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Indoor wintertime PM mass concentrations and their relationship with outdoor meteorological conditions

A case study of a University Campus building in Zagreb, Croatia

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CONTENTS

1. INTRODUCTION

In the atmosphere a large number of pollutants exists that produce different effects on the environment and human health. These include particulate matter (PM), also known as suspended particulates, airborne particles or aerosols. The particulate matter is a complex and highly diverse mixture of solid particles, liquid particles and particles having a solid core surrounded by liquid, which are suspended in the atmosphere, such as dust, soot, salt, smoke or ash (Wilson et al., 2002). The primary particles come from a collection of very different emission sources both natural and anthropogenic, and the secondary particles are formed by chemical and photochemical processes in the atmosphere (Galindo et al., 2013). Some examples of emission sources are the combustion of fossil fuels in vehicles and industries, volcanoes, sea spray, or dust emission from soils. Therefore, particles are characterized by a high variety of morphological, physical, chemical and thermodynamic characteristics, as well as by variable spatial and temporal distributions (Wilson et al., 2002). For example, among particles there are found inorganic ions, metal components, elemental carbon or organic compounds, which are specially complex (the organic fraction which contains hundreds of different compounds) (Wilson et al. 2002).

Due to this diversity, particles have been classified according to different criteria such as their formation mechanisms, chemical composition, and size. After the use of different classifications in relation to the size of the particles, mainly the one in Whitby (1978), many recent studies have taken as reference the categories of PM10 (particles with an aerodynamic diameter < 10 μm), PM2.5 (particles with an aerodynamic diameter < 2.5 μm) and PM1 (particles with an aerodynamic diameter < 1 μm) (Zhang et al., 2006; Kampa and Castanas, 2008; Yin and Harrison, 2008; Polichetti et al., 2009; Titos et al. , 2014). It is very common to find in the literature the label of coarse particles associated with those particles with diameters between 10 μm and 2.5 μm, and that one of ultrafine particles with diameters < 0.1 μm (Zhang et al., 2006; Polichetti et al., 2009; Titos et al., 2014). The denomination of fine particles is in some articles associated with particles of diameter < 2.5 μm (Zhang et al., 2006; Yin and Harrison, 2008; Polichetti et al., 2009), while in others is associated with diameters < 1 μm (Braniš et al., 2005; Kampa and Castanas, 2008; Titos et al., 2014). As indicated in Pérez et al. (2008), according to the US-EPA (2004): "Over the years, the terms fine and coarse, as applied to particles, have lost the precise meaning given in Whitby's (1978) definition. In any given article, therefore, the meaning of fine and coarse, unless defined, must be inferred from the author's usage... Fine particles and PM2.5 are not equivalent terms ".

In urban atmospheres finer particles originate mainly from industrial activities and combustion in vehicles, where the influence of traffic increases with decreasing particle size. The coarser fraction originates mainly from mechanical processes related to human activities (i.e., dust re-suspended by vehicle traffic), the wind-blown dust, sea spray, plants, or volcanoes. Other identified particle sources in urban environment are fossil fuel/wood/biomass burning central heating, agriculture (through the use of fertilizers and pesticides), emissions from mines near the cities, and regional pollution due to large-scale transport of particles (Kertész et al., 2010; Titos et al., 2014).

Depending on their size and chemical composition, suspended particles have an influence on the global climate through effects on the overall balance of radiative forcing. These include heating or cooling of the atmosphere, through direct mechanism of solar radiation scattering or absorption and indirect mechanisms such as acting as cloud condensation nuclei (Wigley, 1989; Charlson et al., 1992; Buseck and Adachi, 2008). Particles have also an effect on human health, in which the particle size determines the point in the respiratory tract that particles can reach. PM10 particles are deposited mainly in the upper respiratory tract, and the fine and ultrafine particles can reach the pulmonary alveoli (Kampa and Castanas, 2008). Indeed, PM10 and PM2.5 are also known as the thoracic and alveolar fractions, respectively, and PM4 (particles with aerodynamic diameter < 4 μm) corresponds to the respirable fraction (Pérez et al., 2008). A large number of studies have linked exposure to particulate matter with adverse effects on human health, which include respiratory and circulatory problems, allergies, toxicity, and even mortality (Nielsen et al., 2007; Kampas and Castanas, 2008; Polichetti et al., 2009; Tobías et al., 2011; Kelly and Fussell, 2012; Perez et al., 2012; Volkoff et al., 2013; Pascal et al., 2014).

Since in the big cities people spend more than 85% of their time indoors (Diapouli et al., 2008), for example, at home, at work, at school, or in the car, in recent times have proliferated studies on air quality in enclosed spaces, with particular attention to the suspended particles (Thatcher and Layton, 1995; Kildesø et al., 1998; Long et al., 2000; Luoma and Batterman, 2001; Braniš et al., 2005; Diapouli et al., 2008; Toftum, 2010; Rovelli et al., 2014). Airborne particles have demonstrated to be important components of indoor environmental air (Nazaroff, 2004), although the understanding of processes involved is still not complete. Particles generated in the outside environment can enter a building through ventilation and infiltration, and with an absence of indoor sources, the outdoor concentration is what determines the indoor concentration (Toftum, 2010). Smaller particles (below approximately PM2.5) penetrate easier through cracks in the building than larger ones (Braniš et al. 2005). Indoor particle sources are cooking, cleaning (dusting, sweeping, vacuuming, use of cleaning chemicals), smoking, indoor heating by burning stoves, other combustion-related activities (such as using candles), and chemical processes in the indoor air (Thatcher and Layton, 1995; Long et al., 2000; Toftum, 2010). The movements and activities of the occupants of enclosed spaces, which generate re-suspension of the particles previously deposited on surfaces, are also indoor particle sources (Thatcher and Layton, 1995; Long et al., 2000; Luoma and Batterman, 2001; Nazaroff, 2004; Braniš et al., 2005; Toftum, 2010; Ollier, 2013; Klaić et al., 2014; Rovelli et al., 2014). Re-suspension is primarily related to the coarse fraction. Some studies show no significant relationship between re-suspension and the fine fraction (Thatcher and Layton, 1995; Braniš et al., 2005), while other suggest the opposite (Long et al., 2000; Luoma and Batterman, 2001; Rovelli et al., 2014). In any case, it is found that the rate of re-suspension increases with particle size increasing (Thatcher and Layton, 1995; Rovelli et al., 2014). Emissions due to occupants in offices and houses include fibers, dust, skin cells, bacteria, fungi, and emissions produced by materials and machinery used indoors (Luoma and Batterman, 2001). In indoor environments without significant and specific emission source, the activities of occupants may represent the main source of dust (Braniš et al. 2005). Emissions from indoor sources can significantly alter the size distribution and composition of particles indoors (Long et al., 2000; Rovelli et al., 2014). As indicated in Long et al. (2000), due to

contributions from indoor sources, indoor exposure can have significant impacts on personal short-term exposures to airborne particles. The impact of intermittent events indoors on personal exposures are expected lower if spaces are routinely ventilated by opening windows and doors (Long et al., 2000).

Several studies show higher concentrations of suspended particles indoors in comparision to outside concentrations (Janssen et al., 1997; Wheeler et al., 2000; Mohammadyan and Shabankhani, 2013; Ollier, 2013). Lower indoor compared to outdoor concentration are also found (Braniš et al., 2005; Diapouli et al., 2008; Rovelli et al., 2014). Multiple studies show a trend to higher particulate matter concentrations both outdoors (Viana et al., 2006; Viana et al., 2007; Vecchi et al., 2008; Massey et al., 2012; Kim et al., 2013; Li et al., 2014) and indoors (Wheeler et al., 2000; Fromme et al., 2007; Massey et al., 2012) during the winter compared to summer months. This is due to an increased emission during the winter because of increased human activity and the use of heating during cold months, reduced ventilation of enclosed spaces, and a combination of certain weather conditions. These weather conditions include higher relative humidity, high air pressure fields (promoting a stable atmosphere and longer residence time of the particles in the atmosphere), lower temperatures (affecting the height of the atmospheric boundary layer and dispersion of pollutants), or relatively low surface wind speeds (promoting lower dispersion of pollutants) (Viana et al., 2006; Viana et al., 2007; Silcox et al., 2012; Massey et al., 2012; Grange et al., 2013; Klaić, 2012; Galindo et al., 2013; Li et al., 2014).

Therefore, the influence of the indoor particulate matter on human health is particularly important during the winter season, when heating is used. In the present study the relationship between particulate concentrations inside a university building in the city of Zagreb and the meteorological conditions outside will be analysed for the period from 01.11.2013 to 28.02.2014. Furthermore, the temporal and spatial variability of these particle concentrations inside the building will be studied. Different PM size fractions (PM1, PM2.5, PM4, and PM10) and Total Suspended Particles (TSP) will be measured. Nevertheless, the analysis will focus on the PM1 fraction because of their smaller diameter and, therefore, their deeper penetration into the respiratory tract.

2. MEASURING SITE AND MEASUREMENTS

Both, indoor concentrations of PM and meteorological variables were measured at the Andrija Mohorovičić Geophysical Institute, Department of Geophysics, Faculty of Science, University of Zagreb. It is located in a residential area in the north of the city of Zagreb, around 1.5 km north-northeast from the city centre and 8-9 km northwest from the industrial zone. It is a relatively hilly area, located on the southeastern slope of Mount Medvednica, at an altitude of 180 m above sea level. This mountain rises approximately 1 km above sea level and extends from west-southwest to east-northeast. To the north and northwest of the building there are several university campus buildings. In the area there are also family houses. Among the buildings there are grassy areas and unevenly distributed trees of around 10 m. The Institute is located near two roads: Horvatovac, at a distance of 50 m from the building, with east-west orientation, a road with a very low traffic, and Bijenička, at a distance of approximately 100 m from the building, with a north-south orientation, which has a weak to moderate traffic (Klaić et al., 2012). More details on the location and surroundings of the Institute can be found in Klaić (2012), Klaić et al. (2012) and Ollier (2013). Measurements were recorded during a total of 120 days, for the period between 01.11.2013 and 28.02.2014.

2.1 Indoor PM mass concentration measurements

PM concentrations are measured inside the building by using two portable monitors/laser photometers (Aerosol Monitors DUSTTRAK^{TM} models 8520 and 8533 - TSI Inc., Shoreview, MN, USA). The DUSTTRAK^{TM} monitor model 8520, located on the ground floor of the building, is set to take measurements of PM1 concentration ([PM1]GF, in mg m^{-3}). The DUSTTRAK^{TM} monitor model 8533, located on the first floor, provides simultaneous measurements of PM1, PM2.5, PM4 (or respirable), PM10 and Total Suspended Particles concentrations ([PM1] FF, [PM2.5] FF, [PM4]FF, [PM10]FF and [TSP]FF, respectively, in mg m^{-3}). Both monitors are configured to provide the 1-min mean values from the records for every second.

The monitors are placed in the hallways next to the stairs that connect both floors, as shown in Ollier (2013). They are suspended in each case at a height of 1.67m above the floor, so instrument inlets are at 1.7 m (the average human breathing height). The stairs layout characterizes the space as an open corridor between the base of the ground floor and the ceiling of the first floor. On the north wall of both floors several windows are located, 6 on the ground floor and 7 on the first, with a radiator under each one. The building entrance is also located on the north wall of the building, on the ground floor, and has two double glass doors.

During weekdays/workdays (from Monday to Friday) average number of people using the building is around 65: 35 employees and 30 students approximately. Arrival, stay and exit times of the building are variable, although busiest moments are from 07:00 to 09:00 LST (Local Standard Time), 12:00 to 14:00 LST and 15:00 to 17:00 LST. Those moments coincide with the arrival of the majority of the staff and students, breaks for lunch, and leaving after the class hours, respectively. From 17:00 LST around 10 people remain in the building, leaving it at

different times until 21:00 LST (one member of staff stays mandatorily in the building up to 21:00 LST) when the building is left empty. The flow of students through the building varies depending on lectures, tutorials, exams and other uses of the facilities, although all students generally have left the building by 16:30.

The building is cleaned once a day during weekdays: ground floor in the morning and first floor during the afternoon. Smoking is forbidden inside the building; therefore, smokers go out to smoke at the entrance of the building at different times throughout the day. There are no cooking facilities either on the ground floor or on the first floor.

Although the identification of individual PM sources inside the building is not part of the scope of this study, it is noteworthy that re-suspension due to the movement of people would probably be the most important one. In this particular case also activities such as printing and copying documents and the use of certain cosmetics and cleaning products could generate effects on PM concentration increase (Klaić et al., 2014).

During the weekend little activity takes place inside the building. On Saturdays one of the staff members is mandatorily in the building from 07:00 to 14:00 LST. Nobody enters the building systematically on Sundays. In any case, staff members may occasionally come at any time of the weekend.

The central heating was put into operation on 11.07.2013. Therefore, the analysis period corresponds almost entirely to the period when the heating inside the building is used, so the windows near aerosol monitors are closed during the study period. The windows in the classrooms and adjacent offices to the hallways are occasionally opened. The heating system comprises various radiators located under the windows in the building. On weekdays the heating is set to 21°C between 06:00 and 21:00 LST, and to 17°C between 21:00 and 06:00 LST. During the weekend the heating is set to 17°C. During the holidays the heating is set to 19°C.

2.2 Meteorological variables measurements

The meteorological variables measured and analysed in this study are: air temperature (T, in °C), air pressure (p, in hPa), relative humidity (RH, in %), global radiation (G, in J cm^{-2}), diffuse radiation (D, in J cm^{-2}), horizontal wind speed (V_h, in m s^{-1}), horizontal wind direction (D_h, in degrees), vertical wind speed (V_w, in cm s^{-1}) and UV radiation (UV-S and UV-Y, in Med h^{-1}, corresponding to the measurements obtained for the same variable - UV radiation - using two different measuring instruments). These variables are measured outside the Institute, except p (which is measured inside the building), using automatic meteorological station META 2000 (AMES, Brezovica, Slovenia). Data provided by the meteorological station are 1-min means obtained from the measurements for each second. Sensors for V_h/D_h, V_w, G/D and UV-S/UV-Y are sited on a roof terrace on the west side of the building. They are at 4.5 m, 4.3 m, 1.8 m, and 0.7 m, respectively, above the roof (that is, 17.3 m, 17.1 m, 14.6 m, and 13.5 m above the ground). The T and RH sensors are located at 2 m above the ground inside a weather shelter, in a grassy area near the entrance of the building. The p sensor is placed inside the building at a height of 7.5 m above the ground, and 187.55 m above sea level.

2.3 Data and methodology

As has been indicated previously, simultaneous measurements of indoor PM concentrations and meteorological variables are taken during a total of 120 days, for the period from 01.11.2013 to 28.02.2014. The data set recorded for the study period consists of 162783 [PM1]GF data and 167903 data for each [PM1]FF, [PM2.5]FF [PM4]FF, [PM10]FF, and [TSP]FF. For each meteorological variable 171557 data were taken, except for the air temperature, for which one least data was obtained, namely 171556 data. The data set was not obtained for all the minutes present in the study period due to several reasons. On the one hand, PM concentrations on both floors stop being registered during data download to computer, once the data storage buffer is full and has no more space to store more records. On the other hand, for the meteorological records, every few months the backup is downloaded, causing a stop in recording data up to one hour. In addition, the measuring is stopped for the maintenance and in case of a malfunction or repair of instruments. Since the moments without record on the ground floor, first floor and for the meteorological variables do not necessarily coincide, and since most of these absences of data are found for relatively large time intervals, filling in the blanks by the linear interpolation method is not performed. Nevertheless the blanks are tracked down for each variable, and for relating variables they are adjusted one to each other by removing any non-concurrent records in both of them.

Vertical wind is analysed in detail. The convection or upward wind (positive values), subsidence or downward wind (negative values) and vertical wind speed in absolute values ($V_{w\text{-Absolute Values}}$) are analysed separately from the original vertical wind speed variable (V_w, which includes the sign/direction of the wind), where 139233 minutes associated with convection, 31572 associated with subsidence and 752 minutes without vertical wind occurred.

To analyse the distribution of PM concentration according to the level of activity inside the building, 5 categories of days are established: 1) Weekdays (Monday-Friday), 2) Saturdays, 3) Sundays, 4) Holidays and 5) Sundays and Holidays. The local holidays comprise 01.11.2013 (All Saints' Day) and the period from 25.12.2014 to 06.12.2014 (Christmas Holidays). The reason why data are separated into categories 3), 4), and 5) is, on the one hand, to make the results comparable to those presented in Ollier (2013), and, on the other hand, to follow the recommendation given in that report for analysing separately Sundays and Holidays.

For the analysis of the temporal variability of indoor PM concentrations, outdoor PM concentration data measured at a different location are taken as representation of the urban background PM concentration in Zagreb. Specifically, the daily mean outdoor PM1 concentrations values recorded in the vicinity of the Institute of Medical Research and Occupational Health in Zagreb, for the period 01.11.2013 to 31.12.2013, are taken. The Institute of Medical Research and Occupational Health is sited approximately 1 km northwest from the Andrija Mohorovičić Geophysical Institute, and the residential environments in both cases have similar characteristics. As indicated in Ollier (2013), the measuring station is located about 50 m from Ksaverska Road, which has moderate traffic levels. The surroundings are mainly asphalt and grass, and the nearest building is about 30 m away from the site and has a

height of around 5 m. Measurements are performed with an LVS3 sampler (Sven Leckel, Berlin, Germany) with a Whatman TM QMA quartz filter (GE Healthcare Bio-Sciences, Pittsburg, USA). The sampler inlet is at 1.8 m above the ground, and the filters are weighed twice before and twice after sampling, every 48 hours, at a temperature of 20°C.

3. RESULTS AND DISCUSSION

3.1 Indoor PM1 mass concentrations

As has been mentioned above, the PM concentration data provided by the measurement monitors corresponds to the 1-minute mean, from records made for each second. Table 1 shows the descriptive statistics for the different fractions of PM. The mean [PM1] inside the building is between 0.046 to 0.060 mg m^{-3}, corresponding to the ground floor ([PM1]GF) and first floor ([PM1]FF) means, respectively. If these concentrations are compared with those in Klaić et al. (2014) and Ollier (2013), the mean [PM1]GF is practically the same as in the first case (0.047 mg m^{-3}), being 26% lower respect to the second case (0.058 mg m^{-3}). The average [PM1]FF here presented is 17% higher compared to Klaić et al. (2014) (0.052 mg m^{-3}) and 5% lower compared to Ollier (2013) (0.063 mg m^{-3}).

