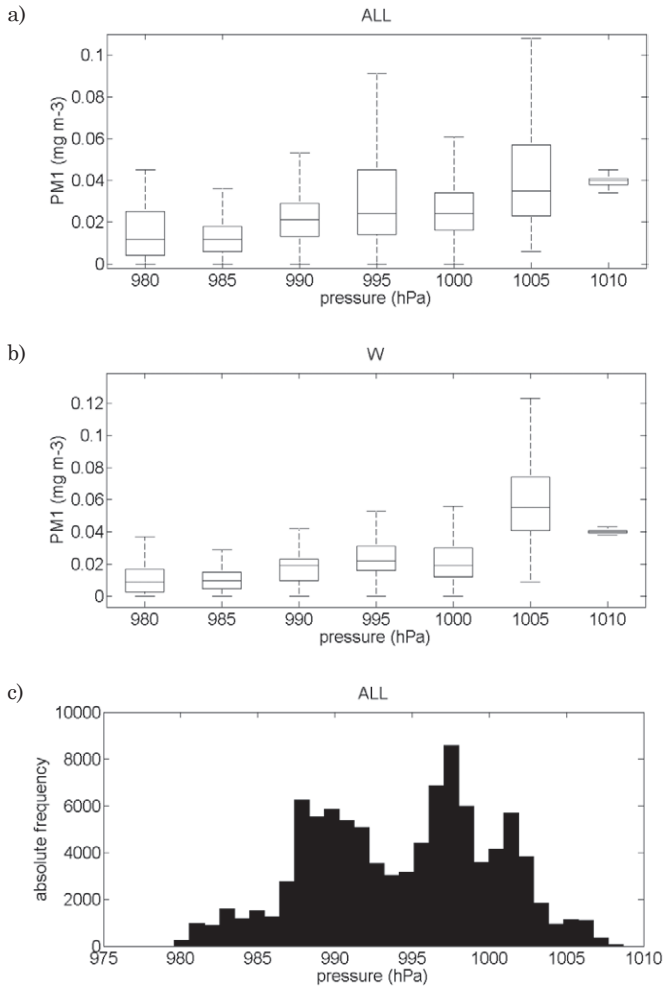


Figure 7. Box-plots of the 1-min mean PM1 mass concentrations *vs.* the 1-min mean air pressure for entire roof data set (a); and for western winds (b). Box-plots show the same statistical parameters as in Fig. 4. Fig. c shows frequency distribution of 1-min mean pressure values for the entire data set.

mass concentrations with temperature was found for temperatures above 19.5 °C, while the results for other directions suggested thresholds of 22.5 °C (4 cases), 16.5 °C (4 cases) or 25.5 °C (3 cases).

Similar to the results of the previous study (Klaić, 2012), the dependence of 1-min mean PM1 mass concentrations on the horizontal wind speed exhibit three prominent patterns (Fig. 10). One of them clearly reflects the effects of local ventilation (Panel a), resulting in a decrease in concentration with an increase

