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Indoor residential and outdoor sources of $PM_{2.5}$ and PM_{10} in Nicosia, Cyprus

Yichen Wang[1] · Petros Koutrakis[1] · Antonis Michanikou[2] · Panayiotis Kouis[2] · Andrie G. Panayiotou[3] · Paraskevi Kinni[2,3,4] · Filippos Tymvios[5] · Andreas Chrysanthou[5] · Marina Neophytou[6] · Petros Mouzourides[6] · Chrysanthos Savvides[7] · Emily Vasiliadou[7] · Ilias Papasavvas[8] · Theodoros Christophides[8] · Rozalia Nicolaou[8] · Panayiotis Avraamides[8] · Choong-Min Kang[1] · Stefania I. Papatheodorou[3,9] · Nicos Middleton[4] · Panayiotis K. Yiallouros[2] · Souzana Achilleos[3,10]

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Abstract

Cyprus is a typical eastern Mediterranean country that suffers from local emissions, transported anthropogenic pollution, and dust storms all year round. Therefore, exposures to PM in ambient and residential micro-environments are of great public health concern. Our study collected indoor and outdoor $PM_{2.5}$ and PM_{10} samples simultaneously in 22 houses in Nicosia, Cyprus, during warm seasons and cold seasons from February 2019 to May 2021. Samples were analyzed for mass and constituents' concentrations. To determine indoor and outdoor sources of PM in residential environments, we used the EPA positive matrix factorization (PMF) model to conduct source apportionment analyses for both indoor and outdoor $PM_{2.5}$ and PM_{10} particles. Generally, six types of residential-level PM sources were resolved: biomass burning, traffic, local or regional secondary sulfate pollution, Ca-rich particles, sea salt, and soil dust. In the source apportionment of $PM_{2.5}$, the main contribution to outdoor levels (33.1%) was associated with sulfate-rich transported pollution. The predominant contribution to indoor levels (48.0%) was attributed to secondary sulfate pollution as a mixture of local- and regional-scale pollutants. Biomass burning and traffic sources constituted the main outdoor sources of indoor $PM_{2.5}$, while the Ca-rich particles were identified to almost originate from indoors. By contrast, the largest fraction (29.3%) of the ambient PM_{10} and a smaller proportion (10.2%) of indoor PM_{10} were attributed to Ca-rich particles. Indoor PM_{10} was associated mainly with outdoor sources, except for the soil dust which originated from indoor activities.

Keywords Source apportionment · PMF · Particulate matter · Eastern Mediterranean · Indoor air quality

✉ Yichen Wang
yichenwang@hsph.harvard.edu

[1] Department of Environmental Health, Harvard T.H. Chan School of Public Health, 401 Park Drive, Boston, MA 02215, USA

[2] Respiratory Physiology Laboratory, Medical School, University of Cyprus, Nicosia, Cyprus

[3] Cyprus International Institute for Environmental and Public Health, School of Health Sciences, Cyprus University of Technology, Limassol, Cyprus

[4] Department of Nursing, School of Health Sciences, Cyprus University of Technology, Limassol, Cyprus

[5] Cyprus Department of Meteorology, Nicosia, Cyprus

[6] Environmental Fluid Mechanics Laboratory, Department of Civil and Environmental Engineering, University of Cyprus, Nicosia, Cyprus

[7] Air Quality and Strategic Planning Section, Department of Labour Inspection, Ministry of Labour and Social Insurance, Nicosia, Cyprus

[8] Department of Cardiology, Nicosia General Hospital, Nicosia, Cyprus

[9] Department of Epidemiology, Harvard T.H. Chan School of Public Health, Harvard University, Boston, MA, USA

[10] Department of Primary Care and Population Health, University of Nicosia Medical School, Nicosia, Cyprus

Introduction

About 99% of the world population is faced with poor air quality that exceeds the World Health Organization (WHO) limits and is subject to health threats (WHO 2022). Particulate matter (PM) is a type of air pollutant that affects more people than others and is responsible for almost 9 million deaths every year worldwide (Burnett et al. 2018). A large number of toxicological and epidemiological studies have shown a range of adverse health outcomes attributed to short- and long-term exposure to PM with aerodynamic diameter ≤10 μm (PM_{10}) and/or 2.5 μm ($PM_{2.5}$), such as premature mortality, cardiovascular and respiratory diseases, and neurological disorders (Delfino et al. 2008; Brook et al. 2010; Ristovski et al. 2012; North et al. 2018; Kyung and Jeong 2020; Shi et al. 2020; Bu et al. 2021; Grande et al. 2021).

Ambient PM_{10} originates mainly from industrial and traffic emissions, crustal minerals, sea salt, and biologically derived materials. Of a smaller particle size, $PM_{2.5}$ is particularly hazardous to human health as it is able to penetrate deep into the lungs. In general, $PM_{2.5}$ is considered more toxic than PM_{10} on an equal-mass basis due to its long residence in the air and deeper penetration inside the lungs (Harrison et al. 2017; Memhood et al. 2018; Zhao et al. 2019). Experimental evidence suggests that $PM_{2.5}$ may have higher cytotoxicity and cause more harm to human through oxidative stress than PM_{10} (Choi et al. 2019). Outdoor $PM_{2.5}$ compositions arise from various sources such as fossil fuel combustion, biofuel combustion, and biomass burning, as well as naturally derived dust (Philip et al. 2014; Thangavel et al. 2022). Secondary particles generated by chemical reactions between precursor gases (i.e., ammonia, sulfuric acids, and nitric acids) are mainly concentrated in the $PM_{2.5}$ fraction (Harrison et al. 1997). PM composition is a major important determinant of PM toxicity (Kelly and Fussell 2012). For example, endotoxin, a component of gram-negative bacteria's cells, has been associated with proinflammatory effects (Dong et al. 1996; Donaldson and MacNee 2001; Donaldson et al. 2003). Black carbon is a byproduct of the incomplete combustion of fossil fuel and biomass burning (Goldberg 1985), which is associated with adverse health effects and reduced life expectancy (Roemer and van Wijnen 2001; Janssen et al. 2011; Grahame et al. 2014). Heavy metals (e.g., zinc, nickel, vanadium), originating from industrial activities, have shown a strong relationship with cardiopulmonary morbidity and mortality (Ostro et al. 2007; Bell et al. 2009; Badaloni et al. 2017).

