



Chemical Characterization of Indoor and Outdoor Particulate Matter (PM_{2.5}, PM₁₀) in Doha, Qatar

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ABSTRACT

The extreme weather conditions in Middle East Area led to the construction of tightly sealed, air conditioned buildings, characterized by indoor air quality deterioration. This study presents the results of chemical characterization of outdoor and indoor PM_{2.5} and PM₁₀ in Doha city, over a two-month period including normal days and dust events, aiming at identifying the factors affecting the indoor air of an office building. The WHO guideline values were exceeded in 100% of the outdoor measurements. 49% of the days of the sampling campaign were characterized as non-dusty (PM₁₀ < 200 µg m⁻³), 49% as minor-dusty (200 < PM₁₀ < 1000 µg m⁻³) while in one case (2%) there was a major-dusty day (PM₁₀ > 1000 µg m⁻³). The contribution of both dust and anthropogenic emissions sources is depicted in particles' mass and chemical composition. The elevated –especially outdoor- levels of carbonate carbon indicate the presence of crustal matter originating from the surrounding crustal material. OC/EC values reveal possible combined contribution from secondary organic aerosol, traffic-related sources and re-suspended dust. The influence of anthropogenic emissions is implied by the predominance of nitrate and sulfate ions, which constitute a substantial percentage of the particle mass. The crustal origin of particles is also depicted in metals. However, the higher enrichment factor values which may imply anthropogenic activities of both the outdoor and indoor environment were determined sequentially for Cd, Pb, As, Cu and Zn, suggesting the role of infiltration. Concluding, the indoor to outdoor relationship is significantly influenced by particles infiltration and penetration into the building mainly through the ventilation system and to a lesser extent, through windows or cracks in the building envelope. Although the low indoor to outdoor ratio underlies the predominance of outdoor levels compared to the indoor ones, there is positive correlation between indoor and outdoor PM, during the days that the building was open to the public and employees.

Keywords: Middle East; Dust events; I/O PM; PM chemical composition; Infiltration.

INTRODUCTION

Middle East Area (MEA) is characterized, excluding specific parts, by an arid, semi-arid or desert land that occupies the heart of world dry land, with its soil being consequently susceptible to Aeolian erosion (Tolba and Saab, 2008). The predominant atmospheric pollutant in MEA is airborne particulate matter (PM) (Tsiouri *et al.*, 2014). Wind-blown dust is a major air pollution source, with

transportation, energy production, industry and construction sources being also present (Kumar *et al.*, 2012a, b, Hansen *et al.*, 2008). Additionally, the severe dust storms that occur frequently in MEA contribute significantly to the large-scale transport of air pollution and particularly to the ambient PM pollution (Akbari, 2011; Saliba and Massoud, 2011; Al-Awadh, 2013). The chemical composition of particulate matter in the atmosphere of arid areas has been thoroughly studied during the last years (Shaltout *et al.*, 2013; Alghamdi *et al.*, 2015; Alhabri *et al.*, 2015; Habeebullah, 2016). Nevertheless, studies which simultaneously characterize the chemical composition of indoor and outdoor PM are limited; thus, further investigation on the field may shed valuable light on the role and contribution of airborne dust as a potentially significant source of air pollution.

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Although air quality in private or public indoor environments is progressively recognized as a critical issue for human health, comprehensive global data are still missing. While health risks from traditional household fuel combustion (Abadi *et al.*, 2017) tend to decrease globally, modern threats from airtight buildings (Ng *et al.*, 2015) as well as building materials (Zhang and Smith, 2003; Missia *et al.*, 2010; Xiong *et al.*, 2016) appear. Representative of the latter is the case of Middle East region, where the prevailing extreme weather conditions, led to the construction of a newly built environment including tightly sealed, air conditioned buildings. Such an environment can be characterized by indoor air quality deterioration due to pollutants' accumulation inside the building envelope. This issue becomes crucial for public health, since people spend approximately 95% of the day indoors -home, office, public buildings- (Klepeis *et al.*, 2001). During summer, the figure is bound to reach 98%, as outdoor average temperature is close to 50°C (Gevao *et al.*, 2006). Irrespective of this fact, there is no sufficient regulatory framework for indoor air pollution and associated health impact in MEA countries. There are only a few recent studies regarding indoor air quality assessment and its associated health risks in MEA and the Arab Gulf region, therefore there is a need for comprehensive studies especially in Qatar which is a MEA country undergoing rapid economic development.

One of the earliest studies was published by Jaradat *et al.* (2003), where heavy metals (Pb, Cd, Cu, Zn, Cr, Fe, and Al) and water-soluble anions (F⁻, Cl⁻, Br⁻, NO₃⁻) and cations (Li⁺, Na⁺, K⁺, Mg²⁺, and Ca²⁺) in indoor (office environments) and outdoor dust fall in the petroleum refinery area of Jordan were determined. Another early study regarding the influence of industrial sources on the level of total suspended particulate matter and heavy metals was conducted in Oman by Abdul-Wahab *et al.* (2004). The first research work on indoor particulate matter (PM₁₀) in Beirut -a city which experiences elevated outdoor PM levels- was conducted by Saliba *et al.* (2009) and indicated specific ionic tracers that infiltrate to indoor environments. A study focusing on the investigation of indoor PM_{2.5} levels in different house parts (living room, kitchen and bedroom) and on the assessment of PM_{2.5} indoor levels in different residential areas of Kuwait was conducted by Yassin *et al.* (2012) who highlighted the aggravated domestic air quality compared to that of houses in other countries. Another study that took place in Kuwait and Pakistan aimed at identifying the profiles of several classes of organic contaminants in indoor dust (Ali *et al.*, 2013) and in parallel, at estimating the exposure of toddlers and adults to these chemicals through dust ingestion. Recently, a cross-sectional population-based study of indoor air pollution and health conducted in the United Arab Emirates (UAE) was published by Yeatts *et al.* (2012), revealing a unique profile of indoor pollution sources in this region, including intense burning and cooking in gas stoves.

All the above evidences call for comprehensive studies focusing on air quality assessment inside modern, insulated buildings in MEA countries, characterized by extreme weather conditions, in order to obtain a clear and

comprehensive picture of the pollutant sources, contributing to the indoor microenvironment quality. To the best of the authors knowledge, recent monitoring studies providing information on the current pollution levels of areas with hot and arid climate such as Qatar (or MEA in general) are limited. For the indoor environment in particular, there is lack of indoor sources identification, consequently accompanied by insufficient mitigation actions. To fill this gap, the present study aims at the identification of factors affecting indoor PM levels and at the evaluation of the impact of outdoor environment on the indoor air of office buildings. In particular, the present work presents the results of the chemical characterization of outdoor and indoor PM_{2.5} and PM₁₀ in Doha city, over a two-month period including normal days and dust events. The outcome of this study would support decision making on environmental and health issues, in areas facing the natural dust phenomenon.

