

Full Length Article

Characterization of particulate matter (PM_{2.5} and PM₁₀) in an urban area in Amman by PIXE, PESA, optical and gravimetric measurements

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ABSTRACT

Atmospheric particulate matter (PM) samples in two fractions (PM_{2.5} and PM₁₀) were collected simultaneously on Teflon filters in an urban area in Amman, Jordan. Gravimetric and light absorption measurements were performed to calculate total mass and black carbon concentrations, respectively. Two ion beam analysis (IBA) techniques were employed to obtain elemental characterization: particle induced X-ray emission (PIXE) and particle elastic scattering analysis (PESA). Chemical components (particulate organic matter, sea salt, ammonium sulfate and soil) were estimated and mass reconstruction was performed. The average PM_{2.5} and PM₁₀ concentrations were $20 \pm 9 \mu\text{g}/\text{m}^3$ and $40 \pm 19 \mu\text{g}/\text{m}^3$, respectively. It was shown that soil was the dominant component in coarse particulates, fine particulates were dominated by ammonium sulfate, black carbon was more abundant in fine particulates (about 20 %), and sea salt was a minor source (1–2 %) in both fractions.

1. Introduction

The demand for characterizing atmospheric aerosols or particulate matter (PM) is increasing worldwide due to the harmful effects of air pollution on the ecological system and human health. It is important to study the elemental content and chemical composition of PM in order to understand air pollutants and identify pollution sources. Over the past few years, the University of Jordan, Amman, Jordan, has been interested in studying the characteristic features of atmospheric aerosol particles in the framework of the International Atomic Energy Agency (IAEA) regional technical cooperation projects (IAEA TC projects RAS0072, RAS0076, and lately RAS0078) for ARASIA member states (Arab states in Asia who are members of the IAEA) [1–4]. Elemental composition of PM samples from Jordan was typically determined using particle-induced X-ray emission (PIXE) technique, either under vacuum using the PIXE-RBS beamline in the University of Jordan Van de Graaff Accelerator (JUVAC) [1] in Amman, Jordan, or using the external beam PIXE setup of the Tandemtron accelerator of INFN-LABEC laboratory in Florence, Italy [2–4].

PIXE technique has been widely used since its birth for the study of PM composition due to its well-known advantages for elemental analysis [5,6] and is still on the cutting edge for the study of aerosol composition due to its potential in giving data for major, minor and trace elements

[7]. Although PIXE can detect easily with good accuracy elements from Na to Pb, it gives only part of the information needed about aerosol chemical composition. It is therefore important to be complemented by information attained through other techniques.

Previous studies of PM samples from Amman [2,3], exclusively based on PIXE, did not obtain the chemical mass closure (mass reconstruction yielded ~7 % [2] and 48–60 % [3]). The deficiency in PM mass reconstruction was attributed to the absence of carbon and/or organic compounds measurements. In this contribution we investigate the composition of a selected set of fine and coarse PM samples collected in Amman, using two IBA techniques, PIXE and particle elastic scattering analysis (PESA), along with gravimetric and light absorption measurements. PIXE provides data on elements from Na to Pb, PESA provides data on H (a useful marker for organic compounds that are undetected by PIXE), and light absorption measurements quantify black carbon (BC) content. A more complete reconstruction of the aerosol mass is thus attempted thanks to the increased range of elemental concentrations measured (H, Na to Pb, and BC).

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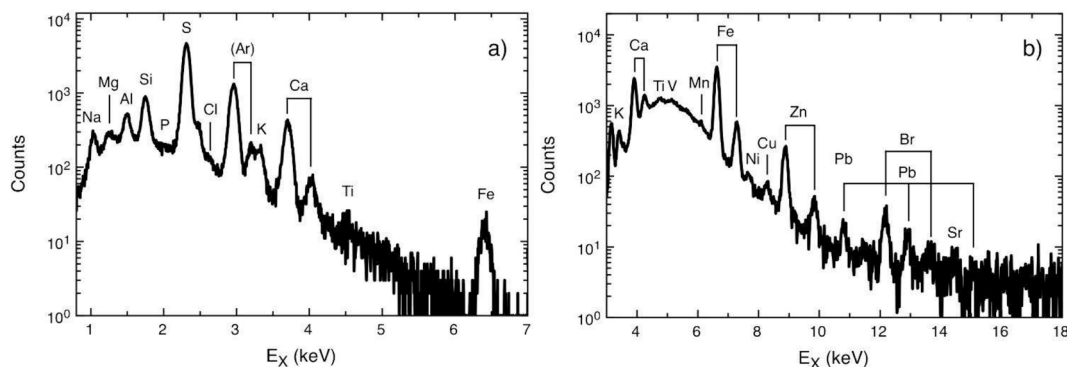