Nowadays, people spend approximately 90% of their time in indoor environments, while the most susceptible population (i.e., the elderly, children, and people with pre-existing conditions) may spend even more. Indoor PM levels experience high variability across microenvironments and seasons, and that depends on indoor sources and activity patterns, building envelope, weather conditions, and ventilation (Tan et al. 2013). There is a considerable amount of infiltrated outdoor pollution constituting indoor PM due to air exchange. Even under poor ventilation conditions, exhaust and non-exhaust traffic tracers, and biomass burning-related species could penetrate indoors (Tofful et al. 2021). Meanwhile, indoor activities of inhabitants serve as other primary origins of indoor airborne particles, including cooking (Jeong et al. 2019; Kim et al. 2020), smoking (Ni et al. 2020), cleaning (Zhao et al. 2006), heating (Zhu et al. 2012), candle or incense burning (Bootdee et al. 2016), and electric appliances equipped with motors (Szymczak et al. 2007; Tofful et al. 2021). The movement of occupants at home could also enhance the resuspension of indoor PM, which mainly elevates the levels of particles with size up to 10 μm (Qian et al. 2014). The potential detrimental outcomes of indoor human activities have been proposed these years. For example, the combustion of domestic solid fuels for cooking can also trigger respiratory effects and further increase the risk of stroke and cardiovascular diseases (Kilabuko et al. 2007; Fatmi and Coggon 2016; Guercio et al. 2022). Polycyclic aromatic hydrocarbons and carbonyls derived from burning of tobacco and incense smoke have been demonstrated to trigger oxidative-DNA damage and inflammatory reactions in human respiratory systems (Friborg et al. 2008; Niu et al. 2021).

Like other countries in the Eastern Mediterranean basin, Cyprus experiences high PM levels due to local (e.g., traffic emissions, biomass burning, resuspended dust) and long-range transported air pollution from Africa, Europe, and Asia (Achilleos et al. 2016, 2020; Lelieveld et al. 2002). The unique dry Mediterranean climate further prevents the washout of PM and exacerbates the pollution level to some extent, contributing to the great challenge of meeting the EU limit values (50 $\mu g/m^3$ PM_{10} for the 24-h mean) (Bari et al. 2009; Mouzourides et al. 2015; Pikridas et al. 2018; Achilleos et al. 2020). Several research teams have investigated the potential contributors of PM in Cyprus (Achilleos et al. 2014, 2016, 2020; Konstantinou et al. 2022; Mouzourides et al. 2015; Pikridas et al. 2018). However, the knowledge regarding the composition and sources of PM in Cyprus remains rather limited. In addition, to the best of our knowledge, no previous studies have investigated the sources of residential indoor PM in Cyprus until now. Still, little is also known about sources of indoor particles in the Eastern Mediterranean area in general, limiting the information to enact effective PM mitigation strategies. To fill the research gap, our study aims to provide a detailed characterization of indoor and outdoor PM composition and to demonstrate for the first time their profiles and

sources utilizing data collected from 22 homes in Nicosia, Cyprus. Moreover, we also intend to investigate the indoor-outdoor relationships of sources.

Materials and methods

Study design

This study is a part of the MEDEA (Mitigating the Health Effects of Desert Dust Storms Using Exposure-Reduction Approaches) project co-funded by the European Union's LIFE program under Grant Agreement LIFE16 CCA/CY/000041, aiming to provide effective and sustainable recommendations for exposure reduction during desert dust episodes. We used PM data sampled during dust storm days and non-dust storm days in warm seasons (February-June) and cold seasons (September-December) from 2019 to 2021, from 22 participants' houses in Cyprus, where six houses were occupied by participating schoolchildren with asthma and 16 houses were occupied by participating adults with atrial fibrillation. More specifications and details about the study design have been documented before (Kouis et al. 2021).

$PM_{2.5}$ and PM_{10} samples were collected inside and outside of these houses simultaneously. The samples were collected on Teflon filters (Gelman Sciences 47-mm) by using Harvard particle samplers (Harvard High Volume Cascade Impactors, Harvard University, USA) at a sampling flow rate of 5 L/min (Marple et al. 1987). Particles with aerodynamic diameters exceeding 10 microns and 2.5 microns were efficiently removed by the impaction substrate made of polyurethane foam on a slit acceleration jet, respectively (Lee et al. 2005). The indoor samplers were positioned on a table in a primary activity room (generally in a living room), and the outdoor samplers were placed at least 3 m away from any vertical exterior wall of the houses. Samples were collected during dust storm days (for as many hours the event lasted, n=15) and non-dust days (7-day sample, n=20).

Our dataset included 91 indoor PM_{10}, 91 outdoor PM_{10}, 89 indoor $PM_{2.5}$, and 90 outdoor $PM_{2.5}$ samples. For the scope of this study, the samples were analyzed for mass, black (BC) and brown (BrC, ultraviolent absorbing particulate matter) carbon, and trace elements including sodium (Na), magnesium (Mg), aluminum (Al), silicon (Si), sulfur (S), chlorine (Cl), potassium (K), calcium (Ca), titanium (Ti), vanadium (V), iron (Fe), zinc (Zn), bromine (Br), lead (Pb), chromium (Cr), manganese (Mn), and strontium (Sr).

For each $PM_{2.5}$ and PM_{10} filter sample, the concentrations of PM mass, BC, BrC, and trace elements were determined using specific methods. Following the 48-h equilibration in a room of controlled temperature (20–22 °C) and relative humidity (40±5%), the PM mass concentration was determined by weighting the Teflon filters using an electronic microbalance (MT-5 Mettler Toledo, Columbus, OH). The BC and BrC concentrations were measured using the SootScan Optical Transmissometer (Model: OT21, Magee Scientific). BC was measured by the optical attenuation of an aerosol-loaded filter (the sample) and a reference filter at a wavelength of 880 nm, while BrC was measured at a wavelength of 370 nm, which may indicate the presence of BrC aerosol derived from biomass combustion. To derive the elemental composition of PM, the filters were analyzed using the high-sensitivity X-ray fluorescence (XRF) spectrometer (XRF, Model Epsilon 5, PANalytical, The Netherlands). These measurements were blank-corrected. All laboratory analyses were carried out at the Environmental Chemistry Laboratory, Harvard T.H. Chan School of Public Health. A more detailed description on the PM sampling and analysis can be found elsewhere (Achilleos et al. 2023).

Information on building characteristics, indoor regular activity patterns, and neighborhood environments in participants' homes was obtained from occupant questionnaires administrated at the time of enrollment into this study. The asked questions ranged from basic building features and nearby traffic conditions to utility energy, cleaning frequency, ventilation, etc.

Data analysis

Ratio analysis

The $PM_{2.5}/PM_{10}$ mass and elemental ratios were calculated focusing on identifying the predominant mode for mass and each element as well as inferring the particle formation processes. The $PM_{2.5}/PM_{10}$ ratio is given as follows:

$$\left(\frac{PM_{ij2.5}}{PM_{ij10}}\right)_{IN} = \frac{C_{ij2.5IN}}{C_{ij10IN}} \quad (1)$$

$$\left(\frac{PM_{ij2.5}}{PM_{ij10}}\right)_{OUT} = \frac{C_{ij2.5OUT}}{C_{ij10OUT}} \quad (2)$$

where $C_{ij2.5IN}$ and C_{ij10IN} are the indoor concentrations of PM mass or species j in sample i; $C_{ij2.5OUT}$ and $C_{ij10OUT}$ are the outdoor concentrations of PM mass or species j in sample i.

