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Influences of outdoor meteorological conditions on indoor wintertime short-term PM_1 levels

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We investigated the relationship between wintertime 1-min mean urban indoor particulate matter mass concentrations for particles with aerodynamic diameter of < 1 µm (PM_1) and outdoor atmospheric conditions. Particle concentrations were measured by two light-scattering laser photometers. Aerosol monitors were placed in the ground and first floor corridors of an university building, with inlets at heights of 1.7 m above the floor. The building is located in residential area of Zagreb, Croatia. During the experiment usual student and employee activities were occurring within the building. Surface meteorological data were collected at a nearby outdoor location. Results show the dependence of indoor PM_1 on outdoor meteorology, with the strongest responses to air temperature and relative humidity, whilst global radiation impacts were almost negligible. Response times varied from 1.2 hours (for relative humidity) to 2.7 days (for global radiation). Furthermore, elevated mean concentrations point to the 8–9 km distant industrial zone. Both, PM_1 and meteorological data series exhibited semidiurnal, diurnal and the long-term (about 10–11 days and about 21 day) periodicity. The long-term periodicity of PM_1 time series might be associated with Rossby waves. Possible association with Rossby waves needs to be investigated further.

Keywords: cross-correlation, DUSTTRAK Aerosol Monitor, 1-min mean, residential, spectral analysis, time lag, urban

1. Introduction

During the last couple of decades numerous papers have reported significant effects of particulate matter (PM) on human health (e.g., Pope and Dockery, 2006; Politis et al., 2008; Jahn et al., 2011; Kelly and Fussell, 2012; Massey et al., 2013; Shields et al., 2013). As most people spend over 80% of the time indoors (e.g., Ramachandran et al., 2000; Klepeis et al., 2001; Bernstein et al., 2008;

Massey et al., 2012), indoor concentrations are important in determining personal exposure and respiratory tract deposition. Other significant factors influencing personal exposure are persons' activities and life style (e.g., Klepeis et al., 2001; Moschandreas and Saksena, 2002; Braniš et al., 2009; Singh and Sharma, 2009).

Major direct indoor sources of PM are smoking (e.g., Wallace, 1996; Jones, 1999; Wallace et al., 2003; Calvo et al., 2013), cooking, particularly frying (e.g., Wallace, 1996; Watson et al., 2002; Wallace et al., 2003; Abdullahi et al., 2013; Calvo et al., 2013), cleaning, vacuuming, dusting and sweeping (e.g., Wallace, 1996; Wallace et al., 2003; Raaschou-Nielsen et al., 2011), and dust re-suspension (e.g., Viana et al., 2011), which is more intense in carpeted spaces (Stranger et al., 2007). Other specific activities associated with working environments, for example, in chemistry laboratories and mechanical workshops (e.g., Žitnik et al., 2010) or three-dimensional printing (Stephens et al., 2013) also contribute to indoor PM. On the other hand, a decreased use of coal and wood for heating in developed societies has resulted in lower particulate pollution than in the past (Jones, 1999). In low-income and developing countries however, particularly in rural areas, indoor concentrations are still elevated due to use of unprocessed biomass as the primary fuel (e.g., Begum et al., 2009; Massey et al., 2009, 2013; Gurley et al., 2013).

Outdoor PM also affects indoor concentrations. Wallace et al. (2003) studied indoor PM of aerodynamic diameter < 2.5 μm ($PM_{2.5}$) in several US cities. They concluded that 25% of the indoor concentrations originate from outdoors. Martuzevicius et al. (2008) measured $PM_{2.5}$ concentrations in six residential houses located near major highways in Los Angeles, USA, and outside of these houses. Authors concluded that the structure of the house envelope and ventilation patterns were more important for indoor traffic-related aerosol than distance from the highway and traffic intensity. However, when Viana et al. (2011) investigated mass concentrations of nanoscaled particles (< 1 μm, PM_1) in a workplace in urban area of Barcelona, Spain, they detected a major impact of traffic emissions on indoor levels. At all times, including periods with closed windows, at least 73% of PM_1 concentrations originated from the outdoor. They attributed such high influence of outdoor concentrations to inadequate building insulation. Recently, Chithra and Shiva Nagendra (2013) also confirmed the importance of vehicular emissions on urban indoor PM concentrations.

Previous study of the relationship between short-term outdoor mass concentrations of PM_1 and meteorological conditions showed a clear dependence of 1-min mean PM_1 levels on concurrent relative humidity, wind speed and direction and air pressure (Klaić et al., 2012). In the present study we inspect if the signature of the outdoor meteorological conditions could be seen in the urban wintertime short-term indoor PM levels. Apart from investigation of relationships between PM_1 levels and concurrent meteorological variables, in the present study we also investigate a delayed response of indoor PM_1 to outdoor atmospheric conditions.

To our knowledge, results of the present study are the first research into indoor PM concentrations in Croatia. Additionally, the present research is associated with a very fine temporal resolution (1 min) of observed data. Ramachandran et al. (2000) also investigated the short-term indoor PM levels. However, they investigated 15-min mean $PM_{2.5}$ concentrations for spring and summer months, while here we focus on a smaller size fraction during the wintertime heating season. Compared to Schneider et al. (2004), who analyzed the role of meteorological conditions in indoor particle size distributions in an uninhabited apartment in order to develop a model for predicting the indoor concentrations of 0.5–4 μm PM fraction, here we investigate PM_1 levels observed in university campus building during everyday presence and common movements of students and employees. Further, the model of Schnieder et al. performed less well for both the wintertime and the fine (0.5–1.2 μm) fraction. Thus, we believe that results of the present study might be useful for modellers dealing with wintertime indoor fine fraction.

2. Measurements

2.1. Particle mass concentrations

Indoor 1-min mean PM_1 mass concentrations were measured in the building of the Department of Geophysics, Faculty of Science, University of Zagreb, Zagreb, Croatia (hereafter DG) during 12 November 2012 – 26 April 2013. The measuring site is in a residential area of northern Zagreb, approximately 1.5 km north-northeast from the city centre and 8-9 km northwest of Zagreb's industrial zone. More details on DG measuring site, its surroundings and placement with respect to the town geometry and closest roads can be found in Klaić et al. (2012).

Two light-scattering laser photometers (DUSTTRAK Aerosol Monitors,TSI, Inc., Shoreview, MN, USA; models 8520 and 8533, respectively), were placed 1.67 m above the floor, so their inlets corresponded to the average breathing height of 1.7 m. Prior to the experiment, both instruments were calibrated by the factory. During the experiment instruments were zeroed, inlets were cleaned and internal filters replaced on regular basis.

While Shields et al. (2013) pointed to some uncertainty in DUSTTRAK aerosol photometer measurements of $PM_{2.5}$, Ramachandran et al. (2000) argued that for finer aerosols such as $PM_{2.5}$, the DUSTTRAK Aerosol Monitor response can be 5-10 times higher than the true value depending on aerosol characteristics. On the other hand, Fromme et al. (2007) compared indoor $PM_{2.5}$ concentrations determined gravimetrically with a laser aerosol spectrometer data (Dust monitor 1.108, Grimm technologies, Ainring, Germany). By gravimetry they obtained generally higher values, but these were strongly correlated with the laser aerosol spectrometer data. Therefore, we calibrated both DUSTTRAK Aerosol Monitors against gravimetric data after the indoor experiment. Namely, during 3 March – 7 April 2015 both instruments were placed next to the closest available (about 1 km distant) gravimetric sampler (more details on gravimetric measuring site are given in the last paragraph of this Section). A comparison between daily mean light-scattering laser photometers data and gravimetric data resulted in correlation coefficients of 0.976 and 0.985 for models 8533 and 8520, respectively, while obtained correction functions are as follows:

$$[PM_1]_{\text{corrected}} = 0.320 \times [PM_1]_{\text{observed}} + 2.434 \qquad \text{(model 8533)}$$

and,

$$[PM_1]_{\text{corrected}} = 0.354 \times [PM_1]_{\text{observed}} + 4.414 \qquad \text{(model 8520)},$$

where all concentrations are given in $\mu g\,m^{-3}$.

Both aerosol monitors were located in building corridors, adjacent to the stair case as described in Ollier (2013) and shown in Fig. 1a, and they were run simultaneously. The Model 8520 was placed at the ground floor, while the Model 8533 was at the first floor. Concentrations were not recorded during instrument maintenance and while downloading the data to a computer. Recorded concentrations were afterwards corrected as described in the previous paragraph. After omitting extremely high PM_1 concentrations recorded at both floors during the working hours of 3–5, 10–11 and 13 December 2012 that were caused by construction works within the building, in total 233011 1-min values per each floor remained.