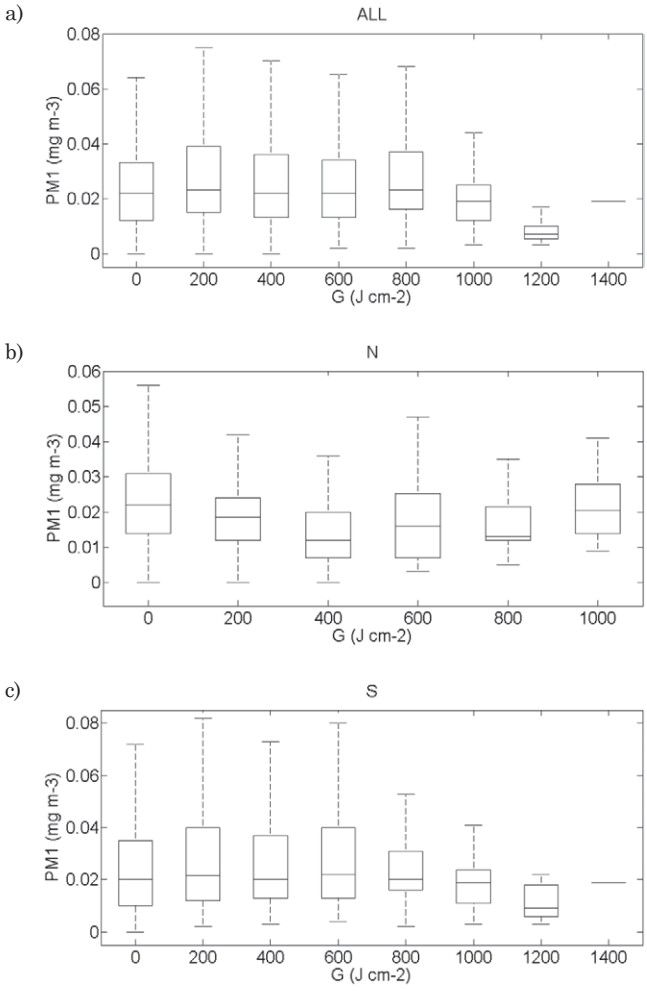
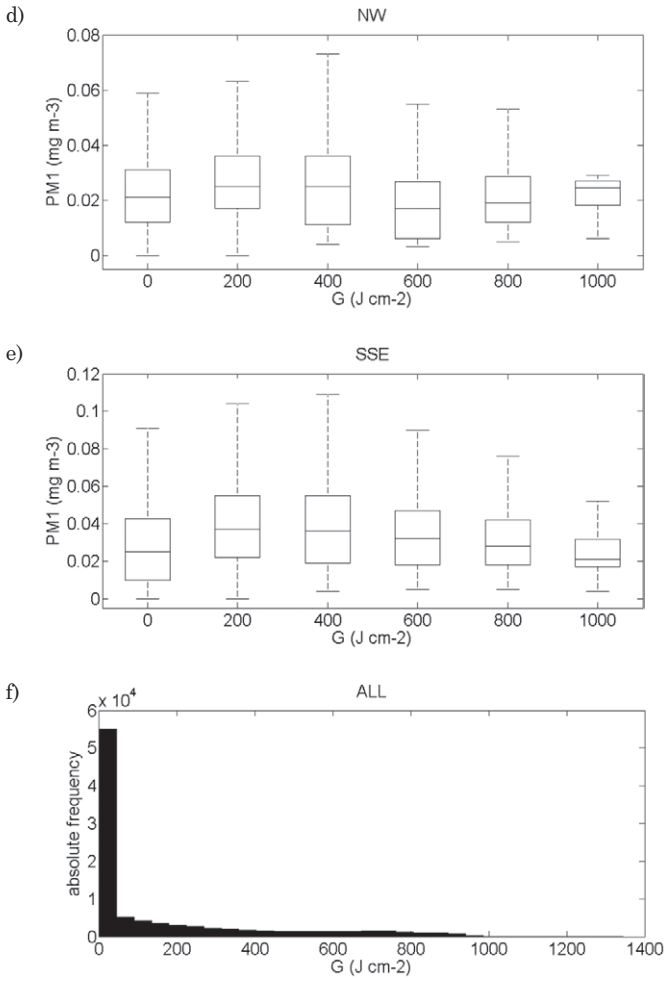


Figure 8. Box-plots of the 1-min mean PM1 mass concentrations vs. the 1-min mean global radiation for the entire roof data set (a); northern (b), southern (c), northwestern (d) and south-southeastern (e) flows. Panel (f) shows frequency distribution of 1-min mean global radiation values for the entire data set.