Given the positive or right skewed distribution of the data (Mean > Median > Mode), the median seems to be the most descriptive of the three statistics for this data set. The median for [PM1]GF is 0.031 mg m^{-3}, being essentially the same in Klaić et al. (2014) (0.033 mg m^{-3}) and around 39% lower than in Ollier (2013) (0.043 mg m^{-3}). The median for [PM1]FF stands at 0.042 mg m^{-3}, being 17% higher than in Klaić et al. (2014) (0.036 mg m^{-3}) and about 19% lower than in Ollier (2013) (0.050 mg m^{-3}). In these studies the location, type and frequency of activity in the building, as well as measuring instruments and its configuration, are the same as in the present study. The variations seen would be mainly due to seasonal/meteorological differences between the studied periods of the year and the size of the data set analysed in each study. In Klaić et al. (2014) the months analysed (12.11.2012-26.04.2013) comprise almost all the months analysed in this study, extending them; in Ollier (2013), the analysed year period (18.11.2012-25.01.2013) is a fragment of the months included in this study, and a specific fragment of the data set in Klaić et al. (2014).

Lower mean values of indoor PM concentration than those here exposed were found in classrooms in Italy in Rovelli et al. (2014) (0.019 mg m^{-3} for PM1, 0.033 mg m^{-3} for PM2.5), and in Iran in Mohammadyan and Shabankhani (2013) (0.018 mg m^{-3} for PM1, 0.047 mg m^{-3} for PM2.5) and Hassanvand et al. (2014) (0.011 mg m^{-3} for PM1, 0.018 mg m^{-3} for PM2.5, 0.047 mg m^{-3} for PM10). However a mean value of 0.400 mg m^{-3} for PM10 was found in Mohammadyan and Shabankhani (2013). In Kildesø et al. (1998), mean values between 0.054-0.133 mg m^{-3} for indoor PM4, and 0.064-0.267 mg m^{-3} for indoor TSP, are measured in an administration building, a school and a kindergarten in Denmark. In Tasić et al. (2011), values between 0.027-0.051 mg m^{-3} for PM2.5, and 0.033-0.061 mg m^{-3} for PM10, were found in an institute, kindergarten and a hospital in Serbia. The difference between the mean PM concentration values in the different studies can be due to several factors, mainly to the different locations with different meteorological and pollution conditions, different study periods, different experiment designs and measuring instruments, and different uses of the study buildings. As pointed in Klaić et al. (2014), some uncertainty is founded in the literature in relation to DUSTTRAK™ aerosol photometer measurements, and maybe that is partly the reason for higher values in the present study than in the aforementioned studies. The application of

gravimetric method in order to compare with the results from the aerosol monitors is not available for this study, so no correction factor is applied. In addition, we note that the focus here is on the variations of indoor PM concentrations and their relation with meteorological conditions outside, rather than on the particular PM concentration levels. Anyway, it is interesting to comment that the measurements in Rovelli et al. (2014), indicated above, were performed with an Aerosol Monitor DUSTTRAK^{TM} 8530, which belongs to the family of models used in the present study.

As a reference of PM1 concentrations obtained outside the building during the winter, the mean of 0.034 mg m^{-3} and the median of 0.028 mg m^{-3} obtained in Ollier (2013) show that outdoor concentration is lower than indoor one. If we take the means obtained in Klaić (2012) (0.028 mg m^{-3}) and Klaić et al. (2012) (0.027 mg m^{-3}) for periods ranging warmer months (from 17.03.2010 to 12.05.2010 and the same period plus from 14.09.2011 to 26.09.2011, respectively), we find a lower outdoor PM1 concentration for warm months compared to winter, as widely seen in the literature (Viana et al., 2007; Vecchi et al., 2008; Massey et al., 2012; Kim et al., 2013; Li et al., 2014).

Table 1: Descriptive statistics for indoor concentrations of the different PM size fractions and TSP (mg m^{-3}) measured on the ground floor (GF) and on the first floor (FF), for the study period (01.11.2013-28.02.2014).

	mean	max	min	median	mode	st.dev	variance
[PM1]GF	0.0461	0.398	0.000	0.031	0.006	0.045	0.0020
[PM1]FF	0.0603	0.452	0.001	0.042	0.019	0.053	0.0028
[PM2.5]FF	0.0604	0.452	0.001	0.042	0.019	0.053	0.0028
[PM 4]FF	0.0607	0.453	0.001	0.042	0.019	0.054	0.0029
[PM 10]FF	0.0618	0.458	0.001	0.043	0.020	0.054	0.0029
[TSP]FF	0.0628	0.517	0.001	0.044	0.020	0.055	0.0030

The mean and median [PM1] GF, as its other statistics, are lower than those for [PM1] FF, being the average variation between both statistics for each floor of approximately 33%. This variation is found to be 3.3 times higher than in Klaić et al. (2014), and nearly half of the variation in Ollier (2013). The statistics of the other fractions of PM measured on the first floor are highly similar to each other and to [PM1] FF, coinciding exactly in many cases. The statistics calculated for [TSP] are also similar to them, although the maximum value of [TSP] exceeds up to 0.060 mg m^{-3} the maximum values for the different fractions. Similarly, the maximum value of [TSP] in Ollier (2013) exceeds 0.100 mg m^{-3} the rest.

3.2 Meteorological conditions

In table 2 the statistics that describe the different meteorological variables are shown, and Figure 1 displays the diurnal variation of these same variables. As for PM concentration data, the data obtained for the different meteorological variables correspond to the 1-minute mean, provided by the meteorological station META 2000 from every second records. The average air

temperature for the study period (5.34°C) is above the values shown in both Klaić et al. (2014) (4.93°C) and Ollier (2013) (3.81°C). The mode value is much higher (7.20°C) compared to those obtained in the above studies (about 0.20°C in both cases). Considering that mode is a very unstable statistic and those data present a negative or left skewed distribution (Mode> Median > Mean), the median turns out to be the most descriptive statistic for this data set. The median air temperature (6.10°C) is also higher than the ones found in Klaić et al. (2014) (3.90°C) and Ollier (2013) (3.00°C).

Considering the air pressure, the mean, the median and the mode show very similar values, so without presenting a symmetrical distribution, data are very close to it. Being the mean value between the median and the mode, it is the most descriptive statistic for the results here presented. However, noting the symmetry of distribution of the air pressure data in the aforementioned studies, the median is the most representative of them. So, the medians are compared, observing that the one obtained here (995.5 hPa) exceeds the one in Klaić (2014) (992.8 hPa) and Ollier (2013) (994.6 hPa). In the case of the relative humidity, and for the same reason, medians are taken to do comparisons, seeing that the value obtained here (90.00%) is higher than in Klaić et al. (2014) (87.00%) but lower than in Ollier (2013) (94.00%). The average global radiation (39.72 J cm^{-2}) is higher than those in the aforementioned studies (35.97 J cm^{-2} and 30.47 J cm^{-2}, respectively). The average diffuse radiation (21.77 J cm^{-2}) is lower than that one for the global radiation, as expected. The averages of UV-S (0.04 Med h^{-1}) and UV-Y (0.05 Med h^{-1}) are within the same order of magnitude, as expected since they represent one single variable (UV radiation) measured by two different instruments.

The median of vertical wind speed (6.20 cm s^{-1}) is also higher than those presented in the aforementioned studies (5.60 cm s^{-1} and 3.90 cm s^{-1}, respectively), as well as the median of horizontal wind speed (0.90 m s^{-1}), compared to 0.60 m s^{-1} and 0.50 m s^{-1}, respectively.

Table 2: Descriptive statistics for meteorological variables, for the study period (01.11.2013-28.02.2014). T (temperature), RH (Relative Humidity), p (air pressure), G (global radiation), D (diffuse radiation), UV-S and UV-Y (UV radiation), V_h (horizontal wind speed), V_w (all vertical wind speed data with direction), $V_{w\ Absolute\ values}$ (all vertical wind speed in absolute values, this means without direction), $V_{w\ Convection}$ (vertical wind speed, only convection data), and $V_{w\ Subsidence}$ (vertical wind speed, only subsidence data).

	mean	max	min	median	mode	st.dev	variance
T (°C)	5.34	20.70	-6.30	6.10	7.20	4.52	20.44
RH (%)	85.88	100.00	34.00	90.00	100.00	13.07	170.93
p (hPa)	995.75	1017.40	973.90	995.50	998.00	9.02	81.36
G (J cm^{-2})	39.72	778.00	0.00	0.00	0.00	97.19	9446.30
D (J cm^{-2})	21.77	502.00	0.00	0.00	0.00	46.59	2170.60
UV - S (Med h^{-1})	0.04	0.88	0.00	0.00	0.00	0.10	0.01
UV - Y (Med h^{-1})	0.05	0.99	0.00	0.00	0.00	0.11	0.01
V_h(m s^{-1})	1.29	29.70	0.00	0.90	0.10	1.66	2.76
V_w(cm s^{-1})	11.63	166.30	-101.00	6.20	0.60	18.89	356.69
$V_{w\ Absolute\ values}$ (cm s^{-1})	14.06	166.30	0.00	8.10	0.60	17.15	294.27
$V_{w\ Convection}$ (cm s^{-1})	15.83	166.30	0.10	9.90	0.60	18.26	333.29
$V_{w\ Subsidence}$ (cm s^{-1})	-6.60	-101.00	-0.10	-4.10	-0.10	7.42	55.08

In regard to vertical wind speed in absolute values, table 2 shows that its median (8.10 cm s^{-1}) is greater than the one obtained considering the sign of vertical wind. This is due to neglecting the negative sign of subsidence data when statistics are performed. Median of convection data (9.90 cm s^{-1}) suggests that when the vertical wind rises it generally does with more speed than when it descends during the subsidence (4.10 cm s^{-1}, median in absolute values). From vertical wind speed total available data, 139233 data convection, 31572 subsidence data and 752 minutes with vertical wind speed equal to zero were obtained. Convection occurs in 81.2% of cases, subsidence in 18.4%, and minutes without vertical wind occurred in 0.4% of the cases. Thus, convection would be approximately 4.4 times more frequent than subsidence, and it generally would occur with 2.4 times more intensity.

Related to Ollier (2013) and Klaić et al. (2012), variables that are common to both and to the present study (T, p, R, G, V_w and V_h) generally have a similar diurnal pattern but with different values. Note that in Klaić et al. (2012) the study period comprises from 17.03.2010 to 12.05.2010 and from 14.09.2011 to 26.09.2011 intervals (both in the warm season). Temperature graph (fig.1.a) shows that, during the night, temperature drops gradually to its minimum (3.90°C) around 08:00 LST from which it begins to climb more markedly. Temperature reaches its maximum (7.34°C) at 15:00 LST and descends after that in a smoother way during the afternoon and the evening. The diurnal pattern of the average relative humidity (fig.1.b) shows how during the night it is increasing until it reaches its maximum (91.13%) at 08:00 LST. From this moment it decreases rapidly during the morning, reaching its minimum (77.83%) at 15:00 LST. Finally, it increases from this point during the afternoon and evening until the next morning (in a more pronounced way until 19:00 LST more or less, and smoother from this time).

Considering that the hours when major changes occur for both temperature and relative humidity are approximately the same, and also considering that the patterns described by their diurnal distributions are very similar but in opposite directions, an inverse relationship is found between the two variables, as expected. Table 3 shows the Pearson´s correlation coefficients between the different meteorological variables and respect to [PM1]GF and [PM]FF. The proposed classification in Salkind (1999) is taken as reference, where the Pearson´s correlation coefficients are classified as follows: 0-0.2 very weak, 0.2-0.4 weak, 0.4-0.6 moderate, 0.6-0.8 strong and 0.8-1 very strong. Taking this into account, most of the calculated coefficients would have a weak or very weak character (table 3). It must not be forgotten, however, that even a weak correlation may be statistically significant if the sample is large enough. The vast majority of Pearson´s coefficients show a correlation between the variables that is statistically significant ($p < 0.05$), being the most frequent p-value zero. That would indicate that a linear relationship exists between these variables, although it is mostly a weak relation. The exceptions are the coefficients found for the relationships between the horizontal wind direction and air pressure, and between horizontal wind speed and global radiation, diffuse radiation, and UV-Y, which do not exhibit statistical significance. The last case, the relation between horizontal wind speed and UV-Y, is remarkable, since a p-value = 0 is obtained for the relationship between horizontal wind speed and UV-S, although the

correlation coefficient value is very small (0.018). At this point it should be remembered that UV-S and UV-Y represent the measurement of UV radiation by two different instruments.

The highest correlation coefficient shown for temperature is the one which relates it to the relative humidity, and vice-versa. There are also closer relations between relative humidity and the variables related to solar radiation, and also with the convection, than between relative humidity and the other variables. A significant relationship between temperature and air pressure is also found, and much lower with the variables of solar radiation (though the temperature increases in the presence of solar radiation, it does not decrease abruptly after the sunset).

The mean diurnal air pressure variation is more uniform (fig.1.c). A slight decrease is noted from midnight until 05:00-06:00 LST, moment from which the air pressure begins to rise softly to its maximum (996.4 hPa) at 11:00 LST. After that, it descends to its minimum (995.2 hPa) around 15:00 LST, remaining more or less stable until 17:00 LST. Later air pressure increases very slightly up to midnight. Global radiation and diffuse radiation graphical representations (fig.1.d and fig.1.e) show, as expected, the complete absence of radiation for the night time hours. In both cases it is noted that on average sunrise occurs in the study period around 07:00 LST, and the sunset around 18:00 LST (though around 18:30 LST there is still seen some diffuse radiation). The peaks of maximum radiation are 182.90 J cm^{-2} and 102.00 J cm^{-2}, respectively, and occur about 12:00 LST. UV radiation graphics (fig.1.f and fig.1.g) show a distribution very similar to each other (and respect to global and diffuse radiation ones), but with lower values in the case of UV-S. The maximum is 0.22 Med h^{-1} for UV-S and 0.26 Med h^{-1} for UV-Y, coinciding with the noon, as in the case of global and diffuse radiation. Likewise concurrence with the moments of sunrise and sunset is observed. As seen in table 3 and as expected, all variables related to solar radiation have high mutual coefficients of correlation.

In the case of the horizontal wind speed (fig.1.h), diurnal average is higher than in Ollier (2013), and the mean diurnal distribution, although follows a similar pattern, is smoother. After horizontal wind speed stayed roughly constant overnight, 3 peaks of smooth increase are seen: at 05:00 LST (1.28 m s^{-1}), at 12:00 LST (1.46 m s^{-1}) and at 20:00 LST (1.36 m s^{-1}), all of them followed by a slight decrease. In Ollier (2013), these peaks occur most markedly but with lower values, and are located at around 04:30 LST (0.83 m s^{-1} approx.), 14:30 LST (1.35 m s^{-1} approx.) and 20:15 LST (1.00 m s^{-1} approx.), respectively. The minimum in the present study is 1.20 m s^{-1}, reached two times during the day, at 07:00 LST and 17:00 LST, while in Ollier (2013) the minimum is found around 0.65 m s^{-1}, at 02:00 LST. For the horizontal wind speed, the highest correlation coefficients obtained are those that relate it to the different variables of the vertical wind speed.

Table 3: Pearson´s correlation coefficients between indoor PM1 concentration (mg m^{-3}) on the ground floor (GF) and on the first floor (FF) and meteorological variables. For the most of correlations p-value=0; for the cases in which the p-value is non-zero, the corresponding value is shown in parenthesis. T (temperature), RH (Relative Humidity), p (air pressure), G (global radiation), D (diffuse radiation), UV-S and UV-Y (UV radiation), V_h (horizontal wind speed), D_h (horizontal wind direction), V_w (all vertical wind speed data with direction), $V_{w\ Absolute\ values}$ (all vertical wind direction in absolute values, this means without direction), $V_{w\ Convection}$ (vertical wind speed, only convection data) and $V_{w\ Subsidence}$ (vertical wind speed, only subsidence data).