On weekdays, the average number of persons using the two floors is 65 (35 staff and 30 students). Since classrooms are located on the first floor, students mainly stay there. The staff offices are at both floors. The arrival and leaving times of both staff and students are varying. The busiest periods of movement are from 07:00 to 09:00 local standard time (LST), from 12:00 to 14:00 LST and from 15:00 to 17:00 LST, and they correspond to the arrival of most of the staff, lunch breaks and leaving the building, respectively. After 17:00 there are around 10 persons in the building, leaving at varying times up to 21:00 LST when the building is empty (on weekdays one member of staff is mandatorily in the ground floor office until 21:00 LST). Students' movement within the building is variable, depending on lectures and exams schedules and use of facilities. However, all students generally leave the building by 16:30. Floors are cleaned once a day, in the morning (ground floor) and the afternoon (first floor). Smoking is forbidden in the building but smokers gather outside the entrance of the building at various times. During the weekend, activity is generally low. On Saturdays, one member of the staff is mandatory in the ground floor office 07:00–14:00 LST, and no one on Sundays. However, staff members come to the building occasionally at any time over the weekend, and this is not recorded.

Central heating was turned on days prior to the first day of experiment, and it was turned off on 15 April 2013. Thus, the majority of collected PM_1 data (218985 out of 233011, that is, ≈ 94 %) corresponded to the heating season. The gas unit for central heating is placed in the DG building basement. On weekdays the heating is set to 21°C from 06:00 to 21:00 LST and to 17°C from 21:00 to 06:00 LST. On weekends the heating is set to 17°C throughout the day. For Christmas holiday period the temperature was set to 19°C from 21:00 LST 22.12.2012. to 06:00 LST 02.01.2013. In the immediate vicinity of the monitors windows were closed during the entire experiment. Yet windows of the offices on the adjoining corridors were occasionally opened.

Since DUSTTRAK monitors cannot operate at low wintertime temperatures (temperatures below 0°C) and DG premises are not equipped for gravimetric measurements, we could not perform simultaneous outdoor measurements in the vicinity of indoor site. Nevertheless, in order to check the consistency of collected indoor concentrations, we compared them with the nearest available outdoor data, that is, with daily mean outdoor PM_1 concentrations routinely observed by Institute of Medical Research and Occupational Health. The outdoor measuring site is in a similar residential environment of the northern part of Zagreb, approximately 1 km north-northeast of the indoor site, and it is considered representative for urban background levels. It is 50 m away from the road with moderate traffic (although the traffic there is somewhat denser than the traffic in the vicinity of the indoor measuring site) and at least 30 m away from closest high obstacles, such as buildings and trees. The sampler is placed on an asphalt surface surrounded by grass. The inlet is 1.7 m above the ground. PM_1 samples are collected daily on quartz fibre filters (Whatman QMA, diameter of 47 mm). Samplings are performed by low volume samplers with a flow rate of 2.3 $m^3 h^{-1}$ according to referent gravimetrical methods described by European standards EN 12341 and EN 14907. Filter conditioning and weighing is performed according to EN 14907; where the sampling filters are conditioned for 48 hours before being weighed, conditioned for an additional 24 hours, and then, weighed again. The same procedure (that is, two weighing once after 48-h conditioning, and again after an additional 24 h of conditioning) is repeated after the sampling.

2.2. Meteorological data

Meteorological variables (surface air temperature, air pressure, relative humidity, global radiation, and three-dimensional wind) are measured routinely at DG premises, as described in Klaić et al. (2012). Precipitation amounts accumulated over 1-minute time intervals are also measured. The precipitation sensor (META 2000, AMES, Brezovica, Slovenia) is placed in the vicinity of DG building. Occasionally, in the case of sensors' malfunctions and instruments reparations for example, meteorological data are not recorded. Additionally, every few months, measurements are stopped for up to one hour due to backup downloading of the data. During experiment period, in total 229586 of 1-min mean data per each meteorological variable were recorded simultaneously with PM data. (The actual number of collected data was somewhat larger, but meteorological data that were not accompanied with concurrent PM data were omitted.)

3. Methods

Apart from standard statistical methods, here we performed spectral analysis. Spectral analysis is widely used in meteorology to characterize time series in frequency domain and to separate scales between large (low frequency), synoptic (medium frequency) and turbulent (high frequency) signals (e.g., Penzar et al., 1980; Večenaj et al., 2011; Babić et al., 2012). Frequency analysis is also applied in PM studies (e.g., Marr and Harley, 2002; Choi et al., 2008; Tchepel and Borrego, 2010).

Here spectral analysis was performed based on Fourier series. This enables an assessment of contributions of different frequencies to the data variance. According to Fourier theorem a function $x(t)$, which is a time series of variable in question, can be represented by a system of trigonometric functions with appropriate phase and amplitude.

$$x(t) = \sum_{j=1}^{N/2} \left[A_j \cos(2\pi f_j t) + B_j \sin(2\pi f_j t) \right], \qquad t = 1, 2, \ldots, N$$

where N is the length of the time series, f_j is j^{th} frequency and $1 \leq j \leq N/2$.

The Fourier transform (which transforms between the time and frequency domain) of an integrable function is:

$$\hat{x}(f) = \int_{-\infty}^{+\infty} x(t) e^{-2\pi i f t} dt.$$

Here we employed the temporal resolution of 1 hour (except for outdoor PM_1, where a 1 day resolution was used). This eliminates waves with periods less than 2 hours (less than 2 days in the case of outdoor PM_1), which corresponds to Nyquist frequency of $(2\ \text{h})^{-1}$ (the Nyquist frequency is defined as the half of the sampling rate of a discrete time signal).

During 3–13 December 2012 construction works were performed within the building on some of the working days. Accordingly, indoor PM_1 levels for some hours during this period were considerably higher than for any other time during the experiment. Therefore, we applied the spectral analysis on time series from 00:00 LST 15.12.2012 to 09:00 LST 26.04.13. A standard pre-processing (e.g., Tchepel et al., 2010) included the following:

1) 1-min mean data were block averaged to get time series of 1-hour mean values (except for outdoor PM1, where the time series comprising of observed daily means was used).
2) The missing data were computed by simple linear interpolation from the data preceding and succeeding missing data.
3) In order to capture the underlying periodicity in the data the overall mean was subtracted from the time series.
4) Linear trend, that is, the least-squares fit of a straight line to the data was removed in order to obtain stationary data sets.

Lisac (1984) investigated multi-year airflow characteristics over the greater Zagreb area. She found four prominent scales in the wind energy spectra for both scalar wind speed and wind speed components. These include: 1) synoptic disturbances (periods between 2 and 12 days); 2) diurnal oscillations (20–30 hours); semidiurnal oscillations (10–20 hours); and 4) short-term oscillations (2–10 hours). Therefore, here we focused on variations from about couple of hours to couple of weeks. Accordingly, we calculated the spectra of power densities (periodogram) for overlapping (by 50%) 42-day segments of a datasets. The spectra are calculated by multiplying the time series with a hamming window with 2^{10} data points (1024 h ≈ 42.67 days). A fast Fourier transform algorithm was used afterwards to perform the Fourier transform. The periodogram for a discrete time series was calculated from a Fourier transform which was multiplied by its complex conjugate and the real part was maintained:

$$\Phi(f_k) = \left|\hat{x}(k)\right|^2 = \left|\frac{1}{\sqrt{N}}\sum_{t=0}^{N-1} x(t)e^{-2\pi i f_k t}\right|^2$$

where $k = 0, 1, 2, \ldots, N–1$, N is the number of observations, $x(t)$ is the time series segment and $f_k = \frac{k}{N}$.

Spectral density functions indicate the strength of the signal as a function of frequency. Also, the time integral over frequency spectrum corresponds to the variance of the time series data (Marr and Harley, 2002). Highest values in the spectra reveal the most important periodic components that contribute to the total variance of inspected time series.

4. Results and discussion

A comparison of daily mean indoor and outdoor PM_1 concentrations (not shown here) reveals that indoor daily values on both floors are consistent with outdoor values. All three time series (two indoor and one outdoor) exhibit similar patterns, that is, concentration peaks coincide throughout entire investigated period, although outdoor values are generally somewhat higher. We note however, that indoor and outdoor data correspond to two different 1 km distant locations, and that they are determined by different measurement methods. Thus, apart from results discussed in the Section 4.4, in the present discussion we will focus solely on indoor PM_1 results. Furthermore, the influence of meteorological conditions on outdoor 1-min mean PM_1 levels at the nearby location is already described in the previous study (Klaić et al., 2012).