Fig. 1. PIXE spectra of a $PM_{2.5}$ sample obtained with an external beam of 3 MeV protons, as collected by the small-area silicon drift detector (a) and by the twin large-area silicon drift detectors, after summing the single spectra (b).

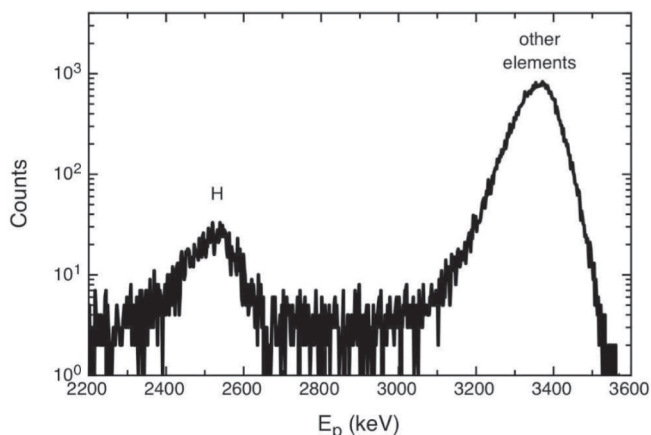


Fig. 2. PESA spectrum of a $PM_{2.5}$ sample obtained in vacuum with 3.6 MeV protons.

2. Experimental methods

2.1. $PM_{2.5}$ and PM_{10} sample collection

Atmospheric PM samples in two fractions ($PM_{2.5}$ and PM_{10} , aerosol particles with aerodynamic diameter less than 2.5 μm and 10 μm , respectively) were collected simultaneously in an urban area in Amman, Jordan, namely on the rooftop of the Physics building at the University of Jordan (32°00'52.27" N, 35°52'25.41" E). Sampling was performed on a sequential basis (24 hr-sampling, 2–3 times a week), during workdays and weekends, in the period from June 2017 to October 2018. Samples were collected using two low-volume (2.3 m^3/h) samplers: ISAP 1050e sampler [8] collected fine particulate matter ($PM_{2.5}$) and Sven Leckel sampler [9] collected coarse particulate matter (PM_{10}). Both PM fractions were collected on 47-mm diameter thin Teflon filters (46 μm thickness and 2 μm pore size) supplied by PALL corporation [10]. A total of fifty-one pairs of $PM_{2.5}$ and PM_{10} samples were investigated in this study, among them ten pairs of samples were collected during weekends

Table 1

Mean, maximum and minimum total mass concentrations of the investigated particulate matter samples. All concentrations and standard deviations (SD) in parenthesis are given in $\mu\text{g}/\text{m}^3$.

Samples	Total mass concentration, [PM] ($\mu\text{g}/\text{m}^3$)								
	All days			Workdays			Weekends		
	Mean (SD)	Max	Min	Mean (SD)	Max	Min	Mean (SD)	Max	Min
$PM_{2.5}$	20 (9)	62	9	21 (10)	62	11	16 (3)	19	9
PM_{10}	40 (19)	130	17	42 (21)	130	20	32 (9)	48	17

(i.e., on Fridays).

2.2. Gravimetric measurements

Gravimetric measurements were performed to calculate the total mass concentrations of aerosol particles, following a routine procedure used in a previous work [3]. Briefly, filters were stabilized during a 24–48 h period in a controlled environment (i.e., stable temperature and relative humidity), prior to being weighed before and after sampling using a digital microbalance (with 1 μg precision) and a static charge eliminator. Daily mass concentrations ($PM_{2.5}$ and PM_{10} , in $\mu\text{g}/\text{m}^3$) were calculated for each sample by dividing the total mass of deposited particles by the volume of air sampled.