	[PM1] GF/	[PM1] FF/	T (°C)	RH (%)	p (hPa)	G (J cm^{-2})	D (J cm^{-2})	V_w(cm s^{-1})	$V_{w\ Absolute\ values}$ (cm s^{-1})	$V_{w\ Convection}$ (cm s^{-1})	$V_{w\ Subsidence}$ (cm s^{-1})	V_h(m s^{-1})	UV - S (Med h^{-1})	UV - Y (Med h^{-1})	D_h
/[PM1]GF	1	0.991	-0.455	0.254	0.265	0.050	0.023	-0.079	-0.116	-0.135	-0.102	-0.135	0.024	0.016	-0.090
/[PM1] FF		1	-0.456	0.249	0.274	0.059	0.035	-0.067	-0.104	-0.123	-0.098	-0.132	0.046	0.028	-0.101
T (°C)			1	-0.364	-0.324	0.158	0.187	0.015	0.045	0.066	0.046	-0.063	0.173	0.192	0.159
RH (%)				1	-0.173	-0.296	-0.200	-0.181	-0.228	-0.254	-0.148	-0.185	-0.210	-0.278	-0.066
p (hPa)					1	0.052	-0.023	0.024	0.025	0.017	0.047	0.023	-0.006 (0.013)	0.013	-0.001 (0.606)
G (J cm^{-2})						1	0.733	0.214	0.247	0.255	0.162	-0.003 (0.277)	0.783	0.937	-0.021
D (J cm^{-2})							1	0.172	0.202	0.210	0.142	-0.003 (0.189)	0.703	0.8121	-0.050
V_w(cm s^{-1})								1	-	-	-	0.202	0.184	0.211	0.066
$V_{w\ Absolute\ values}$ (cm s^{-1})									1	-	-	0.287	0.215	0.244	0.039
$V_{w\ Convection}$ (cm s^{-1})										1	-	0.315	0.224	0.252	0.028
$V_{w\ Subsidence}$ (cm s^{-1})											1	0.311	0.141	0.166	-0.047
V_h(m s^{-1})												1	0.018	-0.003 (0.225)	-0.084
UV - S (Med h^{-1})													1	0.835	-0.036
UV - Y (Med h^{-1})														1	-0.026

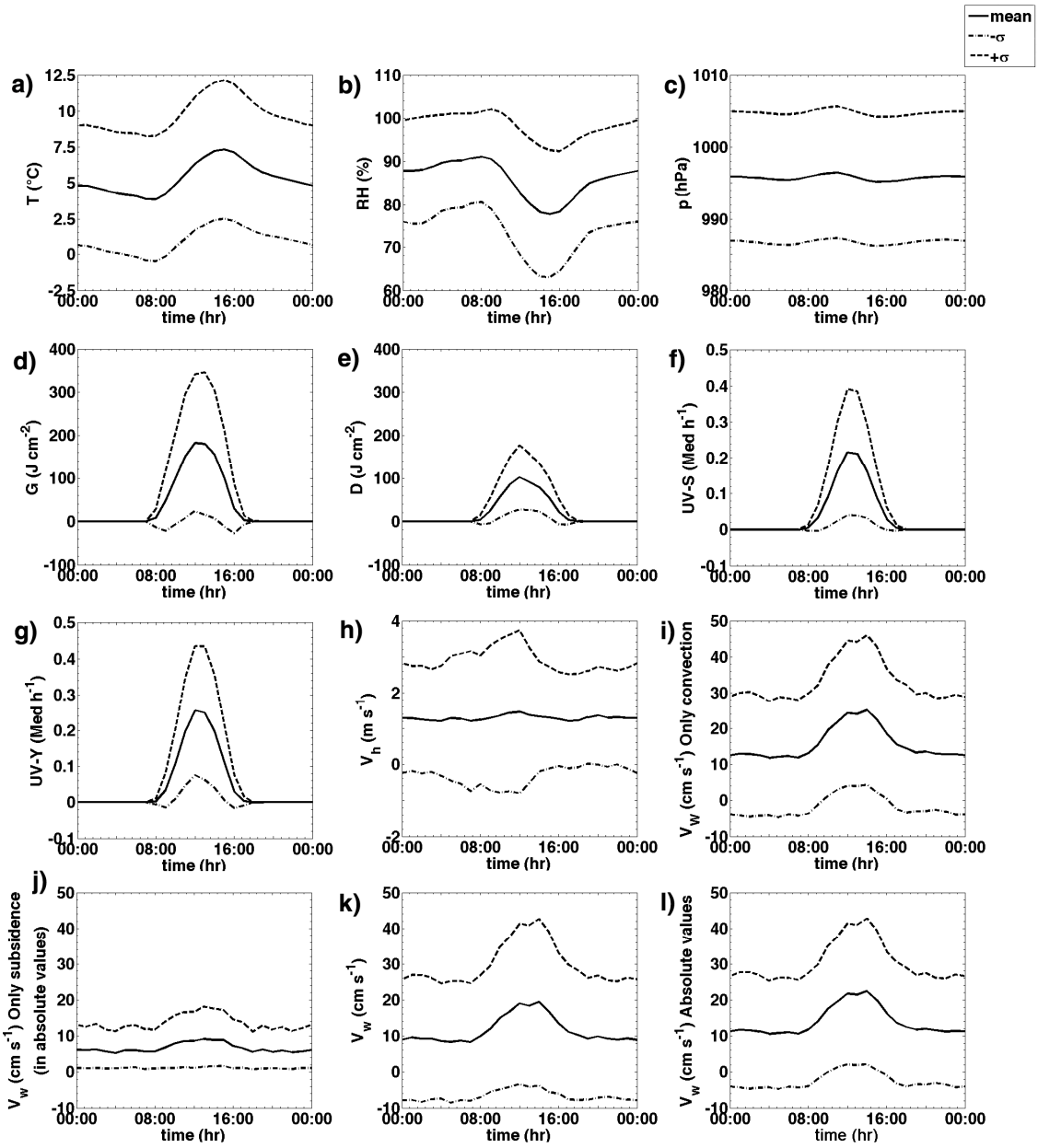

Figure 1: Mean diurnal variation (solid line) +/- standard deviation (dashed and dot-dashed lines, respectively) of meteorological variables, for the study period (01.11.2013-28.02.2014). a) Temperature, b) Relative humidity, c) Air pressure, d) Global radiation, e) Diffuse radiation, f) UV radiation (UV-S), g) UV radiation (UV-Y), h) Horizontal wind speed, i) Convection, j) Subsidence, k) all vertical wind speed data with direction (V_w), and l) all vertical wind direction in absolute values, this means without direction ($V_{w\ Absolute\ values}$).

For the various graphics that describe the behaviour of vertical wind speed (fig.1.i, fig.1.j, fig.1.k and fig.1.l), it is observed that those which represent the average of V_w (fig.1.k) and $V_{w\text{-}Absolute\ values}$ (fig.1.l) have almost the same distribution. They have two marked peaks, which match two maximums, of 18.98 cm s^{-1} and 21.73 cm s^{-1}, respectively, at 12:00 LST, and 19.50 cm s^{-1} and 22.45 cm s^{-1}, respectively, at 14:00 LST. From midnight, vertical wind speed varies slightly. An increase, that clearly starts around sunrise (07:00 LST approx.) and extends until just after the noon, occurs. Vertical wind speed decreases thereafter until after sunset (19:00

LST approx.), and then oscillates slightly during the night hours. Minimum values during the day are seen at 07:00 LST (8.32 cm s^{-1}) for V_w and at 04:00 LST (10.52 cm s^{-1}) for $V_{w\text{-Absolute values}}$.

As shown in table 3, the correlation coefficients for V_w and $V_{w\text{-Absolute values}}$ in relation to the different variables of solar radiation (G, D, UV-S and UV-Y) are between the highest correlation values for these variables of vertical wind. Table 3 also shows a relationship of the same intensity between V_w and $V_{w\text{-Absolute}}$ and horizontal wind speed, and between those variables and relative humidity, although regarding the relative humidity there is an inverse relation.

Regarding the mean diurnal variation of convection (fig.1.i) and subsidence (fig.1.j), it is observed that the distribution of convection follows the same pattern as V_w and $V_{w\text{-Absolute}}$, although with higher values. The mean diurnal variation of convection shows two peaks at 12:00 LST and 14:00 LST, of 24.25 cm s^{-1} and 25.17 cm s^{-1}, respectively. Though the graph of subsidence follows a different distribution, it seems to be an attenuated version of the V_w, $V_{w\text{-Absolute values}}$ and $V_{w\text{-Convection}}$ patterns. For the subsidence, the major increase starts around 08:00 LST, and reaches its maximum at 13:00 LST (9.18 cm s^{-1}), being this one the maximum for the whole day. After that, the mean speed of subsidence decreases until 18:00 LST and oscillates lightly during the night. Given the dominance, both in frequency and magnitude, of convection over subsidence, it´s easily explained why the graphics for the entire vertical wind speed dataset (both V_w and $V_{w\ Absolute\ values}$) are as similar to the distribution of convection.

The patterns for all the vertical wind variables suggest a relationship between the vertical wind speed and the solar radiation, since the main rise and fall of vertical wind speed follow those of the global and diffuse radiation (and UV). Correlation coefficients show that vertical wind and solar radiation variables are effectively interrelated, where this relationship is more intense for convection. The most significant correlation for vertical wind speed is that one with the horizontal wind speed, being the highest correlation of the presented (table 3) for both convection and subsidence, with almost the same value. This connection with the horizontal wind speed is specially remarkable for the subsidence, with a correlation coefficient value of about double or higher than the rest of correlation coefficients values for the subsidence. The coefficients that define the relation between different vertical wind speed variables and horizontal wind direction are of small magnitude, but it is remarkable that the sign of this relationship changes for subsidence to negative.

3.3 Relationship between PM1 concentration and meteorological variables

Table 3 shows that correlation coefficients between [PM1]GF and different meteorological variables, regardless of their sign, are, in more cases than not, lower than those for [PM1]FF (from the total of coefficients reported for 13 exposed meteorological variables, 7 are lower for [PM1]GF and 6 are higher for [PM1]GF compared to [PM1]FF). In Klaić et al. (2014), however, the correlation coefficients between meteorological variables and indoor PM1 concentration are found to be higher in all the cases for the ground floor in comparison to the first floor.

Still being statistically significant, most of the observed relationships between PM1 concentration on both floors and meteorological variables are of very weak character. The higher correlation is noted for temperature, showing a moderate coefficient of around -0.460 for PM1 concentration on both floors. Below this one, the highest coefficients are found to air pressure (around 0.270) and relative humidity (around 0.250). That differs from the results in Klaić et al. (2014), where the values for the first floor are, in general, much smaller (becoming even zero in the case of air pressure), albeit, for the relative humidity, a value of the same order for the ground floor is obtained. The remaining meteorological variables have a very weak correlation with PM1 concentration, comprising coefficients with values between 0.016 and 0.135. The lower correlations are those that relate PM1 concentration to solar radiation variables (G, D, UV-S and UV-Y).

The graphical representations of the relationship between [PM1]GF and [PM1]FF and meteorological parameters are presented in Figure 2. Patterns in these relationships have the same character on both the ground and the first floor, although the median of concentration for each class in the boxplots is generally higher for the first floor, and there are also differences in the boxplot whiskers, in the interquartile ranges, and in the position of the median within them. So, in terms of relations with meteorological variables, we will discuss from now about PM1 concentration without regard to whether it is from the ground or from the first floor.

Figure 2.a shows that PM1 concentration decreases as the temperature increases. As indicated in Ollier (2013), this could be partly due to the influence of outdoor concentrations, which would be higher at lower temperatures given the increased emissions from heating plants located in the west and southeast of the city. It could be also due to the fact that the residence of contaminants in the atmospheric boundary layer is promoted at lower temperatures (Braniš et al., 2009), when the raising of warm air dragging contaminants to higher layers is not favoured. The boundary layer generally has lower altitude and reduced turbulence at low temperatures. Lower temperatures generally favour partition into the particle phase and formation of secondary species (Braniš et al., 2009; Van Drooge et al., 2010; Pandis et al., 2010). This relationship is expected to be positive in summer (Braniš et al., 2009; Ollier, 2013), because with summer conditions the photochemical production of PM would be facilitated. So, given the variety of chemical compositions of the particles, the relationship with temperature may depend on the temperature range involved (Ollier, 2013). In addition, in Kim et al. (2014), it is mentioned that active vertical mixing due to increase of solar heating may contribute to decrease aerosol concentrations near surface.

This non-linear effect of temperature on PM1 concentration is pointed out in Ollier (2013), in relation to outdoor concentrations, for which the pattern is less clear than in the case of indoor concentrations, perhaps due to the interaction with other meteorological variables. We should add that, as argued in Ollier (2013), the interaction between different meteorological variables and variability of PM emissions may distort the results for individual meteorological variables, making them less clear. However, the combinations of different meteorological variables in relation to indoor PM1 concentrations are outside the scope of this study.

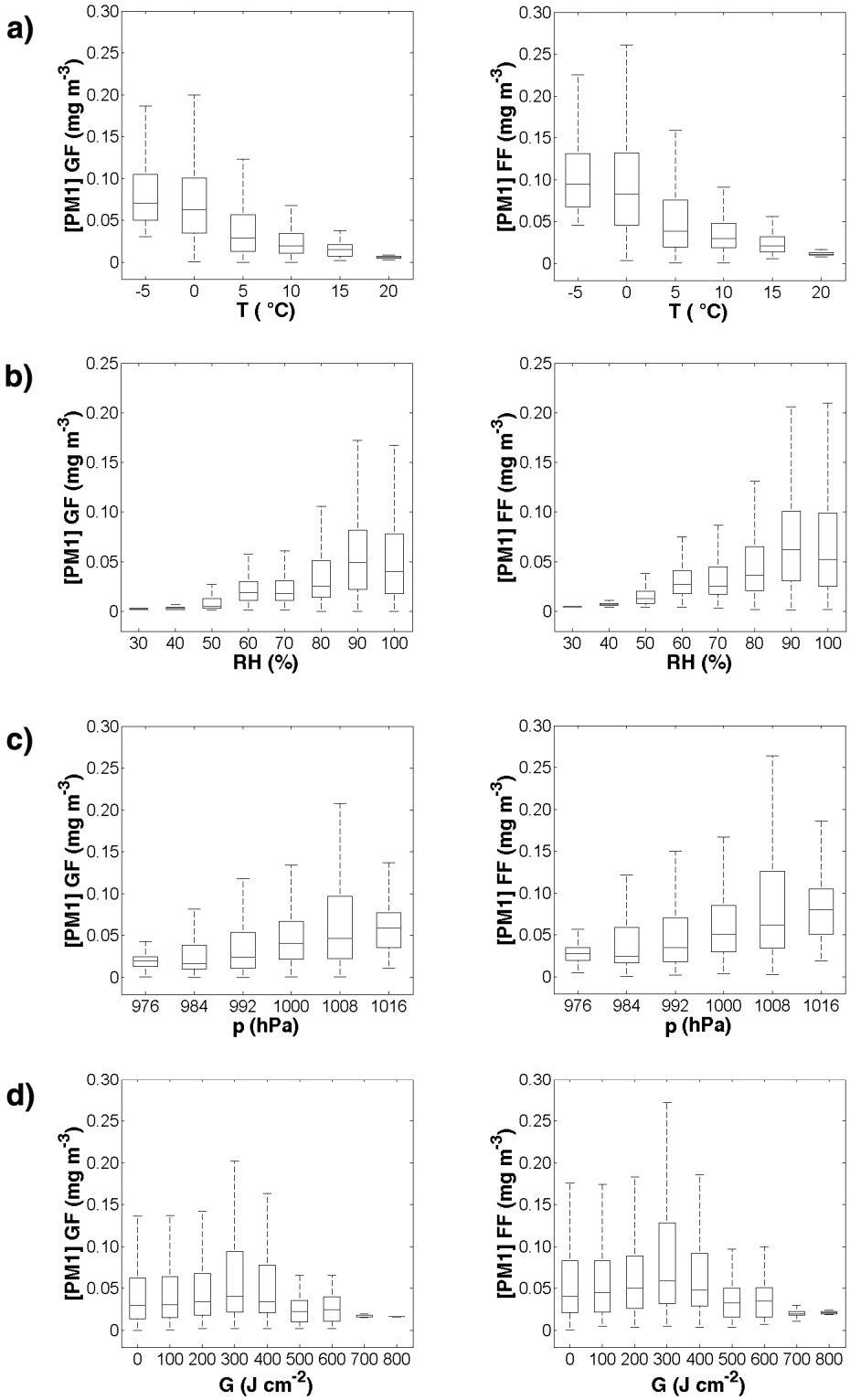

Figure 2: Boxplots for the relationships between indoor PM1 concentration (mg m^{-3}) on the ground floor (GF) and on the first floor (FF) and meteorological variables, for the study period (01.11.2013-28.02.2014). a) Temperature, b) Relative humidity, c) Air pressure, d) Global radiation, e) Diffuse radiation, f) UV radiation (UV-S), g) UV radiation (UV-Y), h) Horizontal wind speed, i) Convection, j) Subsidence, k) all vertical wind speed with direction (V_w), l) all vertical wind direction in absolute values, this means without direction ($V_{w\ Absolute\ values}$), and m) V_w equal to zero.

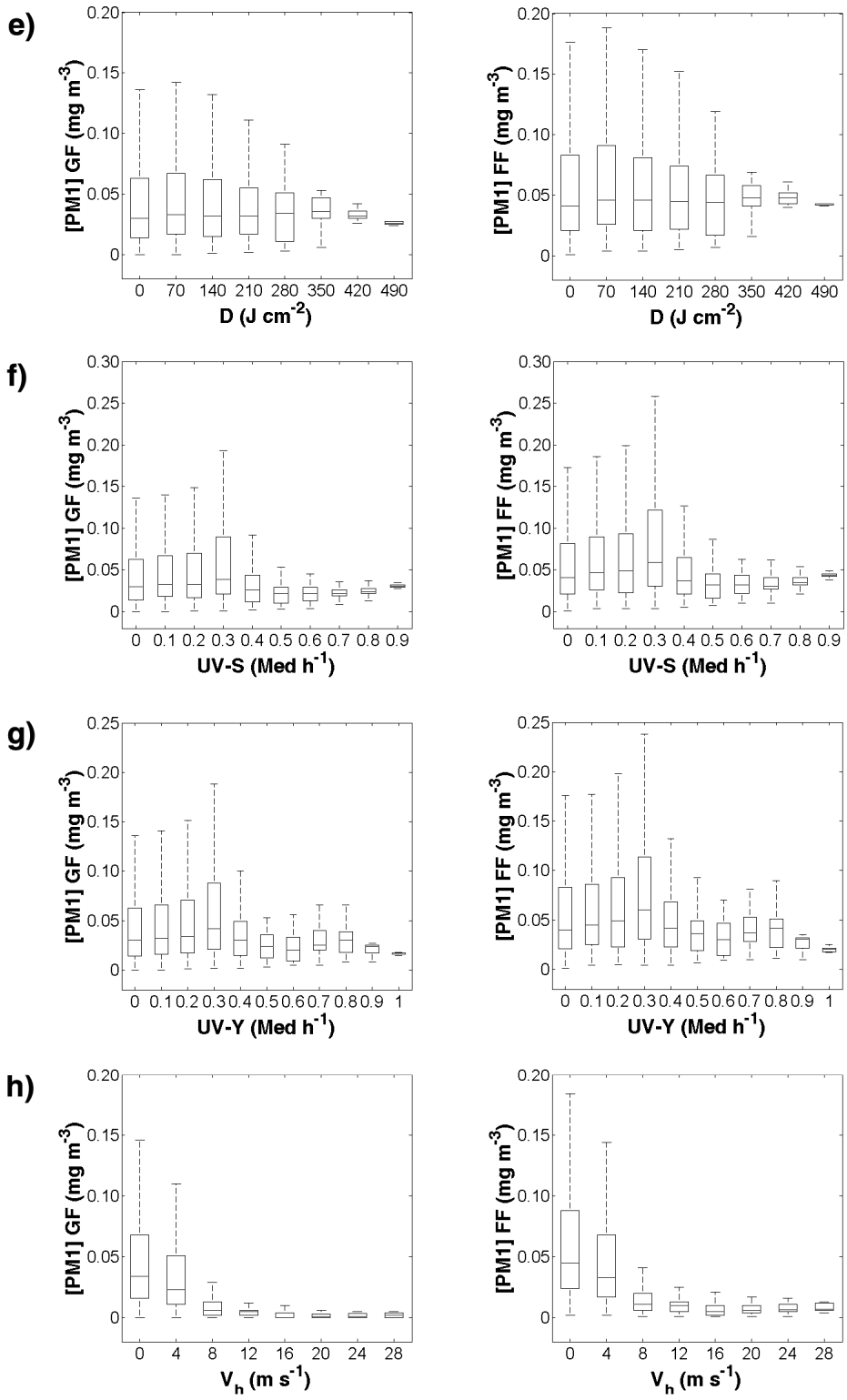

Figure 2. Cont.