4.1. Basic statistics of measured data

Table 1 shows basic statistics for 1-min mean measured variables. It is seen that on average, the PM_1 concentrations on the first floor are lower than concurrent ground floor concentrations. The instrument placed on the ground floor is closer to the building entrance and thus, it is more exposed to the entrainment of the air from outdoors due to door openings. Therefore, we hypothesize that the nearby outdoor PM_1 levels are likely to be higher compared with the indoor concentrations. Thus, the ground floor concentrations may be higher than the first floor concentrations due to more efficient mixing with the ambient, more polluted air. The identification of individual indoor sources is beyond the scope of this study (particulate chemistry was not investigated). Nevertheless, it is generally well known that some particles are exclusively emitted indoors. Indoor fine particles sources that can be relevant for this particular measurement site are cosmetic products (e.g., Conner et al., 2001), waxes and cleaners/polishers (e.g., Geller et al., 2002), and printing and photocopying (Viana et al., 2011). Additionally, dust resuspension due to people's passages can also be an important source of indoor PM_1 (Viana et al., 2011).

4.2. Temporal variations of indoor concentrations

Figure 1b shows temporal variations of indoor PM_1 concentrations. Again, it confirms that wintertime concentrations for the first floor are generally lower compared with ground floor values. The difference between the two floors is the largest for March (0.003 mg m^{-3}), while it is lowest for the December (0.001 mg m^{-3}). Further, for both floors the average concentrations are the highest for January (0.028 mg m^{-3} for the ground floor and 0.026 mg m^{-3} for the first floor, respectively), while they are lowest for April (0.012 and 0.010 mg m^{-3} for the ground and the first floor, respectively). This is in accordance with results showing that summertime $PM_{2.5}$ mass concentrations in classrooms of greater Munich area are significantly reduced in comparison with wintertime values (Fromme et al., 2007).

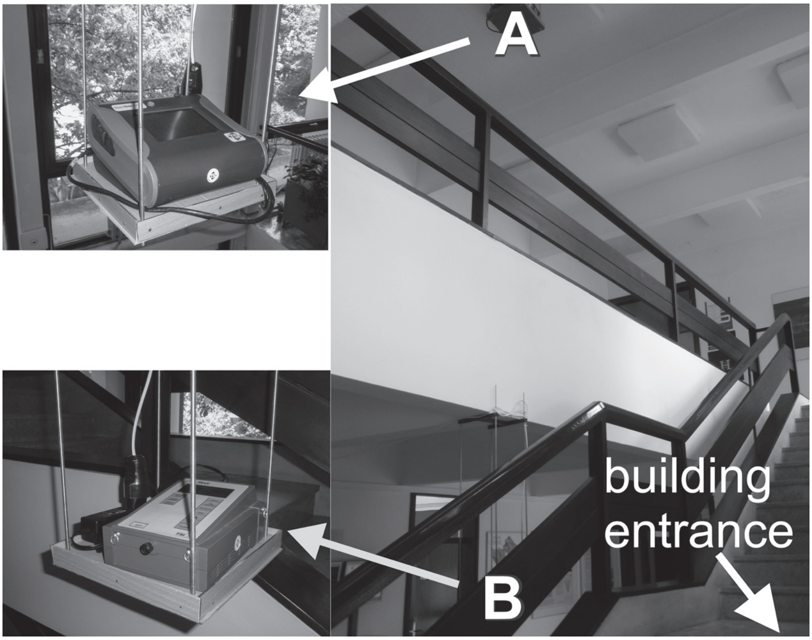
Table 1. Basic statistics for 1-min mean values measured 12 November 2012 – 26 April 2013. PM_1 and meteorological datasets have 233011 and 229586 values, respectively.

Quantity	Mean	Median	St. dev.	Max.	Min.	Most frequent value
PM_1 ground floor (mg m^{-3})	0.021	0.016	0.016	0.172	0.004	0.008
PM_1 first floor (mg m^{-3})	0.019	0.014	0.017	0.642	0.002	0.007
Global radiation (W m^{-2})	35.97	0	72.129	553	0	0
Precipitation (mm)	0.02	0	0.099	5	0	0
Air pressure (hPa)	992.3	992.8	8.59	1012.9	969.1	995.1
Relative humidity (%)	82.0	87	17.89	100	19	100
Air temperature (°C)	4.93	3.9	5.59	26.0	–12.4	0.3
Horizontal wind speed (m s^{-1})	1.05	0.6	1.32	20.6	0	0.1
Vertical wind speed (cm s^{-1})	11.76	5.6	19.73	181.0	-89.3	0.6

Diurnal variations of PM_1 concentrations (Fig. 1b, centre) exhibit elevated values during working hours, with a maximum between 12:01 and 13:00 LST for the first floor (0.021 mg m^{-3}) and between 10:01 and 12:00 LST (0.023 mg m^{-3}) for the ground floor. The lowest average values for both floors are observed between 05:01 and 06:00 LST (0.017 and 0.019 mg m^{-3} for the first and the ground floor, respectively). We note that the time period associated with elevated concentrations is longer for the first (from 09:01 to 16:00 LST) compared with the ground floor (from 09:01 to 13:00 LST). This may be due to particle resuspension, which should be more intense and enduring on the first floor due to students' and teachers' movements during breaks between lectures. Finally, diurnal variation exhibits a secondary maximum between 21:01 and 22:00 LST (0.019 and 0.021 mg m^{-3}, for the first and the ground floor respectively). We note that at the beginning of this hour the one member of the staff that has to be in the office at the ground floor until 21:00 LST leaves the building. However, bimodal diurnal variation was also obtained for summertime outdoor PM_1 concentrations observed at the nearby location (Klaić et al., 2012), although the secondary outdoor maximum occurred earlier (at 19:00 LST), and, for the outdoor PM_{10} fraction (< 10 μm) at other urban locations (Jelić and Klaić, 2010).

Weekly variations of PM_1 concentrations (Fig. 1b, bottom) show the highest values for Monday (0.023 and 0.025 mg m^{-3}, for the first and ground floor, respectively), while the lowest average concentrations are recorded on Saturday (0.017 and 0.019 mg m^{-3}). It is interesting that on Sunday, when the building is generally empty, average concentrations for both floors are about 0.001 mg m^{-3} higher in comparison with Saturday.

a)



b)

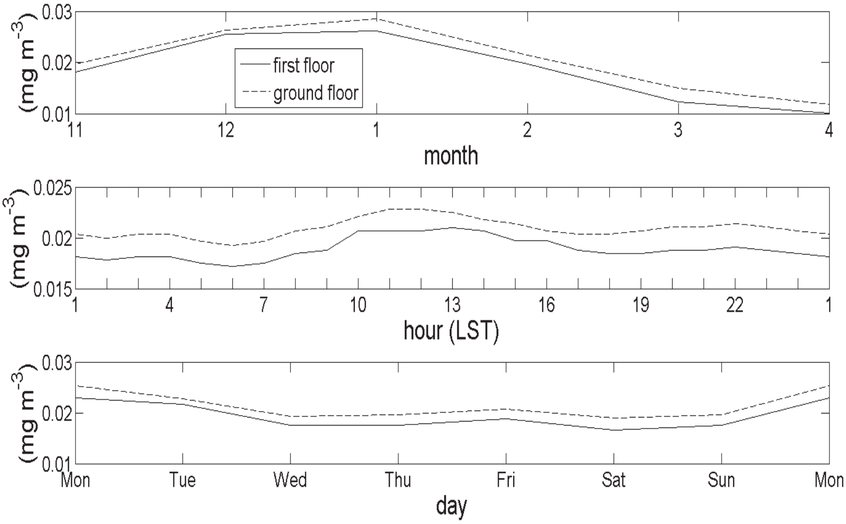


Figure 1. *a*) Location of aerosol monitors. Positions of models 8533 and 8520 are denoted by A and B, respectively. The building entrance is in front of the stair case (not seen at the present photo). *b*) Average temporal variations of indoor PM_1 mass concentrations. Diurnal variations in concentrations (middle graph) correspond to hourly means calculated for the time interval starting at the 1^{st} and ending at the 60^{th} minute of particular hour. For example, the average concentration shown for 07:00 LST corresponds to the time interval from 06:01 to 07:00 LST.

4.3. Relationships between indoor PM_1 concentrations and concurrent outdoor meteorological conditions

Figure 2 and Tab. 2 show relationships between the indoor PM_1 concentrations and concurrent outdoor meteorological conditions, while frequencies of the observed meteorological values are depicted in Fig. 3. As seen from Tab. 2, correlations between the short-term indoor PM_1 levels and short-term outdoor meteorological conditions are stronger for the ground floor in comparison with the first floor. This is in accordance with our previous hypothesis that the ground floor is more exposed to outdoor influences. That is, ground floor concentrations are more affected by the outdoor PM_1, and outdoor PM_1 depends on meteorological conditions (e.g. Klaić et al., 2012). On the other hand, the influence of outdoor meteorology on first floor PM_1 levels is dominated by indoor sources, particularly by resuspension (compared with the ground floor, this floor is used by more people). Nevertheless, outdoor air temperature still influences the first floor PM_1 levels ($R = -0.26$).