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in wind speed. Such a pattern is obtained for the majority of investigated wind directions (specifically, N, NE, NNE, NNW, NW, WNW and S) and for the entire dataset analyzed without considering wind direction. The second pattern (Panel b), is characterized by an increase in PM1 concentration with an increase in wind speed for weak winds (speeds below 2.5 m s^{-1}) and a decrease in concentration with an increase in wind speed for stronger winds (speeds above 2.5 m s^{-1}), which suggests that for the weak winds, the advection of pollution dominates over ventilation. For stronger winds, the effects of advection are dominated by ventilation. Apart from the SSE winds shown in the Panel b, such a pattern is also found for SE and ESE winds. Because the industrial area of Zagreb is southeast

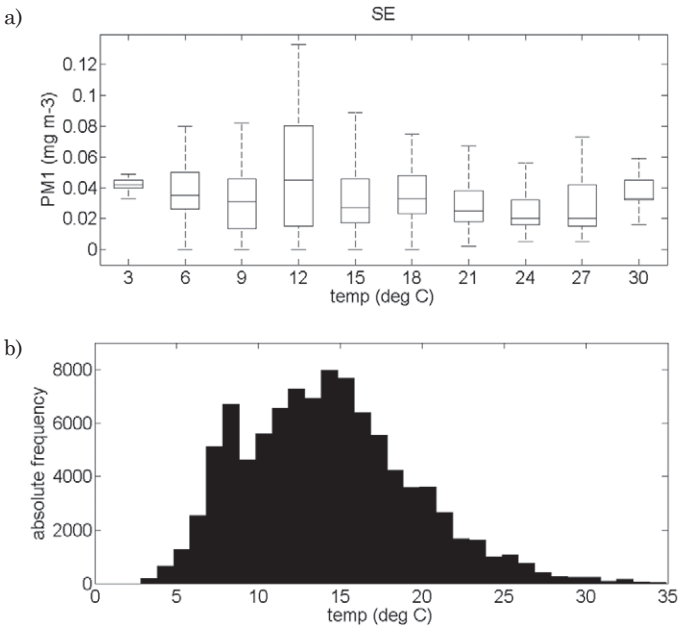


Figure 9. Box-plots of the 1-min mean PM1 mass concentrations vs. the 1-min mean air temperature for southeastern flows (a), and frequency distribution of 1-min mean temperature values for all flows (b).

of the measuring site (approximately 8–9 southeastward, Fig. 1a), we believe that the role of the industrial area in the establishment of the second pattern is substantial. Finally, the third pattern (Panel c) exhibits a decrease in PM1 concentration with an increase in wind speed for weaker winds, and simultaneously, an increase in concentration with an increase in wind speed for strong winds (above 5.5 m s^{-1}). Apart from the western winds shown in Panel c, a similar U-shaped behavior is observed for E, ENE, SSW, SW, W and WSW flows. As previously mentioned (Klaić, 2012), stronger winds should begin with large-scale (synoptic) forcing, and they should be accompanied by large-scale advection. Thus, the U-shaped pattern suggests a noticeable transport of pollution from larger distances (i.e., regional or long-range transport).

Similar to the road results (Panel f in Fig. 4), the majority of the 16 wind directions and the entire dataset as analyzed without respect to wind direction show a ∩-shaped dependence of the 1-min mean PM1 mass concentrations on the vertical wind speed (not shown). The majority of the results for individual wind directions and the results for the entire data set exhibited maximum PM1 concentrations for the weak vertical component of the airflow (namely, for the vertical wind component between –10 cm s^{-1} and +10 cm s^{-1}). Exceptions are south-southeastern winds with maximum concentrations for the weak convection

(vertical velocities between 10 and 30 cm s^{-1}), and southeastern, west-southwestern and southern winds where maximum concentrations were related to air subsidence (with the centers of classes of vertical components at –20, –40 and –60 cm s^{-1}, respectively).