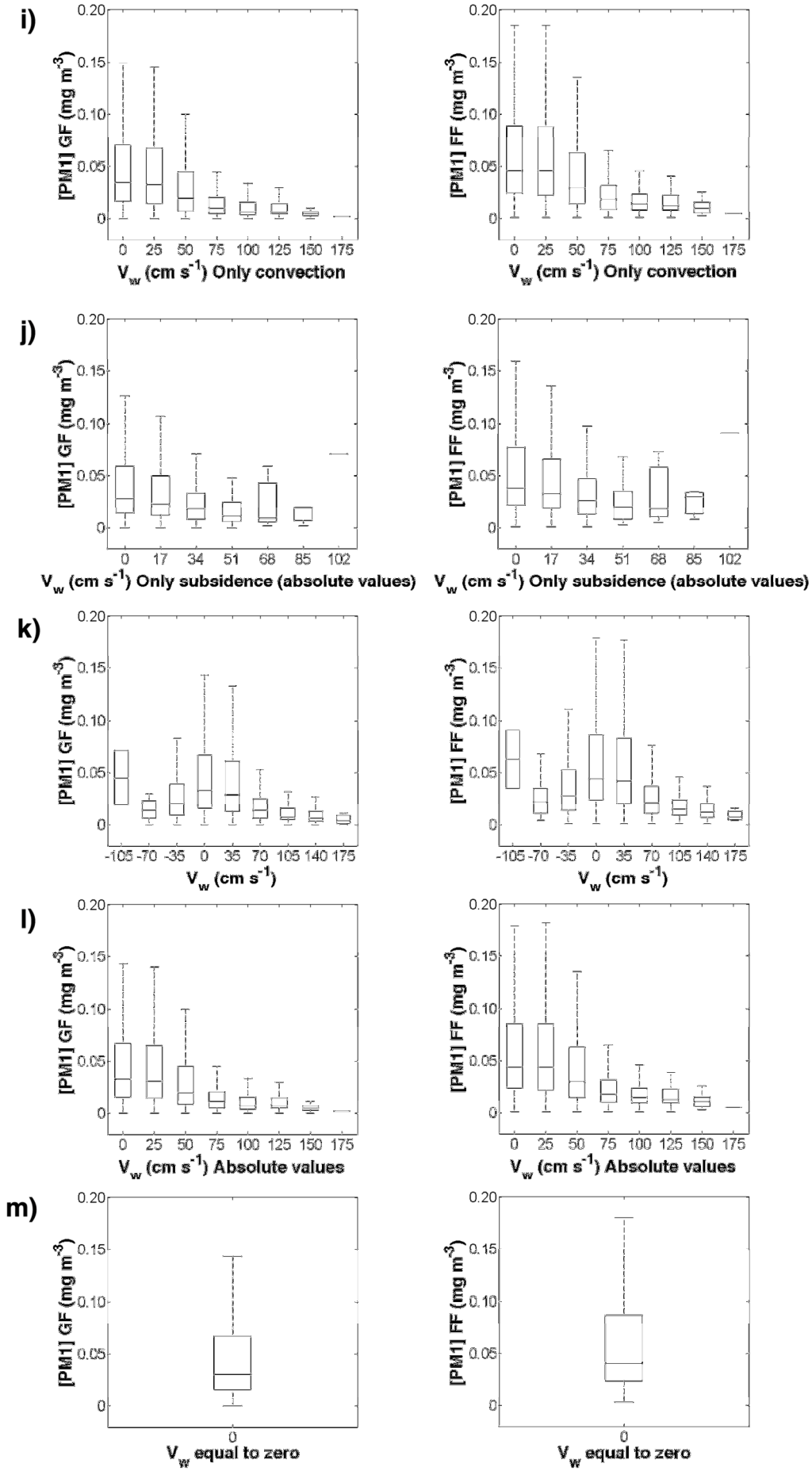

Figure 2. Cont.

The results obtained for the temperature and indoor PM1 concentration are, in general, similar to those in Ollier (2013) and Klaić et al. (2014). Similarly, in Braniš et al. (2009), a significant negative correlation between outdoor temperature and both indoor and outdoor PM2.5 levels is found during the winter, with a correlation coefficient for indoor concentration of -0.387. It shows a close magnitude to the coefficients obtained in the present study and in Klaić et al. (2014). However, different relations with temperature are seen in Klaić (2012) and Klaić et al. (2012), where PM1 concentrations are measured outdoors, and where, albeit they present a wave pattern not so clear, the concentrations at large increase with temperature. These differences with the last two cited studies are probably due to their correspondence with the warm season. In addition, in Braniš et al. (2009), the correlation between outdoor temperature and both indoor and outdoor PM2.5 levels during the summer results positive. Therefore, it would be interesting to carry out studies on the same site and under the same measuring conditions for different periods and seasons. Consequently, different temperature ranges would be analysed, and these data could be compared with each other. The latter would be recommended for the total of meteorological variables.

PM1 concentration increases, as the relative humidity does (Fig.2.b), until a point (90.00% of RH), from which it descends. It may occur because at high relative humidities the phenomena of precipitation or fog are more likely (Klaić, 2012; Klaić et al., 2012; Ollier, 2013). Thus, it may cause removal of particles from the air outdoor by washing mechanisms related to the precipitation and/or fog. The presence of rainfall is associated, in Kim et al. (2014), with low outdoor PM10 concentrations. In Galindo et al. (2013), low outdoor PM concentrations were related to an increase of rainfall, not only because of the removal of particles by wet deposition, but also because of the washout of street, which prevents road dust re-suspension. The reduction of this concentration outdoors would cause a decrease in the amount of particles that penetrates by infiltration into the building, and, thus, a decrease of the indoor PM concentration. Similar results in relation to relative humidity are found in Ollier (2013) (both inside and outside the building), Klaić (2012) and Klaić et al. (2012) (concentrations measured outside), though for cases in which the PM1 concentration is measured outside, the pattern is more wavy. The general trend is the same in Klaić et al. (2014), an increase of indoor PM1 concentration with the increase of relative humidity on the outside. Nevertheless, in the boxplots shown in that study, it doesn´t seem to occur a decrease in concentration with highest relative humidities. It may be due to the rounding used to generate the classes in those boxplots, where bloxpot classes for 80.00% and 100.00% of relative humidity are found. In the results presented here, there is a trend to decreased PM1 concentration from values of around 90.00% of relative humidity (in the same way this trend is observed from around 80.00-90.00% in Klaić (2012) and Ollier (2013)). Maybe for this reason, because of the rounding performed, part of the concentration values still rising are grouped with declining values of concentration for highest relative humidities, resulting in an increased median for the class 100.00% of relative humidity. In Braniš et al. (2009), however, both in winter and in summer, the correlation between the relative humidity in the ambient air and PM2.5 concentration, both indoors and outdoors, is negative. Although the sign is the opposite, in winter and in relation with indoor PM2.5 concentration, the correlation coefficient for relative humidity in Braniš et al. (2009) presents a numerical value (-0.300) of the same order of the one here presented for PM1 indoors (around 0.250). In the cited study, higher correlation coefficients relating to the

relative humidity outdoors are obtained in summer than in winter. Nevertheless, in Braniš et al. (2005), a positive correlation was found between indoor relative humidity and indoor PM1, PM2.5 and PM10 concentrations.

With respect to air pressure (Fig.2.c), it is noted that as it increases, indoor PM1 concentration also does. As indicated in Klaić (2012), this relationship probably is due to the low pressure fields that typically accompany the passage of fronts or cyclones, and with them high wind speeds and precipitation, resulting in ventilation and removal of outdoor particles. On the contrary, as is also indicated in Klaić (2012), the presence of high pressure fields is associated with anticyclones, and thereby with stable atmospheric stratification, winds of low speed, and low rainfall, and so with an increase of the residence time of particles in the air. In addition, in Li et al. (2014), it is mentioned that a high pressure system would be unfavourable for the dispersion of pollutants, as well as in Galindo et al. (2013), and in Grange et al. (2013) high outdoor PM10 concentrations are founded specially during periods dominated by anticyclonic situations. Consequently, the particles concentrate in the air outdoors, and so in the inside with its entry into the building. This behaviour, for both PM1 concentrations measured inside and outside, is seen in Ollier (2013) (although with a more wavy distribution). In Klaić (2012) and Klaić et al. (2012) similar results are obtained for outdoor PM1 concentration, though in Klaić et al. (2012), when the concentration data are analysed separately for each wind direction, in one of the cases the concentration decreases with greater values of atmospheric pressure. It is explained in the cited study as a result of a small number of values associated with higher values of air pressure, which would distort the results for these concentrations. Similarly, in Klaić et al. (2014), PM1 concentration decreases with highest levels of air pressure (in boxplots, class of air pressure of 1010 hPa), which could be explained by the same aforementioned reason.

The relationships of indoor PM1 concentration with global (Fig.2.d) and diffuse radiation (Fig.2.e) do not have a similar distribution. In neither case the pattern is very clear, and, in addition, both variables present very low correlation with PM1 concentration on both floors. Having regard solely to their correlation coefficients, this relation generally would go in the direction of a relative increase in the concentration of PM1 with increasing the amount of radiation. However, fig.2.d might suggest an increase of PM1 concentration with the increase of the global radiation up to 300.00 J cm^{-2}, and a drop of it with higher values of global radiation. The fig.2.e suggests that PM1 concentration would remain similar without major changes until a diffuse radiation of about 350.00 J cm^{-2} and would decrease thereafter. Clear wavelike patterns for global radiation are also not obtained in Klaić (2012), Klaić et al. (2012), Ollier (2013) and Klaić et al. (2014). In the aforementioned studies, in some cases there is a tendency of the increase in PM1 concentration with increasing radiation values (e.g., Klaić et al. (2014), where the maximum concentration matches maximum global radiation, although the almost negligible correlation coefficient is negative), and in the other cases the opposite (e.g., Klaić (2012), minimum concentration matches maximum global radiation). It should be noted that some of these discrepancies are due to seasonal differences between the studies, the size of the data set used in them, and also due to the fact that, in some of those studies, outdoor concentrations are measured instead of indoor ones.

Something similar happens with UV-S (Fig. 2-f) and UV-Y (Fig.2.g), for which PM1 concentration also exhibits an unclear pattern and a very weak correlation. In a way, it is reminiscent of the distribution in the global radiation boxplot (Fig.2.d), which would be reasonable given that the UV radiation is one of the components of the solar radiation spectrum. This unclear relationship between solar radiation variables and PM1 concentration could be related to the different photochemical behaviours of the various suspended particles that are measured. This matter, which has a great variability, is not approached in this study. As mentioned in Ollier (2013), we note that during the winter the time intervals with solar radiation are short and cloudy days are frequent. This would decrease the number of data and the range of solar radiation values, and ,thereby, hinder further the obtaining of clear patterns. In any case, regarding the global radiation, in Klaić (2012) and Klaić et al. (2012), unclear results are also obtained. Those studies include warmer periods of the year, and, accordingly, higher incidence of solar radiation is analysed. Therefore, for solar radiation variables, a deeper study covering all seasons throughout the year would be desirable.

As seen in the fig.2.h, there is a clear pattern in the way how horizontal wind speed and the PM1 concentration relate with each other. As the wind speed increases, the concentration decreases. This is due to efficient outdoor ventilation, through which the wind transports particles elsewhere. In Li et al. (2014), low surface wind speeds are considered as a factor which wouldn´t favour the dispersion of contaminants. In Diapouli et al. (2008), a negative correlation between PM concentrations and horizontal wind speed was also found. Similarly, the effect of ventilation may occur directly inside the building by wind inlet and outlet when the access door or windows are opened. Another possible effect of low horizontal wind speeds is reduced contribution of the wind erosion to atmospheric particles. Similar results to those present here are obtained for indoor PM1 concentrations related to horizontal wind speed in Ollier (2013) and Klaić et al. (2014). In other studies in the area (Klaić, 2012; Klaić et al., 2012), a similar trend is generally observed for concentrations outdoors, although in some cases it is a distribution in an inverse "U" shape.

Interesting differences in the patterns are obtained in Klaić (2012) and Klaić et al. (2012) by analysing the wind speed together with the direction from which the wind blows. For weak winds, according to these two studies, increased speed involves that in some cases the ventilation dominates over the advection of pollutants (mainly for southwestern and eastern winds) and in other cases the opposite occurs (for those winds with southeastern component, since the industrial area of Zagreb is located southeast from the measurement site). For the above directions, for strong winds the trend would be the opposite. The advection would dominate over the ventilation for strong winds mainly from southwest and east (due to these strong winds are generated by a long-scale forcing and would be accompanied by pollution generated at long distances). Conversely, ventilation would dominate over advection for the strong winds with southeastern component. In any case, for most investigated directions and when the wind direction is not taken into account, the trend in those studies shows a PM1 concentration decrease as the horizontal wind speed increases.

The absolute frequency distribution of the different wind directions shown in Fig.3.a is similar to the one in Ollier (2013). The most frequent winds in this case are those from the east (11.0%), north (9.3%), south-southeast (8%), east-northeast (7.9%), northwest (7.9%), and

west (7.7%). The similarity in frequency of the wind directions may be because in both studies the chosen period of the year is similar (although in different years). Presumably, because of different times of the year, wind direction rose here presented does not look at all like the ones in Klaić (2012) and Klaić et al. (2012). In those studies the major winds happen to be those from north-northwest, northwest, north and west.

The mean PM1 concentration rose based on horizontal wind directions is presented in fig 3.b, where it can be seen how indoor PM1 levels are heavily dependent on the wind direction. Highest mean concentrations of PM1 on both the ground floor and the first floor are obtained for winds from the southeast, with values of 0.070 mg m^{-3} and 0.091 mg m^{-3}, respectively. High values are also obtained for both floors for winds from the south-southeast (0.069 mg m^{-3} and 0.088 mg m^{-3} for the ground floor and the first floor, respectively), east (0.066 mg m^{-3} and 0.085 mg m^{-3}, respectively) and east-southeast (0.064 mg m^{-3} and 0.080 mg m^{-3}, respectively), followed by winds from the east-northeast (0.057 mg m^{-3} and 0.075 mg m^{-3}, respectively). It is expected that the maximum concentration for both floors were those in relation to southeastern winds (and also high for directions with south and east component), due to the influence of the industrial zone of Zagreb. The air from the industrial area also would run across the city, which results in an extra gain of particles on the way.

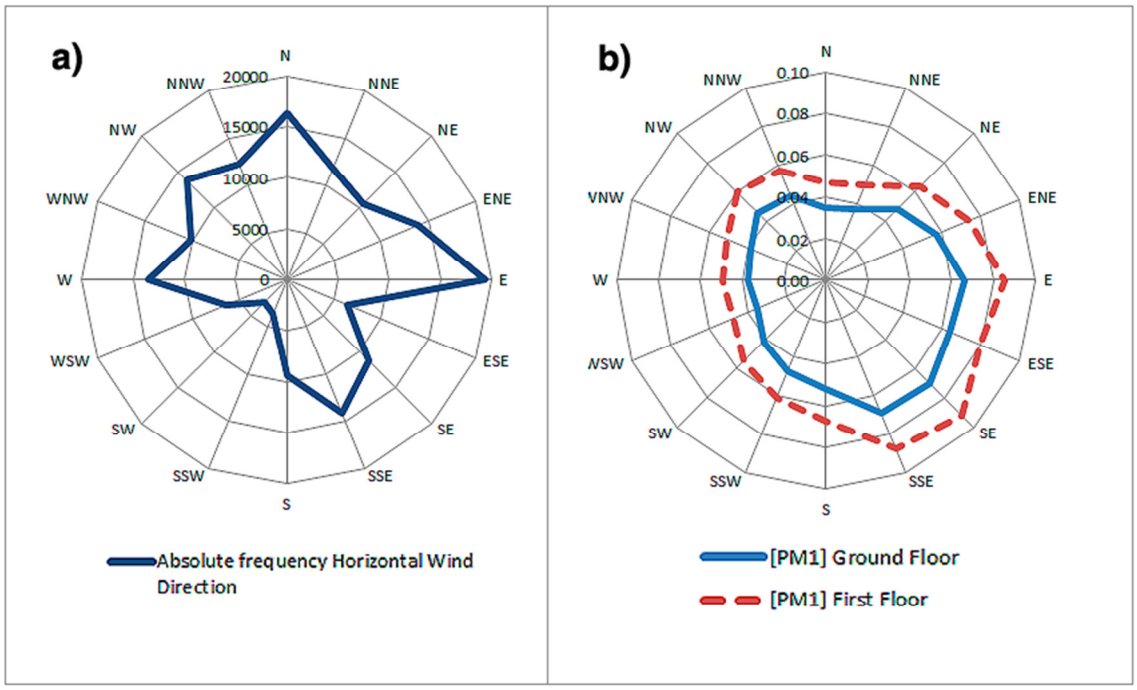

Figure 3: Horizontal wind directions at 22.5° intervals. a) Absolute frequency of horizontal wind directions for the study period (01.11.2013-28.02.2014) and b) Indoor PM1 concentration (mg m^{-3}) on ground floor (GF) and on first floor (FF), solid and dashed lines, respectively, in relation to horizontal wind direction, for the study period (01.11.2013-28.02.2014).

