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Research Article

Characterization of size segregated PM_{10} at a rural site in the Eastern Mediterranean

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ABSTRACT

Size-resolved PM_{10} samples ($PM_{10-2.5}$ and $PM_{2.5}$) were collected at a rural site located in the northeastern Mediterranean between August 2010 and April 2013. A total of 461 PM_{10} aerosol samples (461 $PM_{2.5}$ and 461 $PM_{10-2.5}$) were analysed for major and trace elements (Na, Mg, Al, Si, S, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Zn, As, Ba and Pb) and anions (Cl^-, NO_3^- and SO_4^{2-}). Crustal elements and sea salt particles were found to be mainly associated with the coarse fraction (>60%), whereas SO_4^{2-} and NO_3^- were predominantly found in the fine mode (>80%). The crustal originated species exhibited very strong correlations (r>0.9) in the coarse fraction and PM_{10}, while their correlations were slightly lower in the fine fraction (0.5<r<0.8). Concentrations of most aerosol species exhibited well-defined seasonal variation with higher concentrations in summer than winter. The difference between summer and winter concentrations were not significant for As, Ca, Cr, Pb and NO_3^-. During the sampling period, 11 specific Saharan dust events were identified.

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INTRODUCTION

Ambient particulate matter (PM) in the atmosphere has been great concern of scientific research mainly due to its impact on health [1], climate [2], ecosystems [3], visibility [4] and materials [5]. PM can be emitted into the atmosphere from numerous natural and anthropogenic sources. Therefore, particle population in the atmosphere is composed of mixture of particles between 0.01 and 100 µm with varying chemical composition. Due to chemical complexity, PM is generally characterized by its size (aerodynamic diameter). PM with an aerodynamic diameter less than 10 µm and 2.5 µm are called as PM_{10} and $PM_{2.5}$ (also known as fine particles), respectively. The fraction between 2.5 and 10 µm comprised the coarse particles and designated as $PM_{10-2.5}$ ($PM_{2.5}$ + $PM_{10-2.5}$ =PM_{10}).

Mediterranean Basin (MB), characterized by particular atmospheric dynamics [6], can be considered as a crossroad of air masses originating from Europe, Africa and Asia [7]. Anthropogenic emissions over Europe, frequent dust intrusions from Northern Africa and the intensity of solar radiation result in complex and peculiar aerosol phenomenology over the Mediterranean region [8]. Owing to different atmospheric dynamics, climatic patterns, geographic characteristics and various natural and anthropogenic sources, air quality across the MB, exhibits remarkable spatial and temporal variability [6]. Therefore, there are considerable number of studies on air quality degradation by PM in different locations across the MB [8-28].

In this study, comprehensive long term size segregated PM_{10} measurements were performed at a rural site in the south eastern coast of Turkey. Up to our knowledge, reported long term PM_{10} measurements were performed at least two decades ago in the Eastern Mediterranean (EM) Basin of Turkey [17], [19]. This study will focus on PM_{10} chemical composition to understand PM_{10} composition and its size

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distribution, and to evaluate the impact of Saharan dust intrusions on PM_{10} chemical composition between 2010 and 2013 years. Furthermore, this study will provide assessment of variation of PM_{10} chemical composition across the EM by providing comprehensive data set for comparison purposes.

MATERIALS AND METHODS

Study Area

Daily size segregated PM_{10} samples were collected between 10 August 2010 and 10 April 2013 at a rural site in the Eastern Mediterranean (36^0 58' 12" N and 30^0 26' 2" E), which is approximately 20 km from the coast and 26 km from city of Antalya (one of the largest cities on the Mediterranean coast of Turkey, with population of two million) and further away from other important settlement centers. There are no adjacent anthropogenic sources around the station. Altitude of the station is 484 m above sea level. Location of the sampling station is depicted in Figure 1. The climate, classified as typical Mediterranean climate, is characterized by warm and rainy winters and hot and dry summers. The mean temperature is 9.6 °C and 28 °C in winter and summer, respectively. Precipitation occurs in winter and randomly seen in summer.