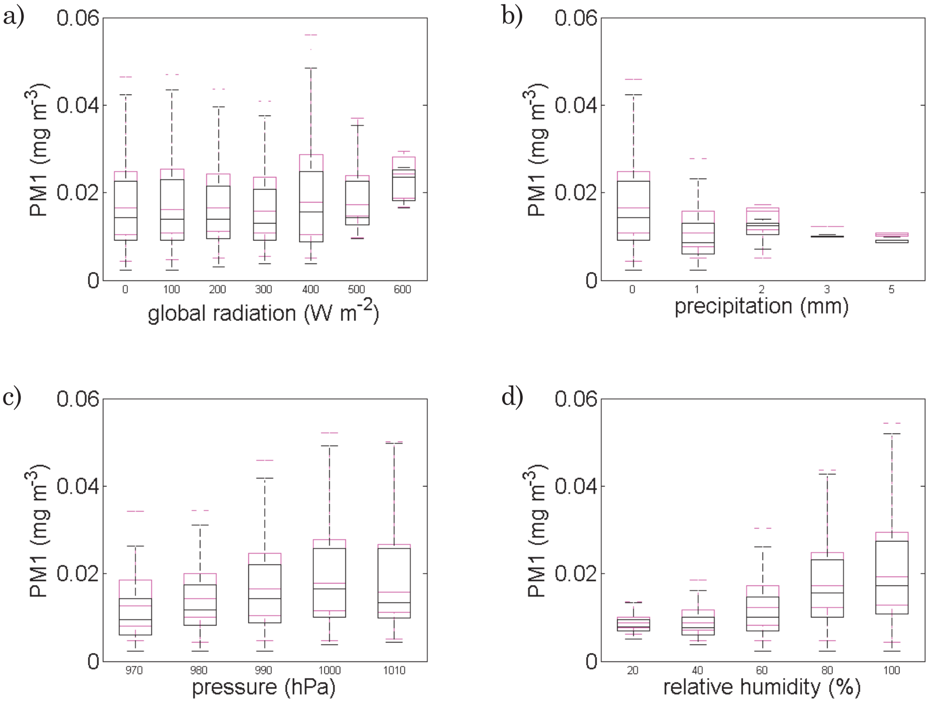


Figure 2. Box plots of the 1-min mean indoor PM_1 mass concentrations *vs.* concurrent 1-min mean ambient meteorological variables for the period 12 November 2012 – 26 April 2013. Each box shows the 25th and 75th percentile and median. The whiskers show the most extreme values that are less than 1.5 times the inter-quartile range away from the top or the bottom of the box. Magenta and black colors correspond to the ground floor and the first floor data, respectively.

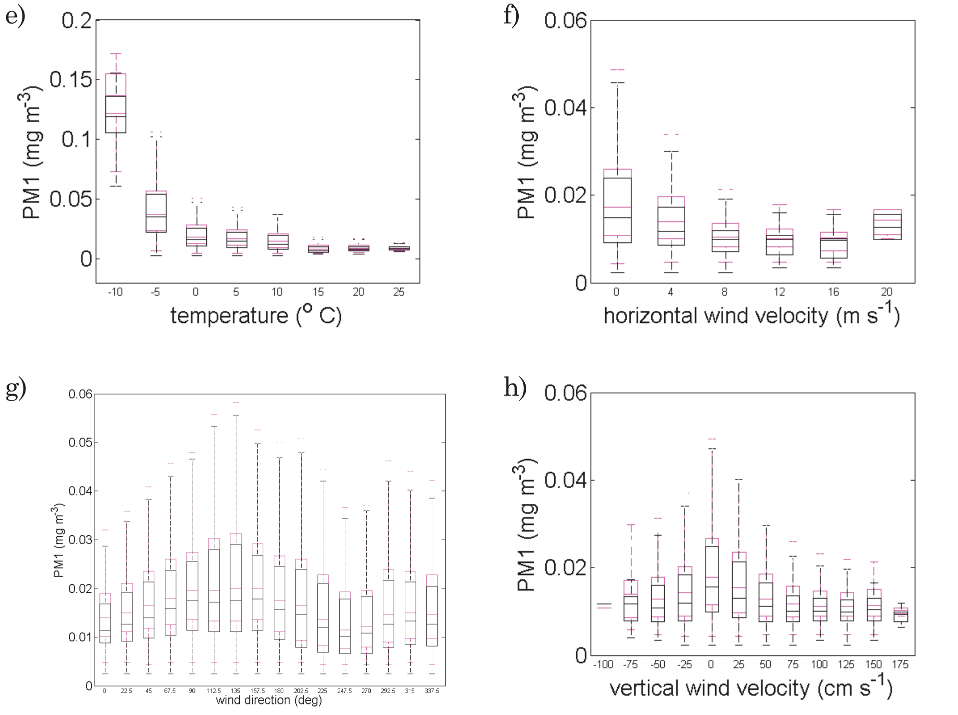


Figure 2. Continued.

Generally, the correlation between indoor PM_1 and concurrent outdoor meteorology (Tab. 2) is the strongest for temperature (both floors) and relative humidity (ground floor). (We note that for a large sample, the low value correlation coefficient R may be statistically significant (e.g., Tompkins, 1992). For example, for the sample comprising of 'only' 250 pairs of data R as low as 0.13 is significantly different from zero at 0.05 level. In the present study we analyzed 229586 pairs of data.) Correlation is almost negligible for the 1-min mean precipitation amount and for the global radiation at both floors, while there is no correlation for the air pressure for the first floor. Still, an increase of the median concentration with the increase of air pressure (except for the pressure class of 1010 hPa, Fig. 2c) is clearly seen for both floors.

While PM_1 concentrations decrease with an increase of outdoor temperature (Fig. 2e, Tab. 2), they increase with an increase of outdoor relative humidity (Fig. 2d, Tab. 2). We note that these are opposed to results for the indoor $PM_{2.5}$ and indoor temperature and relative humidity (Fromme et al., 2007). On the other hand, the results for relative humidity obtained by the present study are similar to those for the outdoor short-term PM_1 concentrations (Klaić et al., 2012).

Table 2. Correlation coefficients (R) between 1-min mean PM_1 mass concentrations and concurrent 1-min mean values of meteorological variables for the period 12 November 2012 – 26 April 2013. R is calculated from 229586 pairs of data. PM_{1GF} and PM_{1FF} denote concentrations on the ground and first floor, respectively, while G, P, p, RH, T, V_h and W are global radiation, precipitation, air pressure, relative humidity, air temperature, and horizontal and vertical wind speed, respectively. Except for the correlation coefficient between p and PM_{1FF}, all listed correlation coefficients are statistically significant at the 0.05 level.

R	PM_{1GF}	PM_{1FF}	*G*	*P*	*p*	*RH*	*T*	V_h	*W*
PM_{1GF}	1	0.92	–0.02	–0.02	0.12	0.29	–0.37	–0.13	–0.10
PM_{1FF}		1	–0.01	–0.03	0.00	0.06	–0.26	–0.09	–0.08
G			1	–0.05	0.03	–0.30	0.20	0.01	0.19
P				1	–0.22	0.16	–0.04	0.05	0.01
p					1	–0.26	0.12	–0.10	–0.02
RH						1	–0.53	–0.06	–0.19
T							1	–0.17	0.08
V_h								1	0.16
W									1

As seen from the Fig. 2f, an increase in outdoor horizontal wind velocity results in decreased indoor PM_1 levels, except for the strongest wind speeds (20 m s^{-1} class). This suggests the importance of outdoor ventilation, which results in generally lower outdoor PM_1 levels and, consequently lower indoor levels. The exception (an increase of PM_1 levels for the 20 m s^{-1} wind speed class in comparison with PM_1 for 8, 12 and 16 m s^{-1}) is most probably the random result of a very small number of data. Namely, out of the total 229586 wind data, only 6 had horizontal wind speeds above 16 m s^{-1} (Fig. 3).

As seen from the Figs. 2g and 4, indoor PM_1 levels strongly depend on the wind direction. Concentrations on both floors are highest for south-eastern winds (average values of 0.026 and 0.024 mg m^{-3} for the ground and the first floor, respectively) and south-south-eastern winds (0.026 and 0.023 mg m^{-3}), while they are lowest for west-south-western (0.016 and 0.015 mg m^{-3}) and northern (0.017 and 0.015 mg m^{-3}) flows. Additionally, although indoor wintertime concentrations are roughly 1.5 times larger in comparison with summertime outdoor values at nearby location (Fig. 4), the shapes of concentration roses are fairly similar. Especially prominent is the association of the highest concentrations for both wintertime indoor and summertime outdoor levels with south-eastern winds, which points to the influence of the industrial zone of Zagreb (which is placed southeast of the measuring site).