In the examinations of the normality of data, the Shapiro-Wilks tests indicated significant deviations from the normal distributions in each group of the above ratios. Therefore, we applied the paired Wilcoxon signed-rank tests to examine the presence of statistically significant differences between median indoor and outdoor $PM_{2.5}/PM_{10}$ ratios. A two-sided p-value $\leq$0.05 indicates a statistically significant level.

Positive matrix factorization analyses

We employed the US Environmental Protection Agency Positive Matrix Factorization (EPA PMF 5.0) model to conduct source identification analyses and determine the chemical profiles of potential sources of residential indoor and ambient $PM_{2.5}$ and PM_{10}. Positive matrix factorization (PMF) is a widely used receptor modeling method that resolves the source profiles based on observations of PM species (Paatero and Tapper 1994). Its mathematical expression is:

$$x_{ij} = \sum_{k=1}^{p} g_{ik} f_{kj} + e_{ij} \quad (4)$$

where x_{ij} is a data matrix of the concentration of species j in sample i, p is the number of factors apportioned to the samples, g_{ik} is the score of factor k responsible for sample i, f_{kj} is the loadings of species j in factor k, and e_{ij} is the error estimate of species j in sample i. PMF model estimates the unknown factor contributions (G) and profiles (F) by minimizing the function Q, which is defined as:

$$Q = \sum_{i=1}^{n} \sum_{j=1}^{m} \left(\frac{e_{ij}}{u_{ij}} \right)^2 \quad (5)$$

where e_{ij} and u_{ij} are estimated error and uncertainty of species j in sample i, respectively; n is the number of samples; m is the number of species. In the PMF analyses, the uncertainties of PM elements were calculated based on the equation $u_{ij}=s_{ij}+DL_{ij}/3$ (Reff et al. 2007), where s_{ij} is the analytical uncertainty and DL_{ij} is the method detection limit (DL) for each PM sample i and species j.

Before executing the models, we excluded the elements with more than 50% of their concentrations below the DL. We also distinguished the species that maintain a significant signal from those predominated by noise judging from the signal-to-noise ratio (S/N), a measure of uncertainty and data quality. Species with $S/N<0.2$ were categorized as "bad" and excluded from the analysis; species with $0.2 \leq S/N<2$ were defined as "weak" and their uncertainties were tripled; $S/N \geq 2$ were labeled as "strong" species and stayed the same. Exceptions to this criterion were decided to increase the goodness-of-fit and guarantee stable solutions: Na for indoor and outdoor PM_{10} samples was recategorized from bad to weak species and Cl for outdoor $PM_{2.5}$ samples was recategorized from weak to strong species. The mass concentration was specified as a total variable in the PMF program and its uncertainty values were automatically tripled. Due to the limited number of samples, we used the data both on the dust days and non-dust days together. We identified and excluded extreme events from the abnormal peaks of PM mass or species that may lead to false and erratic source profiles. To avoid introducing bias, zero or below DL concentrations remained the same in the analysis without any manipulations (Paatero 1997). The percent contribution of each source to the total PM mass was derived from PMF factor contributions output.

The optimal number of contributing sources for the PM was chosen from a range of 4 to 7 based on a comprehensive consideration of the goodness of model fit, error estimation, and interpretability of solutions. The models were validated from the Q values (Q_{true}, Q_{robust}, $Q_{expected}$), bootstrap analysis, displacement analysis, and bootstrap-displacement analysis. The processes of model optimization are provided in supplementary materials (SI 1; Table S1 and S2; Fig. S1).

Lastly, for the common sources of residential indoor and outdoor PM, the indoor/outdoor (I/O) ratios of source contributions were further computed, which gives complementary information to the PM species ratio analyses.

The rest of data analyses were performed in R statistical software, version 4.1.2 (R Foundation for Statistical Computing, Vienna, Austria).

Results and discussion

Home characteristics

The summary of home characteristics is presented in Table 1. About half of the houses were located near main streets and most of them were at crossroads. Eight out of 22 houses reported to have at least one source of dust (e.g., construction, industry, commercial garage) located within 100 m of the dwelling. Information on heating fuels indicated that 50.0% of the houses used electricity or electric heater, while 27.3% used central oil or oil heater and 36.4% utilized natural gas or gas heater. Approximately half of the houses had scented candles or incense indoors, while the minority of the houses had at least one fireplace (n=4). According to the indoor recommendations formulated by the MEDEA clinical intervention project, the participants (n=8) assigned into the intervention group were urged to use air cleaners (AP-1516D, Coway, Korea) with HEPA (High Efficiency Particulate Air) filter.

Indoor and outdoor $PM_{2.5}$ and PM_{10} concentrations

Table 2 gives the descriptive statistics of indoor and outdoor concentrations of $PM_{2.5}$ and PM_{10} mass and their respective constituents. In the participating households in Cyprus, the average outdoor concentration of $PM_{2.5}$ (15.1±6.2 μg/m^3) was only 9.4% higher than its average indoor concentration (13.8±8.7 μg/m^3) while the distinguishment between indoor and outdoor levels for PM_{10} mass was more pronounced (24.6±19.4 μg/m^3 and 30.4±10.8 μg/m^3, respectively). A tendency of higher levels for ambient $PM_{2.5}$ and

Table 1 Summary for house information and indoor activity patterns

Activity	Main street	Crossroad	Any sources of dust	Parking lots	Usual use of vacuum cleaner	Scented candles or incense	Central oil/oil space heater	Central electricity/ electric space heater	Central natural gas/ gas space heater	Fireplace
Frequency (*n*)	12	18	8	16	13	10	6	11	8	4
Percentage (%)	54.5	81.8	36.4	72.7	59.1	45.5	27.3	50.0	36.4	18.2

PM_{10} in Southern Europe ($PM_{2.5}$: 14.7–29.3 μg/m^3, PM_{10}: 35.6–43.1 μg/m^3) has been reported as compared to Western/Central Europe ($PM_{2.5}$: 9.8–22.6 μg/m^3, PM_{10}: 17.6–30.6 μg/m^3) and Northern Europe ($PM_{2.5}$: 8.5–11.1 μg/m^3, PM_{10}: 14.8–16.1 μg/m^3) (Eeftens et al. 2012). Our outdoor result was roughly comparable to their observations in Switzerland, Spain, and Greece, especially for $PM_{2.5}$. However, the ambient PM levels monitored in Cyprus were obviously lower compared to some Middle East countries such as Kuwait ($PM_{2.5}$: 44.3±8.8 μg/m^3, PM_{10}: 116.8±18.4 μg/m^3) (Yuan et al. 2020) and Israel ($PM_{2.5}$: 23.1±25.3 μg/m^3, PM_{10}: 55.5±98.0 μg/m^3) (Achilleos et al. 2020; Kloog et al. 2015), which would likely be due to the different frequencies of dust storms (Ginoux et al. 2012; Gherboudj et al. 2017). The evidence of household PM concentrations in Europe, however, is very limited and geographically dispersed (Vardoulakis et al. 2020). According to the current studies, in general, the households in Cyprus that we investigated appeared to have a higher $PM_{2.5}$ level but a similar PM_{10} level compared to Northern European countries, including Demark ($PM_{2.5}$ median: 6.3–12.2 μg/m^3) (Olsen et al. 2014; Karottki et al. 2015), Lithuania ($PM_{2.5}$: 9.0±17.9 μg/m^3, PM_{10}: 22.5±32.9 μg/m^3), and Finland ($PM_{2.5}$: 9.0±17.9 μg/m^3, PM_{10}: 22.5±32.9 μg/m^3) (Du et al. 2015). Likewise, the pattern of ambient PM, our indoor PM data were also lower than those reported in residential homes in Kuwait ($PM_{2.5}$: 28.5±17.9 μg/m^3, PM_{10}: 40.6±23.0 μg/m^3) (Yuan et al. 2020).