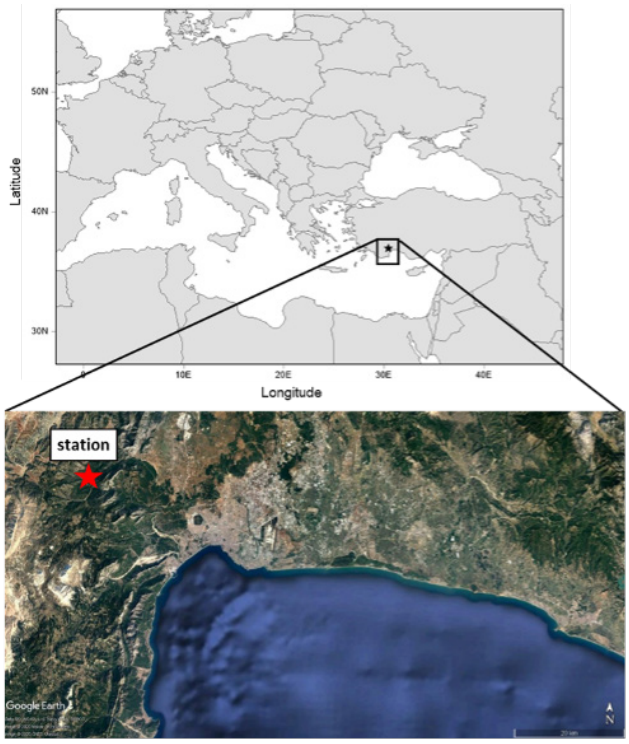

Figure 1. Study area and location of the sampling station

Sampling

A Gent-PM_{10} stacked filter unit (SFU) was used to collect PM_{10} samples in two size ranges, namely coarse ($PM_{10-2.5}$) and fine ($PM_{2.5}$), with a temporal resolution of 24 h. The details of the SFU design and performance characteristics can be found in [29]. Forty-seven mm diameter polycarbonate filters (Nuclepore) with 8.0 μm and 0.4 μm pore sizes, placed sequentially in the filter holder, were used to collect coarse and fine fractions, respectively. A preimpactor with 10 μm cut-point was used to stop particles with diameters larger than 10 μm before they reach to the coarse filter. A pump with a mass flow controller was used to keep the flow rate constant (≈ 16.7 L/min) during the sampling. Before and after sampling, filters were conditioned at constant relative humidity and temperature for at least 24 h before they were weighted using a microbalance (Sartorius, Model MC-5). Between 10 August 2010 and 10 April 2013, a total of 461 daily fine ($PM_{2.5}$) and 461 daily coarse ($PM_{10-2.5}$) samples were collected. As PM_{10} samples are the sum of fine ($PM_{2.5}$) and coarse ($PM_{10-2.5}$) samples, during the sampling period 461 daily PM_{10} samples were collected in this study.

Analysis

Elemental composition of collected samples (922 samples = 461 fine + 461 coarse) were analysed using an energy-dispersive X-ray fluorescence (EDXRF) at the Ankara Nuclear Research and Training Center. Samples were analyzed for a total of 17 elements including Na, Mg, Al, Si, S, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Zn, As, Ba and Pb. A silver anode X-ray tube (I_{max}= 1000 μA, V_{max}=50 kV), a sample chamber, an amplifier, and a liquid nitrogen-cooled lithium-drifted silicon solid-phase detector was used. Four different measuring conditions were applied with an analysis time of 17 minutes/sample as given in Table 1. NIST certified reference filter (SRM 2783-Air Particulate Matter on Filter Media) was used for calibration of EDXRF and checking its accuracy. QA/QC results corresponding to EDXRF measurements are given in Table 2.

After EDXRF analysis (a non-destructive technique), filters were extracted ultrasonically with 15 ml of MQ-grade water and then analyzed by Dionex DX-120 Model Ion Chromatography (IC) to determine major anions (Cl^-, NO_3^- and SO_4^{2-}). The separation was performed using Dionex AG9-HC separation column and an ASRS-ULTRA suppressor with a 10 mM solution of Na_2CO_3 at a flow rate of 1.0 mL min^{-1}. The analytical protocol is described in [30].

Field and laboratory blanks were analysed by EDXRF and IC. Blank subtraction was applied before calculating atmospheric concentrations for each element and anions. Field blanks were collected by air drawn into sampler only for 1 min.

Table 1. Measurement conditions in the EDXRF analysis

Parameter	Condition			
	VLE	S	St	ME
Tube Voltage (kEV)	2.5	10	15	22.6
Tube Current (mA)	900	900	1000	494
Analysis Time (s)	150	100	100	100
Energy Range Analysed	Na - K	K - Cr	Cr - Fe	Fe - Mo
Elements Analysed	Na, Mg, Al, Si, S	K, Ca	Ti, V, Cr, Mn	Fe, Ni, Zn, As, Ba, Pb

VLE: Very light elements; S : Solids; St : Steels; ME : Medium elements

Table 2. QA/QC data for EDXRF analysis

Element	Certified (ng filter^{-1})	Measured (ng filter^{-1})	Error %	MDL (ng/cm^2)
Na	1860 ± 100	1760 ± 70	5	40
Mg	8620 ± 520	9800 ± 1170	14	35
Al	23210 ± 30	24138 ± 483	4	122
Si	58600 ± 600	59186 ± 720	1	256
S	1050 ± 260	915 ± 165	13	15
K	5280 ± 520	4900 ± 590	7	59
Ca	13200 ±1700	12400 ± 372	6	136
Ti	1490 ± 240	1400 ± 98	6	13
V	48.5 ± 6	53.8 ± 7	11	2.3
Cr	135 ± 25	147 ± 17	9	7.5
Mn	320 ± 12	330 ± 13	3	6
Fe	26500 ± 1600	28100 ± 1680	6	89
Ni	68 ± 12	73 ± 17	7	4.6
Zn	1790 ± 130	1660 ± 232	7	16
As	11.8 ± 1.2	12.4 ± 1.5	5	1.1
Ba	335 ± 50	309 ± 32	8	3.7
Pb	317 ± 54	325 ± 23	3	10

RESULTS AND DISCUSSIONS

General Characteristics of Data

The average and median concentrations of measured pollutants in the fine ($PM_{2.5}$) and coarse ($PM_{10-2.5}$) particles, and PM_{10} (sum of fine and coarse fraction) are presented in Table 3. Elemental S concentration (EDXRF) was found to be about one third of SO_4^{2-} (IC) and indicating that almost all S is in the form of SO_4^{2-} salts. Therefore, only SO_4^{2-} concentration is reported in Table 3.