2.3. Black carbon measurements

Black carbon (BC) content in PM can be investigated using optical methods that rely on the ability of PM samples to reflect and absorb light [11]. In this work multi-wavelength absorption black carbon instrument (MABI) from ANSTO [12] was used. Teflon filters were exposed to a multi-wavelength light source and the intensity of the transmitted light was measured for each filter before and after sampling by a photodiode detector. The BC concentration ([BC] in $\mu\text{g}/\text{m}^3$) for every sample (i.e., loaded filter) was calculated by dividing the aerosol optical absorption coefficient, σ_a , by the specific mass absorption coefficient, ϵ (also known as the absorption efficiency) [13], using the formula:

$$[BC] = \sigma_a / \epsilon$$

where the aerosol optical absorption coefficient (σ_a , in m^{-1}) was obtained using the well-known Lambert-Beer law [14]. There were many different values for ϵ reported in literature due to using different reference techniques, different types of filters, and even performing measurements using different wavelengths [15]. Following the work presented by Cohen et al. [14] and Taha et al. [15] and exploiting the multi-wavelength capability of MABI (namely, 405 nm, 465 nm, 525 nm, 639 nm, 870 nm, 940 nm, and 1050 nm), ϵ for PM samples collected in Amman on Teflon filters was obtained empirically at each wavelength [16]. More details on MABI calibration, measurements and related

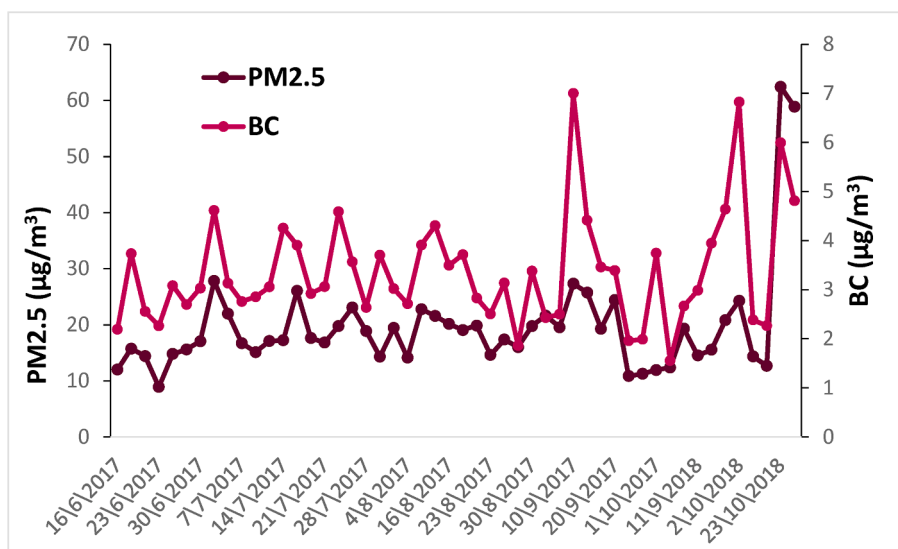


Fig. 3. The variation of black carbon (BC) content in $PM_{2.5}$ samples during the sampling period in Amman.