Among the analyzed species, both BC and BrC took up the highest percent contribution to indoor $PM_{2.5}$ mass, accounting on average 8.2% each to the total mass. The abundance of S was merely second to the carbon-containing particles in indoor $PM_{2.5}$ and accounted for 6.5% of its total mass, which suggested evidence of outdoor secondary pollution sources such as the combustion of sulfur-containing fuels in power plants and industrial activities (Long and Sarnat 2004; Tang et al. 2018). In addition, Ca, Si, K, Na, Al, and Fe had contributions varying from 1.1 to 2.1% to the $PM_{2.5}$ mass, following the downward order. In contrast, indoor PM_{10} consisted mainly of Ca, BC, and BrC, whose contribution percentages were 5.6, 4.9, and 4.9% compared to the total mass. A slightly lower contribution was observed for S, Si, Al, and Fe, which were estimated to be 4.0, 3.2, 2.1, and 1.9% of the indoor PM_{10} total mass, respectively. Other elemental weight percentages in indoor PM_{10} did not appear to deviate much from those in indoor $PM_{2.5}$. Since S is preferably present in sulfate particles (SO_4^{2-}) (Marcazzan et al. 2001) and SO_4^{2-} exists mostly as ammonium sulfate ($(NH_4)_2SO_4$) in the atmosphere (Hassan et al. 2013; Masri et al. 2015), it is reasonable to estimate the average concentrations of $(NH_4)_2SO_4$ in indoor $PM_{2.5}$ and PM_{10} mass to be 3.71 μg/m^3 (26.9%) and 4.03 μg/m^3 (16.4%), respectively. Assuming that the crustal elements are present in the form

Table 2 Summary statistics of indoor and outdoor concentrations of $PM_{2.5}$ mass, PM_{10} mass, and their components

Species	Indoor $PM_{2.5}$ (N=89)		Indoor PM_{10} (N=91)		Outdoor $PM_{2.5}$ (N=90)		Outdoor PM_{10} (N=91)	
Unit: ng/m^3	Mean±SD	%	Mean±SD	%	Mean±SD	%	Mean±SD	%
Mass*	13.8±8.7	100.0	24.6±19.4	100.0	15.1±6.2	100.0	30.4±10.8	100.0
BC*	1.1±0.6	8.2	1.2±0.6	4.9	1.3±0.6	8.7	1.4±0.6	4.7
BrC*	1.1±0.6	8.2	1.2±0.7	4.9	1.3±0.5	8.6	1.4±0.6	4.7
Na	179.7±119.6	1.3	341.1±234.3	1.4	228.2±93.7	1.5	484.0±248.6	1.6
Mg	86.8±108.5	0.6	212.4±248.9	0.9	102.2±68.1	0.7	277.7±128.3	0.9
Al	178.2±205.1	1.3	515.7±625.9	2.1	253.8±184.9	1.7	756.0±465.6	2.5
Si	247.0±316.1	1.8	783.3±938.1	3.2	301.3±275.8	2.0	1110.2±730.1	3.7
S	899.2±545.8	6.5	978.4±544.6	4.0	1176.7±569.1	7.8	1287.4±565.5	4.2
Cl	50.5±128.0	0.4	217.9±295.3	0.9	44.9±101.2	0.3	334.6±391.6	1.1
K	177.0±157.8	1.3	260.3±190.2	1.1	204.3±117.7	1.4	336.0±151.7	1.1
Ca	290.8±785.0	2.1	1381.8±3286.8	5.6	249.5±155.5	1.7	1583.3±974.3	5.2
Ti	11.6±16.5	0.1	34.4±33.5	0.1	13.7±9.3	0.1	48.3±27.8	0.2
V	1.9±1.6	<0.1	2.1±2.0	<0.1	2.5±1.9	<0.1	3.6±2.4	<0.1
Mn	5.1±4.9	<0.1	10.7±10.5	<0.1	5.6±4.4	<0.1	13.7±8.1	<0.1
Fe	147.5±243.8	1.1	468.4±783.8	1.9	188.3±111.9	1.2	685.3±373.0	2.3
Zn	19.3±38.0	0.1	26.2±45.9	0.1	26.0±56.4	0.2	35.2±69.2	0.1
Br	2.4±2.7	<0.1	3.2±3.2	<0.1	3.1±3.0	<0.1	4.2±3.3	<0.1
Pb	13.7±13.6	0.1	14.5±15.8	0.1	18.9±19.1	0.1	19.5±19.2	0.1
Cr	1.2±1.2	<0.1	1.8±1.5	<0.1	1.1±1.1	<0.1	2.1±1.3	<0.1
Sr	5.0±28.4	<0.1	8.5±30.0	<0.1	4.8±23.2	<0.1	9.5±24.0	<0.1

*In μg/m^3

of metal oxides (i.e., Al_2O_3, SiO_2, K_2O, CaO, TiO_2, MnO_2, Fe_2O_3), the average dust mass concentrations in $PM_{2.5}$ and PM_{10} indoors were estimated to be 1.72 μg/m^3 (12.5%) and 5.64 μg/m^3 (22.9%), respectively (Malm et al. 2004).

In terms of the chemical components, the indoor and outdoor patterns of elemental loadings were generally similar. BC, BrC, and S had the highest levels in outdoor $PM_{2.5}$, responsible for approximately 8.7, 8.6, and 7.8% of the total mass. Si, Al, Ca, Na, K, and Fe accounted for 1–2% of the mass, whereas the residual trace elements contributed little to the mix of particles. In the cases of relative abundance of components in outdoor PM_{10}, the significances of BC, BrC, Ca, S, Si, Al, and Fe were highlighted with their accumulative contributions of up to 27.3%. S mainly as SO_4^{2-} is representative of long-range atmospheric transportation. A previous trans-European study on $PM_{2.5}$ speciation pointed out that S was the most abundant element accounting for about 4.6–8.8% of $PM_{2.5}$ mass (Götschi et al. 2005), which was similarly found in our results. Assuming S to be in the form of $(NH_4)_2SO_4$, the estimated concentrations of $(NH_4)_2SO_4$ in outdoor $PM_{2.5}$ and PM_{10} were 4.85 μg/m^3 (32.1%) and 5.31 μg/m^3 (17.5%), respectively. The dust mass represented as metal oxides in outdoor $PM_{2.5}$ and PM_{10} was estimated to be 5.30 μg/m^3 (35.1%) and 23.6 μg/m^3 (77.9%), respectively (Malm et al. 2004). The observed large amount of crustal metal oxides supported the evidence of a remarkably larger contribution of mineral dust to $PM_{2.5}$ and PM_{10} in Southern Europe compared to Northwestern and Central Europe monitored over the past decade (Putaud et al. 2010).