METHODS

Sampling Location

A monitoring campaign of PM measurements was performed at an office building (Ministry of Public Health), Al Rumaila West, Doha City, Qatar. Simultaneous indoor and outdoor PM (PM_{2.5}, PM₁₀) sampling took place from 22nd April to 21st June 2015. The duration of the campaign (two months in spring period) could be an important limitation in terms of comparison of measured concentrations with limit values (WHO guideline; EC Directive 2008/50/EC). However, this period can be considered as representative of the dusty period in Doha (<http://qweather.gov.qa>; Tsiouri *et al.*, 2014 and references within). The city of Doha is the capital of Qatar situated at the Doha bay with a population of more than 1,7 million people (2010 census; estimated to 2,5 million today). Temperatures range from 14°C to 22°C during winter, reaching 31°C to 42°C in summer period. Rainfalls are scarce with an average annual precipitation less than 70 mm. The building is located at the city of Doha (UTM 39R 551180 E 2797980 N) next to a very busy road, at a distance of 500 m from the beach front and at 18 m above the mean sea level. It has a rectangular shape (130 m long and 30 m wide) and seven floors; all used as office space and it is aligned along the NNW to SSE axis. The nearby area, which is representative of the whole metropolitan area, is characterized by intense construction activities and low rise buildings. The building is mechanically ventilated (8–10 air changes per hour (ACH) using 75%–90% fresh air and with minimum diurnal/weekly variations). Indoor conditions are very well regulated, temperature at 22.5°C ± 1°C and relative humidity at 51% ± 9%. It is also quite busy over the weekdays (Sunday to Thursday) and from 07:00 to 14:00, while it is closed to the public and employees during the weekend (Friday–Saturday).

For the purposes of the campaign, outdoor and indoor sampling sites were established. The outdoor site is located at the NNW fence line, ten meters from the narrow building facade. The indoor one is at the 3rd floor reception area, in the middle of the building length. The whole floor is carpeted and it includes two rows of offices and meeting

rooms, along the length of the building and one at each side, while in the middle there are open areas with office cubicles. The selected floor was considered representative of the whole building. The reception area is also quite busy over the day, it connects the office areas with the utilities rooms (restroom, prayer room and exits), but it is not directly exposed to the elevators area and staircase.

During the study period, the mean temperature ranged from 27°C to 40°C, no precipitation occurred, wind speed averaged at 5.1 m s⁻¹ with a maximum of ~15 m s⁻¹ (data collected for Hamad International Airport from the National Centers for Environmental Information, National Oceanic and Atmospheric Administration, US), and wind was blowing mainly from NNW (Figs. S1–S3). At the same period, Qatar Meteorological Department issued ten warnings for strong winds accompanied by medium or slight dust events (24–Apr; 2, 5, 23–May; 2, 4, 8, 13, 21–June)

Filter Sampling, Weighing and Chemical Analysis

PM mass sampling was carried out by four identical high volume (nominal flow 2.3 m³ h⁻¹, instantaneous value within 5% of nominal value) samplers (LVS16 by WB Engineering GmbH) over periods of 24 hours. Two samplers were equipped with a PM_{2.5} sampling head and the other two with a PM₁₀, all according to EN 12341. Each sampler was loaded periodically with seven 47 mm quartz fiber filters (Tissuequartz 2500QAT-UP by Pall Life Sciences) and filter replacement took place in the afternoons, during which all previous flow rates (average, standard deviation, minimum, and maximum) were extracted. Quartz filters are the only type of filter that can be used for further chemical analysis (OC/EC) and are recommended by EN12341:2014 as well. PM mass concentration gravimetric determination according to standard EN 12341:2014 is described in the supplementary material.

After sampling, conditioning and weighing, each filter was divided in four parts: a quarter for organic, elemental and carbonate carbon (OC, EC, CC) analysis, a quarter for ionic analysis and two quarters of the filters for elemental analysis. A detailed description of each analysis method can be found in the supplementary material.

Data Analysis

For the statistical analysis of the data, IBM SPSS Statistics 22.0 software has been used. As values of our data were continuous, normally distributed and a linear relationship was examined, the Pearson test was selected for the correlation analysis.

RESULTS AND DISCUSSION

PM Mass Concentration

PM mass concentration variation during the sampling campaign is shown in Figs. 1(a)–1(b). PM_{2.5} concentration ranged between 4.95 and 79.4 µg m⁻³ indoors (average 15.5 µg m⁻³) and between 35.8 and 1569 µg m⁻³ outdoors (average 172 µg m⁻³). Correspondingly, PM₁₀ ranged between 6.92 and 141 µg m⁻³ indoors (average 21.2 µg m⁻³) and between 108 and 1781 µg m⁻³ outdoors (average 267 µg m⁻³).

The particle mass concentration results were indicatively compared to the World Health Organization guideline values for the outdoor air (PM₁₀: 50 µg m⁻³ for 24-h average; 20 µg m⁻³ for annual average, PM_{2.5}: 25 µg m⁻³ for 24-h average; 10 µg m⁻³ for annual average) for a health risk assessment as well as to the European Commission Directive 2008/50/EC limits for ambient air (PM₁₀: 50 µg m⁻³ for 24-h average; 40 µg m⁻³ for annual average, PM_{2.5}: 25 µg m⁻³ for annual average). In particular, both daily limits for outdoor PM₁₀ (50.0 µg m⁻³) and PM_{2.5} (25.0 µg m⁻³) were exceeded in 100% of the outdoor measurements and in one and two cases of the indoor PM₁₀ and PM_{2.5} samples, respectively. An identification of dust events has been proposed by Draxler *et al.* (2001) and Al-Dabbous and Kumar (2014), with the ambient levels of PM₁₀ being < 200 µg m⁻³, laying between 200 and 1000 µg m⁻³ and being > 1000 µg m⁻³ respectively characterized as non-, minor- and major-dust periods. According to the aforementioned categorization and on the basis of PM₁₀ outdoor levels, 49% of the days during the sampling campaign are characterized as non-dusty and 49% as minor-dusty, while in single case (2%) a major-dusty day (May 4th). The average outdoor PM₁₀ concentration during these days was 165, 298 and 1781 µg m⁻³, respectively. Compared to similar studies performed in MEA, the average outdoor values found in the present study are quite similar to those (PM_{2.5} and PM₁₀) reported by Krasnov *et al.* (2015) during a dust period in Israel. Since Israel is also an area frequently impacted by dust storms, dust events can increase the daily levels of PM₁₀ in the center of city (Tel Aviv) to as high as 2100 µg m⁻³ (Ganor *et al.*, 2009; Kalderon-Asael *et al.*, 2009). Our average PM_{2.5} concentration was lower compared to the corresponding value (283 µg m⁻³) measured in Chengdu, China, during a dust storm (Wang *et al.*, 2013). On the contrary, our values were significantly higher when compared to PM levels recorded during dust storm days in Iran (highest mean outdoor PM₁₀ and PM_{2.5} concentrations reached 360 and 118 µg m⁻³ respectively, Hassanvand *et al.*, 2014) and Cyprus (highest mean outdoor PM₁₀ and PM_{2.5} values were 165 and 38 µg m⁻³ respectively, Achilleos *et al.*, 2016). It has to be noted that the comparison of this study's results with corresponding from literature, is only indicative as the dust storms occurring in several areas (e.g., Middle East, Asia, Mediterranean basin areas) are not expected to have similar characteristics (from the aspect of frequency, intensity, physicochemical parameters etc.). The maximum outdoor concentration for both PM_{2.5} (1569 µg m⁻³) and PM₁₀ (1781 µg m⁻³) was observed on May 4th, 2015, reaching over 60 and 30 fold values, compared to those of corresponding EU directives. Indoor PM₁₀ and PM_{2.5} levels followed the same trend, reaching their maximum levels (141 and 79.4 µg m⁻³ respectively).