The lower mean indoor PM1 concentrations are obtained for winds from the north (0.035 mg m^{-3} on the ground floor and 0.047 mg m^{-3} on the first floor), followed by those from the west-southwest (0.036 mg m^{-3} and 0.048 mg m^{-3}, respectively), west (0.037 mg m^{-3} and 0.049 mg m^{-3}, respectively), north-northeast (0.037 mg m^{-3} and 0.050 mg m^{-3}), and west-northwest (0.039

mg m^{-3} and 0.051 mg m^{-3}). The winds from the north, north-northeast and west-northwest are among those listed in Klaić (2012) that are expected to transport clean air from the Mount Medvednica. This would explain the low mean concentrations associated with them. For the low concentrations associated with winds from the west-southwest and west, the explanation could be in air masses coming from the opposite side of Mount Medvednica (from its north-northwest slope). Those air masses surround the mountain to its southwestern end and join the airflows coming from the west of Zagreb. This supply of clean air dilutes the concentration of particles in these airflows from the west, which would reach the building site at its western and west-southwestern side. In addition, these airflows coming with a west and south-west component could be more or less clean, in special for weak winds, because they could transport clean air from the Adriatic sea. Conversely, higher concentration is expected for strong winds with this direction, as seen in Klaić (2012) and Klaić et al. (2012), since they can transport large-scale pollution from industrial zones in the north of Italy.

Mean concentration rose shape (Fig.3.b) resembles the ones in the other studies in the area (Klaić, 2012; Klaić et al., 2012; Ollier, 2013; Klaić et al., 2014), albeit the mean PM1 concentrations obtained for different wind directions are higher in the present study (much higher compared to the first three studies, which have a maximum between 0.035 mg m^{-3} and 0.045 mg m^{-3} for outdoor PM1 concentration; not so large compared to the fourth study, which has a maximum of 0.060 mg m^{-3} and 0.067 mg m^{-3} for indoor PM1 concentration on the ground floor and first floor, respectively). These differences between concentrations could be due to seasonal differences and to the fact that PM tends to concentrate more indoors than outdoors, as seen in several studies (Janssen et al., 1997; Wheeler et al., 2000; Mohammadyan and Shabankhani, 2013; Ollier, 2013). In all cases, the maximum concentration is found for winds from the southeast, as well as high values for the south-southeastern and east-southeastern winds. However, minimums are not concurrent. In those studies, when outdoor PM1 concentration is analysed, the lower values are those for wind from west, west-northwest and north. For those studies analysing indoor PM1 concentrations, the lower values are obtained in relation to the winds from north and west-southwest. Compared to the others, in the present study a slight narrowing is noted in the concentration rose shape related to the winds from the north, north-northeast and northeast. This could be somehow related to airflows that come from the north of Mount Medvednica and surround it by its northeastern end. Thus, a stream of clean air may incorporate into the airflows that reach the building site at its northeastern side. This effect may be more enhanced in the particular period analysed here compared to the other studies.

Considering figures 2.i, 2.j, 2.k, 2.l, and 2.m, it could be concluded that with increased vertical wind speed, regardless of its direction, it is favoured the dilution of concentration of the particles. The highest indoor PM1 concentrations are obtained for the minutes without vertical wind (0 cm s^{-1}) (fig.2.m). Patterns of PM1 concentration relative to vertical wind speed in absolute values (fig.2.l) and convection (fig.2.i) are very similar. They are so similar that sometimes the values obtained for medians in the boxplot are identical. Furthermore, the correlation coefficients that define their relationship with indoor PM1 concentration are very close, being in fact the greater ones for the set of variables describing the vertical wind. This would point again (as discussed in Section 3.2) the dominance, in frequency and magnitude, of convection over subsidence in the data set recorded. Subsidence presents noticeable

differences in its relation with indoor PM1 concentration (fig.2.j) comparing to convection and vertical wind speed in absolute values. Indoor PM1 concentration decreases with increasing subsidence speed, albeit with a smoother path than in the cases of the vertical wind speed in absolute values and convection. An exception is observed for the class of subsidence set for 102.00 cm s^{-1}, for which one the PM1 concentration increases. The latter is explained by a very small number of data (just 3 measurements, from a total of 31572 subsidence data, are above 85.00 cm s^{-1}), which could generate a random result for this class.

These results for indoor PM1 concentration and its relationship with the vertical wind speed are similar to those found in Klaić et al. (2014). Furthermore, in Klaić (2012) and Klaić et al. (2012), similar results are found, although the PM1 concentration measurements are performed outdoors. Conversely, the results obtained in Ollier (2013) for outdoor PM1 concentration show no decrease with an increased rate of subsidence.

This reduction in the concentration of PM1 related to vertical wind speed increase could be due to an effective dilution of the particle concentration. Particles are transported by convection to greater heights in the atmosphere, taking them away from the air layer in contact with the ground. It allows efficient vertical mixing and dilution of pollution. With the raising of these particles, the concentration in the air entering into the building would decrease. Therefore, the PM1 concentration decreases in the air in contact with aerosol monitors, as discussed in Klaić (2012) for outdoor measurements. In the case of subsidence, this decrease in indoor PM1 concentration could be associated with winds from the north coming down-slope from the southern slope of Mount Medvednica. These are usually clean winds with low pollutant concentrations, given the non-urbanization of the slope and the absence of major sources of pollutants (Klaić, 2012; Klaić et al., 2012; Klaić et al., 2014). These winds could drag downward particles into a thin layer of air near the ground, an effect enhanced at higher wind speeds. Due to dilution of the particles concentration by the contribution of clean air and to the drag of the particles to the bottom air layer, particles which would enter into the building would do it in the lower layers of air. Thus, particles maybe stay below the aerosol monitors inlet, as indicated in Klaić (2012).

For the other fractions of PM measured on the first floor (PM2.5, PM4, PM10 and also TSP), similar relationships with meteorological variables to the ones here presented for PM1 are found. In most cases, boxplots (not shown here) exhibit almost identical distributions to those for PM1 concentration on the first floor (fig. 2). Slight differences in the value of the median of concentration, interquartile ranges, in the position of the median within the boxes, and in whiskers path, are found.

3.4 Temporal variation of indoor PM1 concentration

Figure 4.a shows the average diurnal distribution of the PM1 concentration for the entire study period, for both the ground floor and the first floor. The pattern throughout the day is very similar in both cases, but smoother on the ground floor, and the average concentrations are higher on the first floor. Three raises of the PM1 concentration are seen starting around 06:00 LST, 09:00 LST and 16:00 LST, respectively. In the first case, the beginning of PM1

concentration increase could be associated with the temperature increase inside the building, due to the configuration of the central heating. This increase in temperature may result in the re-suspension of the particles because of the thermal uplift from the air layer closest to the ground (Lau and Chen, 2007; Ollier, 2013). Furthermore, early in the morning some employees start arriving and the opening of the door in the main entrance would favour the infiltration of outdoor air. The particle concentration is supposed to have started increasing in the outdoor air due to the traffic.

After this initial increase from 06:00 LST, a slight stagnation around 08:00 LST is observed. Another increase is noted from 09:00 LST, coinciding with the arrival of the majority of employees and students. This increase continues throughout the morning due to the movement of people through the hallways, resulting in a more or less constant re-suspension of particles. In addition, the PM1 concentration outside increase related to traffic and human activity in the city (e.g., emissions in the industrial area), favouring by infiltration the increase of particle concentrations inside the building. The increase would peak around 12:00 LST (0.053 mg m^{-3} and 0.070 mg m^{-3}, on the ground floor and first floor, respectively), coinciding with the maximum observed for the whole day. After reaching the maximum, the PM1 concentration on the ground floor clearly begins to descend. On the first floor, however, this peak remains almost at the same concentration until 13:00 LST, and thereafter it begins to descend. This difference would be due to a greater use of the first floor compared to the ground at that time of day, where transit of students and teachers through the hall between classes. Hence, re-suspension would remain PM1 concentration more or less stable at its highest level. The drop of concentration after this peak, maybe due to the activity decrease in the hallways and students leaving the building, is gently slowing until shortly before 16:00 LST, when a minimum is reached. In Rovelli et al. (2014), the highest PM2.5, PM5, PM10 and TSP mean concentrations are also observed in the classrooms when they are occupied by students.

The third increase, observed from 16:00 LST-17:00 LST, would be related to the movement of people prior to the gradual leaving. The concentration then increases to the second maximum for the whole day on both floors (0.049 mg m^{-3}, at 20:00 LST, on the ground floor, and 0.064 mg m^{-3}, at 19:00 LST, on the first floor).

The difference in the time when this second maximum is reached on one and another floor, could perhaps be due to increased use of the ground floor in this moment of the day, on which one employee remains mandatory until 21:00 LST. In any case, although the second maximum is reached on the first floor before than on the ground floor, at 20:00 LST PM1 concentration has decreased very slightly. A larger decrease is seen as of 21:00 LST on the ground floor. Thus, throughout the day the time period associated with the moments of higher concentrations is wider on the first than on the ground floor. As of 20:00 LST, the concentration begins to decrease clearly and gradually on both floors. We note a slight increase in concentration around 03:00-04:00 LST. The minimum average concentration on both floors occurs around 06:00 LST (0.040 mg m^{-3} and 0.052 mg m^{-3}, for the ground floor and first floor, respectively). Diurnal amplitude of the average concentrations is relatively low, of 0.013 mg m^{-3} for the ground floor and of 0.018 mg m^{-3} for the first floor. Similar diurnal amplitude is found in Ollier (2013) for the first floor.

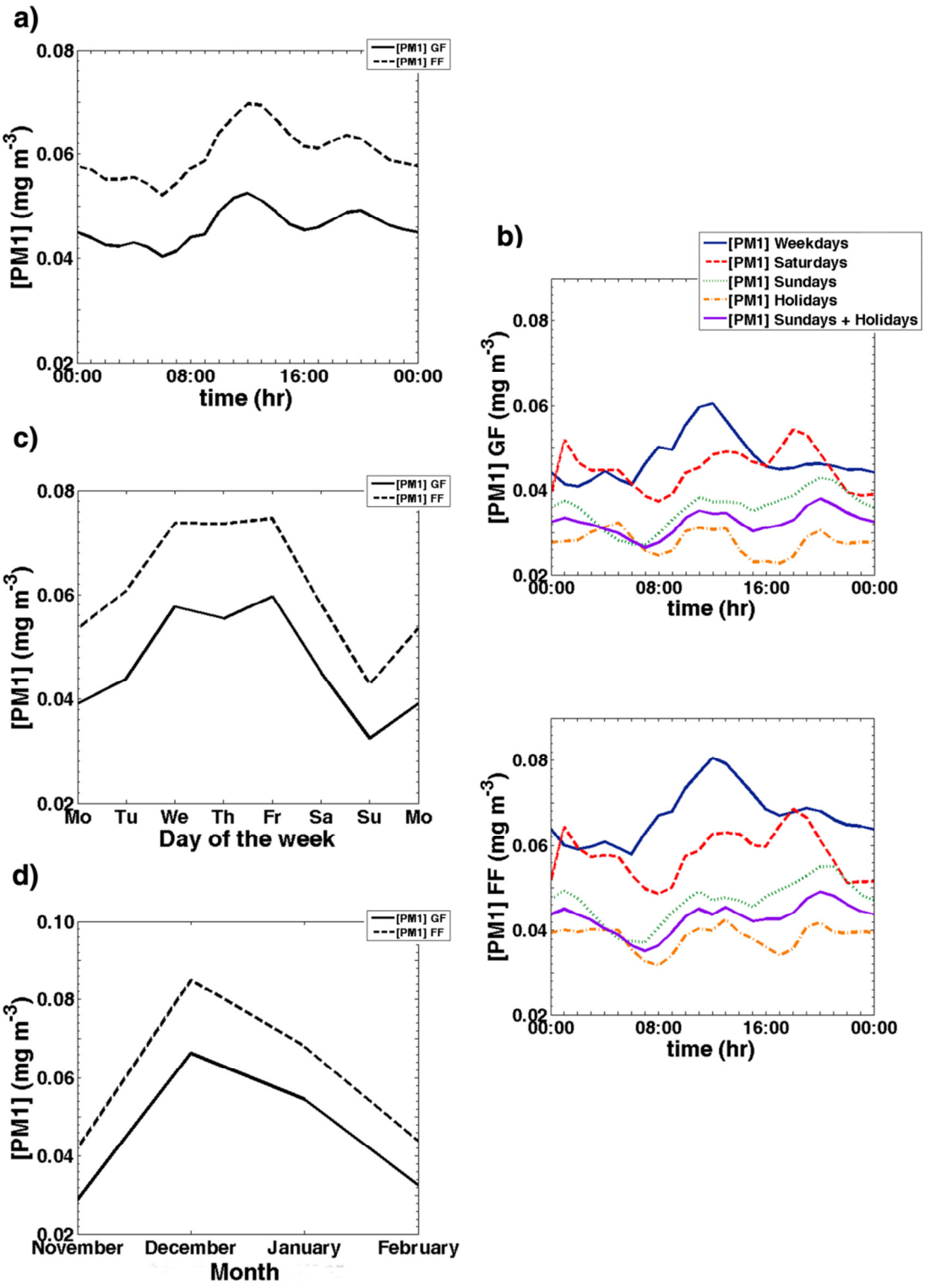

Figure 4: Temporal variations of indoor PM1 concentrations (mg m^{-3}) on ground floor (GF) and on first floor (FF), for the study period (01.11.2013-28.02.2014). a) Mean diurnal variation of indoor PM1 concentration on both floors, for the study period, b) Mean diurnal variation of indoor PM1 concentration for weekdays (blue solid line), Saturdays (red dashed line), Sundays (green dotted line), holidays (purple solid line) and Sundays+holidays (orange dot-dashed line), on both floors, for the study period, c) Mean weekly variation of indoor PM1 concentration on both floor, for the study period, and d) Mean monthly variation of indoor PM1 concentration on both floors, for the study period.

These results are compared with those obtained for the same building and under the same wintertime conditions, although for slightly different periods of a different year (Ollier, 2013; Klaić et al., 2014). It is observed that, although distributions are similar, peaks do not always coincide exactly. In Ollier (2013), on the first floor the first peak of maximum PM1 concentration is between 11:00LST and 12:00 LST (0.063 mg m^{-3}) and the second maximum is around 21:00 LST (about 0.062 mg m^{-3}). Another set of intermediate peaks are observed around 14:00 LST and 18:00 LST. As in the study is argued, these peaks correspond to the entry and exit of employees due to the lunch break and leaving the building. The minimum in this study occurs at 05:00 LST (0.053 mg m^{-3}). In Klaić et al. (2014), the maximum is also around 11:00-12:00 LST (0.052 mg m^{-3}) on the ground floor, and at 13:00 LST on the first floor (0.058 mg m^{-3}). It is followed by a second peak around 22:00 LST (0.048 mg m^{-3} and 0.052 mg m^{-3} for ground floor and first floor, respectively). The minimum in the study is found at 06:00 LST approximately (0.0420 mg m^{-3} and 0.0460 mg m^{-3} for the ground floor and first floor, respectively) and a slight increase in concentration is also observed during night hours, from 02:00 LST to 03:00 LST in this case. The nocturnal peaks seen here and in Klaić et al. (2014) could be related to some extent to the decrease of nocturnal boundary layer height, which results in an increase of PM concentrations outside. In the study period some episodes of larger infiltration might occur at night, due to the meteorological conditions outside. In Grange et al. (2013), it is mentioned how during wintertime, with cold and calm conditions during anticyclonic situations, nocturnal temperature inversions limit the vertical and horizontal dispersion of pollutants. Furthermore, in Silcox et al. (2012) peaks of outdoor PM2.5 concentration were found during night time, due to surface temperature inversions. However, in Ollier (2013), the effect of typical shallow nocturnal boundary layer is not apparent for outdoor PM concentrations, most probably because of the predominance of source reduction and interaction of several meteorological variables. Some differences observed for PM concentrations between afternoon and night in Klaić et al. (2012) were attributed to stronger static stability during the night time. This stronger stability results in a lower boundary layer and weaker pollutant dilution. Klaić et al. (2012) concludes that effects of boundary layer depth were dominated by the effects of advection, and that ,since experiments correspond to wintertime period, a generally shallow boundary layer without important temporal variations of its depth was expected.

Considering that the diurnal variation of PM1 concentration for the entire period not only takes into account the weekdays, but also weekends and holidays, we analyse the variations between the groups of days described in section 2.3. Those groups of days characterize more precisely the impact of human activities on the indoor PM1 concentration. The first thing to note is that the diurnal variation for weekdays (Fig.4.b) closely resembles that one for the diurnal variation for all data (Fig. 4.a). That is reasonable considering that from the entire period of 120 days, 76 days are weekdays/working days. The mean diurnal variations of concentration of groups of days (fig.4.b) are very similar on both floors, with higher mean concentrations on the first floor. In the graphics for weekdays only (fig.4.b), higher concentrations, on average, are found for both floors than in the diurnal variation for the entire period (fig.4.a). This is more evident for the first floor, for which one an almost identical patterns are observed, but with higher magnitude. In addition, on the ground floor weekday

peaks rising early in the morning and at noon are more enhanced, while the peak around 20:00 LST is quite lower.