$PM_{2.5}/PM_{10}$ mass and elemental ratios

The box plots of the $PM_{2.5}/PM_{10}$ ratios of mass and elements measured inside and outside participants' houses are shown in Fig. 1. The corresponding median values are presented in Table S4 and Fig. S2, and the scatterplots of the comparison between indoor and outdoor ratios is presented in Fig. S3. The median $PM_{2.5}/PM_{10}$ mass ratios corresponding to indoor and outdoor levels were 0.58 and 0.49, respectively. In our study, the PM in Cyprus displayed a lower $PM_{2.5}/PM_{10}$ ratio compared to the levels in Europe (0.5–0.9) (Putaud et al. 2010), which is comparable with previous studies (Saliba and Massoud 2011; Achilleos et al. 2016). This finding may reflect the importance of primary particles emitted from dust storms or non-dust coarse particles such as sea salt controlled by resuspension (Sugimoto et al. 2016). The frequent dust storm outbreaks over North Africa and Arabian Peninsula can also contribute a great fraction of coarse particles to the nearby Eastern Mediterranean regions (Kubilay et al. 2000; Perez et al. 2008; Dimitriou and Kassomenos 2017). Moreover, the roles of dry climate and less vegetation cover in resuspension of particles may explain further for accumulated coarse

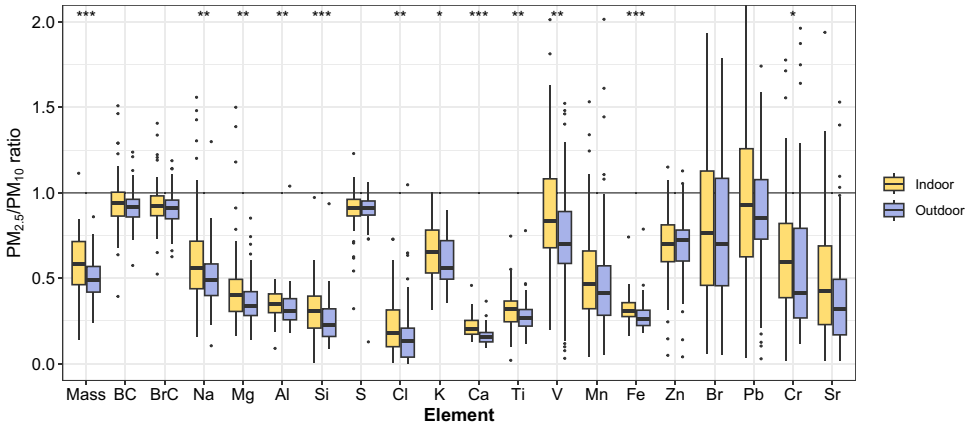


Fig. 1 $PM_{2.5}/PM_{10}$ ratios for mass and elemental concentrations in residential indoor and outdoor environments. Extremes of the whiskers indicate 25th and 75th percentiles. * $p < 0.05$, ** $p < 0.01$, *** $p < 0.001$

particles. Nevertheless, the ratios measures in the dwellings in Cyprus were generally higher than previously reported in other Eastern Mediterranean sites (~0.25) (Saliba and Massoud 2011), which can be accounted for by site type, underlying surface, anthropogenic activities, and meteorological conditions (Fan et al. 2021). For instance, a higher level of anthropogenic aerosols is expected to be present in the residential areas near the streets than the rural sites or urban background sites due to denser hotspots of traffic and domestic activities.

At the same time, the different PM constituents manifested apparent discrepancies in their preferential modes. BC, BrC, S, Pb, Zn, and V that may point to anthropogenic emissions from combustion, traffic, and industry had the highest median $PM_{2.5}/PM_{10}$ ratios (≥0.70), suggesting their predominant fine mode. The ratios of K, Na, Cr, and Mn were modest with the medians of 0.35–0.70. The rest of the elements (Mg, Sr, Al, Ti, Fe, Si, Ca, and Cl) that exhibited relatively lower ratios were of terrestrial and marine origin, and tended to be predominantly in the PM_{10} mode. Similarly, the presence of crustal elements such as Ca and Si in the coarse mode in Cyprus was previously reported by Achilleos et al. (2016). Overall, data suggests that $PM_{2.5}$ is more related to human activities, whereas particles of terrestrial and marine origin are more likely found in PM_{10}. Furthermore, the indoor $PM_{2.5}/PM_{10}$ ratios for mass and most of the elements (Na, Mg, Al, Si, Cl, K, Ca, Ti, V, Fe, Cr) were significantly higher than the corresponding ratios outdoors.