A remarkable difference between weekdays (Sunday–Thursday) and weekends (Friday and Saturday) was observed only in the case of outdoor PM_{2.5} (on average 199 µg m⁻³ and 100 µg m⁻³, respectively). This can be attributed to the traffic loads, which are significantly lower during the weekend. The corresponding difference for PM₁₀ was lower than 10% (on average 275 µg m⁻³ and 245 µg m⁻³, respectively). On the

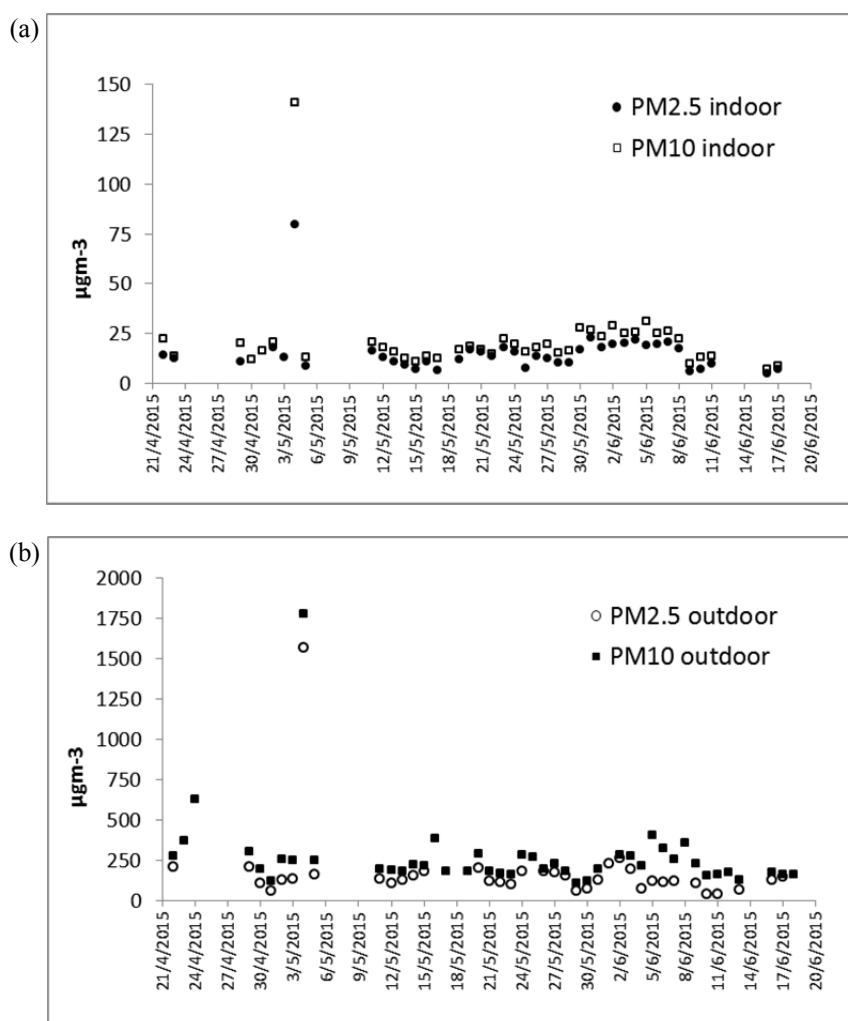


Fig. 1. PM_{2.5} and PM₁₀ mass concentration variation during the sampling campaign a) indoors b) outdoors.

other hand, a significant reduction in both indoor and outdoor levels has been recorded during the week before the Ramadan holiday (18th June–17th July, 2015). Specifically, indoor and outdoor PM_{2.5} demonstrated a 54% and 50% reduction during these days. Correspondingly, PM₁₀ levels diminished by 51% and 38% respectively.

The two particle size fractions examined (PM₁₀, PM_{2.5}) follow identical trends, presenting very strong correlation both indoors and outdoors (Pearson $r = 0.98$ for both), revealing their common origin. PM_{2.5}/PM₁₀ ratio was found to be 0.73 ± 0.12 indoors and 0.62 ± 0.18 outdoors. The outdoor PM_{2.5}/PM₁₀ ratio values are comparable to those found in other studies reporting dust events in Israel, Taiwan and Spain (values ranging between 0.5–0.6, Fang *et al.*, 2002; Querol *et al.*, 2009; Krasnov *et al.*, 2015). The outdoor ratio value presents a decrease during weekends and Ramadan days (PM_{2.5}/PM₁₀ = 0.54 and 0.51 respectively), possibly due to the reduced vehicles circulation in the area and the consequently lower concentration of fine particles (Vasilakos *et al.*, 2005).

Organic, Elemental and Carbonate Carbon Concentration

Carbonaceous species constitute major components of

atmospheric particles and are classified in three categories: elemental, organic and carbonate carbon. Elemental carbon (EC) is primarily produced by incomplete combustion. In contrast, organic carbon (OC), anthropogenic or biogenic, can be emitted either directly into the air or formed in the atmosphere through reactions of organic precursors with oxidative species and subsequent gas-to particle partitioning of the less volatile products. Carbonate carbon (CC), a primary carbonaceous species is present in natural ground and building materials demolition dust (Karanasiou *et al.*, 2011). However, CC concentrations may be significant in certain areas and/or under specific meteorological conditions such as soil dust outbreaks and severe dust storms episodes (Cao *et al.*, 2005; Sillanpaa *et al.*, 2005; Gerasopoulos *et al.*, 2006). OC, EC and CC concentration values for PM_{2.5} and PM₁₀ determined in the present study are presented in Table 1. According to the mass concentration trend, all carbon fractions (OC, EC and CC) were on average higher outdoors, for both PM_{2.5} and PM₁₀. The average OC and EC values were observed to be similar to those reported recently in Athens and Thessaloniki, Greece (Saraga *et al.*, 2015; Tolis *et al.*, 2015), Kuwait (Alolayan *et al.*, 2013) and Chengdu, China (Wang *et al.*, 2013).