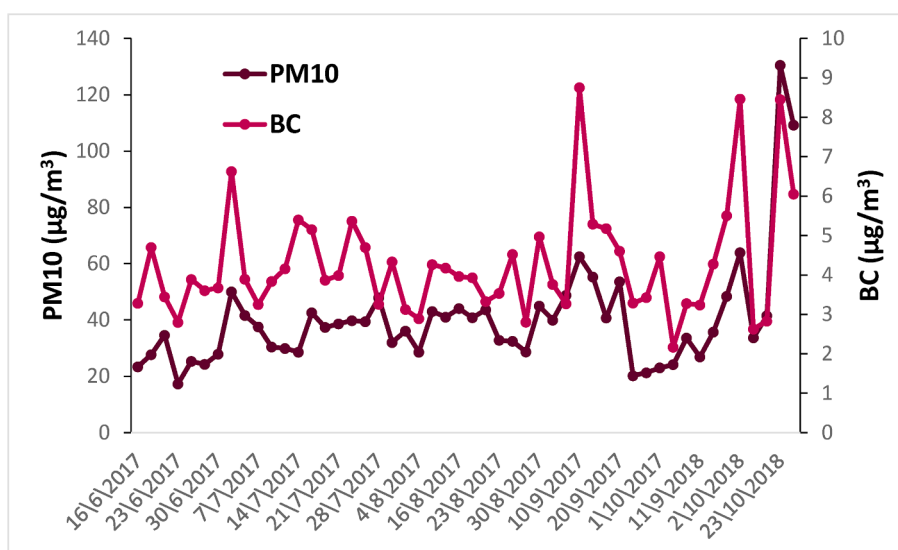


Fig. 4. The variation of black carbon (BC) content in PM_{10} samples during the sampling period in Amman.

calculations have been recently documented by Ryś and Samek [17].

2.4. PIXE measurements

PIXE measurements were performed using the external beam PIXE set-up of the 3 MV Tandatron accelerator of the LABEC laboratory [18] of INFN, Florence, Italy. Each sample was irradiated with a proton beam extracted into ambient pressure, with an energy of 3.00 MeV on the target and a beam current ranging from 30 to 50 nA, depending on the loading of the filter, for about 45–60 s. The samples were continuously scanned in the plane perpendicular to the impinging beam direction to analyse as much aerosol deposit as possible to average out possible local inhomogeneities arising from the sampling. The low-Z elements (Na to Ca) were detected by a small-area silicon drift detector (SDD), with helium flowing in front of it, while the medium–high-Z elements (Ca and above) were detected by two identical large-area SDDs to double the statistics of the acquired spectra, covered by a 450 μm thick Mylar absorber. Examples of PIXE spectra (where elements from Na to Pb were detectable) are shown in Fig. 1. Following the same approach employed previously [1], PIXE spectra were fitted and analyzed using the

GUPIXWIN code [19], and a set of thin elemental standards from MicroMatter [20] containing known amounts of elements relevant for aerosol particles analysis was analyzed in the same geometry and used for quantitative analysis [7]. Typical experimental uncertainties ranged from about 5 % for the more abundant elements to a few tens of percent for the minor elements. The accuracy of the obtained elemental concentrations was guaranteed by analyzing NIST standard reference material SRM2783 [21]. A comprehensive description of the theoretical basis of the PIXE technique are available in literature [22,23]. A detailed description and characteristics of the external beam PIXE set-up at the LABEC laboratory are reported elsewhere [7].

2.5. PESA measurements

In-vacuum PESA measurements were carried out at LABEC laboratory [24,25] using a proton beam at an energy of 3.60 MeV, with a beam current of 10–15 nA and lasting 150 s. The total hydrogen content was obtained from the scattered/recoiled protons detected by a silicon pin diode detector placed at a 30° scattering angle (forward scattering), collimated to a small solid angle of about 0.3 msr. The H-peak area was

Table 2

Mean, maximum and minimum values of PM_{2.5} mass concentrations, the corresponding elemental composition, the calculated sea salt, ammonium sulfate, soil, particulate organic matter (POM), black carbon (BC) components; percentage of the sea salt, ammonium sulfate, soil, POM, BC and of the reconstructed mass (RCM) over the PM_{2.5} mass (in italic) of the collected samples. All the concentrations are given in ng/m³.