PM sources in residential environments

Source apportionment

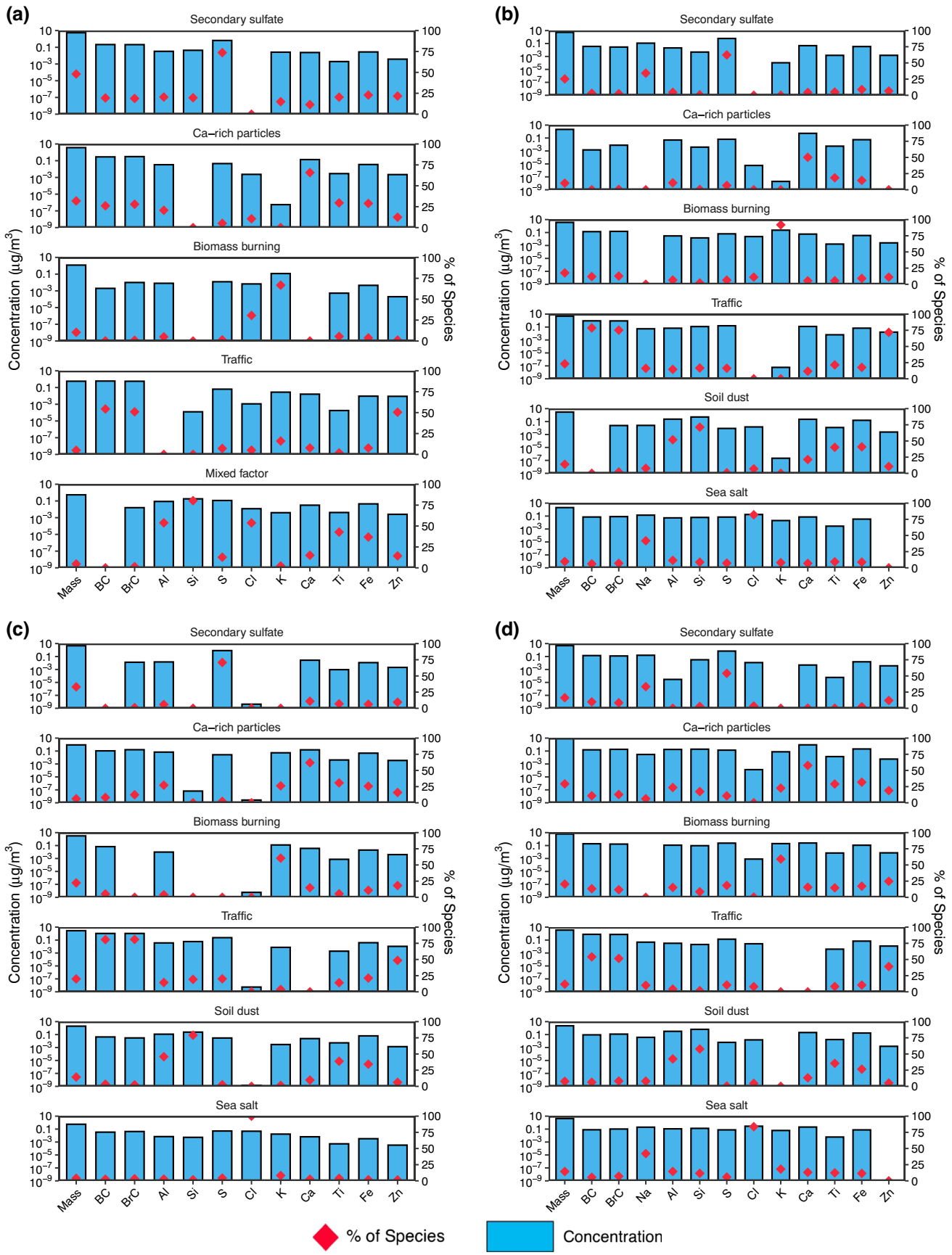
The apportioned source profiles of residential indoor and outdoor PM are shown in Fig. 2, and the specific source contributions are presented in Fig. 3. The comparison between measured and predicted concentrations of mass and elements is provided in Table S3. Secondary sulfate pollution, Ca-rich particles, biomass burning, traffic, soil dust, and sea salt were identified as the main sources of $PM_{2.5}$ and PM_{10}. As can be seen, these sources showed a highly similar profile across the $PM_{2.5}$ and PM_{10} modes, but the relative source contributions vary between the indoor and outdoor PM, and between the two PM fractions.

Specifically, the source apportionment of indoor $PM_{2.5}$ identified five sources with secondary sulfate pollution as the major contributing factor, followed by Ca-rich particles, biomass burning, traffic, and mixed factor.

The secondary sulfate pollution source, which was the major contributor to the indoor $PM_{2.5}$ mass (48.0%), was characterized by high levels of S, which is an important tracer of regional pollution (Lall and Thurston 2006). Therefore, our results indicate the importance of transported pollution from other regions.

The Ca-rich particles source was the second largest source and represented 31.9% of indoor $PM_{2.5}$ mass. Calcium carbonate is well known as the main filler ingredient of wall putty and is commonly seen in paint and coating applications. Therefore, the Ca-rich component may come from the deterioration of building materials, furniture coatings (Suryawanshi et al. 2016; Carrion-Matta et al. 2019), showers, or humidifiers (Anderson et al. 2007; Lau et al. 2021; Yao et al. 2020). Accompanied by some contents of Fe, Ti, and Zn, we cannot rule out the potential penetrations of resuspended road dust. It is also likely for the occupants to bring the Ca-rich component attached to clothes indoors after outdoor activities.

The biomass burning source was characterized by K (66.8%), and to a less extent by Cl (31.6%), which was responsible for 10.4% of the total mass. K has been widely used as an indicator of biomass burning (Kim et al. 2003; Pachon et al. 2013; Masri et al. 2015; Saggu and Mittal 2020). The use of fireplaces indoors (18.2%) and scented candles or incense (45.5%) may partially comprise the biomass burning source of indoor PM. However, since the study residents rarely used biomass fuels such as wood for heating or cooking, the biomass contribution should be mostly of outdoor origin.


(a)
(b)
(c)
(d)
Secondary sulfate
Ca–rich particles
Biomass burning
Traffic
Soil dust
Sea salt
Mixed factor
Concentration (µg/m3)
% of Species
Mass
BC
BrC
Na
Al
Si
S
Cl
K
Ca
Ti
Fe
Zn
% of Species
Concentration

◂**Fig. 2** Source profiles of **a** indoor $PM_{2.5}$, **b** indoor PM_{10}, **c** outdoor $PM_{2.5}$, and **d** outdoor PM_{10}

The traffic source was characterized by high loadings of BC, BrC, and Zn, which were associated with motor vehicle exhausts and non-exhausts. This source accounted for 54.4% of BC, 51.0% of BrC, and 50.5% of Zn for the mass concentration. BC in urban areas originates mainly from diesel engines (Gray and Cass 1998). In traffic-dominated cities, secondary photochemical reactions of traffic emissions can be the primary source of substantial BrC in air (Zhang et al. 2021). Zn is commonly used as an additive in lubricating oil in engines, and it is also present in vehicle tire wear (Fergusson and Kim 1991). In our study, most of the sampled representative households in Cyprus are located on main streets or at crossroads and nearby a parking lot, which can provide some traffic origins. Nevertheless, the traffic factor was the second least contributor to indoor $PM_{2.5}$ and only responsible for 5.0% of the mass concentration.

There appeared to be a mixed factor of crustal and marine particles, only accounting for 4.7% of the indoor $PM_{2.5}$ mass. This factor was dominated by high concentrations of Si and Al, and Cl. As aforementioned, the potential indoor sources of Cl were probably mixed with sea salt and had a small contribution. As described in the supplementary materials, the mixture of two sources in the same factor was apportioned with the consideration of better model physical interpretability.

Similarly, six sources were identified for indoor PM_{10}. Consistent with the findings for indoor $PM_{2.5}$, secondary sulfate pollution (25.3%) was the most significant source of the indoor PM_{10} mass; meanwhile, the biomass burning factor characterized by K and the traffic factor characterized by BC, BrC, and Zn also had comparable proportions, accounting for 17.6% and 23.2% of the total PM_{10} mass. As expected, the contributions of sea salt and soil dust to indoor PM_{10}, 9.7 and 13.9%, respectively, were higher as compared to those for indoor $PM_{2.5}$. Ca-rich particles contributed to 10.2% of indoor PM_{10}. The soil dust source was a major contributor to several crustal elements in indoor PM_{10} (71.3% of Si, 51.9% of Al, 41.0% of Fe, and 40.2% of Ti). The sea salt factor contributed to a large fraction of Cl (82.0%) and Na (41.9%). A relatively lower Na content in the sea salt factor can be explained by the existence of Na in regional sulfur pollution, which has been reported in other studies (Masri et al. 2015; Achilleos et al. 2016). It is likely that sulfuric and nitric acids can react with sea salt (NaCl) suspended in the atmosphere; consequently, the product Na_2SO_4 is deposited on the filters while another product HCl is released into the air.