Size distributions of measured species in the coarse and fine fractions are presented in Figure 2. Crustal (Ca, Mg, Ti, Al and Si) and marine elements (Cl^- and Na) were associated mainly with coarse particles (> 60%), while SO_4^{2-} and NO_3^- were predominantly found in the fine mode (> 80%). Concentrations of V, Fe, Mn and Ba in the coarse fraction were higher (by a factor of 1.3 to 1.9) than the corresponding concentrations in the fine fraction. Cr, K, Zn and Ni were equally distributed in the coarse and fine fractions with $PM_{2.5}/PM_{10}$ ratios from 0.47 to 0.53. Cr, Zn and Ni are well-known anthropogenic pollutants and expected to be confined in the fine mode, however soil along the Mediterranean coast of Turkey is highly enriched with these elements [31] and shifting their mass median diameter to > 2.5 μm. Significant proportion of Pb and As measured in this study were found in the coarse fraction. Occurrence of such high fractions of As and Pb in the coarse mode was also reported by [32] and attributed to sticking of fine anthropogenic particles onto coarse particles.

Table 3. Statistical descriptions of the elemental and ionic concentrations (ng m^{-3}) measured in $PM_{10-2.5}$, $PM_{2.5}$, and PM_{10} collected over the period of August 2010 and April 2013

	$PM_{10-2.5}$			$PM_{2.5}$			PM_{10}		
	N	**Mean**	**Median**	**N**	**Mean**	**Median**	**N**	**Mean**	**Median**
SO_4^{2-}	460	614 ± 745	365	460	2990 ± 1985	2462	460	3603±2249	2989
NO_3^-	367	228 ± 261	134	367	1261 ± 1164	897	367	1489±1285	1113
Cl^-	423	1179 ± 1198	866	423	616 ± 740	401	423	1795±1734	1410
Na	461	920 ± 1045	607	461	450 ± 538	260	461	1370±1458	906
Mg	381	366 ± 462	248	381	136 ± 170	82	381	502±597	347
Al	459	310 ± 462	183	459	141 ± 252	74	459	451±660	273
Si	459	1053 ± 1667	608	459	481 ± 724	265	459	1533±2245	929
K	461	135 ± 157	93	461	125 ± 133	88	461	260±267	193
Ca	456	1058 ± 1206	781	456	295 ± 362	199	456	1353±1515	1017
Ti	461	16 ± 25	11	461	7.0 ± 11	3.8	461	23±34	15
V	461	1.9 ± 2.0	1.3	461	1.1 ± 1.2	0.66	461	3.0±2.9	2.1
Cr	433	3.1 ± 9.7	1.5	433	2.7 ± 13	1.3	433	5.8±21	3.1
Mn	438	4.0 ± 5.0	2.7	438	2.8 ± 3.3	1.8	438	6.8±7.6	5.2
Fe	456	153 ± 264	92	456	94 ± 124	60	456	247±355	169
Ni	380	1.2 ± 1.6	0.73	380	1.4 ± 2.2	0.91	380	2.6±3.3	1.7
Zn	460	5.5 ± 5.1	4.3	460	5.0 ± 4.4	3.9	460	10.5±8.2	8.7
As	461	0.23 ± 0.21	0.19	461	0.33 ± 0.25	0.26	461	0.56±0.40	0.47
Ba	459	4.3 ± 5.0	3.0	459	3.2 ± 3.3	2.2	459	7.4±7.5	5.5
Pb	301	5.2 ± 4.9	4.5	301	6.9 ± 5.5	5.7	301	12±9.7	10

Concentrations of the measured species in this study were compared to the ones reported in the literature over the Mediterranean Basin. Table 4 presents the studies conducted in the same region of Turkey and Eastern and Western Mediterranean [8, 10, 17, 19, 24, 28, 33]. Note that rural and regional background stations with long-term PM_{10} measurements were selected for comparison purpose.

Na and Cl^- concentrations in this study were lower than those conducted near the coast. Sea spray emissions (Na and Cl^-) are consisted of large particles, hence they are quickly scavenged out from the atmosphere. With this rapid removal process, sea spray contribution can vary substantially from one place to another depending on the distance to the coast. The sampling station in this study is nearly 20 km away from the coast with an altitude of 484 m a.s.l. which may limit the arrival of sea spray.

Very high SO_4^{2-} concentrations (5-6 μg m^{-3}) were reported for the EMB [17, 19, 33], however those studies conducted nearly 2 decades ago. Due to application of effective SO_2 emission control measures in Europe, SO_4^{2-} levels in the EMB has been decreasing [34, 35, 36]. SO_4^{2-} level in this study is still high (3.6 μg m^{-3}) but lower than those reported decades ago. NO_3^- level in this study which is 1.5 μg m^{-3} is comparable to those reported in the EMB. Although there are precautions to reduce NO_x emissions throughout the Europe, decrease of NO_3^- levels is not strong as in SO_4^{2-} [37, 38, 39, 40].

Levels of trace elements, especially those related with anthropogenic activities, namely V and Ni, were comparable and even slightly lower than those measured in the EMB. $PM_{2.5}$/PM_{10} ratios of V and Ni were 0.36 and 0.53, respectively. This indicates that majority of V and significant portion of Ni were confined in the coarse mode which is inline with the previous studies conducted in the EMB and other regions of Turkey [32, 41, 42, 43].