	All days			Workdays			Weekends		
	Mean	Max	Min	Mean	Max	Min	Mean	Max	Min
PM _{2.5}	20,000	62,000	9000	21,000	62,000	11,000	16,000	19,000	9000
H	570	1100	310	580	1100	310	510	660	320
Na	110	320	38	100	120	38	130	320	42
Mg	92	680	16	100	680	17	59	130	16
Al	300	2200	39	340	2200	59	160	460	39
Si	660	5000	100	740	5000	140	370	1000	100
P	23	88	4.2	24	88	4.2	18	28	6.9
S	1300	3100	420	1400	3100	420	1200	1900	750
Cl	29	130	7.8	29	130	10	27	110	7.8
K	180	720	59	190	720	60	130	260	59
Ca	1400	6800	280	1500	6800	280	770	1500	300
Ti	29	190	6.2	32	190	6.2	18	41	6.2
V	2.9	8.9	0.48	3.1	8.9	0.48	2.1	3.8	0.80
Cr	1.1	5.9	0.30	1.2	5.9	0.38	0.85	1.4	0.30
Mn	5.5	33	1.1	6.1	33	1.1	3.3	5.6	1.8
Fe	260	1800	58	290	1800	58	160	360	58
Ni	1.2	4.0	0.50	1.2	4.0	0.50	0.91	1.3	0.52
Cu	2.2	7.1	0.75	2.3	7.1	0.75	1.7	3.2	1.0
Zn	17	54	6.1	17	54	6.1	15	21	9.1
As	1.1	4.5	0.09	1.2	4.5	0.09	0.79	1.2	0.21
Se	0.52	1.1	0.08	0.52	1.1	0.08	0.52	0.99	0.20
Br	5.5	15	1.8	5.4	13	1.8	5.8	15	2.1
Rb	0.79	3.4	0.11	0.91	3.4	0.11	0.48	0.89	0.12
Sr	2.3	9.1	0.12	2.4	9.1	0.12	1.6	2.7	0.41
Y	0.82	2.5	0.22	0.82	2.5	0.22	0.82	1.3	0.32
Zr	1.7	7.6	0.22	1.8	7.6	0.22	1.4	3.3	0.30
Mo	2.2	5.8	0.48	2.3	5.8	0.56	1.8	4.2	0.48
Pb	2.9	6.8	1.1	3.0	6.8	1.8	2.6	4.3	1.1
BC	3400	7000	1600	3500	7000	1600	3000	4300	2200
Sea salt	18 %	31 %	8.2 %	18 %	31 %	8.2 %	19 %	25 %	14 %
	270	810	97	260	500	97	320	810	110
	1.5 %	4.8 %	0.3 %	1.4 %	3.5 %	0.3 %	2.0 %	4.8 %	0.8 %
Ammonium sulfate	5600	13,000	1700	5700	13,000	1700	5000	8000	3100
	33 %	54 %	2.9 %	31 %	54 %	2.9 %	33 %	48 %	16 %
Soil	5200	32,000	980	5800	32,000	1200	2900	6700	980
	23 %	52 %	7.9 %	25 %	52 %	9.3 %	18 %	35 %	7.9 %
POM	4000	8900	1200	4100	8200	1200	3900	8900	2300
	22 %	53 %	11 %	21 %	53 %	11 %	25 %	47 %	14 %
RCM	98 %	120 %	78 %	97 %	120 %	78 %	99 %	115 %	85 %

calculated by integration of the peak counts and subtraction of a linear background. Apart from the PM samples, a thin Upilex (C₂₂H₁₀N₂O₄) foil, containing known amounts of hydrogen, was repeatedly analyzed and used as standard for the quantitative analysis to determine hydrogen concentrations, and to check reproducibility and repeatability. Typical experimental uncertainties were about 7–8 %. An example of PESA spectrum (with a clear indication of the isolated peak due to forward scattered/recoiled protons) is shown in Fig. 2.

The H concentration obtained by PESA was used to calculate the organic component of PM (particulate organic matter, POM) to account for the unmeasured C, N, and O in organic compounds. POM is typically estimated from organic carbon (OC) as obtained by thermo-optical analysis methods [26,27] applying a conversion factor. The conversion factor, POM/OC, varies depending on the sampling site category (urban or non-urban site) as suggested by Turpin and Lim [26]. In this work, we followed a similar approach [25], starting from organic hydrogen (H_{org}, hydrogen contained in organic compounds) instead. The hypothesis is that H can be apportioned just to ammonium and organic compounds, and that ammonium is neutralized by sulfate; this simple approach is reasonable, assuming loss of water during the proton beam irradiation and in the absence of any other major sources of hydrogen contribution (i.e., negligible bisulfate contribution) [25]. This method was originally proposed by Eldred et al. [28] and the group of the University of California, Davis [29] and then it has been largely used (e.g., [13,30,31] and references therein), particularly when IBA measurements are not

combined to OC measurements with thermo-optical analysis methods.