In the outdoor environment, $PM_{2.5}$ mainly came from secondary sulfate, biomass burning, and traffic, whereas PM_{10} mainly originated from Ca-rich particles, biomass burning, and secondary sulfate. An evidently higher contribution of Ca-rich particles and sea salt was seen in outdoor PM_{10} as compared to outdoor $PM_{2.5}$.

Indoor to outdoor ratios for source contribution

We explored the indoor-outdoor relationships for corresponding source contributions by calculating the Spearman's rank correlation coefficients (Table 3). The distributions of I/O ratios of each common source of $PM_{2.5}$ and PM_{10} are shown in Fig. 4, with their median values presented in Table S5.

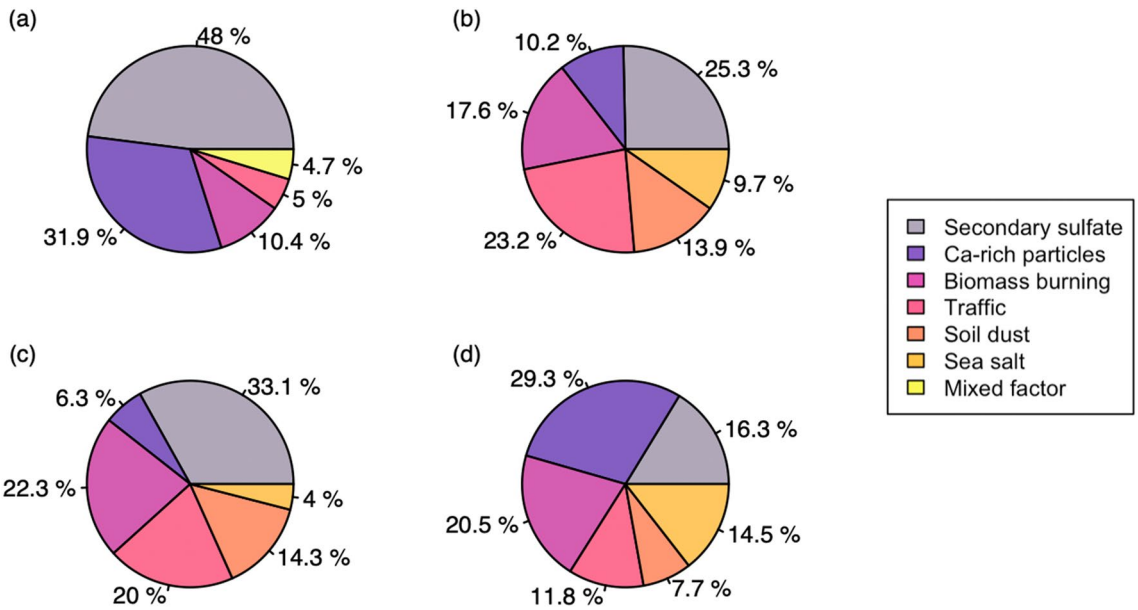


Fig. 3 Source contributions to **a** indoor $PM_{2.5}$, **b** indoor PM_{10}, **c** outdoor $PM_{2.5}$, and **d** outdoor PM_{10}

Table 3 Spearman's rank correlation coefficients of source contributions in indoor environments and in outdoor environments to $PM_{2.5}$ and PM_{10}

Source	Secondary sulfate	Ca-rich particles	Biomass burning	Traffic	Soil dust	Sea salt
$PM_{2.5}$	0.70	0.60	0.56	0.69	–	–
PM_{10}	0.74	0.18	0.53	0.36	−0.30	−0.06

As expected, the correlation between the contributions of common sources to indoor and outdoor PM_{10} was evidently lower than that of $PM_{2.5}$, especially for Ca-rich particles, soil dust, and sea salt. This finding indicates a higher infiltration rate of outdoor $PM_{2.5}$ than PM_{10}. Overall, the highest indoor-to-outdoor correlation of secondary sulfate and I/O ratios close to 1 for both $PM_{2.5}$ and PM_{10} samples indicated a considerable amount of outdoor pollution penetrated indoors. In detail, the I/O source contribution ratios for $PM_{2.5}$ suggest that the indoor levels were influenced by outdoor sources such as traffic, secondary sulfate pollution, and wood burning. The fine Ca-abundant fraction indoors was associated with indoor sources (median I/O ratio=3.89) such as degradation of walls and furniture coatings (Suryawanshi et al. 2016; Carrion-Matta et al. 2019). The fine particles could be also generated from aerosolized water during the operation of humidifiers or the showers (Anderson et al. 2007; Lau et al. 2021; Yao et al. 2020). Meanwhile, a small fraction of fine Ca particles in the household may come from construction activities in neighboring areas (de Moraes et al. 2016; Saggu and Mittal 2020). In PM_{10}, the median values of I/O source contribution ratios for Ca-rich particles, biomass burning, traffic emissions, soil dust, and sea salt were lower than one. This indicates that these sources were mostly affected by outdoor sources.

For traffic emissions, a few I/O ratios above unity could be accounted for by some resuspended soil and road dust constituents (i.e., Ca, Si, Al, Ti, and Fe) in the source profile (Fig. 2). Cl that characterized the sea salt factor is another example, given that it can also be produced from indoor activities, such as cooking (Ozkaynak et al. 1996; Habre et al. 2014), cleaning using bleach-containing products and bathing in chlorinated municipal water (Zhao et al. 2006, 2007; Habre et al. 2014).

We further restricted the examination to the non-dust days to avoid the potential influence of dust events. The sensitivity results (Fig. S4 and Table S5) were in good agreement with that derived from all sampling days (Fig. 4b).