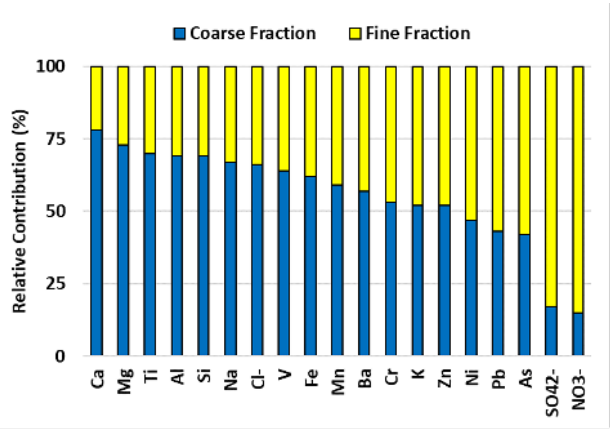

Figure 2. Relative contribution of aerosol species in the fine ($PM_{2.5}$) and coarse ($PM_{10\text{-}2.5}$) fraction to PM_{10}

Crustal Enrichment Factor (EF_c)

Enrichment factor (EF) which is a double normalization technique, is generally used for the preliminary source apportionment of aerosol species in the atmosphere. The EF of an element is calculated by Equation 1 [44, 45] in where C_x represents the concentration of the target element and C_R represents the concentration of the reference element which can be used as a marker of its source. The ratio of the C_x to C_R in aerosol sample to that in the reference source constitutes the double normalization. In order to determine the extent of contribution of anthropogenic sources to elemental composition of PM_{10} and its size resolved components (i.e. fine ($PM_{2.5}$) and coarse ($PM_{10\text{-}2.5}$)), crustal enrichment factor (EF_c) values for each element were calculated. For the EF_c calculations, Mason's soil composition [46] was used as the reference crustal source and Al as the reference soil element assuming that the only source of Al is soil.

$$EF = \frac{\left(\frac{C_x}{C_R}\right)_{aerosol}}{\left(\frac{C_x}{C_R}\right)_{Reference}} \quad (1)$$

The calculated EF_c values are presented in Figure 3. Elements with EF_c values ≈ 1.0 were considered as purely crustal originated and hence non-enriched. Deviations from unity indicates contribution of non-crustal sources on measured concentrations of that element. Elements with EF_c values between 1.0 and 10 were considered as moderately enriched. These moderately enriched elements are called as elements with mixed origin due to comparative contributions of crustal and non-crustal sources to their observed concentrations. Elements with EF_c >10 were regarded as highly enriched as these elements are associated with non-crustal sources.

The EF_c values calculated for each element showed slight to large variation in each size range, but these variations did not change their classifications (Figure 3). Therefore, throughout the manuscript sum of size resolved fractions, namely PM_{10} is discussed. Based on the above threshold values, Fe, Si and Ti were observed to be entirely of crustal origin. Mn, K, Ba, Mg, V, Ni, Cr, Ca and Na were found to be mixed- originated, while Zn, As, Pb, S and Cl^- were exclusively originated from non-crustal sources. Note that the mineralogy of Mediterranean coast of Turkey is characterized by high Cr and Ni levels due to ophiolitic rocks hence high concentrations of Cr and Ni were reported not only in the atmosphere but also in sediments [31, 47, 48].

Correlations Between PM_{10} Chemical Components

The interrelationships of pollutants measured in PM_{10} and its size resolved components (i.e. fine ($PM_{2.5}$) and coarse ($PM_{10\text{-}2.5}$)) were assessed by Pearson Correlation Analysis. The elements displaying significant correlations (at 99 or 95% confidence interval) with each other is assumed to have common sources and/or they are exposed to similar atmospheric conditions and chemical processes. For this analysis, 4 categories are defined based on absolute values of correlation coefficients:

$$|r| = \begin{Bmatrix} \leq 0.3 & weak \\ 0.3 - 0.5 & moderate \\ 0.5 - 0.8 & strong \\ \geq 0.8 & very\ strong \end{Bmatrix}$$

The correlations between crustal related elements were either strong or very strong in PM_{10} and the coarse fraction ($PM_{10\text{-}2.5}$), while these correlations are strong and/or moderate in the fine fraction. In the fine fraction, no pollutants showed very strong correlations with each other. Apart from these, no significant differences were found in the correlation analysis of size resolved fractions. Therefore, the interrelationships of pollutants measured in PM_{10} are discussed and presented in Table 5.

Crustal related elements, Mg, Al, Si, K, Ca, Ti, Mn, Fe and Ba depicted strong correlations (r>0.6) among themselves. Na showed highest correlation with Cl^- (r=0.64) and indicating that sea spray emission is the main source of Na and Cl^-.

Table 4. Comparison of PM_{10} chemical composition with the literature reported values (concentrations are in ng m^{-3})

	EMB						WMB		
	Studies in Turkiye								
	This study	Ozturk, 2012	Kocak ,2007	Andree, 2002	Theodosi et al., 2011	Querol et al., 2009a	Cerro et al., 2020	Querol et al., 2009a	Rodriguez et al., 2002
Type of site	Rural	Rural	Rural	Arid	RB	RB	RB	RB	Rural
	Antalya	Antalya	Erdemli	Israil	Greece	Greece	Spain	Spain	Spain
Period	2010-2013	1993-2001	2001-2002	1995-1998	2005-2006	2004-2006	2010-2012	2004-2007	1999-2000
SO_4^{2-}	3603	7927	4953	8000		5500	3000	2600	3877
NO_3^-	1489	1285	1857			1700	1200	1700	2177
Cl^-	1795	1885	5492	1127			2400		349
Na	1370	2241	3434a	1187			1200		294
Mg	502	538	489a	919			200		117
Al	451	661		1867					398
Si	1533			3786					
K	260	536	360	589			100		203
Ca	1353	1422	1888	6538			500		930
Ti	23	32	27	123			19		20
V	3.0	3.2	8.7	9.9	6	8	3.6	4	2
Cr	5.8	6.8	5.7		7	9	2	1	
Mn	6.8	8.9	7.6	24.2	17	12	4.2	5	5
Fe	247	400	351	1245	987		200		222
Ni	2.6	2.5	3.7	5.7	4	4	2.5	2	
Zn	10.5	17	9.7	32.5		29	10.8	12	
As	0.56	0.82					0.2	0.3	
Ba	7.4	11					3.6	6	
Pb	12	52		21.3	12	9	2	4	10