Fig. 11 shows the dependence of 1-min mean PM1 mass concentrations on the 1-min mean relative humidity values. The majority of wind directions and the entire data set show an increase in concentration with an increase in relative humidity, except at the highest values of relative humidity (Panels a and b). As in the previous study (Klaić, 2012), we argue that the highest values of relative humidity are most likely related to precipitation or fog events. Thus, the decrease in concentration for the highest values of relative humidity is most likely due to the removal of particles through rainout and/or washout processes. Such a decrease is found for relative humidity above 65% (for SSW winds, not shown), 75% (for all data, N, NE, NNW, NW, SE and SSE flows) or above 85% (for E, ENE, ESE, NNE and S winds). In contrast to the previous study, in which all directions exhibited patterns similar to those shown in Panels a and b, here SW, W, WNW and WSW winds do not show a decrease in PM1 concentrations for the highest class of relative humidity (see, for example, concentrations for relative humidity over 95% for WSW winds, Panel c). The pattern shown in Panel c corresponds to

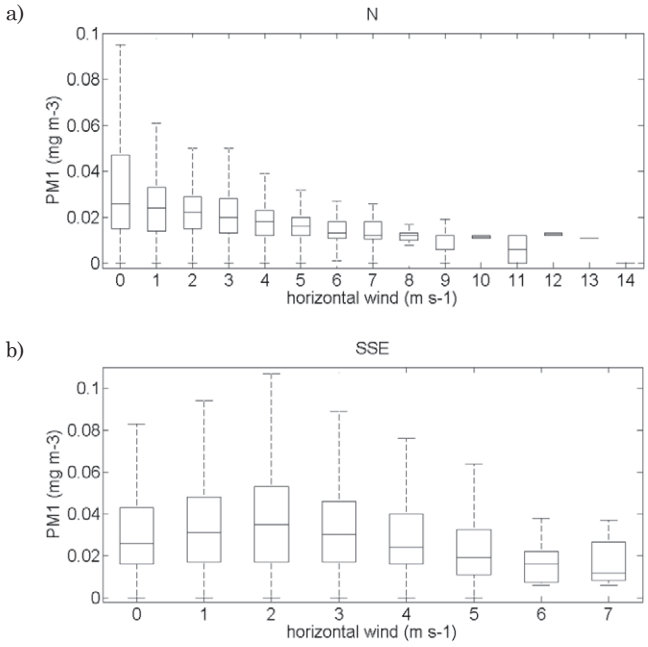
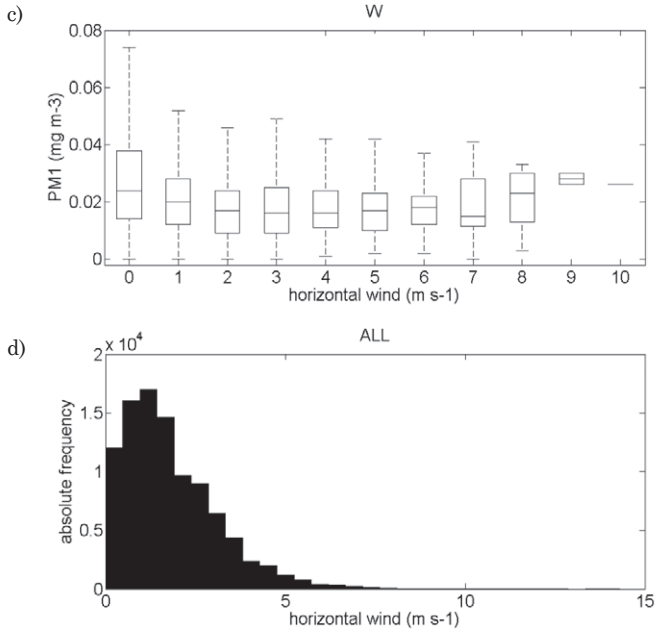


Figure 10. Box-plots of the 1-min mean PM1 mass concentrations vs. the 1-min horizontal wind speeds for the northern (a), south-southeastern (b), and western (c) flows. Panel (d) shows frequency distribution of 1-min mean horizontal wind values for the entire data set.