For Saturdays (Fig 4.b), the first increase of indoor PM1 concentration in the morning starts later than on weekdays (Fig. 4.b), at 09:00 LST, coinciding with the second increase of the concentration for the weekday mornings. This PM1 concentration increase occurs in a staggered mode until 12:00-13:00 LST, reaching a value of 0.049 mg m^{-3} and 0.063 mg m^{-3} (ground floor and first floor, respectively). Afterwards, concentration stays roughly constant until between 14:00 LST and 16:00 LST, and then it starts to decrease very slightly. This could be linked with the fact that heating is maintained at a constant temperature during the weekend. So, the observed rise early in the morning for weekdays (Fig.4.b) and for the diurnal average for the whole period (Fig.4.a), which is partly associated with the temperature increase at 06:00 LST due to the heating, would not occur on Saturdays. Furthermore, on Saturdays the traffic is weaker and starts later. The increase of concentration observed around 09:00 LST could be associated with the arrival of the staff member who comes mandatorily on Saturdays, although his/her schedule is from 07:00 LST to 14:00 LST. The midday peak of concentration would be due to the activity of the employee in the building, the opening of the entrance door, and/or the possible arrival of other employees. Another factor to consider may be the traffic increase in the city throughout the Saturday morning, which, though, is weak, could contribute to increase indoor PM1 concentration by infiltration. On Saturdays, a peak with a maximum around 18:00 LST (0.054 mg m^{-3} and 0.069 mg m^{-3}, for the ground floor and first floor, respectively) is also observed. This peak happens to be the maximum mean concentration on both floors on Saturdays. In Ollier (2013), a lower peak around the same time (18:00 LST) is seen on Saturdays, although much less than the one observed at noon in the same study. Furthermore, the pattern for the diurnal variation of indoor PM1 concentration on Saturdays in Ollier (2013) presents a generally different shape in comparison with the here presented.

This peak in the late afternoon could be due to several factors or combination of them: more regular access of other employees to the building on Saturday afternoon during the study period (perhaps because of the need to work overtime to finish a project), increased infiltration through cracks in the building due to meteorological conditions outside, or maybe increased traffic in the afternoon on Saturdays (perhaps related to recreational activities or shopping). In any case, it is still remarkable, in the present study, that this increased concentration in the afternoon represents the maximum average concentration for Saturdays, over that obtained when the building is occupied for sure. Another important factor is the possibility that the unusual pattern of a particular day or of a few days can alter the mean pattern for the entire data set for Saturdays. It´s quite possible, given the limited amount of data for Saturdays in this study: 15 Saturdays, which makes 12.5% of the total number of days studied, this is 5 times less data than for weekdays. Therefore, the diurnal variation of indoor PM1 concentration for each of the 15 Saturdays is separately analysed, and compared with each other (graphics not presented here). It is noted that the peak of increased concentration at noon occurs very clearly in the 100% of the cases. In many cases, this increase is very significant and occurs in the specific time slot between 11:00 LST and 13:00 LST (i.e., this increase in the case of Saturday 21.12.2013 is the highest, where the concentration value at 12:00 LST becomes 16 times greater than at 11:00 LST, and the concentration value at 13:00

LST is again similar to the one at 11:00 LST). However, in the afternoon, only for two Saturdays, a high peak around 17:00 LST is seen. This peak is specially prominent for one of them (Saturday 30.11.2013), whilst on other Saturdays some considerably less pronounced peaks are observed at different times of the afternoon and evening. On some Saturdays, no significant peaks in the afternoon or evening are found. Given the magnitude of the peak at 17:00 LST on Saturday 30.11.2013, and the location of other peaks at different hours of the afternoon on other Saturdays, it is not surprising that on average for Saturdays a peak around 18:00 LST appears. Nevertheless, this peak does not represent a clearly common trend around that specific time on all or majority of the Saturdays. Hence, it would be highly recommendable in future studies to expand the number of Saturdays taken into account (and similarly for Sundays and holidays). It could be done perhaps taking Saturday data from the same period of year, but from different years. That would provide a more definite pattern of the general trend in the heating/winter season and an understanding more precise of factors affecting.

To characterize the diurnal distribution of indoor PM1 concentration inside the building in the absence of activity, three groups of days are set: Sundays, holidays and a joint group of Sundays and holidays (all represented in Fig.4.b). In extraordinary occasions an employee could have come to the building during these days, although it is not the common trend and there are not available records of it. The reason why the days are divided in this way is, on one hand, to generate comparable results with Ollier (2013), where Sundays and holidays were analysed together. On the other hand, we follow the recommendation given in that study for analysing Sundays and holidays separately, looking for possible differences between them. It is observed for these three groups of days that, even in the absence of activity in the building, a diurnal variation of the indoor PM1 concentration takes place. The pattern of this variation is very similar in all three cases, although with higher magnitude on Sundays separated from holidays. The latter would suggest that, at least for the study period, the hypothesis of Ollier (2013) is not confirmed. In that study, it was hypothesized that data of Christmas holidays could have raised the mean concentrations that would have been obtained if only Sunday data would have been analysed.

It is noteworthy in figure 4.b that, even in the absence of activity in the building, PM1 concentration increases around the same hours that are observed for increases when the building is occupied. In the absence of activity inside the building, the average concentrations throughout the day are, however, of lower magnitude. These peaks seen for Saturdays and holidays may be related to increased infiltration. Perhaps it could be also related to the re-suspension of particles. The re-suspension in this case would be due to the warming of the air layer adjacent to the ground along the day, which would uplift the particles, which remain in the air for a while. The highest indoor PM1 concentrations on Sundays comparing with the holidays period could be due to the effect of the concentration on Saturdays. After the activity in the building on Saturdays, on Sundays would not have been eliminated by deposition such amount of airborne particles as in the case of a longer period of inactivity such as holidays. Furthermore, during the holidays ventilation and cleaning don´t occur in the building, which would lead to have inside the building a greater amount of the particles that have penetrated by infiltration. Anyway, in the absence of major sources of re-suspension, the deposition of

particles would dominate over re-suspension, and no such high concentrations would be observed.

The fact is that under conditions of reduced activity inside the building, or even in the absence of it, peaks of increased concentration occur around same time slots as for weekdays, overlapping in some cases. Thence is inferred that the activity in the building helps and contributes to magnify the increased concentrations seen, but it´s not the only factor involved in it, and does not fully explain the increases that occur when the building is occupied. In Long et al. (2000), the indoor/outdoor correlations were modified due to the contributions of indoor particle sources. This demonstrates that indoor generated particles can mask the underlying relation between outdoor PM concentrations and indoor PM concentrations that originate outdoors. Ollier (2013) suggests that the main control of indoor PM1 concentration could be the outdoor concentration with an overprint of indoor sources, such as re-suspension or weak ventilation, which leads to a deficient removal of accumulated infiltrated PM from outdoors. Since, as for Saturdays, for Sundays and holidays a limited number of data was available compared to the data available for weekdays, it would be recommendable to use more data in future studies.

In Figure 4.c the weekly mean variation of PM1 concentration on both floors is presented. It is observed on both floors that concentration increases from Monday to Wednesday. On the first floor the mean concentration remains more or less constant from Wednesday to Friday, though with a little increase on Friday, when the maximum average concentration for the whole week (0.075 mg m^{-3}) is reached. Thereafter, PM1 concentration declines progressively until Sunday, when the minimum for the entire week (0.043 mg m^{-3}) is observed. On the ground floor, pattern is, in general, similar to the one on the first floor (again with lower concentrations than on the first floor), although the concentration drops from Wednesday to Thursday and it is not constant as in the case of the first floor. Again, the weekly maximum occurs on Friday (0.060 mg m^{-3}). Similarly to what happens on the first floor, over the weekend the average concentration on the ground floor decreases linearly until its minimum on Sunday (0.032 mg m^{-3}). The concentration increase seen between Sunday and Monday could be explained by the re-suspension produced by employees returning to the building after the weekend, the temperature rise configured for heating system, and increased activity outdoors (traffic, industrial area, etc.).

Since the kind of activity and the approximate number of people in the building do not vary throughout the weekdays (from Monday to Friday), we expect similar re-suspension rates in these days. Furthermore, cleaning tasks are performed daily, and the residence time for particles in the indoor air is not very long (Qian et al., 2008). Thus, one might expect a similar average indoor PM1 concentration for every weekday. However, an increase of concentration between Monday and Friday is observed, although concentrations tend to be more or less constant in midweek. The observed weekly variation could maybe be largely explained by the infiltration of particles from the outside. The outdoor PM concentration could follow a pattern with increased concentrations at the end of the weekday period, given the human activity rate in the city throughout the week. To check this, the diurnal mean outdoor PM1 concentration values recorded in the vicinity of the Institute of Medical Research and Occupational Health in Zagreb, for the period from 01.11.2013 to 31.12.2013, are taken, as indicated in section 2.3.

For this particular period, figure 5 shows weekly variation of PM1 concentration for both floors and for outdoor measurements at Institute of Medical Research and Occupational Health. The weekly variation of indoor PM1 concentration for this period (fig. 5) varies comparing to the one for the whole study period (fig. 4.c). In any case, the concentration rises along the weekdays reaching its maximum on Friday and descends after that throughout the weekend, as also seen for the entire study period. Comparing with the whole study period, the maximum in this particular period is much greater in magnitude (on the ground floor 0.098 mg m^{-3}, in comparison with 0.060 mg m^{-3} for the whole study period; on the first floor 0.120 mg m^{-3}, in comparison with 0.075 mg m^{-3} for the whole study period). This maximum on Friday is also much higher than the values of the rest of the days of the week. Though the minimum value, for this particular period, is reached on the first floor on Sunday (0.049 mg m^{-3}), as for the entire study period, it is reached on the ground floor on Tuesday (0.033 mg m^{-3}), but it is very similar to the one on Sunday.

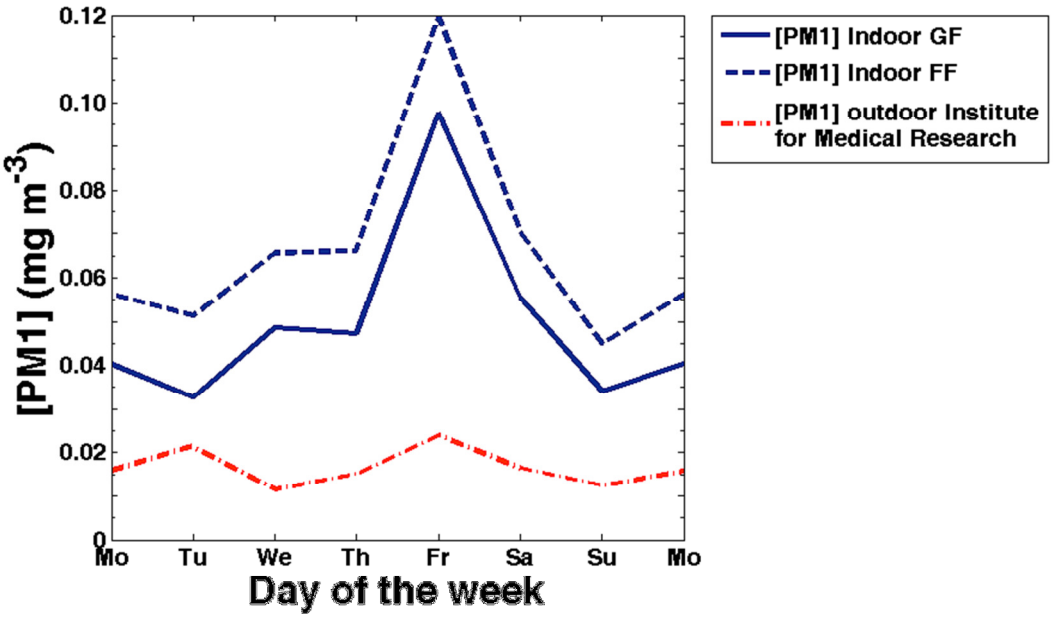

Figure 5: Mean weekly variation of indoor PM1 concentration (mg m^{-3}) on ground floor (GF) and first floor (FF), and of outdoor PM1 concentration (mg m^{-3}) measured at Institute for Medical Research and Occupational Health, for the period 01.11.2013-31.12.2013.

The maximum outside PM1 concentration (fig. 5) also occurs Friday (0.024 mg m^{-3}). It is also noted that the concentration decreases during the weekend. Although the pattern has a different shape for the days in the midweek than the patterns for indoor PM1 concentration on both floors, generally outdoor PM1 concentration also increases from Monday to Friday. The minimum mean outdoor PM1 concentration is found on Wednesday (0.011 mg m^{-3}), though it is almost the same value that the one found on Sunday. Generally increased outdoor PM1 concentration at the end of weekday period together with the decrease during the weekend could partly explain the patterns observed for indoor PM1 concentrations. These somewhat similar patterns outdoors and indoors would suggest that activity outdoors and consequent concentration derived from it would influence, by infiltration, indoor PM1

concentrations observed during the week. It has to be mentioned that these outdoor PM1 concentrations measured at Institute of Medical Research and Occupational Health represent a sample of concentrations that could be found outside the study building, since both locations are not too far one from the other, as explained in section 2.3, and the residential environments around them have similar characteristics.

Mean PM1 concentrations outside the building are lower than those found inside the building (fig. 5). This phenomenon of higher PM concentrations indoors than outdoors, even in the absence of major indoor sources of particles, is often observed in the literature (Janssen et al., 1997; Mohammadyan and Shabankhani, 2013; Ollier, 2013). It occurs mainly due to the poor ventilation in the building in winter (Ollier 2013), when the windows are not often opened because of the low temperatures outside.

The weekly variation found in the present study for indoor PM1 concentration (fig. 4.c) does not correspond to those in Ollier (2013) and Klaić et al. (2014). In these studies, the PM1 concentration in the building decreases from Monday to Friday, contrary to what is observed in this study. In Klaić et al. (2014), a decrease between Friday and Saturday occurs. After that, the concentration slightly increases on Sunday, and rises rapidly between Sunday and Monday. In Ollier (2013), nonetheless, no significant decrease between Friday and Saturday occurs and the mean concentration between these two days is almost constant. In that study, concentration rises more steeply between Saturday and Sunday than in Klaić et al. (2014), and less between Sunday and Monday. The greater similarity between these two studies is because the period analysed in Ollier (2013) is part of the period analysed in Klaić et al. (2014). Conversely, lower mean PM2.5 and PM10 concentrations were found during the weekend in Diapouli et al. (2008) in comparison with the weekdays, as in the present study. In Braniš et al. (2009), during the winter time, the highest concentrations of particles are also found on weekdays.

Regarding the monthly variation for the study period (fig. 4.d), it is noted that the average monthly indoor PM1 concentration is the highest during December (0.066 mg m^{-3} and 0.085 mg m^{-3}, for the ground floor and the first floor, respectively). The minimum average monthly concentrations occur in November (0.029 mg m^{-3} and 0.042 mg m^{-3}, ground floor and first floor, respectively). So, the winter months mean indoor PM1 concentration values are higher than those for November. They are also higher than the values obtained in Klaić et al. (2014) for November and for the spring (March, April), analysed in the same location under the same measuring features although for a different year. The distribution is again similar on both floors, with the highest difference between concentrations on both floors for December and the lowest for February (0.014 mg m^{-3} and 0.011 mg m^{-3}, ground floor and first floor, respectively). Such distribution of indoor PM1 concentration may be partly due to a more intensive use of heating in the city (related enhanced emissions) in months from December to January. This distribution may be also due to the combination of certain meteorological conditions, which could favour higher concentrations outside and enhanced infiltration in the building.

During the Christmas holidays (from 25.12.2013 to 06.01.2014), that cover part of both December and January, the building is unoccupied (with possible exceptions which anyway are

not recorded). Therefore, the high increases of the average monthly concentration of these months compared to November and February (when the building is occupied more days) are remarkable, in particular the maximum in December, of up to 25% of increase on the first floor and 21.8% on the ground floor compared to the mean values for January.

In general, the differences between monthly averages in Klaić et al. (2014) are less abrupt than in the present study. In that study, the maximum mean monthly indoor PM1 concentrations correspond to January (0.068 mg m^{-3} and 0.074 mg m^{-3}, for the ground floor and first floor, respectively), although the concentrations obtained for December are similar to them. So, it is not observed such difference between these two months, as it is in the present study. The mean concentrations obtained for December in the aforementioned study are lower than the ones presented here, the mean concentrations obtained for January are higher (because of the different place for the maximum concentrations in each study). The mean concentrations obtained for November and February are above those obtained in this study. In a similar way as in the present study, in Klaić et al. (2014) the difference between the concentrations for both floors is the highest for the month of December (0.010 mg m^{-3}). However, the difference between concentrations on both floors in that study is, in general, lower than here.

3.5 Relationship between the different indoor PM size fractions measured

As indicated in section 3.1, table 1 shows that the statistics obtained for the different PM size fractions on the first floor are highly similar to each other, coinciding exactly in many cases. For example, the mean values for the entire period are found in a very narrow range between 0.060 mg m^{-3} for [PM1]FF and 0.062 mg m^{-3} for [PM10]FF. It is noted that the statistics calculated for [TSP]FF concentrations are likewise similar to those calculated for the different PM fractions on the first floor, although the maximum value measured of [TSP]FF exceeds up to 0.060 mg m^{-3} the maximum values of the different fractions. This small variation between the mean concentrations measured on the first floor for the different PM size fractions and TSP is clearly observed in figure 6.a, where the mean concentrations of the different fractions for the entire study period are virtually indistinguishable.

Figure 6.b shows that the frequency of occurrence of the different concentrations values for the different PM size fractions on the first floor decreases with increasing concentration value. For all fractions, the lowest concentrations (below 0.100 mg m^{-3}) are the most common, and among them are more frequent those below 0.033 mg m^{-3}. Histograms for [PM1]FF and [PM2.5] FF (fig. 6.b) are virtually indistinguishable. As the size of the fraction increases, a slight decrease is observed in the frequency with which the concentrations below 0.033 mg m^{-3} occur. Furthermore, a slight increase is noted in all frequencies for concentrations between 0.033 mg m^{-3} and 0.200 mg m^{-3}.

Table 4: Ratios (Pearson´s correlation coefficients) between the different indoor PM size fractions measured on the ground floor (GF) and on the first floor (FF). Ratios calculated as (larger fraction/smaller fraction). P=0 for all the coefficients.

	[PM1] GF/	[PM1] FF/	[PM2.5]/	[PM4]/	[PM10]/	[TSP]/
/[PM1]GF		1.500 (0.991)				
/[PM1] FF	0.703 (0.991)		1.002 (1.000)	1.011 (0.999)	1.038 (0.998)	1.068 (0.995)
/[PM2.5]				1.008 (0.999)		
/[PM4]					1.027 (0.999)	
/[PM10]						1.025 (0.999)
p=0 for all R^2 values						

Histogram for [TSP]FF (fig. 6.b) also shows a low increase in the frequency of concentrations between 0.300 mg m^{-3} and 0.500 mg m^{-3}, comparing with the different PM fractions. It is also frequent to find in the literature higher indoor concentration values for the larger PM fractions than for the smaller ones (Kildesø et al., 1998; Tasić et al., 2011; Mohammadyan and Shabankhani, 2013; Rovelli et al., 2014; Hassanvand et al., 2014).