[a] Ionic concentrations were reported; RB: Regional background; EMB: Eastern Mediterranean Basin; WMB: Western Mediterranean Basin

Table 5. Correlation coefficients among PM_{10} components

	Na	Mg	Al	Si	K	Ca	Ti	V	Cr	Mn	Fe	Ni	Zn	As	Ba	Pb	Cl^-	SO_4^{2-}	NO_3^-
Na	1																		
Mg	.203**	1																	
Al	.104*	.874**	1								very strong (>=0.8)								
Si	.104*	.874**	1.000**	1							strong (0.5-0.8)								
K	.159**	.798**	.774**	.774**	1						moderate (0.3-0.5)								
Ca		.898**	.922**	.922**	.860**	1					weak (<0.3)								
Ti	.093*	.796**	.962**	.962**	.650**	.944**	1				no significant								
V	.310**	.347**	.494**	.494**	.437**	.445**	.501**	1											
Cr		.423**	.222**	.222**	.697**	.311**			1										
Mn	.133**	.790**	.862**	.862**	.634**	.785**	.794**	.444**	.154**	1									
Fe	.093*	.888**	.980**	.980**	.886**	.954**	.996**	.497**	.252**	.808**	1								
Ni		.174**	.208**	.208**	.265**	.245**	.215**	.381**	.328**	.223**	.225**	1							
Zn	.247**	.178**	.294**	.294**	.296**	.251**	.268**	.696**		.346**	.275**	.419**	1						
As	.104*	.132*	.156**	.156**	.187**	.216**	.169**	.521**		.188**	.174**	.373**	.612**	1					
Ba	.231**	.655**	.734**	.734**	.640**	.757**	.735**	.631**	.143**	.715**	.751**	.308**	.517**	.470**	1				
Pb		.265**	.239**	.239**	.167**	.137*			.289**	.551**		.193**	.242**	.122*	.336**	1			
Cl^-	.635**	.173**			.135**									-.170**			1		
SO_4^{2-}	.152**		.240**	.240**	.265**	.173**	.244**	.638**		.236**	.239**	.220**	.396**	.251**	.279**	.118*		1	
NO_3^-	.202**	.293**	.351**	.351**	.428**	.321**	.328**	.516**	.299**	.358**	.324**	.247**	.340**	.195**	.388**	.203**	.247**	.525**	1

Pb is strongly correlated with Mn (r=0.55) and weakly correlated (r<0.2) with Zn and crustal elements such as Al, Si and Fe. As discussed before, Mn is a mixed originated element and could be both from naturally occurring crustal material and non-crustal sources such as gasoline antiknock additives [49]. Moreover, dust from tire wear contains significant amounts of Zn [50]. This would indicate that Pb in this study is both due to vehicular emissions and non-exhaust vehicle emissions i.e. resuspension of road dust.

SO_4^{2-} depicted strong correlation with V (r=0.64) and NO_3^- (r=0.53), moderate correlation with Zn (r=0.4) and weak correlation with crustal-related and anthropogenic elements. Vanadium showed strong correlation with Zn, SO_4^{2-}, Ba, NO_3^- and As (r>0.5), moderate correlation with crustal elements and Ni. Vanadium and Ni are usually considered as markers for residual/fuel oil combustion, such as power plants and oil refineries or oil burned in marine vessels [51, 52] and hence expected to have strong correlation. Moderate correlation between these two elements is probably due to this common source. Arsenic, a tracer of coal combustion, demonstrated strong correlation with Zn (r=0.6) and V (r=0.52). These elements are also emitted from coal combustion processes [53].

Seasonal Variation of PM_{10} Chemical Components

Statistical summary of the seasonal variations of the pollutants are given in Table 6. Colder months with high precipitation represent the winter season (from October to March) and hotter and dryer months represents the summer season (from April to September). The seasonality of pollutants was tested by the Median test at 95% confidence levels. The median concentration of all pollutants, except As, Ca, Cr, Pb and NO_3^-, showed a clear seasonal variation. The lack of seasonality, especially for those originated from anthropogenic sources, is also reported in Eastern and North-western Mediterranean [16, 54, 55].

The summer to winter (S/W) ratio of pollutant concentrations in PM_{10} and its size resolved components (Figure 4) indicated that emission strength of As, Pb and Cr (S/W ratios are around 1.0 for all size ranges) are higher in winter and/or these pollutants are produced locally so that they are less efficiently scavenged during rain events. Arsenic is a well-known marker for coal combustion [56]. Coal is used in the region for domestic heating and heating the greenhouses during cold months. Local weather conditions in winter, generally characterized by weak surface winds and low mixing heights, may limit the mixing and dispersion of pollutants, thereby favoring the accumulation of local pollution [57]. This could be the reason for observing higher As, Pb and Cr concentration in winter season as Pb and Cr is also emitted from coal combustion processes [58]. Coal combustion is still common in Eastern Europe, Russia and Turkey, therefore long range and regional transport of coal power plants emissions can impact the station in summer when rainfall is less. Note that Pb can also be emitted from non-exhaust traffic emissions [59] while Cr can be emitted from resuspension of local soil during summer months. This duality is probably the main reason for the lack of seasonal variation for As, Pb and Cr.