As a proxy of organic matter, H_{org} was calculated by subtracting from the total H concentration obtained by PESA ([H_{PESA}]) the additional hydrogen from ammonium, assuming that all ammonium ions (NH₄⁺) are balanced by sulfate ions (SO₄²⁻) [25], i.e., 8H atoms for each S atom in the molecule [13], using the formula:

$$[H_{org}] = [H_{PESA}] - 0.25 \times [S_{PIXE}]$$

where [S_{PIXE}] represents sulfur concentration as obtained by PIXE measurements. The concentration of POM was then estimated by multiplying the measured concentration of H_{org} by a factor, i.e., [POM] = Factor × [H_{org}]. Clearly, we had to apply different factors than those POM/OC factors proposed by Turpin and Lim [26]. For the current study (PM collected on Teflon filters in an urban site), taking seasonal dependence into consideration, the values of 19.0 ± 1.0 (urban background, summer) and 15.4 ± 0.5 (urban background, winter) for POM/H_{org} conversion factor [25] were used.

3. Results and discussion

3.1. PM mass and BC concentrations

The values of the mean, maximum, and minimum mass concentrations of PM_{2.5} and PM₁₀ in µg/m³, during the whole sampling period as well as during workdays and weekends, are summarized in Table 1. The

Table 3

Mean, maximum and minimum values of PM₁₀ mass concentrations, the corresponding elemental composition, the calculated sea salt, ammonium sulfate, soil, particulate organic matter (POM), black carbon (BC) components; percentage of the sea salt, ammonium sulfate, soil, POM, BC and of the reconstructed mass (RCM) over the PM₁₀ mass (in italic) of the collected samples. All the concentrations are given in ng/m³.

	All days			Workdays			Weekends		
	Mean	Max	Min	Mean	Max	Min	Mean	Max	Min
PM ₁₀	40,000	130,000	17,000	42,000	130,000	20,000	32,000	48,000	17,000
H	680	1300	440	690	1300	460	620	840	440
Na	280	840	65	250	580	65	380	840	150
Mg	220	1100	56	230	1100	56	180	280	85
Al	590	3500	140	630	3500	140	1300	9700	160
Si	1500	8600	380	1600	8600	380	1000	2100	430
P	39	160	15	41	160	15	31	45	21
S	1300	2800	580	1300	2800	580	1400	2200	750
Cl	210	1200	15	190	1200	27	290	1000	15
K	320	1500	100	330	1500	100	270	460	120
Ca	3900	16,000	1300	4200	16,000	1300	2700	4200	1600
Ti	82	430	24	88	430	36	58	130	24
V	5.1	19	1.3	5.1	19	1.5	5.0	17	1.3
Cr	2.4	9.9	0.96	2.5	9.9	0.96	2.0	3.8	1.2
Mn	14	71	4.4	15	71	5.6	11	22	4.4
Fe	740	3900	230	780	3900	300	580	1200	230
Ni	2.1	6.7	1.0	2.1	6.7	1.0	2.0	6.6	1.1
Cu	5.4	33	1.7	5.1	13	2.2	6.6	33	1.7
Zn	26	100	11	27	100	11	22	45	15
As	1.6	4.6	0.08	1.7	4.6	0.08	1.3	2.2	0.72
Se	0.64	1.4	0.06	0.63	1.4	0.08	0.65	1.0	0.06
Br	7.8	17	2.7	7.7	16	2.7	8.5	17	3.3
Rb	2.3	19	0.10	2.2	19	0.10	2.6	11	0.25
Sr	5.9	31	1.0	6.2	31	1.0	4.7	10	1.6
Y	0.82	1.6	0.19	0.85	1.6	0.28	0.70	1.3	0.19
Zr	3.4	20	0.30	3.6	20	0.30	2.6	4.5	0.71
Mo	2.4	7.3	0.37	2.4	7.3	0.37	2.5	5.2	1.2
Pb	3.9	7.7	1.1	4.5	7.7	2.3	2.7	4.5	1.1
BC	4300	8800	2200	4400	8800	2200	3700	5400	2800
Sea salt	700	2100	170	640	1500	170	950	2100	370
	2.0 %	6.9 %	0.2 %	1.8 %	6.9 %	0.2 %	2.9 %	5.6 %	1.3 %
Ammonium sulfate	5500	11,000	2400	5400	11,000	2400	5800	8900	3100
	15 %	28 %	2.2 %	14 %	25 %	2.2 %	19 %	28 %	6.5 %
Soil	13,000	65,000	4400	14,000	65,000	4400	9300	17,000	4600
	33 %	53 %	16 %	31 %	53 %	16 %	28 %	36 %	21 %
POM	6000	16,000	2500	6300	16,000	3300	5200	7600	2500
	16 %	29 %	8.0 %	16 %	29 %	8.0 %	16 %	21 %	11 %
RCM	78 %	101 %	63 %	78 %	101 %	63 %	80 %	94 %	66 %