Table 1. Carbonate fractions (OC, EC, CC) concentration in PM_{2.5} and PM₁₀.

	PM _{2.5} indoor ($\mu\text{g m}^{-3}$)			PM _{2.5} outdoor ($\mu\text{g m}^{-3}$)		
	OC	EC	CC	OC	EC	CC
average	3.29	0.97	0.38	7.57	2.83	4.82
median	3	0.91	0.18	7.36	2.63	3.13
min	2.16	0.45	0.1	3.83	1.28	0.38
max	8.04	1.76	2.69	32.2	5.27	48.1
stdev	1.24	0.33	0.64	4.45	0.9	7.49
	PM ₁₀ indoor ($\mu\text{g m}^{-3}$)			PM ₁₀ outdoor ($\mu\text{g m}^{-3}$)		
	OC	EC	CC	OC	EC	CC
average	3.49	0.98	0.51	9.62	2.91	7.55
median	3.19	0.96	0.3	8.65	2.72	6.34
min	2.21	0.23	0.11	4.4	0.66	1.15
max	8.39	1.88	4.81	32	5.73	43.2
stdev	1.26	0.36	0.84	4.47	1.03	6.64

Although carbonate carbon is generally not considered in many atmospheric chemistry studies due to its low contribution to the atmospheric PM mass concentration (Karanasiou *et al.*, 2011), its levels were recorded to be rather significant in the present study. Specifically, the CC concentration was recorded to be quite elevated, accounting on average for 29% and 34% of the total carbon concentration in PM_{2.5} and PM₁₀, respectively, reaching outdoor values similar to those in studies carried out during intense dust events. Specifically, CC -which is mainly present as CaCO₃- has been reported ranging between 9.8 and 29.2 $\mu\text{g m}^{-3}$ in Athens and between 0.3 and 6.3 $\mu\text{g m}^{-3}$ in Barcelona (Sillanpaa *et al.*, 2005). In another study (Querol *et al.*, 2009), African dust outbreaks resulted in a 30–40% contribution of CaCO₃ (in the form of carbonate carbon) to PM₁₀ concentration at two sites of the Eastern Mediterranean Basin. The CC levels observed in the present study, are also comparable to those reported in a study conducted at a dust source region (Loess Plateau, China), where the average level of CC in PM_{2.5} during five dust storms was 7.2 $\mu\text{g m}^{-3}$ (Cao *et al.*, 2005). Following the trend of aggravated outdoor mass concentrations and because of its direct association with natural ground matter (Karanasiou *et al.*, 2011 and references within), carbonate carbon was expected to be significant when referring to outdoor particles. Indeed, the outdoor average CC levels were significantly higher than the indoor ones (10 and 13 fold for PM_{2.5} and PM₁₀, respectively).

OC/EC ratio is commonly used to estimate the extent of secondary organic aerosol formation as well as to indicate the dominant fuel type for primary OC emissions. The results of the present study revealed very similar OC/EC ratio values in all cases: 3.5 and 3.1 for indoor and outdoor PM_{2.5}; 3.9 and 4.0 for indoor and outdoor PM₁₀. The same ratio value (3.2 \pm 0.3) for outdoor PM_{2.5} has been reported by Wang *et al.* (2013), referring to a dust storm period in Chengde, China. As reported in literature, OC/EC ratio values varying between 0.5–0.8 have been connected to heavy-duty diesel vehicles, between 1.3–4 to light-duty gasoline vehicles/traffic (Cao *et al.*, 2005; Pio *et al.*, 2011), while emissions from residential heating sources (biomass burning, wood combustion and natural gas home appliances) and road dust have been connected to OC/EC values ranging from

3.3 to 13.1 (Querol *et al.*, 2013; Saffari *et al.*, 2013). In the present study, the highest values of OC/EC ratio (8.4 and 22.4 for indoor and outdoor PM_{2.5}; 11.3 and 28.6 for indoor and outdoor PM₁₀) were observed during the incidents of aggravated PM mass concentration. On the other hand, the ratio values are reduced by 32–40% when calculated for the period of reduced vehicles circulation (Ramadan holidays and weekends), being equal to 2.4 and 2.0 for indoor and outdoor PM_{2.5}; 2.4 and 2.5 for indoor and outdoor PM₁₀, approaching the aforementioned values reported in literature, which correspond to secondary organic aerosol and traffic emissions (further information regarding fuel usage and motorization in Qatar are presented in Supplementary material). Therefore, the OC/EC values reveal a potential combined contribution from traffic-related sources and re-suspended dust, in absence of biomass burning in the area.

Ions Concentration

The average and standard deviation of ionic species in PM_{2.5} and PM₁₀ are summarized in Table 2. Among the measured ions, the most abundant in outdoor air were nitrates (54% for PM_{2.5}; 67% for PM₁₀) and sulfates (27% for PM_{2.5}; 17% for PM₁₀), followed by calcium (13% for PM_{2.5}; 9% for PM₁₀), ammonium (6% for PM_{2.5}; 3% for PM₁₀), chloride (2.5% for both) and the other ions measured (<2%). The outdoor average values of almost all ionic species were found to be elevated compared to those recorded in Thessaloniki (Voutsas *et al.*, 2014; Tolis *et al.*, 2015) and similar or higher of those detected in other Mediterranean cities in Italy, Spain, Greece, France, Lebanon (Saliba *et al.*, 2009; Salameh *et al.*, 2014). In literature, sulfate and nitrate anions are related to anthropogenic emissions (e.g., industrial units, vehicle emissions; Tsitouridou and Samara, 1993; Saliba *et al.*, 2009) and secondary inorganic ions formed mainly by the conversion of gaseous precursors (Wang *et al.*, 2013). A slightly different trend was followed in the case of indoor PM, while presenting lower concentration levels. In particular, among the measured ions, the most abundant were sulfates (40% for PM_{2.5}; 35% for PM₁₀) and nitrates (30% for PM_{2.5} and 35% for PM₁₀) followed by ammonium (22% for PM_{2.5}; 15% for PM₁₀), calcium (8% for PM_{2.5}; 10% for PM₁₀) and the rest of the ions measured.