Calcium is a mixed-originated element and contribution of crustal sources (i.e. soil resuspension) is dominant in dry months. Anthropogenic sources (i.e. road dust, construction activities and mining activities) may also contribute observed Ca concentrations [60]. Mining activities around the region could be the main reason for the non-seasonality observed in Ca and Cr concentrations.

Crustal elements, Si, Al, Fe and Ti, exhibited the highest S/W ratios (>2.0) in all size fractions. Higher summer concentration of these elements can be attributed to both elevated dust resuspensions of local and regional origin and low rainfall in summer season.

Magnesium, Ba and Zn also have high S/W ratio (> 1.0). They showed similar pattern like crust originated species. Except Zn, all have mixed sources. Zinc is an anthropogenic element and can be both emitted from motor vehicles and industrial emissions [61, 62].

Those having mixed sources i.e., Na, Mn, K, Ni and V also had high S/W ratio. Higher S/W ratio of Na and Mn in the coarse fraction could be explained by higher contribution of crustal sources on these elements due to enhanced resuspension of dust as soil is mostly dry in summer. Moreover, lack of precipitation could favour long range transport of K, Ni and V hence increased their summer concentrations in the fine fraction [63, 64].

Calculated S/W ratios for SO_4^{2-} is around 1.5 in each size fraction due to enhanced photochemistry (high solar radiation and temperature) and the potential pollutant transport by long range transport in summer months. Previous studies in EMB and WMB also reported the same seasonal pattern for SO_4^{2-} [8, 19].

The S/W ratio for NO_3^- is around 1.2 in each size fraction and indicating higher concentrations during summer. This distinct seasonal pattern for NO_3^- is also reported in the previous studies conducted in EMB of Turkey [19, 55].

Apart from crustal material, the main source of Na could be sea spray emissions. Sodium and Cl^- in PM_{10} showed S/W ratio of 1.36 and 1.32, respectively. As these constituents are generally used as marker of sea spray emission, they are expected to be higher in winter and in the coarse fraction. Because sea spray emission has the highest source strength in winter due to unsettled weather. However, in this study, the strength of marine emissions is found to be lower due to the long distance between the station and the coast. In winter season, strong correlation (r=0.80, p<0.01) was observed (not presented) between Na and Cl^-, this implies that sea spray is the main source for both Na and Cl^-. The correlation is moderate (r=0.54, p<0.01) in summer. Cl^-/Na ratio in winter and summer is calculated as 2.04 and 1.59, respectively. This indicates that despite chloride depletion, sea spray emission is still common source for Na and Cl^- in summer [65, 66].

Table 6. Statistical summary of the seasonal variations of PM_{10} components (in ng m^{-3})

	Summer			Winter		
	N	**Mean**	**Median**	**N**	**Mean**	**Median**
SO_4^{2-}	281	4168 ±2441	3615	179	2716 ±1542	2387
NO_3^-	263	1644 ±1428	1205	104	1098 ±679	958
Cl^-	270	1820 ±1549	1547	153	1750 ±2025	1168
Na	282	1506 ±1479	970	179	1157 ±1401	711
Mg	241	555 ±706	394	140	410 ±315	312
Al	280	587 ±789	339	179	238 ±265	143
Si	280	1996 ±2683	1154	179	809 ±903	485
K	282	310 ±296	240	179	182 ±192	139
Ca	280	1543 ±1833	1064	176	1052 ±678	928
Ti	282	31 ±41	19	179	12 ±12.5	8
V	282	3.8 ±3.3	2.9	179	1.8 ±1.6	1.3
Cr	272	5.6 ±22	3.2	161	6.3 ±19	2.9
Mn	274	8.2 ±8.8	6.0	164	4.4 ±4.0	3.5
Fe	280	321 ±427	206	176	130 ±123	98
Ni	234	2.8 ±2.5	2.2	146	2.2 ±4.3	1.2
Zn	282	11.6 ±8.9	9.2	178	8.8 ±6.6	7.2
As	282	0.53 ±0.38	0.46	179	0.6 ±0.41	0.53
Ba	280	8.6 ±8.8	5.9	179	5.5 ±4.0	4.7
Pb	190	12.3 ±11.7	10.4	111	11.7 ±4.7	10.2

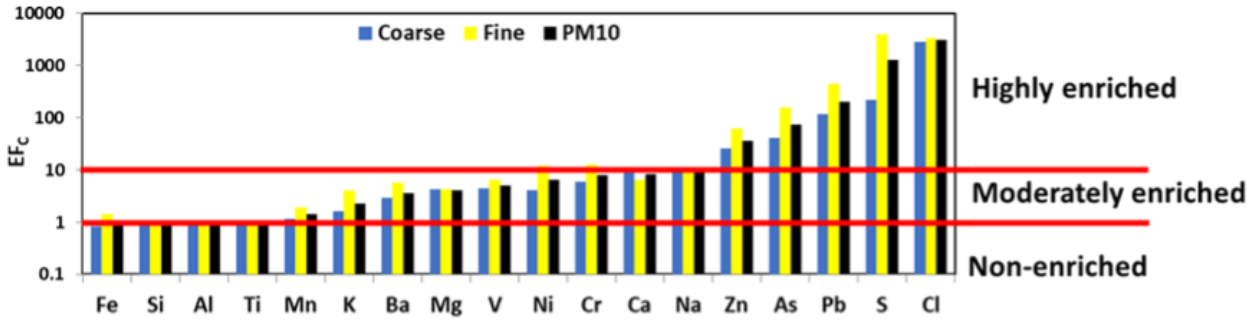

Figure 3. Crustal enrichment factor for the elements measured in $PM_{2.5}$, $PM_{10\text{-}2.5}$, and PM_{10}