Figure 2h illustrates the dependence of indoor PM_1 on vertical wind speed (*W*). Although correlation coefficients for both floors suggest simple decrease of

PM_1 levels with an increase in vertical wind speed (Tab. 2), it is seen that concentrations are the highest for 0 cm s^{-1} vertical wind class, while they decrease with both strengthening of the subsidence ($W < 0$), and strengthening of the convection ($W > 0$). Since the measuring site is located in a moderately hilly environment at the south-facing slope of Mount Medvednica, negative vertical wind speeds can occur in association with night-time down-slope winds. Such winds are generally associated with a weak synoptic forcing (that is, with the high-pressure conditions), which according to Lončar and Bajić (1994) occur over Croatia the most frequently during the wintertime. Under such conditions the night-time down-slope winds over the area of interest have northern directions (e.g., Klaić et al., 2002; 2003). Accordingly, night-time down-slope winds transport clean air from the mountain toward the measuring site. This should result in a decrease in outdoor PM_1 levels and consequently, a decrease in indoor levels. On the other hand, the convection is associated with efficient vertical mixing, dilution of outdoor pollutants and a consequent decrease in indoor levels.

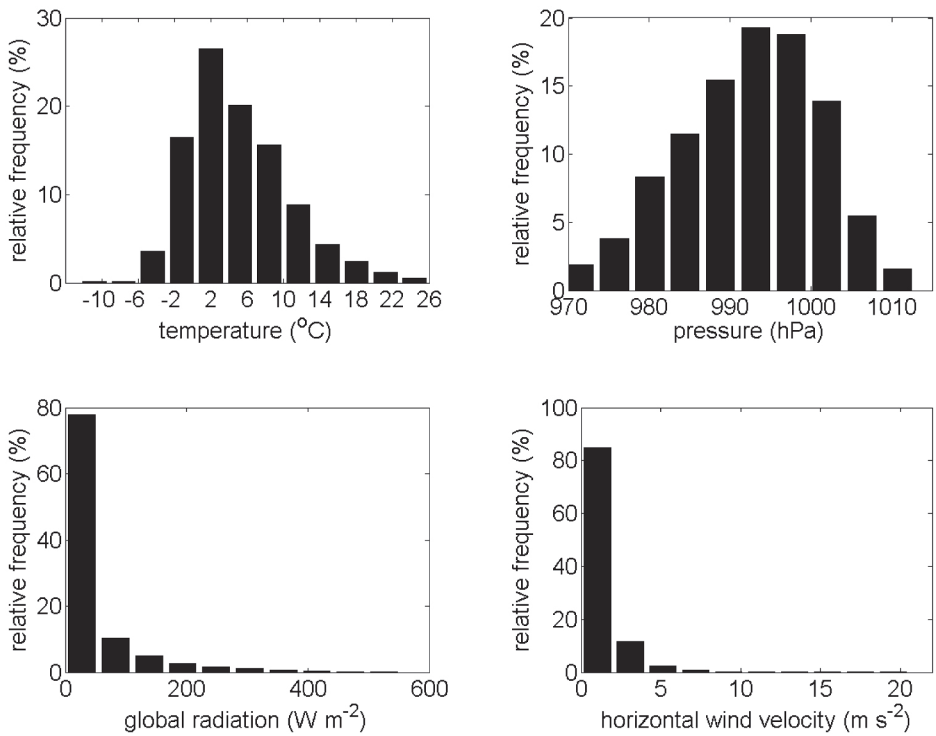


Figure 3. Frequency distributions of the 1-min mean meteorological variables for the period 12 November 2012–26 April 2013.

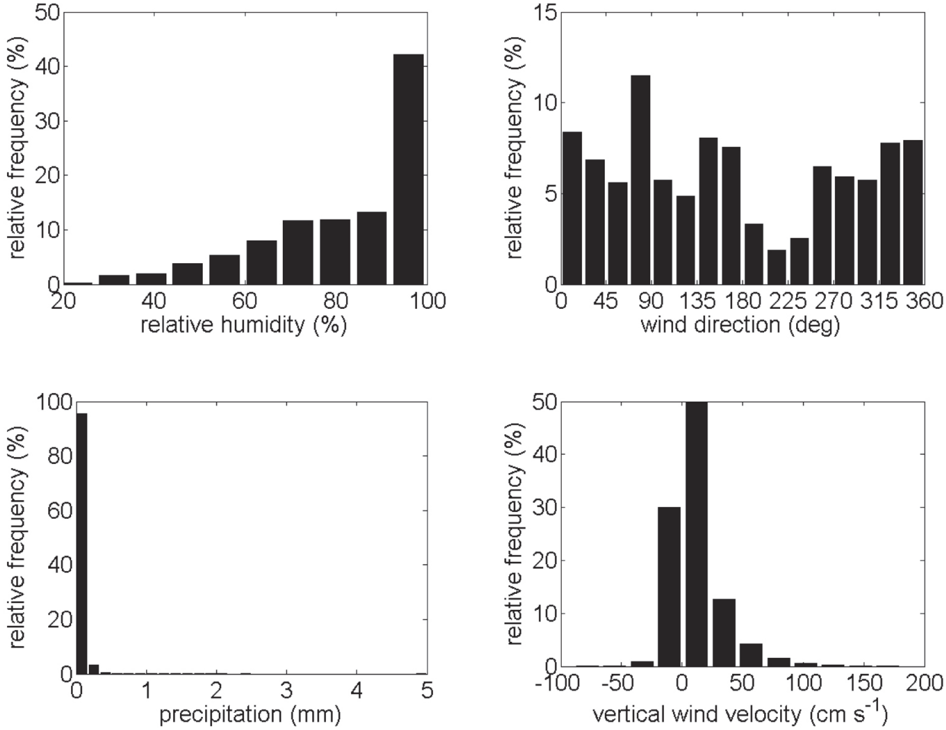


Figure 3. Continued.

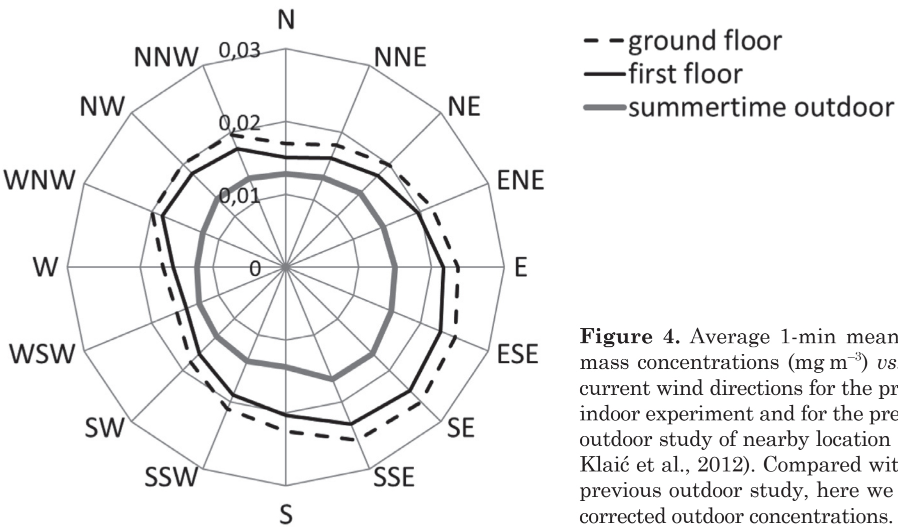


Figure 4. Average 1-min mean PM_1 mass concentrations (mg m^{-3}) *vs.* concurrent wind directions for the present indoor experiment and for the previous outdoor study of nearby location (after Klaić et al., 2012). Compared with the previous outdoor study, here we show corrected outdoor concentrations.

4.4. Cross-correlation

To investigate if there is a delayed response of indoor PM_1 concentrations to the outdoor meteorological conditions we calculated correlations between meteorological time series and time-lagged indoor PM_1 concentration time series for the range of delays from 0 min to 10 days (i.e., 14400 min) (Figs. 5a-g.). When calculating cross-correlation between the concentrations at the ground and the first floor, concentrations at the first floor were time-delayed with respect to the ground floor data (Fig. 5h).