Table 2. Cations and anions concentration in PM_{2.5} and PM₁₀.

average concentration ($\mu\text{g m}^{-3}$)	PM _{2.5} indoors	PM _{2.5} outdoors	PM ₁₀ indoors	PM ₁₀ outdoors
Na ⁺	0.2 ± 0.07	1.26 ± 0.94	0.29 ± 0.12	2.36 ± 1.55
NH ₄ ⁺	2.67 ± 1.54	3.35 ± 2.30	2.19 ± 1.64	2.8 ± 2.01
K ⁺	0.07 ± 0.04	0.23 ± 0.13	0.13 ± 0.29	0.33 ± 0.15
Mg ²⁺	0.03 ± 0.03	0.31 ± 0.15	0.04 ± 0.04	0.52 ± 0.22
Ca ²⁺	0.92 ± 0.73	7.42 ± 3.06	1.47 ± 0.97	9.97 ± 3.37
Cl ⁻	0.2 ± 0.09	1.4 ± 1.67	0.32 ± 0.27	2.75 ± 2.95
NO ₃ ⁻	3.68 ± 1.26	30.5 ± 20.3	5.02 ± 2.07	74.5 ± 36.4
SO ₄ ²⁻	4.82 ± 2.11	15.24 ± 6.15	5.07 ± 2.22	18.7 ± 7.62

The predominant sulfate, nitrate and ammonium ions in indoor air originate from the outdoor environment as they constitute components of aged atmospheric aerosol and are mostly related to large-scale regional transport processes rather than local primary emissions (Saraga *et al.*, 2015). Moreover, it has been found that when transferred from outdoor to indoor environments, nitrate levels are significantly decreased by more than 35% since nitrate is prone to dissipation, evaporation due to temperature changes and/or surface reactions indoors (Sawant *et al.*, 2004). However, it should be noted that differences in particle chemical composition exist even between ambient-generated particles that have infiltrated indoors and their corresponding ambient particles. As Diapouli *et al.* (2013) report, this may be attributed to the physical loss mechanisms influencing the infiltration of particles of different sizes, as well as chemical transformations affecting specific PM constituents, such as changes in gas-to-particle partitioning during the infiltration of organic compounds.

The ionic balance, as a mole equivalence, can be a useful tool for the determination of possible missing ionic species and the study of acid-base of aerosol particles. It is expressed by the ratio of the equivalent cation sum (sum of NH₄⁺, K⁺, Na⁺, Mg²⁺, Ca²⁺ in neq m⁻³) to the equivalent anion sum (the sum of NO₃⁻, Cl⁻, SO₄²⁻, PO₄³⁻ in neq m⁻³). The apparent alkaline character of the aerosols (excess of positive charge) indicates that CO₃²⁻ is most probably the missing anion which has not been measured using ionic chromatography and is expected to associate with Ca²⁺. For that purpose carbonate was calculated by manual integration on the thermograph from OC/EC analysis (Supplementary material) and was included in the sum of ions. On the other hand, the acidic character (excess of anion charge) is attributed to the presence of H⁺ (not measured) mainly associated with SO₄²⁻ (Kocak *et al.*, 2007). To check this possibility, the variation of cations deficit versus the SO₄²⁻ concentration is examined (Fig. S.4). The significant correlation ($r=0.89$) with slope close to unity (1.72), indicates that the cations deficit is most probably due to H⁺ associated with SO₄²⁻. In the current study, the ratio of the equivalent cation to anion sum is higher than unity indicating the alkaline character of indoor aerosols in both PM_{2.5} and PM₁₀ fractions (1.05 and 1.21 respectively). On the other hand, both fractions of outdoor aerosols demonstrate an acidic character (0.45 and 0.69 for PM₁₀ and PM_{2.5} respectively). The acidic character of outdoor dust PM is attributed to the significant higher NO₃⁻ and SO₄²⁻ outdoor concentrations,

compared to the corresponding indoor values, despite the simultaneous increased of outdoor Ca²⁺ concentration.

Metals Concentration

Chemical analysis of 17 metals (Al, As, Ba, Cd, Co, Cr, Cs, Cu, Fe, Ga, Mn, Ni, Pb, Rb, Sr, V, Zn) was performed for PM_{2.5} and PM₁₀ particles in both indoor and outdoor samples (Table 3). Among the elements measured, Fe and Al strongly dominate in PM_{2.5} of both indoor and outdoor air (Fe: 50.1% and 50.3%, Al: 39.0% and 43.0%, respectively), followed by Zn, Mn, V, Ba, Cu and Sr (Zn: 2.8% and 0.7%, Mn: 1.4% and 1.3%, V: 1.4% and 0.4%, Ba: 1.0% and 1.1%, Cu: 1.0% and 0.5%, Sr: 1.0% and 1.7%) and the rest of the metals analyzed, contributing less than 1%. Regarding PM₁₀ in indoor and outdoor air, the contribution of metals demonstrates a similar pattern, with the prevalence of Fe and Al (Fe: 50.4% and 51.3%, Al: 39.2% and 42.0%, respectively) being followed by Zn, Mn, V, Ba, Cu, Sr (Zn: 2.5% and 0.6%, Mn: 1.5% and 1.4%, V: 1.2% and 0.4%, Ba: 1.3% and 1.1%, Cu: 1.0% and 0.5%, Sr: 1.1% and 1.5%) and the rest of the metals analyzed contributing less than 1%. These findings indicate a similar elemental consistency of both PM_{2.5} and PM₁₀ indoor and outdoor particles, with the distribution of metals being independent of the particle size.

Al, Fe, Mn, Sr and Ba are classified among mineral components, Cu among traffic-related components (brake pad abrasion tracers), Ni and V among heavy fuel-oil combustion components, Pb and Co among mineral matter resuspension and metallurgical activities components, as well as among tracers of heavy oil combustion and Cd, Zn, Cr and As among industrial elements (Abdul-Wahab *et al.*, 2004; Rivas *et al.*, 2014). Among the metals studied, Ni and Cd are regulated through WHO guidelines (WHO, 2000) with a maximum value of 1000 ng m⁻³ for Ni and 5 ng m⁻³ for Cd. Pb is regulated by National Ambient Air Quality Standards with a value up to 150 ng m⁻³ (US EPA, 1997) and by the 1999/30/EC European legislation directive with an annual limit value of 500 ng m⁻³. Due to their negative impact on human health, the 2004/107/EC directive establishes the annual limits of 20, 5 and 6 ng m⁻³ for Ni, Cd and As respectively, in the PM₁₀ fraction. Considering toxic trace metals in outdoor air, the mean Ni outdoor concentration measured herewith in the PM₁₀ fraction exceeded by 83% the limit value established by EC but not the corresponding established by WHO. In 29 out of the 45 days of the whole study outdoor Ni concentrations exceeded

Table 3. Mean metals concentration and enrichment factors for PM_{2.5} and PM₁₀.