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airflows with a significant western component, that is, the airflows that are likely governed by large scale westerlies, and thus accompanied by the larger scale transport of pollutants, that is, the arrival of older particles at the measuring site. This assumption is further corroborated for W, WSW and SW winds, which exhibit U-shaped dependence on the horizontal wind speed; this dependence suggests the transport of pollution from more distant areas, such as the industrialized areas of northern Italy, for example (Fig. 10c). We assume, there-

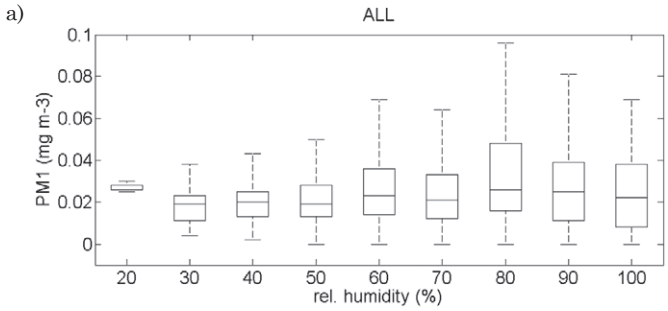
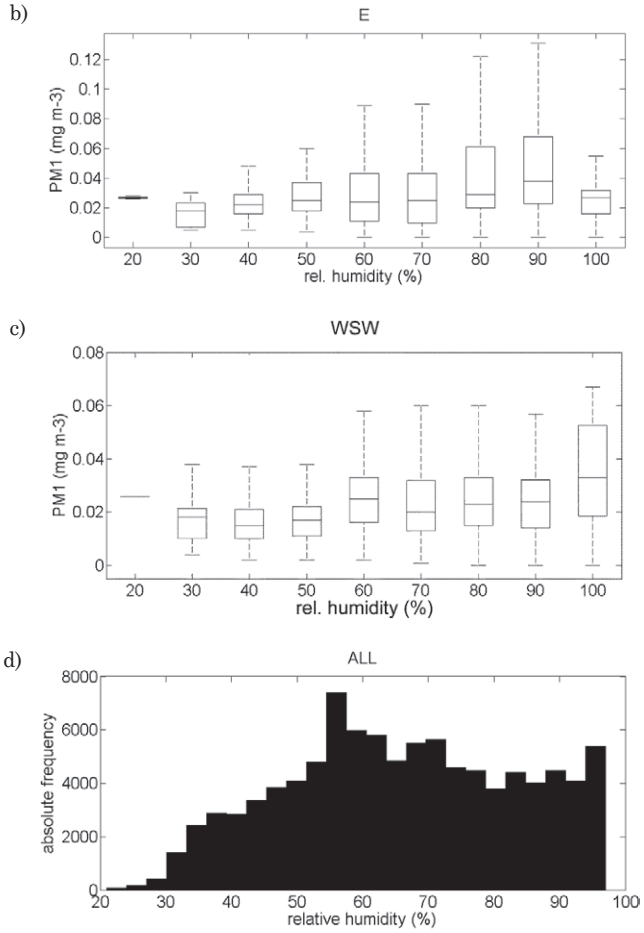


Figure 11. Box-plots of the 1-min mean PM1 mass concentrations vs. the 1-min relative humidity for entire data set (a), and for the south-southeastern (b), and western (c) flows. Panel (d) shows frequency distribution of 1-min mean relative humidity values for the entire data set.



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fore, that compared to the majority of the 16 investigated airflows, the differing dependence of PM1 concentrations on relative humidity for at least W, WSW and SW flows is due to the presence of older particles.

5. Conclusions

The 1-min mean PM1 mass concentrations measured next to a road with weak to moderate traffic at the height of 17 m AGL in the residential area of Zagreb in February 2009 were associated with traffic density only if more than 22 vehicles drove past in both directions per minute. This finding implies that for the weaker local traffic, the PM1 mass concentration was dominated by the

advection of particles from sources other than local traffic. We note however, that above result should be considered as preliminary, since it was obtained from relatively small dataset (439 values). In future, larger road dataset should be investigated, and it should be accompanied with an analysis of PM1 data measured simultaneously at the site placed far away from the road.

The average PM1 mass concentration (38.7 μg m^{-3}) next to the road at a height that roughly corresponds to the height at which pedestrians breath is comparable to the estimated exposure of a cyclist to PM1 in the town of Mol (Belgium) (Berghmans et al., 2009).