Comparison of indoor and outdoor sources in Cyprus with other regions

Our results agree well with the previous findings for European or adjacent areas. Several Eastern Mediterranean studies have reported ammonium sulfate as a significant contributor to $PM_{2.5}$, which varied from 7.8 to 47% (Mantas et al. 2014; Paraskevopoulou et al. 2015; Fadel et al. 2023). A study investigating 16 Eastern Europe and Central Asia cities in urban and sub-urban background showed that biomass burning, traffic, and industry constituted the significant contributors to ambient $PM_{2.5}$ based on PMF results, emphasizing the influences of anthropogenic activities (Almeida et al. 2020). In another review article on PM source apportionment from a global perspective, the authors presented the contributions of natural sources (16%) and traffic emissions (19%) to ambient $PM_{2.5}$ in Central and Eastern Europe (Karagulian et al. 2015). Our results are highly comparable with their findings, despite some differences due to certain geographical and social reasons. To be specific, sea salt tends to contribute a larger fraction to ambient PM_{10} in our study because Cyprus is an island. The significantly lower contribution of biomass burning as compared to the contribution reported for Central and Eastern Europe could be

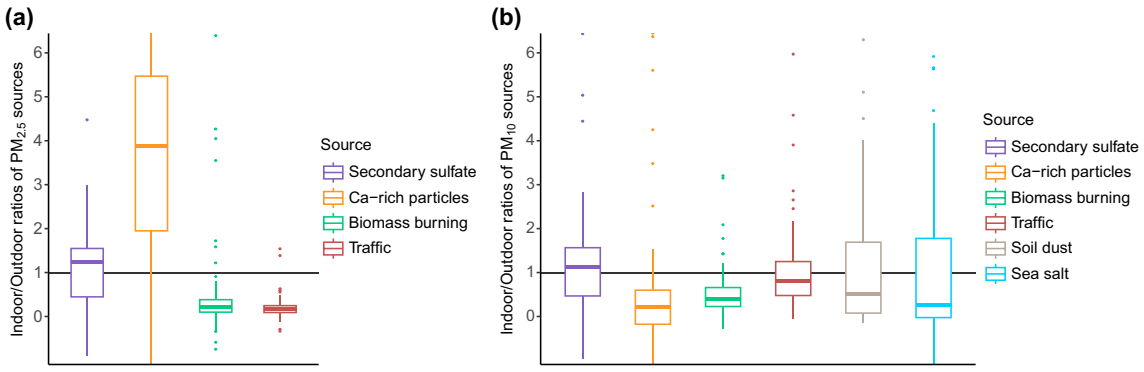


Fig. 4 Source contributions to **a** $PM_{2.5}$ and **b** PM_{10}. Extremes of the whiskers indicate 25th and 75th percentiles

attributed to the discrepancy in domestic heating sources in research areas.

Bordered by the densely populated European cities to the north and the Northern African continent to the south, the Eastern Mediterranean region is severely subjected to air masses originating from Sahara Desert and pollution produced from North and East Europe and transported in the form of tropospheric aerosols (Mihalopoulos et al. 1997). High levels of sulfate and carbonaceous particles in this region have been reported (Luria et al. 1996; Mamane et al. 1980; Guieu et al. 2010; Im et al. 2012). Thus, transported pollution from other countries is in part responsible for the relatively high-level secondary sulfate pollution and its large impact on indoor PM exposures.

As compared to other European regions, the Eastern Mediterranean countries have a lower precipitation rate and are especially typical of arid summer and fall. Therefore, the proportions of crustal and marine aerosols in the atmosphere are likely to overtake those in other regions, enhancing the resuspension of mineral materials and road traffic dust outdoors and indoors. Similar findings have been reported previously in this region (Kubilay and Saydam 1995; Kleanthous et al. 2009; Im et al. 2012; Achilleos et al. 2014, 2016; Faridi et al. 2022). On average, sea salt and dust contributed 20% and 15% to PM_{10} mass, respectively (Im et al. 2012). A recent study involving 22 Eastern Mediterranean countries determined dust to be the most dominant source of ambient $PM_{2.5}$ (7–95%, with an average of 49%) and PM_{10} (8–80%, with an average of 30%) (Faridi et al. 2022). In addition, we identified Ca-rich particles as an important fine PM contributor in residential environments, as its I/O ratios larger than unity indicates the existence of the Ca indoor source, such as wall dust (Suryawanshi et al. 2016; Carrion-Matta et al. 2019), shower, and use of humidifiers (Anderson et al. 2007; Yao et al. 2020; Lau et al. 2021).

However, it should be noted that there are some limitations in our analysis. First, the sampling periods excluded January, July, and August, which may slightly reduce the representativeness of the whole year and the comparability of our findings with other studies. However, the sampled 9 months of the year covered both the cold seasons and warm seasons in Cyprus; we do not expect much bias caused by the deviation from sampling the entire year. Second, the lack of data on SO_4^{2-} and nitrate (NO_3^-) in the source apportionment may underestimate the contribution of secondary sulfate pollution, traffic, and/or biomass burning. The measurement of NH_4^+ was not available as well; however, it may not lead to a considerable bias due to its small proportion of PM (1–3%) as suggested in previous reports in Cyprus (Bari et al. 2009; Achilleos et al. 2016). Third, several months of sampling occurred during the COVID-19 restriction measures, which may decrease the ambient PM level due to reduced human activities such as transportation and industrial and commercial activities compared to normal days (Marinello et al. 2021; Faridi et al. 2021). Moreover, in light of longer stays and more indoor activities in the household during the lockdown, the indoor-originated sources may contribute a larger fraction to indoor PM pollution, such as Ca-rich particles in $PM_{2.5}$. However, the indoor PM levels might demonstrate a more complex and less clear pattern in the context of lockdown, considering the significant impact of outdoor secondary pollution on indoor PM levels as indicated in our results.

Conclusions

In this study, we examined the $PM_{2.5}$ and PM_{10} mass and elemental levels and identified possible sources contributing to indoor and outdoor PM levels at household premises in Nicosia, Cyprus. Indoor PM levels were comparable to outdoor levels, implying penetration of outdoor pollution. Indoor $PM_{2.5}$ was affected by local or transported secondary sulfate pollution, biomass burning, traffic, Ca-rich particles, and mixed factor of marine and crustal origins (i.e., sea salt and soil dust). The source categories showed highly consistent for indoor PM_{10}. In the residential environment, secondary sulfate pollution served as the most significant contributor to indoor $PM_{2.5}$ and PM_{10}, accounting for 48.0% and 25.3% of mass, respectively. Moreover, our results revealed strong evidence of considerable indoors-originated Ca-rich fine particles in residences. Our findings suggest that residents of Cyprus are exposed to significant amounts of indoor and outdoor PM (especially $PM_{2.5}$), and therefore efforts to reduce ambient pollution will greatly mitigate the exposure to PM in indoor microenvironments. This also highlights the importance of air infiltration and urban design in relation to public health.

Our study provides a case for the policy implementation and public health promotion regarding air particulate pollution in Eastern Mediterranean and other European countries under similar climates and economical status. Effective interventions are warranted to reduce exposure to high-level PM and thus mitigate its attributable health burden. Feasible personal strategies include using indoor air purifiers with high-efficiency particulate air filters, properly installing particle filtration systems in heating, ventilation, and air conditioning units, and avoiding outdoor activities where peak pollution may occur. In a broader context, population-level actions should focus on reducing air pollution emissions from transportation and industry by transitioning from conventional fossil fuels to renewables and implementing stricter air pollution abatement policies. In addition, enhancing real-time air quality monitoring and early warning systems should also be prioritized.

Supplementary Information The online version contains supplementary material available at https://doi.org/10.1007/s11869-023-01460-8.

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Data availability Data will be made available on request.

Declarations

Ethics approval Informed consent was obtained from all individual participants included in the study.

Consent to publish The authors affirm that human research participants provided informed consent for publication of the tables and figures.

Competing interests The authors declare no competing interests.

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