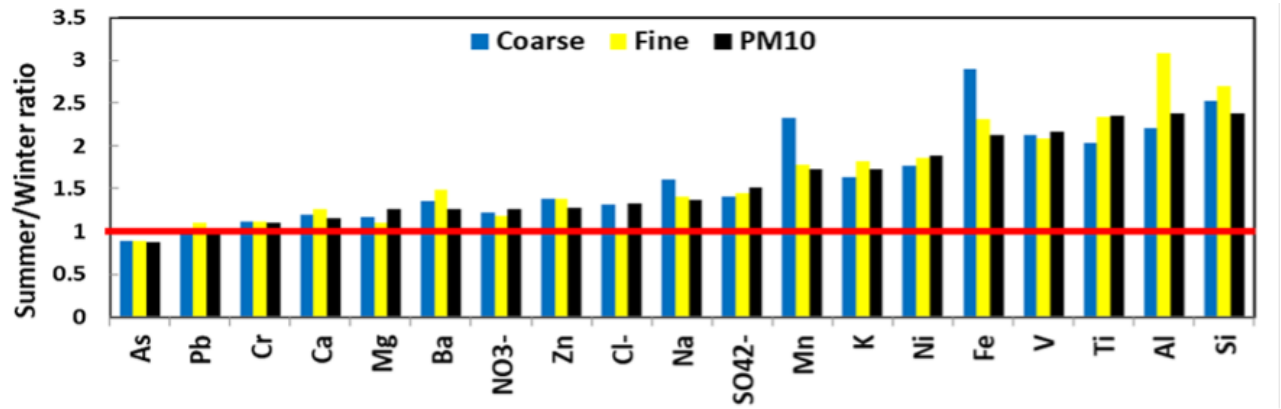

Figure 4. The ratio of summer and winter pollutant concentrations in $PM_{2.5}$, $PM_{10\text{-}2.5}$, and PM_{10}

Influence of Saharan Dust Transport on PM_{10} Chemical Composition

Mineral dust transported from North Africa and occasionally from the Middle East is known to affect the Mediterranean region [67]. Crustal elements, especially Al, Ca, Si, Ti and Fe, have been used as tracer of Saharan dust intrusions over the Mediterranean region [13, 17, 23, 65, 69, 70, 71]. In this study, 48 potential dust episode days were selected based on the temporal variation of Al, Fe and Ti concentrations. To identify dust episodes among these 48 episodes, 5 days backward trajectories (at 500, 1500 and 3000 m arrival heights) were calculated by HYSPLIT model [72] and satellite images were obtained. In order to assign the episode days as Saharan dust transport episode, at least one of the backward trajectories must originate from North Africa and satellite images must indicate dust loading over the sampling region as given in Figure 5. Keeping these criteria, 24 dust days were characterized by 11 dust events varying from 1 day to 6 days.

Saharan dust intrusions significantly increased the levels of crustal elements (Table 7). Dust to non-dust ratio for Al, Si, Ti and Fe which are generally used as tracer for dust transport, were in the range of 6.4 to 6.0. Concentrations of Cr, Ca, Mg, Mn, K and Ba were at least three times higher on dust days. Significant enhancement of these mixed–originated elements during dust events can be explained by their crustal origin. Enhancement of V and Ni were moderate (i.e. 1.9 and 1.3) compared to other mixed-originated elements. Anthropogenically derived species (NO_3^-, Zn, SO_4^{2-}, As and Pb) also showed enhancement on dust days with values ranging from 2 to 1.2. Mineral dust particles are in the coarse fraction and during the transport of air masses they can serve as a surface (reaction and/or adsorption) for the anthropogenically derived species which are dominant in the fine fraction. Previous studies conducted over Eastern Mediterranean region also reported enhancement of anthropogenic species on dust event days [69, 73]. Mild enhancement was detected in Na and Cl^- on dust days with a ratio of 1.2 due to air masses passing over the Mediterranean Sea before intercepting the sampling station.