The average PM1 mass concentration on the roof terrace at a height of 15.8 m AGL in the same area of Zagreb for two periods in 2010 and 2011 (both in the warm part of the year) was substantially lower (26.5 μg m^{-3}) than the average PM1 concentration in the vicinity of the road. The differences between these two are not surprising because the two data sets correspond to different seasons (aside from different meteorological conditions and different boundary layer heights, this difference also implies different pollutant emissions), different measurement heights, and varied distances from the roads (which are major sources of fine particles in urban environments).

The April 2010 eruption of the Eyjafjallajökull volcano (Iceland) did not affect near-ground (15.8 m AGL) PM1 mass concentration levels recorded in the urban, residential area of Zagreb.

The diurnal variation of PM1 concentration has two maximums corresponding to the morning and evening rush hours, suggesting the major role that traffic has on recorded PM1 levels. However, mean PM1 mass concentration rose at the measuring site, which is approximately 100 m east of a road with weak to moderate traffic and approximately 50 m north of a road with weak traffic (i.e., roof concentrations), does not reflect any influence of the two nearest roads on PM1 levels. That is, the concentrations at the residential measuring site are mainly affected by the advection of pollution from other urban sources (i.e., from other roads), among which the industrial area of Zagreb (which is approximately 8–9 km southeast of the site) seems to be the most important. This result is in accordance with the previous findings of Bešlić et al. (2007) for the same area of Zagreb showing elevated PM2.5 and PM10 mass concentrations for southeastern flows.

In contrast to the results of Hussein at al. (2006), in which there was no relationship between relative humidity and the number concentrations of both the accumulation-mode fraction and ultrafine particles in urban environment, our data (both the near-road and roof measurements) exhibit a clear increase in PM1 mass concentration with relative humidity, except at the highest values of relative humidity and for SW, W, WNW and WSW flows. The large values of relative humidity occur during precipitation or fog events. Thus, the decrease in PM1 concentrations at the highest values of relative humidity is likely due to particle depletion in the atmosphere due to precipitation or the rainout process during fog. We believe

that the distinct pattern that is obtained solely for SW, W, WNW and WSW flows, which shows an increase in PM1 concentration at the highest values of relative humidity, might be due to the presence of older particles transported towards the measuring site by regional or long-range transport. The more so, the dependence of PM1 concentrations on the horizontal wind speed for W, WSW and SW flows shows the notable influence of distant pollution sources.

Our results also suggest an increase in PM1 mass concentrations with the air pressure, which was not the case with Hussein et al. (2006). We note, however, that the present study differs from the study of Hussein et al. in the type of concentration investigated (we investigate mass concentration, while Hussein et al. inspect number concentration), measurement height, particle fractions investigated and the temporal scale of analyzed data. However, we emphasize that an increase in PM1 concentration with air pressure is expected because high-pressure values point to a stronger static stability and, consequently, higher near-ground pollutant concentrations.

The relationship between the 1-min mean global radiation and the 1-min mean PM1 mass concentration is not completely clear. Based on the results for different wind directions, which correspond to different advections of pollution), we hypothesize that for newly formed particles, this relationship is U-shaped, while for older particles and higher global radiation values (roughly above 700 J cm^{-1}), PM1 concentrations decrease with an increase in global radiation. We note, however, that this should be investigated further.

While Hussein et al. (2006) found a prominent increase in hourly mean accumulation-mode (particle sizes between 100 nm and 2.5 μm) number concentrations with air temperatures above 15 °C, we found an increase in 1-min mean PM1 mass concentrations with air temperatures at least few degrees higher. These differences in the temperature thresholds are not surprising because the two studies correspond to 1) different particle fractions (though these coincide with a wide range, that is, from 100 nm to 1 μm), 2) different concentration measures (though higher number concentrations should imply higher mass concentrations) and 3) different time scales. We conclude that an increase in PM1 concentration with higher air temperatures, which is seen in the results for all 16 wind directions, should be due to the role of higher ambient temperatures in particle formation. Thus, it is reasonable to assume that a quicker response to temperature increases (i.e., the formation of particles during a short time interval of ~ 1 min) requires higher ambient temperatures than a slower response (i.e., formation of particles during an interval of ~ 1 h).