	Concentration (ng m ⁻³)				EF			
	PM _{2.5} indoors	PM _{2.5} outdoors	PM ₁₀ indoors	PM ₁₀ outdoors	PM _{2.5} indoors	PM _{2.5} outdoors	PM ₁₀ indoors	PM ₁₀ outdoors
Al	273	2876	327	4288				
Fe	351	3357	421	5165	2.57	2.34	2.57	2.41
Mn	9.96	88.8	12	138	3.14	2.66	3.17	2.78
Ba	7.31	72.2	11.2	115	6.45	6.06	8.25	6.47
Sr	6.83	111	9.26	165	5.54	8.59	6.26	8.51
V	9.26	28.6	10.1	35.7	17.4	5.09	15.8	4.27
Cr	4.3	19.8	4.76	39.3	12.9	5.64	11.9	7.52
Rb	0.46	3.64	0.57	5.41	1.53	1.15	1.57	1.15
Ni	4.55	21.1	5.23	36.6	17.1	7.52	16.4	8.75
Zn	19.5	45.8	25.2	63.8	78.2	17.4	84	16.3
Cu	6.92	31.86	8.89	53.7	41.5	18.2	44.5	20.5
Co	0.23	2.24	0.28	3.59	3.52	3.2	3.55	3.43
Ga	0.13	1.22	0.18	1.85	2.17	1.93	2.49	1.96
Pb	6.06	17.77	7.39	20.6	130	36.2	132	28.2
Cs	0.04	0.31	0.05	0.45	4.14	2.92	4.39	2.89
As	0.27	1.39	0.31	2.03	53.3	26.4	52.6	25.9
Cd	0.16	0.17	0.19	0.27	446	44.6	444	47.1

the imposed limits. The highest outdoor Ni concentration was recorded on May 4th, being equal to 507 ng m⁻³. The limit value for As established by EC was exceeded also on May 4th by As outdoor concentration in the PM₁₀ fraction (8.7 ng m⁻³). Considering toxic trace metals in indoor air, the limit values established by US-EPA, WHO and EC were not exceeded.

In order to define the most likely sources of indoor particles, enrichment factors were calculated for individual elements in terms of the average elemental composition of the upper continental crust. For each one of the examined metals, Al was used as reference, assuming minor contributions of Al as a potential pollutant and considering the upper continental crustal composition given by Rudnick and Gao (2003). The enrichment factor (EF) of an element *E* is defined according to Eq. (1):

$$EF = (E/R)_{\text{air}} / (E/R)_{\text{crust}} \quad (1)$$

where *E* and *R* represent the concentrations of the examined and the reference element, respectively. If EF approaches 1, then the earth's crust is the predominant source of the examined element. Operationally, given the local variation in soil composition, EF > 5 suggests that non-crustal sources contribute a significant fraction of the element (Gao *et al.*, 2002). As the EF value increases, the contribution from non-crustal sources demonstrates a similar tendency. Higher EF values which may imply anthropogenic activities of both the outdoor and indoor environment were determined sequentially for Cd, Pb, As, Cu and Zn, suggesting that in these cases, air infiltration affects indoor air quality. Regarding V, Cr and Ni, elevated values of EFs were only determined in indoor samples. In all metals under study, excluding Sr, higher EF values were obtained for both coarse and fine indoor particles, in comparison to outdoor ones, indicating an impact on metal concentrations by indoor

sources (Table 3). The latest appear to significantly affect the indoor concentrations of mainly Cd, Pb, Zn, As, Cu, V, Ni and Cr (Table 3). Since smoking is prohibited everywhere, it is indicated that the specific metals mostly derived from resuspension of dust due to indoor human activities, principally cleaning (dusting, washing and vacuuming), carried out almost every day, as well as the mobility of both personnel and visitors. The fact that the mean outdoor concentrations of these metals for both coarse and fine particles exceed the indoor ones (Table 3), indicates the significant contribution of air infiltration (mainly during days with HVAC system in operation) to the enrichment of the building indoor areas in metals (also supported by the low I/O ratios of the particular metals; Table S1). Other studies have also reported ratios higher than those expected, pointing to the importance of non-apparent indoor sources (Wallace, 1996). It has been demonstrated that some elements (i.e., Zn, Cu) are detectable in printer-emitted particles and toner powder (Pirela *et al.*, 2014). A relevant study regarding urban office areas in China has shown that serious outdoor pollution greatly affected all office environments studied, masking the original differences of indoor characteristics (Zhang *et al.*, 2017). The lack of differentiation of the calculated EF values of metals between coarse and fine particles indicates once more the presence of metal sources independent of the particle size.

Chemical Mass Closure

Chemical components of PM were grouped into ten classes as follows: organic, elemental and carbonate carbon (OC, EC, CC), non-sea salt sulfate (nss-SO₄²⁻), nitrate (NO₃⁻), ammonium (NH₄⁺), sea-salt, mineral dust, non-dust elements and unaccounted mass (Figs. 2(a)–2(d)). The chemical mass closure was calculated followed the methodology described in Salameh *et al.* (2015). The non-sea salt sulfate was calculated from the measured sulfate minus the sea-salt