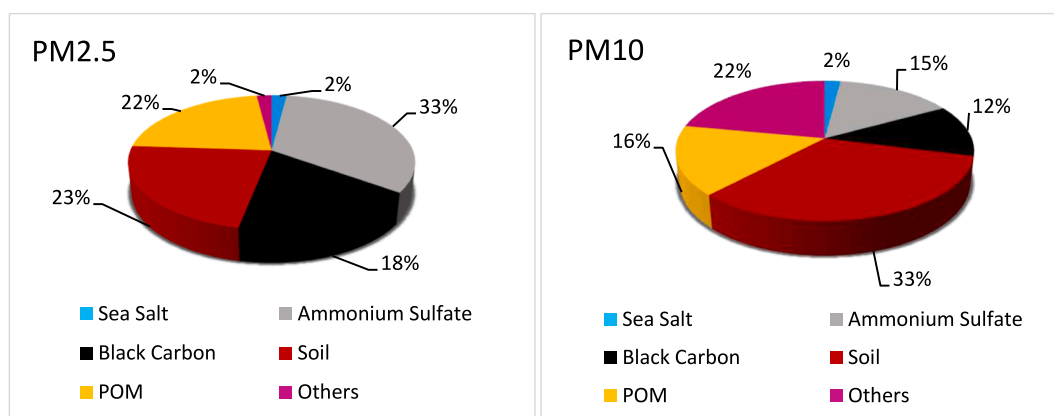


Fig. 5. Identification of the major macro-components for PM_{2.5} (left) and PM₁₀ (right) during the sampling period in Amman.

co-located sampling of the PM₁₀ and PM_{2.5} fractions allowed us to determine the division of the weighted mass: the average fraction ratio and its standard deviation was PM_{2.5}/PM₁₀ = 0.50 ± 0.33. Figs. 3 and 4 show the variation of BC content in PM_{2.5} and PM₁₀ samples, respectively. It is clear that both fractions, as well as their BC content, recorded higher concentrations during workdays than during weekends; and this is expected because human activities are strongly reduced during

weekends. The mass concentrations of both fractions exceeded the standard limits of the annual mean and the 24-h mean, according to the old and new WHO air quality guidelines [32].

3.2. PIXE and PESA results: quantitative elemental analysis

The values of the mean, maximum, and minimum elemental

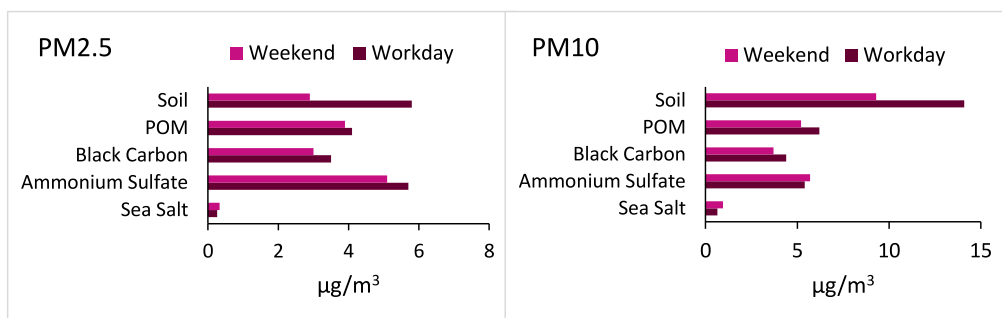


Fig. 6. The variation of mean concentrations of the estimated chemical components in PM_{2.5} (left) and PM₁₀ (right) during weekends and workdays in Amman.

concentrations in ng/m³, in the fine and coarse particulates, during the whole sampling period as well as during workdays and weekends, are summarized in Tables 2 and 3.