Table 4 shows the concentration ratios and Pearson´s correlation coefficients (calculated from 1-min mean values) for the relationships between different indoor PM size fractions. Both ratios and correlation coefficients are high, being all of them around 1, which shows the close relationship between the different fractions on the first floor (note that for all correlation coefficients, the p value is equal to zero). Similar results were obtained in Ollier (2013), where it is argued that this close relationship may be because, in the absence of significant sources of PM emissions inside the building, most of the particles of the larger fractions originate outdoors as smaller fractions. This can be seen in figure 6.c, where though larger fraction concentrations deviate from [PM1] FF more than smaller fraction concentrations, none them deviates too much from [PM1]FF or from concentrations of the other fractions. Braniš et al. (2005) argue that the very high correlation they found (all of correlation coefficients above 0.9) between indoor fractions PM1, PM2.5 and PM10 during the weekends (that is, without people in the university lecturing room, where the measurements are performed) suggests that there is only one source of particles. In the same study, they argue that the lower correlation coefficients (between 0.762-0.872) found during the day time of weekdays, when the room is occupied, would demonstrate the influence of human activities. Again in Braniš et al. (2005), it is pointed that the strong association found between indoor PM2.5 and PM1 and outdoor PM10 suggests that outdoor air may be an important source of fine indoor particles. In Rovelli et al. (2014), the Spearman´s correlation coefficients between the different PM size fractions are also lower when the schools are occupied than for the nights. This suggests that without activity inside the building no indoor PM sources are found, and, therefore, the main determinant of indoor PM concentration is the particle infiltration from outdoors.

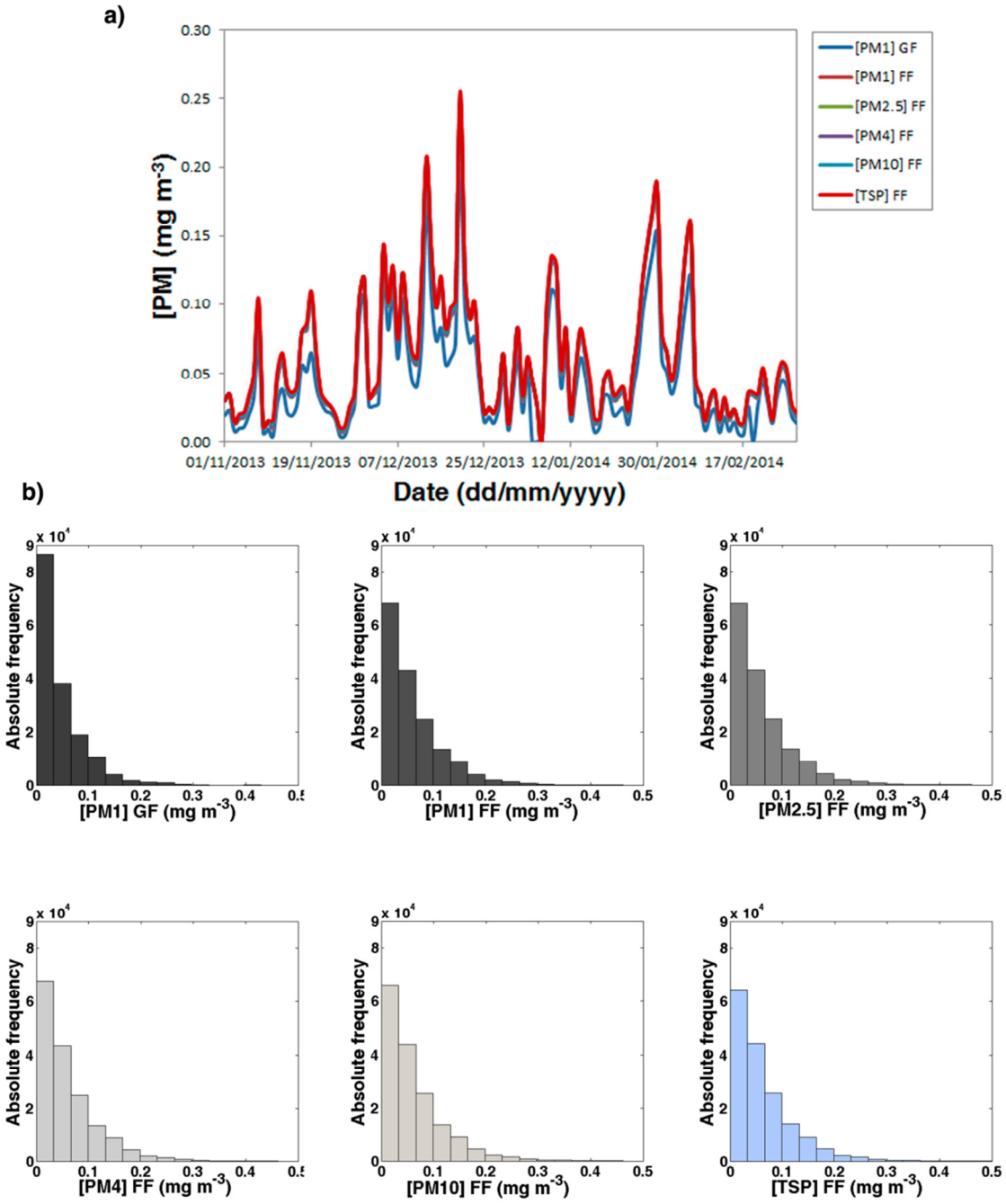

Figure 6: Relationships between different indoor PM size fractions on the ground floor (GF) and on the first floor (FF). a) Times series of concentrations of all PM size fractions (mg m^{-3}), for the study period (01.11.2013-28.02.2014), b) Histograms which show absolute frequency of the concentrations for the different PM size fractions, for the study period, and c) Dependence of larger fractions on PM1 on the first floor, for the study period.

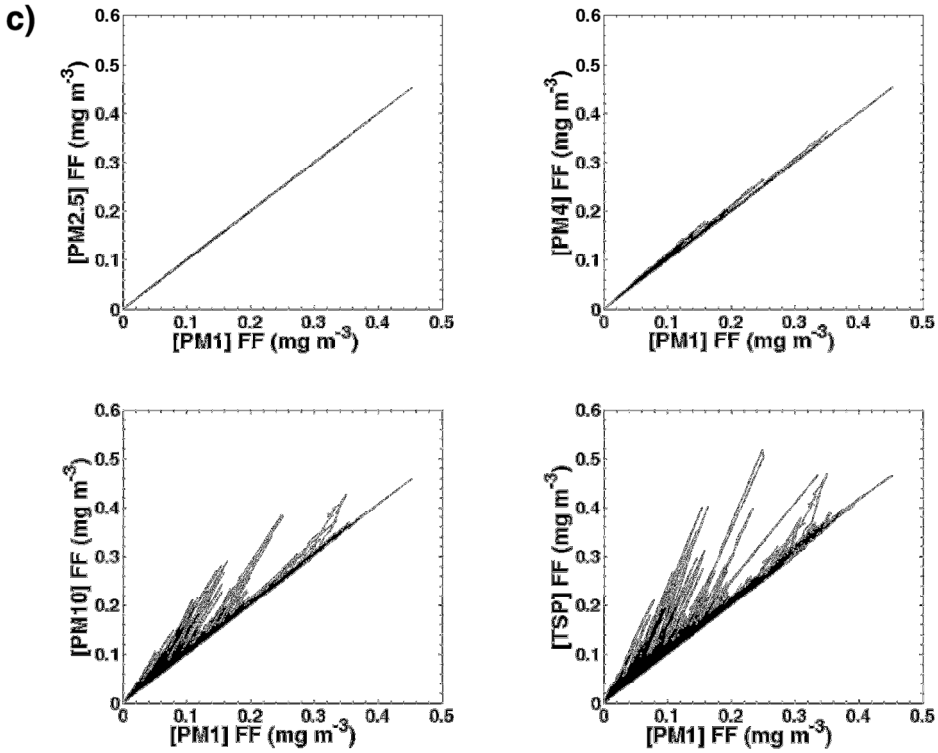

Figure 6 Cont.

As indicated in section 3.3, for all PM size fractions measured on the first floor, similar relationships with meteorological variables are found. Mean diurnal, weekly and monthly variations with similar patterns were obtained for the concentrations of all fractions of PM. The increases and decreases in concentrations generally coincide in the time, although these increases are of a magnitude somewhat greater as the particle size increases. Regarding the mean diurnal variation of the concentration of groups of days (graphics not shown here for fractions higher than PM1), the increases in concentration throughout the day on weekdays are clearly greater the larger the fraction of PM is. These peaks are partly associated with re-suspension by the activity on the first floor. However, peaks of increased concentrations on Saturdays, Sundays, and holidays, not always directly associated with re-suspension, are virtually the same for all fractions. This suggests that re-suspension affects larger particles more than smaller ones, as pointed in Thatcher and Layton (1995) and Rovelli et al. (2014).

3.6 Spatial variation of indoor PM1 concentration. Relationship between ground floor and first floor PM1 concentrations

PM1 concentration is taken into account, since it is the only PM fraction for which concentration data are available for both floors. Time series of indoor PM1 concentration for the study period (fig. 7.a), show that the concentration on the ground floor generally remains below the one on the first floor. The mean and the median for the entire period on the first floor (0.0603 mg m^{-3} and 0.042 mg m^{-3}, respectively) exceed those on the ground floor (0.046

mg m^{-3} and 0.031 mg m^{-3}, respectively), as seen in table 1. Furthermore, we note that PM1 concentrations on both floors show similar patterns in their relationships with the different meteorological variables and in their temporal variations. This would be attributed mainly to the increased activity in the hallway of the first floor, and, probably, to the thermal eddies which uplift the particles to the highest layers of air within the building (remember that the stairs layout characterizes the space as an open corridor between the base of the ground floor and the ceiling of the first floor, allowing the air and particles to move easily between both floors). Furthermore, weak ventilation on the first floor would favour particles concentration.

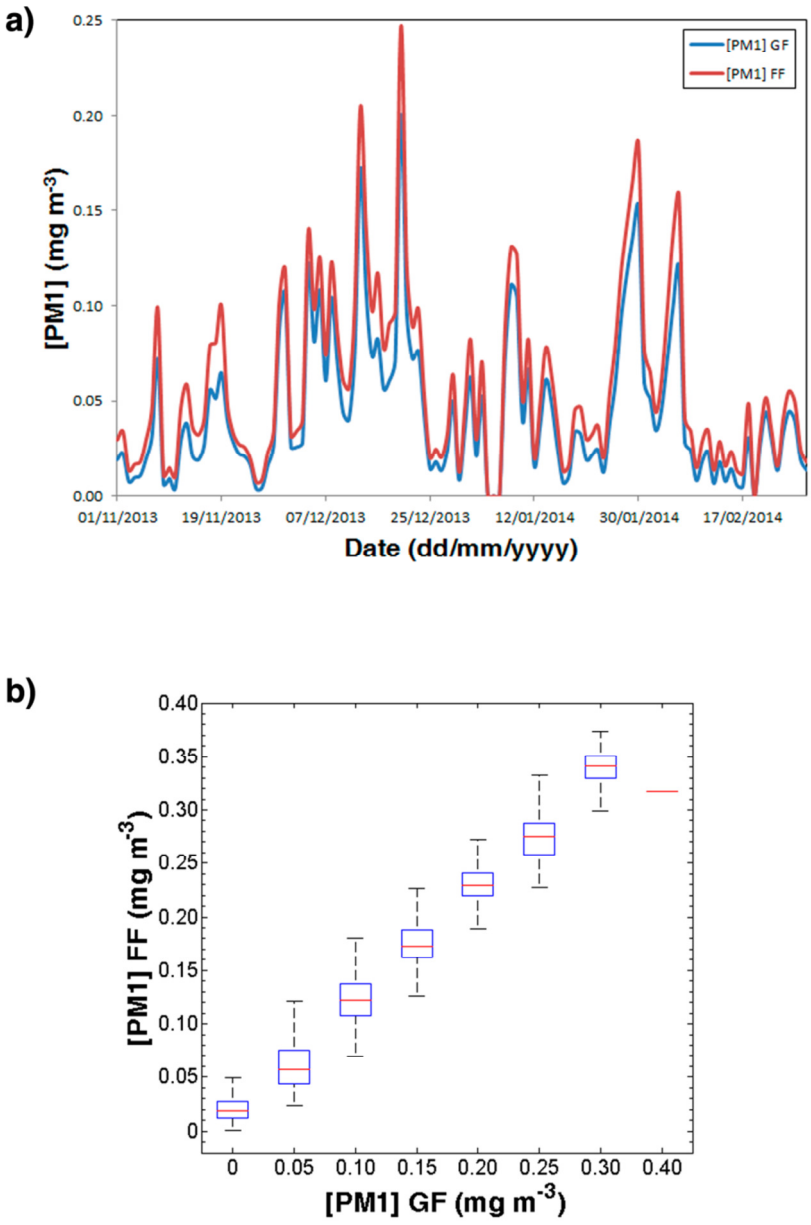

Figure 7: Relationships between indoor PM1 concentration (mg m^{-3}) on the ground floor (GF) and on the first floor (FF). a) Times series of indoor PM1 concentrations on both floors, for the study period (01.11.2013-28.02.2014), and b) Boxplot for the relationship between indoor PM1 concentration on both floors.

As pointed in Ollier (2013) higher PM1 concentration is occasionally observed on the ground floor compared to the first floor. This is due to increased activity at the ground floor and gravitational deposition of particles from the first floor to the ground floor. Furthermore, the measurements are performed on each floor with an aerosol monitor of the same brand but different model, and, hence, the different characteristics of the measuring equipment could also somewhat influence the results.

The correlation between PM1 concentrations on both floors is high, with a Pearson´s correlation coefficient of 0.991 and a ratio [PM1]FF/[PM1]GF of 1.5 (table 4). In the boxplot for the relationship between the two floors (fig. 7.b), a clear linear relationship between the PM1 concentrations is observed. For highest [PM1]GF (0.4 mg m^{-3} concentration class), however, a decrease in [PM1]FF is seen, most probably due to a very small number of data for this concentration class on the ground floor. In the respective histograms (fig.6.b), it is seen that the most frequent concentrations on both floors are those below 0.1 mg m^{-3}. In particular, concentrations below 0.033 mg m^{-3} are quite more frequent on the ground floor than on the first floor, while the other concentration ranges (below and above 0.100 mg m^{-3}) are more frequent on the first floor. This again confirms that concentrations on the first floor are higher in comparison with the ground floor.

In the absence of combustion sources and other significant sources of particle production within the building, the main source of these PM1 particles inside the building would be the infiltration of particles generated outside (Toftum, 2010). This infiltration could occur mainly through the door at the entrance of the building but also through cracks on the building and open windows. Such close relationship between PM1 concentrations on both floors can be attributed to the common origins of particles found on both floors.

4. CONCLUSIONS

After analysing the indoor PM1 concentrations inside a University Campus building in a residential area in Zagreb (Croatia) and their relationships with the meteorological conditions during the winter/heating season, we found a decrease in indoor PM1 concentrations related to outdoor air temperature, horizontal wind speed, and vertical wind speed (both convection and subsidence) increases. Conversely, increased indoor PM1 concentrations are seen with the increase of relative humidity and air pressure. Not very clear wavelike patterns and low correlation are found for relationship between indoor PM1 concentration and solar radiation variables (global, diffuse and UV radiation). Regarding solely the sign of their correlation coefficients, this relation, in very general terms, show some increase in indoor PM1 concentrations with the increase of the amount of solar radiation. With respect to the horizontal wind direction, indoor PM1 concentrations show a large dependence on the direction from which the wind blows. The associations of the industrial zone of the city with increased concentrations and of the clean air coming from the nearby Mount Medvednica with decreased concentrations are particularly noticeable. For indoor PM1 concentration relations with outdoor air temperature, relative humidity, air pressure, horizontal wind speed, and convection, similar results are found in previous studies in the building (Ollier, 2013; Klaić et al., 2014). For subsidence, similar results to those in the present study are found in Klaić et al. (2014), but not in Ollier (2013). Note that the particular study period in Ollier (2013) is a fragment of the one in Klaić et al. (2014). These aforementioned relations, except in the case of air temperature, are also similar to those in previous outdoor studies in the location (Klaić, 2012; Klaić et al., 2012). Again, note that the particular study period in Klaić (2012) is a fragment of the one in Klaić et al. (2012). For global radiation, clear patterns are also not found in Klaić (2012), Klaić et al. (2012), Ollier (2013) and Klaić et al. (2014). Regarding the horizontal wind direction, although the absolute frequency of the different wind directions is not necessarily the same in the aforementioned studies, the mean PM1 concentration roses found in them (indoors and outdoors, depending on the case) resemble the one obtained here. In contrast to Klaić et al. (2014), the correlation between meteorological variables and indoor PM1 concentrations is not always stronger for the ground floor in comparison with the first floor. With respect to the concentrations of the other PM size fractions on the first floor (PM2.5, PM4, and PM10) and also of TSP, similar relationships with the meteorological variables as for PM1 are obtained.