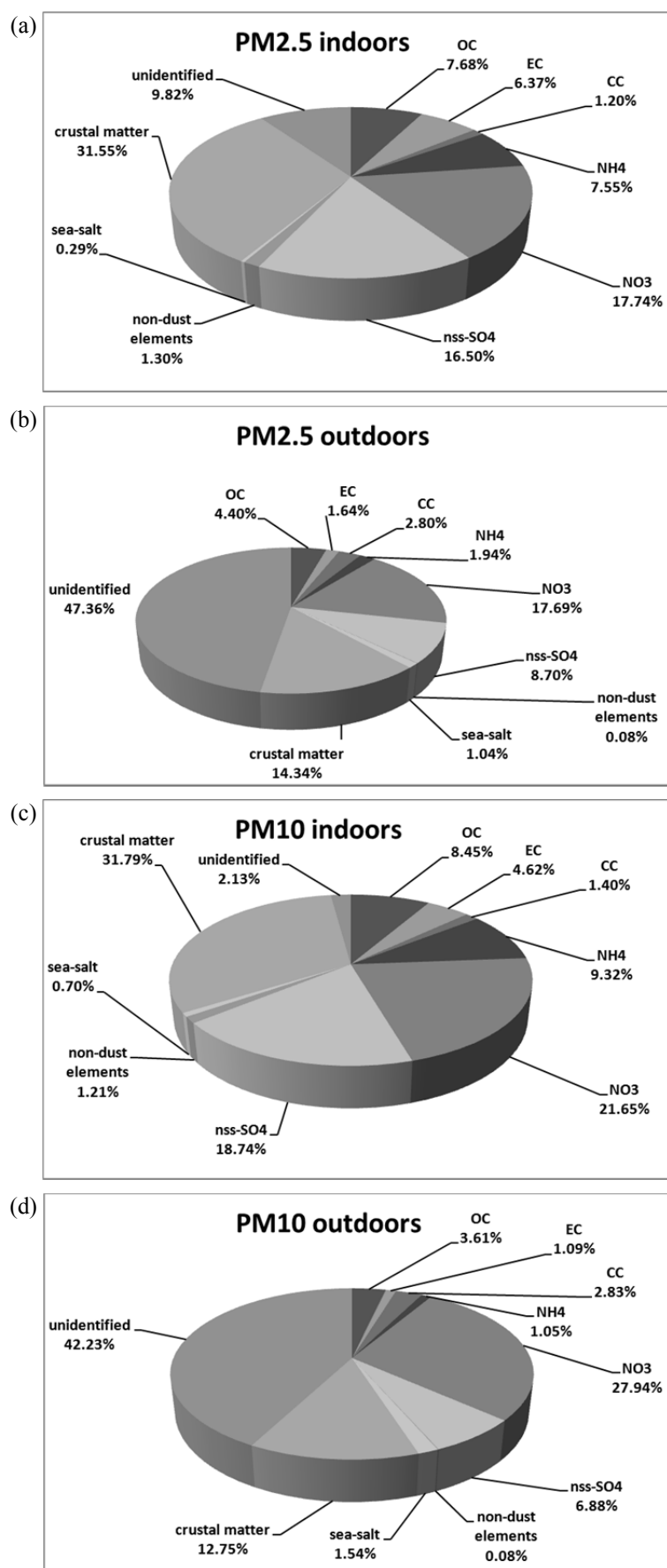


Fig. 2. % Chemical Mass Closure for a) indoor PM_{2.5} b) outdoor PM_{2.5} c) indoor PM₁₀ and d) outdoor PM₁₀.

fraction of SO_4^{2-} ($\text{ssSO}_4^{2-} = 0.252 \times \text{Na}^+$). Sea-salt concentrations were calculated from soluble sodium concentrations, assuming that sea-salt mass is equal to $3.252 \times \text{Na}^+$. Mineral dust was considered as the sum of Al_2O_3 , SiO_2 , CO_3^{2-} , Ca, Fe, K, Mg and Mn, where Al_2O_3 , SiO_2 and CO_3^{2-} were indirectly determined using empirical equations ($\text{Al}_2\text{O}_3 = 1.89 \times \text{Al}$; $\text{SiO}_2 = 3 \times \text{Al}_2\text{O}_3$; $\text{CO}_3^{2-} = 1.5 \times \text{Ca}$). Non-dust elements correspond to the sum of the common measured trace elements (i.e., Cu, Ni, Pb, V, Zn) other than geological ones. The unaccounted mass is obtained as the difference between measured PM mass concentration and the reconstructed mass (i.e., the sum of aerosol component concentrations). The predominance of crustal matter and ionic species is evident for both indoor fractions, followed by carbonate fractions, sea salt and non-dust metals. The unaccounted mass percentages are quite low indoors and can be attributed either to uncertainties in chemical analysis or to undetermined chemical species. In contrast, a different picture is presented for the outdoor $\text{PM}_{2.5}$ and PM_{10} composition. Crustal matter and ionic species remain the predominant species but approximately half of the mass proves to be unaccounted. The high percentage of the unidentified mass in both outdoor fractions is probably attributed to the high particle mass loading during sampling and the possible mass loss during the shipping of samples for the chemical analysis in the laboratory. This indicates that, in terms of mass loading sufficiency, a sampling period of 24 hours may not be appropriate during periods of high particle concentration in the outdoor air (in our case average $\text{PM}_{2.5} = 172 \mu\text{g m}^{-3}$; $\text{PM}_{10} = 267 \mu\text{g m}^{-3}$). This observation is amplified when mass percentage composition is calculated for non-dusty days (PM_{10} levels $< 200 \mu\text{g m}^{-3}$), minor dusty days ($200\text{--}1000 \mu\text{g m}^{-3}$) and major dusty days ($> 1000 \mu\text{g m}^{-3}$). Indeed, the unidentified outdoor PM_{10} mass percentage is 34% for non-dusty days, raises to 44% for minor dusty days and reaches even 80% for the major-dusty day of the campaign, confirming the mass loss in cases of high mass loading, which could be considered as a limitation of the study. In addition, the large unaccounted mass observed during dusty days could also be attributed to the formula used to calculate crustal material, suggesting the need for using site-specific dust compositional data for mass closure analysis.

Indoor to Outdoor Relationship

Indoor PM levels can be affected either by particles infiltration from outdoors into the building through ventilation, windows or cracks of the building envelope or by indoor emissions (Chen *et al.*, 2011; Halios *et al.*, 2013). The robust contribution of the outdoor air to the indoor PM mass concentration levels is implied by the strong positive correlation between indoor and outdoor concentrations of both $\text{PM}_{2.5}$ and PM_{10} (Pearson $r = 0.94$ and 0.97 , $p < 0.05$), during weekdays, when the ventilation system (HVAC) of the building was in operation. On the contrary, during weekends and Ramadan holiday (HVAC not in operation or working less hours, building closed to the public and employees), no correlation between indoor and outdoor levels was observed ($r < 0.3$).

This finding hints at the role of the HVAC system's operation, which can considerably influence the indoor particles' concentration. In contrast, when HVAC system is not in operation, infiltration of ambient particles indoors is expected to occur through cracks or other openings of the building envelope, which in the case of an insulated building are limited. Consequently, an average estimation of the infiltration factor has been attempted, using the correlation method described by Wilson *et al.* (2000) and Hanninen *et al.* (2004). Their methodology concludes in the equation:

$$C_i = C_{ia} + C_{ig} \quad (2)$$

where

$$C_{ia} = F_{inf} C_a \quad (3)$$

where C_i and C_a is the indoor, ambient concentration respectively, F_{inf} is the infiltration factor, C_{ia} is the fraction of outdoor particles infiltrated indoors and C_{ig} is the fraction of particles generated indoors. The parameters F_{inf} and C_{ig} can be solved from the regression of indoor concentration against the ambient concentration (the slope of the regression estimates the F_{inf} and the intercept the average concentration of indoor-generated PM (C_{ig})).