In order to calculate cross-correlation functions based on single, continuous time series that covers the entire investigation period, 1-min mean data that were missing due to occasional instrument malfunctions or due to data being downloaded from the instrument were replaced by linearly interpolated values. While results shown in Tab. 2 correspond to original, discontinuous datasets that comprise of 229586 pairs of 1-min mean data (that is, 229586 minutes for which both

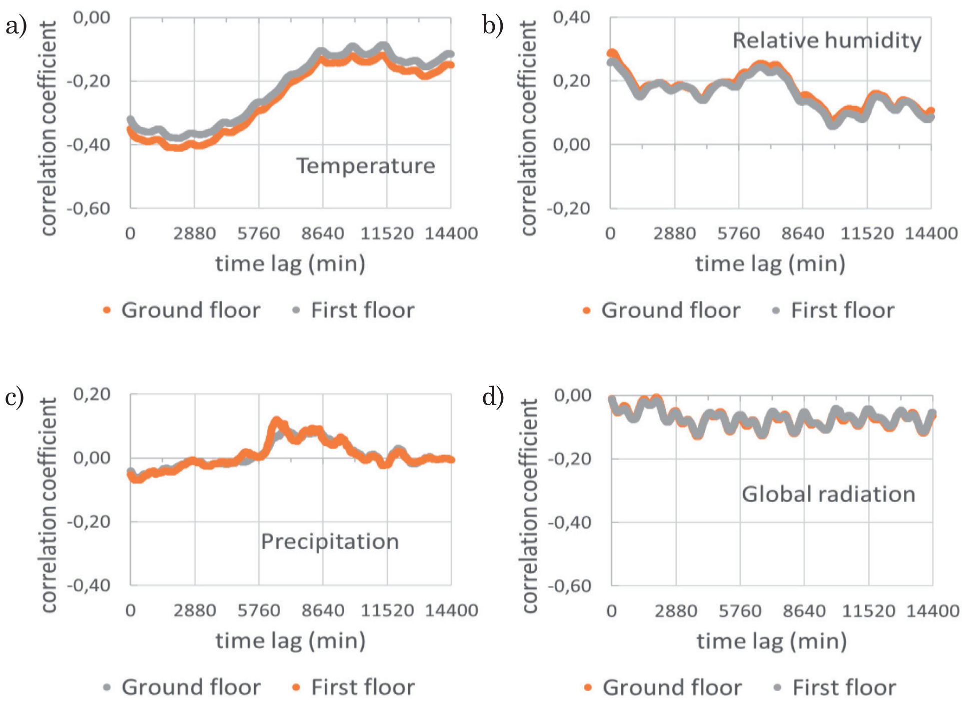


Figure 5. Cross-correlation functions produced by cross-correlating meteorological time series with up to 10 days time-lagged PM_1 time series (panels a-g). Panel h shows cross-correlation between PM_1 concentrations at the ground floor and up to 2 days time-lagged PM_1 concentrations at the first floor.

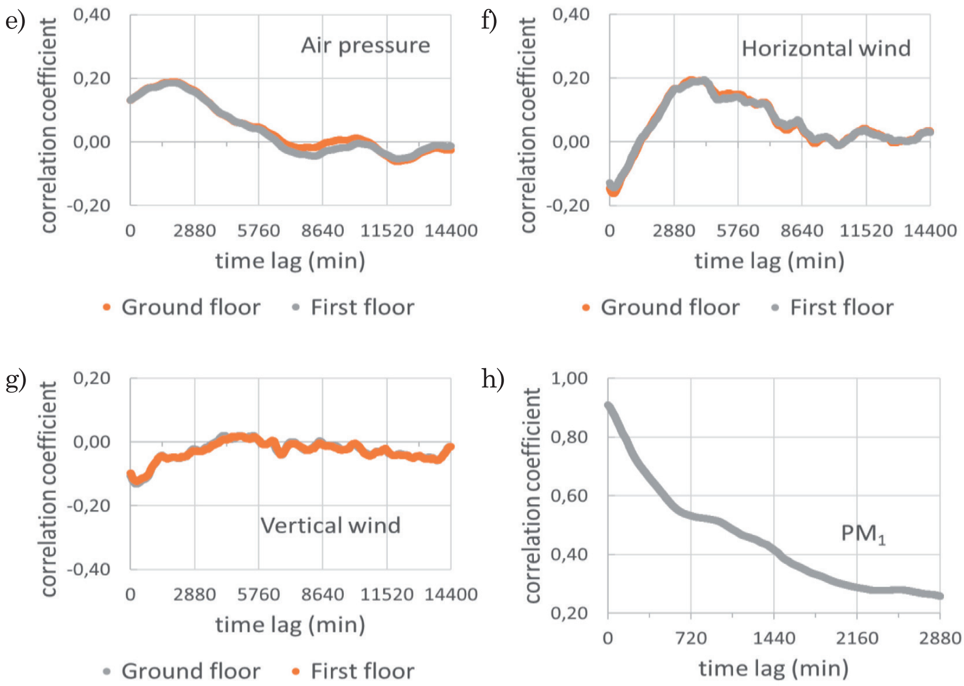


Figure 5. Continued.

concentrations and meteorological data are available), cross-correlation functions shown in Fig. 5 are calculated from continuous, 237366 minutes long time series, where 1.8% of PM_1 values and 3.3% of meteorological values were obtained by linear interpolation. Therefore, some of correlation coefficients obtained for the time lag of 0 min in Fig. 5 somewhat differ from those listed in Tab. 2.

As seen from the Fig. 5 and Tab. 3, for all investigated meteorological variables a time-delayed response of indoor PM_1 concentrations to meteorological conditions is somewhat stronger than the simultaneous response. That is, correlation coefficients between meteorological variables and time-lagged PM_1 concentrations (Tab. 3) are higher than correlation coefficients between PM_1 levels and concurrent meteorological conditions (Tab. 2 or Fig. 5 for time lag equal to 0 min). On the other hand, there is no lagged relationship between PM_1 levels on the two floors (the correlation coefficient between concentrations on the two floors is the highest for the time lag of 0 min, Fig. 5h). Again, the response is the strongest for outdoor air temperature and relative humidity, while it is the weakest for global radiation. The response time (that is, the time-lag needed for correlation coefficient shown in Fig. 5 to attain maximum absolute value) is the shortest for relative humidity – about 1.2 hours (ground floor) and 1.7 hours (first floor), respectively (Table 3). Quicker responses are also found for horizontal (about 2.5 hours for both floors) and vertical wind velocity (4.2 and 4.5 hours for the ground and the first floor, respectively), and precipitation (6.7 and 6.5 hours for the ground and the first floor, respectively). On the other hand, the response times above 1 day are found for the air temperature (about 1.5 days), air pressure (about 1.3 days) and global radiation (about 2.7 days.)

Table 3. Maximum correlation coefficients (absolute values) for cross-correlation functions shown in Fig. 5 (r_{max}) and corresponding response times (RTs, that is, time lags associated with maximum absolute values of correlation coefficients).

Cross-correlated time series		r_{max}	*RT* (min)
PM_1 on the ground floor *vs*. time-lagged PM_1 on the first floor		0.91	0
Outdoor air temperature *vs*. time-lagged indoor PM_1			
	Ground floor	–0.39	2170
	First floor	–0.36	2210
Outdoor relative humidity *vs*. time-lagged indoor PM_1			
	Ground floor	0.29	72
	First floor	0.26	102
Precipitation amount *vs*. time-lagged indoor PM_1			
	Ground floor	–0.06	400
	First floor	–0.07	392
Global radiation *vs*. time-lagged indoor PM_1			
	Ground floor	–0.05	3890
	First floor	–0.05	3890
Air pressure *vs*. time-lagged indoor PM_1			
	Ground floor	0.19	1940
	First floor	0.19	1869
Horizontal wind velocity *vs*. time-lagged indoor PM_1			
	Ground floor	–0.16	150
	First floor	–0.14	147
Vertical wind velocity *vs*. time-lagged indoor PM_1			
	Ground floor	–0.13	254
	First floor	–0.12	270

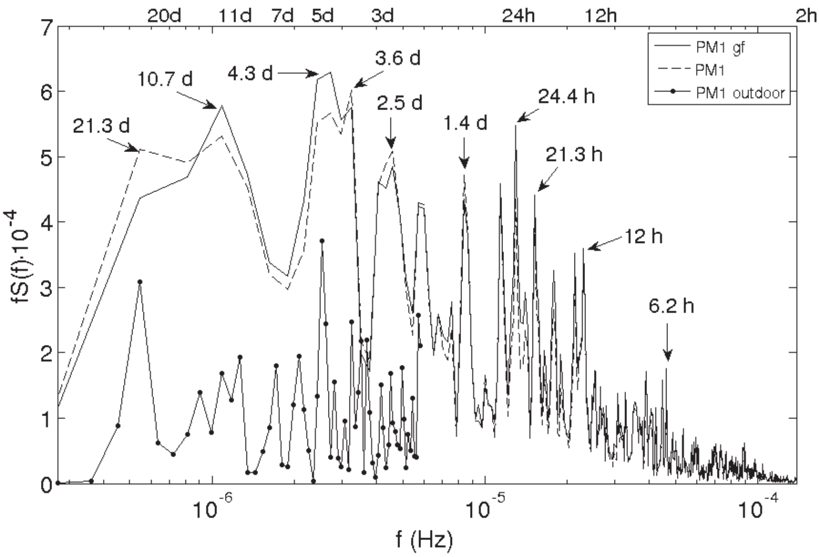
We note that for precipitation (Fig. 5c) and horizontal wind velocity (Fig. 5f) signs of correlation coefficients are changed from negative values obtained for shorter time lags to positive values corresponding to longer time lags. While correlation coefficients between PM_1 and precipitation for shorter time lags are negative as expected (precipitation washes out airborne PM), about 4.5–5 days after the precipitation event coefficients attain maximum positive values. We speculate that this positive correlation might be due to more intense evaporation after precipitation events (due to previous precipitation, there is more liquid water available for evaporation in the soil and/or at impervious urban surfaces), and consequent increase of relative humidity (which is positively correlated with PM_1 levels). However, this hypothesis deserves further investigation.