3.3. Chemical components and PM mass reconstruction

Using concentrations of the detected elements, chemical components can be estimated by multiplying the elemental concentration(s) by a molar correction factor (these factors are usually calculated by dividing the total molar weight of a specific component over the molar weight of an element) [13]. In this work, POM was estimated using H and S concentrations obtained by PESA and PIXE, respectively, with a conversion factor f of 19.0 for summer and 15.4 for winter samples (see Section 2.5), while sea salt, ammonium sulfate and soil [13] were estimated from PIXE measurements using the following equations (brackets indicate the concentration of each element/component):

$$[\text{POM}] = f \{ [\text{H}] - 0.25 [\text{S}] \}$$

$$[\text{Sea salt}] = 2.54 [\text{Na}]$$

$$[\text{Ammonium sulfate}] = 4.125 [\text{S}]$$

$$[\text{Soil}] = 2.20 [\text{Al}] + 2.49 [\text{Si}] + 1.63 [\text{Ca}] + 1.94 [\text{Ti}] + 2.42 [\text{Fe}]$$

The values of the mean, maximum, and minimum concentrations (in ng/m³) for the calculated chemical components in fine and coarse particulates are reported in Tables 2 and 3, respectively. Fig. 5 shows the percentage contribution of the estimated components, and the variation of mean concentrations during workdays and weekends is shown in Fig. 6.

The results reveal that soil is the dominant component in coarse particulates, followed by organics and ammonium sulfate, whereas fine particulates are dominated by ammonium sulfate, followed by soil and organics. BC is more abundant in fine particulates, indicating that most of BC content in PM₁₀ comes from PM_{2.5} (as PM_{2.5} mass is already included in PM₁₀). On average, concentrations of soil, POM, and BC, in fine and coarse particulates, have a similar trend of variation between workdays and weekends, recording higher concentrations in workdays compared to weekends, while ammonium sulfate in PM₁₀ tends to be slightly higher in weekends than in workdays. Sea salt is a minor source (1–2 %) in both fractions, and this is expected as Amman is a non-coastal city.

A typical validation check in aerosol composition studies can be performed by comparing the measured gravimetric mass to a reconstructed mass (RCM) [13,33] defined as the sum of the calculated chemical components and of the remaining elemental concentrations “traces”, for instance trace metals, that were not included in components calculation. As such, RCM is expressed as:

$$[\text{RCM}] = [\text{Sea salt}] + [\text{Ammonium sulfate}] + [\text{Soil}] + [\text{BC}] + [\text{POM}] + [\text{Trace metals}]$$

The difference between the measured and reconstructed mass is

denoted by “others” in Fig. 5 and considered as unexplained mass. The unexplained mass may be attributed to measurement uncertainties, improper conversion factors, missing or underestimated sources, and particle-bound water [33–35]. On average, as can be seen in Table 2, Table 3 and Fig. 5, RCM accounted for about 98 % of PM_{2.5} and 78 % of PM₁₀. Unexplained mass was thus about 2 % and 22 % in fine and coarse fractions, respectively. Lower PM₁₀ RCMs may be attributed, for instance, to the use of H_{org}-to-POM conversion factors that were determined for the PM_{2.5} fraction [25].

4. Conclusions

A set of fine (PM_{2.5}) and coarse (PM₁₀) particulate matter samples collected on Teflon filters from Amman, Jordan, was investigated using gravimetric, BC, PIXE and PESA measurements. Mass, black carbon and elemental concentration values were measured for the samples, and compared during workdays and weekends. Further analysis including estimation of various chemical components (namely, particulate organic matter, sea salt, ammonium sulfate and soil) and mass reconstruction was also performed. A reasonable PM_{2.5} and PM₁₀ mass reconstruction was accomplished: RCM yielded about 98 % of PM_{2.5} and 78 % of PM₁₀. This study reports for the first time the use of PESA techniques for hydrogen quantification, as a useful marker for organic compounds that are undetected by PIXE, and results one of the major components after soil dust in particulate matter samples from Amman. The obtained results, combined with PIXE and BC data, allowed to improve the reconstructed mass of Jordan samples by at least 30 % compared to previous studies. This in turn provides further information on air pollution fingerprints of both fine and coarse particles in Amman’s atmosphere. A proper source apportionment study using receptor models such as Positive Matrix Factorization (PMF) is planned after analysing a larger set of samples.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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