With the purpose of examining the infiltration in the absence of mechanical air exchange, data from the days without HVAC in operation were used. The results revealed quite low values of infiltration factor ($F_{inf} = 0.2$ for $\text{PM}_{2.5}$ and $F_{inf} = 0.1$ for PM_{10}), proving the effective insulation of the buildings. Correspondingly, the C_{ia} and C_{ig} fractions are $9.1 \mu\text{g m}^{-3}$ and $6.1 \mu\text{g m}^{-3}$ respectively for $\text{PM}_{2.5}$ and $8.4 \mu\text{g m}^{-3}$ and $12.8 \mu\text{g m}^{-3}$ respectively for PM_{10} . On the contrary, an average estimation of the infiltration factor during the days of the operation of HVAC, resulted in values of $F_{inf} = 0.9$ and $F_{inf} = 0.7$ for $\text{PM}_{2.5}$ and PM_{10} , respectively.

The relative impact of indoor and outdoor sources on indoor air is indicated by the indoor to outdoor concentration ratio (I/O). In terms of mass concentration, I/O ratio was found to be significantly lower than unity (0.12 ± 0.07 for $\text{PM}_{2.5}$ and 0.09 ± 0.04 for PM_{10}) underlying the predominance of outdoor particle levels compared to indoor ones.

The aforementioned indexes (r , I/O) have been also calculated for PM chemical components (Table S1). Similarly to mass concentration, I/O average values are significantly lower than unity for all carbon fractions as well as for all ions and metals (except for NH_4^+ and Cd where the ratios are close to unity). The similar I/O ratios calculated for fine and coarse particles indicate sources independent of the particle size.

Pearson correlation analysis performed among indoor and outdoor OC, EC and CC concentrations revealed the strong outdoor atmosphere's contribution to indoor levels, for all carbonate fractions, except for the case of organic carbon in PM_{10} (Table S1). A different picture is derived for anions and cations, where moderate or strong correlation ($r > 0.6$) was observed only in the cases of ammonium, magnesium, calcium and sodium, chloride (in $\text{PM}_{2.5}$) and sulfate (in PM_{10}). Regarding metals, Co, Rb, Ga and Al showed a very strong

correlation ($r > 0.80$) between indoor and outdoor concentrations in both $PM_{2.5}$ and PM_{10} fractions, whereas Mn, Cs, Fe and Sr showed a very strong correlation in PM_{10} fraction and a strong correlation ($r = 0.60$ – 0.79) in $PM_{2.5}$. V in both fractions and As in PM_{10} showed a strong correlation between indoor and outdoor concentrations, whereas Cr and Ba in both fractions and As in $PM_{2.5}$ demonstrated a moderate one ($r = 0.40$ – 0.59) (Table S1). These findings suggest that, regardless of the effects of indoor sources of these metals, a rather significant fraction of their indoor concentrations is attributed to infiltration of outdoor particles (mainly through ventilation). Previous studies have indicated that areas in the vicinity of urban and industrial sites are characterized by higher metal concentration in indoor dust, compared to rural and village ones. The metal concentration in indoor dust was altered by the building's dustiness and ventilation pattern (Tan *et al.*, 2016).

CONCLUSIONS

The present work aims at the presentation of the chemical characterization of outdoor and indoor $PM_{2.5}$ and PM_{10} collected in Doha city, Qatar, an area for which recent monitoring studies providing information of the current pollution levels are limited. Further, it aims at the identification of the factors affecting indoor PM levels and at the evaluation of the impact of outdoor environment on the indoor air of office buildings. The results highlighted the effect of dust on outdoor PM concentration, which in 100% of the sampling days exceeded the European Commission limit and WHO guideline values. Specifically, 49% of the sampling days are characterized as non-dusty ($PM_{10} < 200 \mu\text{g m}^{-3}$), 49% as minor-dusty ($200 < PM_{10} < 1000 \mu\text{g m}^{-3}$), while a major-dusty day ($PM_{10} > 1000 \mu\text{g m}^{-3}$) was also recorded (2%). Indoor $PM_{2.5}$ and PM_{10} levels seem to follow the outdoor pattern only during working days, when the building is open to the public and the ventilation system is in operation. In contrast, during weekends and other days of reduced activity (close to the Ramadan holiday), both outdoor and indoor levels were significantly lower, possibly due to reduced traffic loading in the area.

The contribution of dust and anthropogenic emissions sources is also depicted in the chemical composition of particles. Regarding carbonaceous components, the remarkable levels of carbonate carbon indicates the presence of crustal matter originating from the surrounding crustal material (mainly characterized as calcisols: soil based on Ca^+ and Mg^+ carbonates). Further, OC/EC values reveal possible combined contribution from secondary organic aerosols, traffic-related sources and re-suspended dust. The influence of anthropogenic emissions on particles is implied by the predominance of nitrate and sulfate ions, which constitute a substantial percentage of the particle mass. Finally, the crustal origin of particles is also depicted in metals. However, the higher enrichment factor (EF) values which may imply anthropogenic activities of both the outdoor and indoor environment were determined sequentially for Cd, Pb, As, Cu and Zn, suggesting that in these cases air infiltration affects indoor air quality.

Although the indoor to outdoor ratio is very low, underlying the predominance of outdoor levels compared to indoor ones, in the cases of not isolated building (working days, HVAC in operation), a positive correlation between indoor and outdoor PM mass and components concentration was noticed. All the above hint at the role of the HVAC system's operation, which considerably influences the indoor particles' concentration. In contrast, in absence of mechanical ventilation, the infiltration factor is low as it is expected to occur through cracks or other openings of the building envelope, which in the case of this –insulated–building, are limited.

To conclude, the indoor PM mass concentration and composition is highly influenced by the outdoor levels as a result of infiltration mainly through the HVAC system operation. Nevertheless, the contribution from indoor sources which influence the accumulated re-suspended indoor particles cannot be excluded.

The outcome of this study would support decision making on environmental and health issues in areas facing the natural dust phenomenon. For instance, an open-access monitoring network could be established so as to alert the public and related authorities about the short-term air pollution episodes. Future research should include a source apportionment approach with the use of state-of-the-art receptor models aiming at the estimation of the contribution of main sources to indoor and outdoor particle levels. On the top of that, we consider that within a modern building, the combination of the control of the physical parameters (ventilation, temperature, relative humidity) together with an early warning system which notifies the building's occupants about the indoor microenvironment air quality could significantly protect their health, wellness and comfort.

ACKNOWLEDGMENTS

The present study has been supported by NPRP award number NPRP 7-649-2-241 from the Qatar National Research Fund (a member of The Qatar Foundation). The authors would also like to acknowledge the support (contracts number SCH/PA/14/2015 and SCH/PA/18/2015) and assistance from the Ministry of Public Health, State of Qatar. The statements made herein are solely the responsibility of the authors.

DISCLAIMER

Reference to any companies or specific commercial products does not constitute financial and personal conflict of interests.

SUPPLEMENTARY MATERIAL

Supplementary data associated with this article can be found in the online version at <http://www.aaqr.org>.

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Received for review, May 16, 2016

Revised, April 18, 2017

Accepted, April 19, 2017