Finally, we note that patterns of time-lagged correlation coefficients for temperature, relative humidity and global radiation (panels a, b and d) clearly reflect diurnal periodicity of these meteorological variables.

4.5. Spectral analysis

Figure 6 shows the results of spectral analysis. Since the spectral density function for outdoor PM_1 was calculated from daily means, the corresponding line in Fig. 6a shows only periods greater than or equal to 2 days. Not surprisingly, peaks corresponding to periods of about 1 day are present in both indoor PM_1 spectra and for all investigated meteorological variables except for air pressure. The period of 24.4 h is found for PM_1 at floors, while periods of 23.8 h are present for the temperature, horizontal and vertical (not shown) wind components and relative humidity. Diurnal periodicity in meteorological variables can be attributed to solar forcing and consequent variations of other variables governed by or substantially influenced by the solar radiation (air temperature and consequently, relative humidity; vertical wind due to daytime up-slope/night-time down-slope flows; horizontal wind components due to the daily cycle of up- and down-slope winds). Semidiurnal (12 h) or approximately semidiurnal (11.5 and 11.6 h) periods are found for all variables shown in Fig. 6. Two long-term periods are also noticeable. One of about 10–11 days is found for indoor PM_1 for both floors (10.7 days, Fig. 6a) and for outdoor PM1 (about 10 days, Fig. 6a) and for the outdoor temperature (10.6 days, Fig. 6b). Another long-term period of about 21.3 days is seen for the PM_1 on the first floor, outdoor PM_1 and for the air pressure (Figs. 6a and e). It is interesting that the period typical for cyclonal disturbances is present only in the pressure spectrum (5.3 days, Fig. 6e), although in PM_1 concentrations somewhat smaller peaks are found (4.3 days in both indoor spectra and 4.7 days in outdoor PM_1). Finally, we observe that spectral density functions for the outdoor temperature, pressure and relative humidity (Figs. 6b, e and f, respectively) have simple shapes. Spectra of the horizontal wind components are less straightforward (Figs. 6c and d), while the noisiest are patterns associated with PM_1 concentrations for both floors and outdoors.

a)



b)

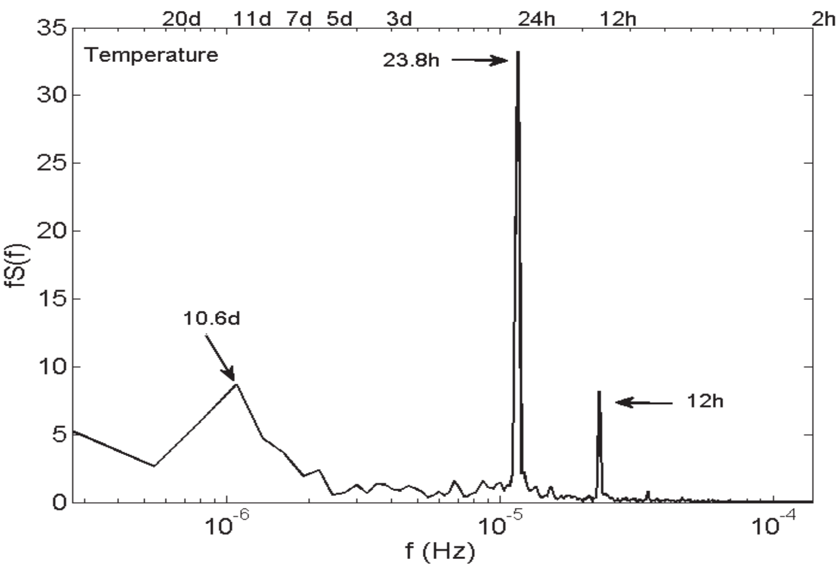
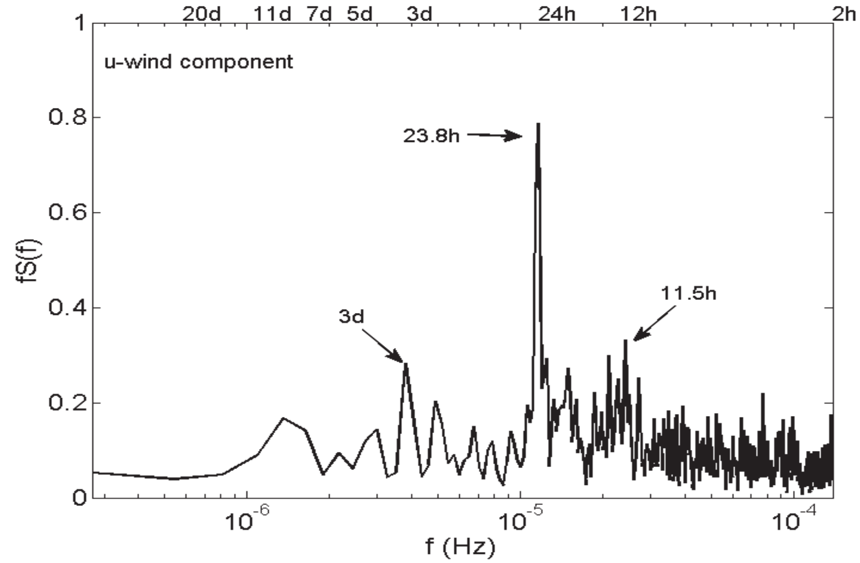


Figure 6. Spectral density functions for *a*) PM_1 concentrations for both floors and for the 1 km distant outdoor measuring site (indoor PM_1 spectra were calculated from hourly means, while outdoor spectrum was determined from daily means); *b*) temperature; *c*) eastward wind component; *d*) northward wind component; e) air pressure; and f) relative humidity. Spectral density functions are calculated from the data observed during 15 December 2012–26 April 2013 period.

c)



d)

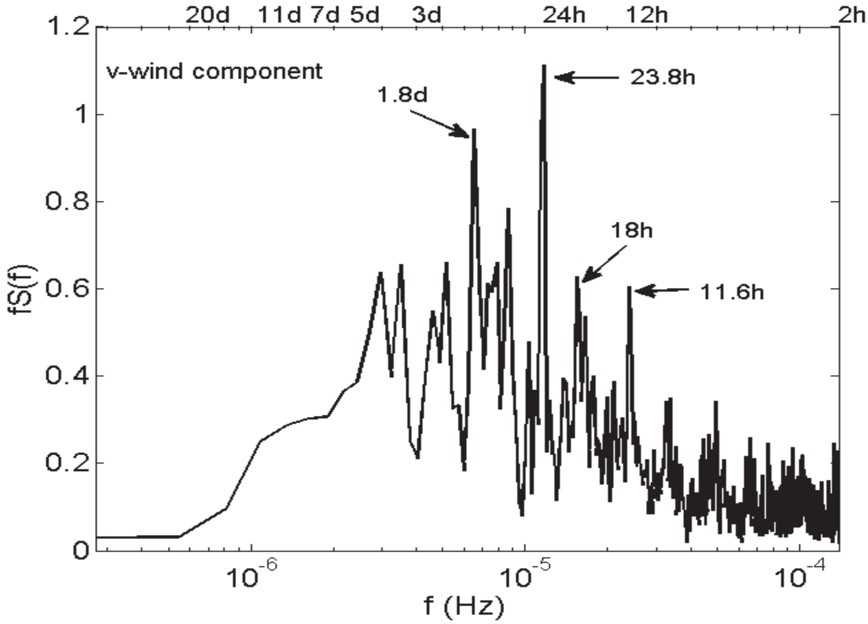
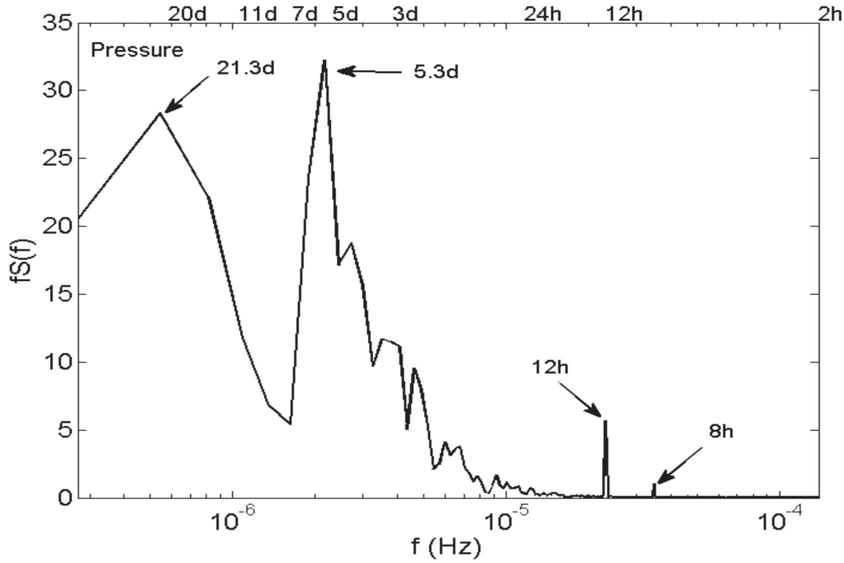