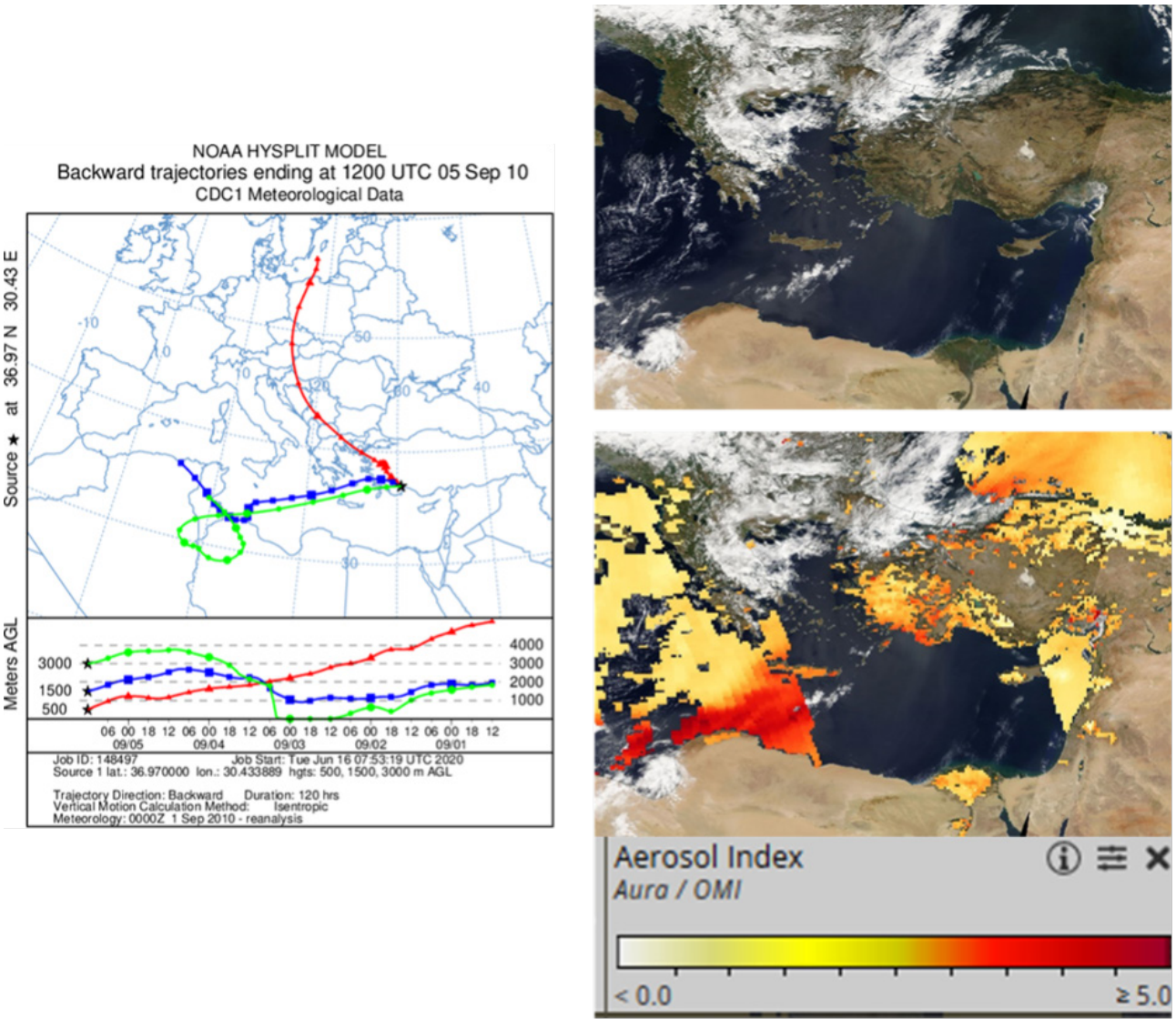

Figure 5. Corresponding backward trajectories and satellite image on episode day of 5 September 2010

Table 7. Summary statistics of PM_{10} on dust and non-dust days (in ng m^{-3})

	Dust day				Non-dust day			
	N	**Mean**	**STD**	**Median**	**N**	**Mean**	**STD**	**Median**
SO_4^{2-}	23	4378	2410	3568	437	3562	2236	2951
NO_3^-	21	2812	1703	2689	346	1409	1213	1069
Cl^-	23	2108	1505	1787	400	1776	1746	1395
Na	24	1596	1248	1361	437	1358	1468	901
Mg	23	1768	1705	886	358	420	299	324
Al	24	2238	1728	1500	435	352	339	266
Si	24	7611	5877	5099	435	1198	1154	906
K	24	816	678	623	437	230	182	183
Ca	23	5062	4596	3057	433	1156	755	991
Ti	24	113	96	77	437	19	17	15
V	24	5.6	3.8	4.8	437	2.9	2.8	2.0
Cr	23	22	75	6.2	410	4.9	12.5	3.0
Mn	24	23	17	17	414	5.8	5.3	4.9
Fe	23	1189	1034	733	433	197	170	161
Ni	21	3.4	2.4	2.9	359	2.5	3.3	1.7
Zn	24	16	8.9	15	436	10.2	8	8.4
As	24	0.69	0.27	0.72	437	0.55	0.4	0.46
Ba	24	20	17	15	435	6.7	5.9	5.3
Pb	18	15	3.6	14	283	12	10	10

CONCLUSIONS

Chemical composition of size segregated PM_{10} samples was investigated at a rural site in the northeastern Mediterranean. Crust (Ca, Mg, Ti, Al and Si) and sea salt (Na, Cl^-) originated species were found to be associated mainly with coarse particles (>60%), whilst secondary inorganic species (SO_4^{2-} and NO_3^-) were primarily found in the fine fraction (>80%). Cr, K, Zn and Ni were found to be equally distributed between coarse and fine fractions. SO_4^{2-} and NO_3^- levels were lower than previous studies (conducted at least two decades ago) and indicated decreasing trend for these pollutants over the Mediterranean region.

NO_3^-, As, Ca, Cr, and Pb concentrations did not present a clear seasonal trend, while crustal elements and SO_4^{2-} indicated significantly higher concentrations during summer. The lack of seasonality in the former pollutants were attributed to their local sources and long-range transport.

11 Saharan dust transport events were identified and found to significantly influence PM_{10} chemical composition. Although all species showed enrichment during Saharan dust incursions, crust-originated elements exhibited highest enhancement (over 6 times).

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DATA AVAILABILITY STATEMENT

The authors confirm that the data that supports the findings of this study are available within the article. Raw data that support the finding of this study are available from the corresponding author, upon reasonable request.

CONFLICT OF INTEREST

The author declared no potential conflicts of interest with respect to the research, authorship, and/or publication of this article.

USE OF AI FOR WRITING ASSISTANCE

Not declared.

ETHICS

There are no ethical issues with the publication of this manuscript.

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