Although we cannot quantitatively distinguish between the contribution of urban sources and the regional and/or long-range transport of pollutants to PM1 levels, the results regarding the dependence of PM1 concentrations on horizontal wind speed show the influence of distant sources on the E, ENE, SSW, SW, W and WSW flows. The same also corresponds to recent findings of Costabile et al. (2009) and Timonen et al. (2008). While Costabile et al. suggest the importance

of the long-range transport in the formation of droplet-mode particles (i.e., particles with diameters from 300 to 800 nm and with a mean aerodynamic diameter of 0.7 μm (John, 2001)) in the urban atmosphere, Timonen et al. point to the importance of the long-range transport in the transport of accumulation-mode particles (particles between 0.1 and 1 μm).

In future work it would be interesting to separate different, typical synoptic setups more firmly, and to investigate their role in the PM1 levels. This could be done by taking into account typical weather types (e.g., Bešlić et al., 2007) or by performing cluster analysis (e.g., Green et al., 2008). Of course, both approaches would require larger datasets.

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SAŽETAK

Utjecaj meteoroloških uvjeta i slabe do umjerene gustoće prometa na koncentracije lebdećih čestica PM1 u rezidencijalnom dijelu Zagreba

Zvjezdana Bencetić Klaić, Kristijan Ružman, Ivan Smiljanić, Miran Stojnić i Philipp Erbe

Analizirane su 1-minutne masene koncentracije lebdećih čestica aerodinamičkog promjera do 1 μm (PM1) i njihova ovisnost o meteorološkim uvjetima (tlaku zraka, brzini horizontalnog i vertikalnog vjetra, smjeru vjetra, globalnom zračenju, temperaturi zraka i relativnoj vlažnosti) te ovisnost koncentracija o gustoći prometa. Koncentracije PM1 mjerene su na dva bliska mjerna mjesta u rezidencijalnom dijelu Zagreba pomoću 8520 DUSTTRAK™ aerosol monitora (TSI Inc., Shoreview, MN, USA). Jedno mjerno mjesto nalazilo se uz samu prometnicu sa slabim do umjerenim prometom, a drugo na krovnoj terasi približno 100 m istočno od prometnice. Visine instrumenta bile su na 1.7 m (uz cestu) i 15.8 m (na krovu) nad tlom. Oba eksperimenta provedena su istim mjernim in-

strumentom, te se stoga odnose na različite vremenske intervale. Rezultati mjerenja na krovu pokazuju da obližnja prometnica s rijetkim do umjerenim prometom ne utječe na koncentracije PM1 u točki udaljenoj oko 100 m od ceste. Preliminarni rezultati mjerenja uz cestu pokazuju da lokalni promet utječe na koncentracije tek pri većoj gustoći prometa, odnosno ako u jednoj minuti pokraj instrumenta prođu bar 22 vozila. Međutim, čini se da na koncentracije PM1 u sjevernom rezidencijalnom dijelu Zagreba jako utječu udaljeniji dijelovi grada. Rezultati dobiveni za neke smjerove vjetra ukazuju na utjecaj regionalnog i/ili daljinskog transporta na razine PM1. Konačno, pokazuje se da 1-minutni srednjaci masene koncentracije PM1 ovise o 1-minutnim vrijednostima meteoroloških varijabli.

Ključne riječi: advekcija čestica, koncentracije, DUSTTRAK™ Aerosol Monitor, gustoća prometa, 1-minutni srednjak

Corresponding author's address: Zvjezdana Bencetić Klaić, University of Zagreb, Faculty of Science, Department of Geophysics, Andrija Mohorovičić Geophysical Institute, Horvatovac 95, HR-10000 Zagreb, Croatia, tel: +385 (0)1 4605 929, fax: +385 (0)1 4680 331, e-mail: zklaic@gfz.hr