Figure 6. Continued.

e)



f)

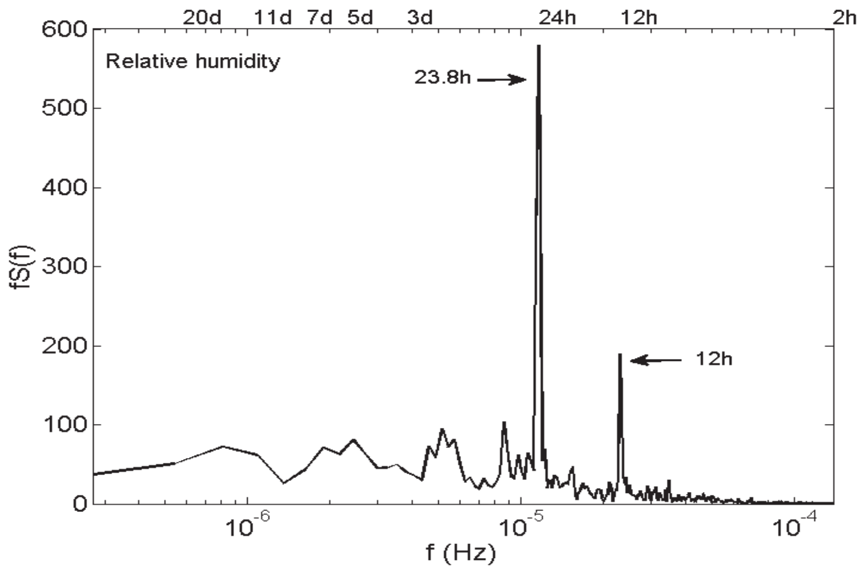


Figure 6. Continued.

5. Conclusions

Investigation of the influence of the wintertime outdoor 1-min mean atmospheric conditions on the concurrent indoor PM_1 mass concentrations observed at ground and first floor of University Campus building located in urban residential area of Zagreb for both floors showed a decrease in PM_1 with increase in concurrent outdoor temperature, precipitation amount, and horizontal wind velocity, while PM_1 increased with outdoor relative humidity. Additionally, for the ground floor PM_1 increased with the air pressure. For vertical wind velocity W, the PM_1 levels were the highest for $W \approx 0$, while they decreased with the strengthening of both upward and downward vertical motions. We note however, that lowering of indoor PM_1 concentrations with the strengthening of downward motions are most likely the result of down-slope winds (which bring the clean air from the nearby mountain toward the measuring site). Indoor PM_1 concentrations also exhibited prominent dependence on the outdoor wind directions. Accordingly, influence of the 8-9 km distant industrial zone could be detected in indoor concentrations. Results of present study associated with the influences of relative humidity, horizontal wind speed and direction, and the air pressure on the concurrent indoor wintertime PM_1 levels are similar to those obtained by the previous outdoor study of nearby location (Klaić et al., 2012).

The correlation between the outdoor meteorological conditions and concurrent indoor PM_1 levels was stronger for the ground in comparison with the first floor. This is due to the fact that the ground floor aerosol monitor was more exposed to intrusions of outdoor air due to persons' entering or leaving the building (and the outdoor PM_1 levels depend on meteorology). On the other hand, PM_1 concentrations at the first floor were dominated by indoor aerosol resuspension, since this floor was on average used by more persons. Ground floor concentrations were generally higher than first floor values. Therefore, we hypothesize that nearby outdoor levels were higher than indoor values, which contributed to pollution of the ground floor air due to the mixing with outdoor air. Our hypothesis is to some extent corroborated by comparison of indoor concentrations with daily mean outdoor values observed at about 1 km distant background measuring site. We note however, that the outdoor data were determined not only at different location, but also by different measurement method. Therefore, a further study of simultaneous indoor and nearby outdoor PM_1 levels, both determined by the same measurement method, is needed.

Investigation revealed a delayed response of the indoor PM_1 levels to outdoor meteorological conditions for all investigated meteorological variables. This response was the strongest for the air temperature and relative humidity, while it was almost negligible for global radiation. The response time varied from about 1.2 hours (for relative humidity and PM_1 at the ground floor) to 2.7 days (for global radiation and PM_1 at both floors). Generally, ground floor PM_1 concentrations responded quicker to the outdoor conditions than concentrations at the first floor, which might be associated with the fact that the ground floor is more exposed to intrusions of outdoor air than the first floor. Therefore, in further work it would be interesting to compare response times of indoor PM_1 concentrations with response times of outdoor PM_1 levels.

Although average concentrations for both floors exhibited weekly variation with the maximum/minimum on Monday/Saturday, expected 7-day periodicity due to human activity (e.g., Choi et al., 2008) was not corroborated by spectral analysis. (Interestingly on Sundays, when the building was generally empty, average concentrations were 0.001 $mg\,m^{-3}$ higher than on Saturdays at both floors). However, the influence of other periodic processes, with semidiurnal, diurnal and long-term (about 11 and 21 days) periods is found for both indoor PM_1 and meteorological fields, while periods of about 10 and 21 days were also present in outdoor PM_1. Periods of 12 h, 24 h and 21 day were also found in the study of urban traffic-related outdoor PM_{10} (Tchepel and Borrego, 2010). While Tchepel and Borrego attribute the 21-day period in PM_{10} fluctuations to the long-range transport of pollutants, we note that long-term disturbances observed in the present study in both atmospheric fields and indoor and outdoor PM_1 (10–11 and 21 days) might be associated with atmospheric planetary (Rossby) waves (e.g., Penzar et al., 1980; Pasarić et al., 2000; Šepić et al., 2012). Therefore, further investigation of possible role of Rossby waves in PM levels would be desirable.

Finally, in comparison with investigated meteorological variables, patterns of spectral density functions for PM_1 on both floors and outdoors were the most complex (that is, "major" peaks were less prominent). This suggests that PM_1 is governed by a large number of comparably important processes occurring at different spatio-temporal scales, which is in accordance with previous findings for outdoor PM_{10} (Hrust et al., 2009).

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SAŽETAK

Utjecaj vanjskih meteoroloških uvjeta na kratkotrajne koncentracije PM_1 u zatvorenom prostoru

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Ispitali smo vezu između jednominutnih srednjaka zimskih masenih koncentracija lebdećih čestica aerodinamičkog promjera < 1 μm (PM_1) izmjerenih u zatvorenom prostoru u urbanom okolišu i vanjskih atmosferskih uvjeta. Koncentracije lebdećih čestica mjerene su dvama laserskim fotometrima. Fotometri su se nalazili u hodnicima u prizemlju i na prvom katu zgrade Sveučilišta, a zrak je u instrumente ulazio na visini od 1.7 m nad tlom. Zgrada se nalazi u rezidencijalnom dijelu Zagreba. Zaposlenici i studenti su se tijekom eksperimenta bavili uobičajenim aktivnostima. Prizemni meteorološki podaci prikupljani na obližnjem vanjskom mjernom mjestu. Rezultati pokazuju ovisnost koncentracija PM_1 u zatvorenom prostoru o vanjskim meteorološkim uvjetima. Dok su vanjska temperatura zraka i relativna vlažnost povezane s najjačim odzivom koncentracija lebdećih čestica u zatvorenom prostoru, utjecaj globalnog zračenja gotovo je zanemariv. Vrijeme odziva varira od 1.2 h (za relativnu vlažnost) do 2.7 dana (za globalno zračenje). Nadalje, povišene prosječne koncentracije PM_1 pri jugoistočnom strujanju ukazuju na utjecaj 8–9 km udaljene industrijske zone. I u meteorološkim nizovima i u nizovima koncentracija lebdećih čestica prisutna je poludnevna i dnevna periodičnost te dugoperiodičnost (približno 10–11 dana te oko 21 dan). Dugoperiodičnost vremenskih nizova koncentracija PM_1 mogla bi biti povezana s Rossbyjevim valovima. Kako bi se potvrdila ta hipoteza, potrebna su daljnja istraživanja.

Ključne riječi: poprečna korelacija, DUSTTRAK monitor aerosola, 1-minutni srednjak, rezidencijalni, spektralna analiza, vremenski posmak, urbani

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