Regarding the temporal variation of indoor PM1 concentrations, several peaks of increased concentration occur throughout the day. These peaks are related mainly to 1) re-suspension of particles by human activity; 2) thermal eddies uplifting the particles with the increase of temperature inside the building; 3) the infiltration of the increased outdoor PM1 concentration associated with the traffic and industrial activity; and 4) outdoor meteorological conditions among others. The maximum mean diurnal concentration of indoor PM1, for the whole study period, is reached on both floors around 12:00 LST, and the minimum around 06:00 LST. The analysis of the indoor PM1 concentration for days grouped based on different activity levels inside the building, shows that re-suspension is an important indoor source of

particles. But it is not the only factor, since the PM1 concentration also varies along the day in the absence of activity in the building (for example, on holidays), although lower increases and generally lower concentrations are seen. The major factor determining indoor PM concentrations seem to be the outdoor PM which enters the building by infiltration. The low ventilation during the winter/heating season also affects indoor concentrations because of the less removal of accumulated infiltrated PM from the outside. The weekly variation of indoor PM1 concentration shows that the concentration generally tends to increase from Monday to Friday, reaching the maximum on Friday, and afterwards descends during the weekend, reaching its minimum on Sunday. Somewhat similar patterns for the outdoor concentrations (measured at Institute of Medical Research and Occupational Health, in Zagreb) and concentrations inside the building suggest that activities outdoors and the consequent concentration derived from them affect, by infiltration, indoor PM1 concentrations observed during the week. The weekly variation found in the present study for indoor PM1 concentration does not correspond to those in Ollier (2013) and Klaić et al. (2014). Regarding the monthly variation, the maximum mean monthly concentration was observed for December and the minimum for November. This is partly due to lower temperatures (and combination of other meteorological variables) and more intensive use of heating in the city, and consequent enhanced emissions.

The different indoor PM size fractions measured on the first floor (PM1, PM2.5, PM4 and PM10), and also TSP, are highly correlated (correlation coefficients around 1). This could be due to the absence of significant indoor sources of particles generation, thus the particles mostly would have a common origin in the outside, and because of that their concentrations are well correlated. All the PM size fractions on the first floor show similar temporal variations with the increases and decreases in concentrations coinciding in the time, but these increases are of a magnitude somewhat greater as the particle size increases. The latter specially occurs during weekdays, when the re-suspension by human activity is the highest. This implies that re-suspension affects larger particles more than smaller ones. Anyway, the results show that re-suspension also affects the smaller particles, given the reduction in mean PM1 concentrations when the building is not occupied.

Spatial variation of indoor PM1 concentration between ground floor and first floor shows that the concentration on the ground floor generally remains below the concentration on the first floor. This is attributed mainly to 1) the greater activity in the hallway of the first floor in comparison to the ground floor; 2) the thermal eddies which uplift the particles to the highest layers of air within the building; and 3) the weak ventilation on the first floor, which favours the increase of particle concentration. Furthermore, PM1 concentrations on both floors show similar patterns in their relationships with the different meteorological variables and in their temporal variations. Both the ratio and the correlation between PM1 concentrations on both floors are very high, which again points to the common origin of the particles inside the building, most probably from combustion activities outside the building (traffic and industrial sources).

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REFERENCES

- Braniš, M., Řezáčová, P. and Guignon, N. (2002). Fine Particles (PM1) in Four Different Indoor Environments. *Indoor and Built Environment*, 11(4), pp.184-190. Available at: http://dx.doi.org/10.1177/1420326x0201100402.

- Braniš, M., Řezáčová, P. and Domasová, M. (2005). The effect of outdoor air and indoor human activity on mass concentrations of $PM_{10,}$ $PM_{2.5}$, and PM_1 in a classroom. *Environmental Research*, 99(2), pp.143-149.

- Braniš, M., Šafránek, J. and Hytychová, A. (2009). Exposure of children to airborne particulate matter of different size fractions during indoor physical education at school. *Building and Environment*, 44(6), pp.1246-1252.

- Buseck, P. R. and Adachi, K. (2008). Nanoparticles in the Atmosphere. *Elements*, 4(6), pp.389-394. Available at: http://dx.doi.org/10.2113/gselements.4.6.389.

- Charlson, R. J., Schwartz, S. E., Hales, J. M., Cess, R. D., Coakley, J. A., Hansen, J. E. and Hofmann, D.J. (1992). Climate forcing by anthropogenic aerosols. *Science*, 255(5043), pp.423-430.

- Diapouli, E., Chaloulakou, A. and Spyrellis, N. (2008). Indoor and outdoor PM concentrations at a residential environment in the Athens area. *Global NEST Journal*, 10(2), pp.201-8.

- Fromme, H., Twardella, D., Dietrich, S., Heitmann, D., Schierl, R., Liebl, B. and Rüden, H. (2007). Particulate matter in the indoor air of classrooms—exploratory results from Munich and surrounding area. *Atmospheric Environment*, 41(4), pp.854-866.

- Galindo, N., Gil-Moltó, J., Varea, M., Chofre, C. and Yubero, E. (2013). Seasonal and interannual trends in PM levels and associated inorganic ions in southeastern Spain. *Microchemical Journal*, 110, pp.81-88.

- Grange, S. K., Salmond, J. A., Trompetter, W. J., Davy, P. K. and Ancelet, T. (2013). Effect of atmospheric stability on the impact of domestic wood combustion to air quality of a small urban township in winter. *Atmospheric Environment*, 70, pp.28-38.

- Hassanvand, M. S., Naddafi, K., Faridi, S., Arhami, M., Nabizadeh, R., Sowlat, M., Pourpak, Z., Rastkari, N., Momeniha, F., Kashani, H., Gholampour, A., Nazmara, S., Alimohammadi, M., Goudarzi, G. and Yunesian, M. (2014). Indoor/outdoor relationships of PM_{10}, $PM_{2.5}$, and PM_1

mass concentrations and their water-soluble ions in a retirement home and a school dormitory. *Atmospheric Environment*, 82, pp.375-382.
Available at: http://dx.doi.org/10.1016/j.atmosenv.2013.10.048.

- Horemans, B. and Van Grieken, R. (2010). Speciation and diurnal variation of thoracic, fine thoracic and sub-micrometer airborne particulate matter at naturally ventilated office environments. *Atmospheric Environment*, 44(12), pp.1497-1505.

- Janssen, N.A.H., Hoek, G., Harssema, H., Brunekreef, B. (1997). Childhood exposure to PM_{10}: Relation between personal, classroom and outdoor concentrations. Occupational and Environmental Medicine, 54, pp.888-894.

- Kampa, M. and Castanas, E. (2008). Human health effects of air pollution. *Environmental pollution*, 151(2), pp.362-367.

- Kelly, F. J. and Fussell, J. C. (2012). Size, source and chemical composition as determinants of toxicity attributable to ambient particulate matter. *Atmospheric Environment*, 60, pp.504-526.

- Kertész, Z., Szoboszlai, Z., Angyal, A., Dobos, E. and Borbély-Kiss, I. (2010). Identification and characterization of fine and coarse particulate matter sources in a middle-European urban environment. *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, 268(11), pp.1924-1928.

- Kildesø, J., Tornvig, L., Skov, P. and Schneider, T. (1998). An intervention study of the effect of improved cleaning methods on the concentration and composition of dust. *Indoor Air*, 8(1), pp.12-22.

- Kim, Y., Kim, S., Yoon, S., Kim, M. and Park, K. (2014). Aerosol properties and associated regional meteorology during winter pollution event at Gosan climate observatory, Korea. *Atmospheric Environment*, 85, pp.9-17.

- Klaić, Z. B. (2012). The Influence of Meteorological Conditions on Fine Particle (PM1.0) Levels in the Urban Atmosphere. In: Fernando, H. J. S., Klaić Z.B. and McCulley, J. L. eds. *National Security and Human Health Implications of Climate Change*. Netherlands: Springer, pp.163-173.

- Klaić, Z. B., Ružman, K., Smiljanić, I., Stojnić, M. And Erbe, P. (2012). The influence of meteorological conditions and weak to moderate traffic density on PM1 levels in a residential area of Zagreb, Croatia. Geofizika, 29, pp.69-102.

- Klaić, Z. B., Ollier, S. J. Babić, K. and Bešlić, I. (2014). Influences of outdoor meteorological conditions on indoor wintertime short-term PM1 levels. *Journal of Applied Meteorology and Climatology*, tentatively accepted.

- Lau, J. and Chen, Q. (2007). Floor-supply displacement ventilation for workshops. *Building and environment*, 42(4), pp.1718-1730

- Li, R., Kou, X., Geng, H., Dong, C. and Cai, Z. (2014). Pollution characteristics of ambient PM2.5-bound PAHs and NPAHs in a typical winter time period in Taiyuan. *Chinese Chemical Letters,* 25(5), pp.663-666. Available at: http://dx.doi.org/10.1016/j.cclet.2014.03.032.

- Long, C. M., Suh, H. H. and Koutrakis, P. (2000). Characterization of Indoor Particle Sources Using Continuous Mass and Size Monitors. *Journal of the Air \& Waste Management Association*, 50(7), pp.1236-1250.

- Luoma, M. and Batterman, S.A. (2001). Characterization of Particulate Emissions from Occupant Activities in Offices. *Indoor Air*, 11(1), pp.35-48. Available at: http://dx.doi.org/10.1034/j.1600-0668.2001.011001035.x.

- Massey, D., Kulshrestha, A., Masih, J. and Taneja, A. (2012). Seasonal trends of PM_{10}, $PM_{5.0}$, $PM_{2.5}$ & $PM_{1.0}$ in indoor and outdoor environments of residential homes located in North-Central India. *Building and Environment*, 47, pp.223-231. Available at: http://dx.doi.org/10.1016/j.buildenv.2011.07.018.

- Mohammadyan, M. and Shabankhani, B. (2013). Indoor PM_1, $PM_{2.5}$, PM_{10} and Outdoor $PM_{2.5}$ Concentrations in Primary Schools in Sari, Iran. *Archives of Industrial Hygiene and Toxicology*, 64(3), pp.371-377.

- Nazaroff, W. W. (2004). Indoor particle dynamics. *Indoor air*, 14(s7), pp.175-183.

- Nielsen, G. D., Larsen, S. T., Olsen, O., Løvik, M., Poulsen, L. K., Glue, C. and Wolkoff, P. (2007). Do indoor chemicals promote development of airway allergy?. *Indoor air*, 17(3), pp.236-255.

- Ollier, S. J. (2013). Indoor and ambient particulate matter relationships during the heating season. Report, *Department of Geophysics, Faculty of Science, University of Zagreb*, 36pp. Available at: http://www.gfz.hr/eng/meteorologija/AQCT/PM_REPORT_SARAH_final.pdf.

- Pandis, S. N., Donahue, N. M. and Robinson, A. L. (2010). Atmospheric Organic Particulate Matter: Revising Its Sources, Properties and Impacts. In: Steyn, D. G and Rao, S.T. eds. *Air Pollution Modelling and its Application XX*. Dordrecht: Springer, pp. 429-437

- Pascal, M., Falq, G., Wagner, V., Chatignoux, E., Corso, M., Blanchard, M., Host, S., Pascal, L. and Larrieu, S. (2014). Short-term impacts of particulate matter (PM_{10}, $PM_{10–2.5}$, $PM_{2.5}$) on

mortality in nine French cities. *Atmospheric Environment*, 95, pp.175-184. Available at: http://dx.doi.org/10.1016/j.atmosenv.2014.06.030.

- Pérez, N., Pey, J., Querol, X., Alastuey, A., López, J. M. and Viana, M. (2008). Partitioning of major and trace components in PM10–PM2.5–PM1 at an urban site in Southern Europe. *Atmospheric Environment*, 42(8), pp.1677-1691.
  Available at: http://dx.doi.org/10.1016/j.atmosenv.2007.11.034.

- Perez, L., Tobías, A., Querol, X., Pey, J., Alastuey, A., Díaz, J. and Sunyer, J. (2012). Saharan dust, particulate matter and cause-specific mortality: A case–crossover study in Barcelona (Spain). *Environment International*, 48, pp.150-155.
  Available at: http://dx.doi.org/10.1016/j.envint.2012.07.001.

- Polichetti, G., Cocco, S., Spinali, A., Trimarco, V. and Nunziata, A. (2009). Effects of particulate matter (PM_{10}, $PM_{2.5}$ and PM_{1}) on the cardiovascular system. *Toxicology*, 261(1-2), pp.1-8. Available at: http://dx.doi.org/10.1016/j.tox.2009.04.035.

- Qian, J. , Ferro, A. R. and Fowler, K. R. (2008). Estimating the Resuspension Rate and Residence Time of Indoor Particles. *Journal of the Air and Waste Management Association*, 58:4, pp.502-516.

- Rovelli, S., Cattaneo, A., Nuzzi, C., Spinazzè, A., Piazza, S., Carrer, P. and Cavallo, D. M. (2014). Airborne Particulate Matter in School Classrooms of Northern Italy. *IJERPH*, 11(2), pp.1398-1421. Available at: http://dx.doi.org/10.3390/ijerph110201398.

- Salkind, N. J. (2000). *Exploring research*. 4th ed. Upper Saddle River, New Jersey: Prentice Hall.

- Silcox, G. D., Kelly, K. E., Crosman, E. T., Whiteman, C. D. and Allen, B. L. (2012). Wintertime $PM_{2.5}$ concentrations during persistent, multi-day cold-air pools in a mountain valley. *Atmospheric Environment,* 46, pp.17-24.
  Available at: http://dx.doi.org/10.1016/j.atmosenv.2011.10.041.

- Tasić, V., Kovačević, R., Milošević, N. (2011). Particulate matter (PM) indoor – outdoor relationships in buildings in Bor, Serbia. (Ban, M., Duic, N., Guzovic, Z. (Eds.)). *CD Proceedings of 6th. International Conference on Sustainable Development of Energy, Water and Environment Systems*, Faculty of Mechanical Engineering and Naval Architecture, Zagreb, 1-32. ISBN 978-953-7738-13-6

- Thatcher, T. L. and Layton, D. W. (1995). Deposition, resuspension, and penetration of particles within a residence. *Atmospheric Environment*, 29(13), pp.1487-1497.

- Titos, G., Lyamani, H., Pandolfi, M., Alastuey, A. and Alados-Arboledas, L. (2014). Identification of fine (PM_1) and coarse (PM_{10-1}) sources of particulate matter in an urban environment. *Atmospheric Environment*, 89, pp.593-602.
  Available at: http://dx.doi.org/10.1016/j.atmosenv.2014.03.001.

- Tobías, A., Pérez, L., Díaz, J., Linares, C., Pey, J., Alastruey, A. and Querol, X. (2011). Short-term effects of particulate matter on total mortality during Saharan dust outbreaks: A case-crossover analysis in Madrid (Spain). *Science of The Total Environment*, 412-413, pp.386-389. Available at: http://dx.doi.org/10.1016/j.scitotenv.2011.10.027.

- Toftum, J. (2010). Air pollution from residential wood combustion in a Danish village. Indoor-outdoor measurements. *International Centre for Indoor Environment and Energy. Department of Civil Engineering. Technical University of Denmark*. 18 pp. (also available at http://orbit.dtu.dk/fedora/objects/orbit:83078/datastreams/file_5208943/content)

- US-EPA (2004). Air Quality Criteria for Particulate Matter (Final report, Oct 2004). *US Environmental Protection Agency*, Washington, DC, EPA 600/P-99/002aF-bF.

- Van Drooge, B. L., Fernández, P., Grimalt, J. O., Stuchlík, E., Torres García, C. J. and Cuevas, E. (2010). Atmospheric polycyclic aromatic hydrocarbons in remote European and Atlantic sites located above the boundary mixing layer. *Environmental Science & Pollution Research*, 17(6), pp.1207-1216. Available at: http://dx.doi.org/10.1007/s11356-010-0296-0.

- Vecchi, R., Chiari, M., D'Alessandro, A., Fermo, P., Lucarelli, F., Mazzei, F., Nava, S., Piazzalunga, A., Prati, P., Silvani, F. and Valli, G. (2008). A mass closure and PMF source apportionment study on the sub-micron sized aerosol fraction at urban sites in Italy. *Atmospheric Environment*, 42(9), pp.2240-2253.
  Available at: http://dx.doi.org/10.1016/j.atmosenv.2007.11.039.

- Viana, M., Chi, X., Maenhaut, W., Querol, X., Alastuey, A., Mikuška, P. and Večeřa, Z. (2006). Organic and elemental carbon concentrations in carbonaceous aerosols during summer and winter sampling campaigns in Barcelona, Spain. *Atmospheric Environment*, 40(12), pp.2180-2193. Available at: http://dx.doi.org/10.1016/j.atmosenv.2005.12.001.

- Viana, M., Maenhaut, W., Chi, X., Querol, X. and Alastuey, A. (2007). Comparative chemical mass closure of fine and coarse aerosols at two sites in south and west Europe: Implications

for EU air pollution policies. *Atmospheric Environment*, 41(2), pp.315-326. Available at: http://dx.doi.org/10.1016/j.atmosenv.2006.08.010.

- Wheeler, A. J., Williams, I., Beaumont, R. A. and Hamilton, R. S. (2000). Characterisation of particulate matter sampled during a study of children's personal exposure to airborne particulate matter in a UK urban environment. *Environmental Monitoring and Assessment*, 65(1-2), pp.69--77.

- Whitby, K. T. (1978). The physical characteristics of sulfur aerosols. *Atmospheric Environment (1967)*, 12(1), pp.135-159.

- Wigley, T. (1989). Possible climate change due to SO_2-derived cloud condensation nuclei. *Nature Publishing Group*, 339, pp.365-367.

- Wilson, W. E., Chow, J. C., Claiborn, C., Fusheng, W., Engelbrecht, J. and Watson, J. G. (2002). Monitoring of particulate matter outdoors. *Chemosphere*, 49(9), pp.1009-1043.

- Wolkoff, P. (2013). Indoor air pollutants in office environments: Assessment of comfort, health, and performance. *International Journal of Hygiene and Environmental Health*, 216(4), pp.371-394. Available at: http://dx.doi.org/10.1016/j.ijheh.2012.08.001.

- Yin, J. and Harrison, R. M. (2008). Pragmatic mass closure study for $PM_{1.0}$, $PM_{2.5}$ and PM_{10} at roadside, urban background and rural sites. *Atmospheric Environment*, 42(5), pp.980-988. Available at: http://dx.doi.org/10.1016/j.atmosenv.2007.10.005.

- Zhang, L., Ninomiya, Y. and Yamashita, T. (2006). Formation of submicron particulate matter (PM1) during coal combustion and influence of reaction temperature. *Fuel*, 85(10-11), pp.1446-1457. Available at: http://dx.doi.org/10.1016/j.fuel.2006.01.009.