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SIMILARITIES AND DIFFERENCES IN COMPOSITIONS OF EASTERN AND WESTERN BLACK SEA AEROSOL

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ABSTRACT

In this study daily aerosol samples were collected at two different sampling stations at the Black Sea Region of Turkey. Western Black Sea sampling station is located at the Bulgarian border of Turkey and approximately 50 km from the Black Sea coast. The station on the Eastern Black Sea is located at a high altitude site which is approximately 75 km from the Black Sea coast. Both stations are situated at relatively remote rural locations. Nearest large settlement area is at least 50 km away from both sites. At both sampling stations daily coarse ($PM_{2.5-10}$) and fine ($PM_{2.5}$) aerosol samples were collected on polycarbonate (Nuclepore) filters using Stacked Filter Units (SFU) between March 2011 and December 2012. Concentrations of eighteen elements were determined in collected aerosol samples by using an energy dispersive X-ray flurescence spectrometry. Measured concentrations of elements and ions varied between 0.46 ng m^{-3} for As and 2071 ng^{-3} for SO_4^{2-} for Eastern Black Sea atmosphere and 0.6 ng m^{-3} for As and 3415 ng^{-3} for SO_4^{2-} for Western Black Sea atmosphere. Concentrations of pollution derived elements were higher at the western part of the Black Sea, which is not surprising when the distribution of industrial sources around sampling location is considered. However, concentrations of some specific elements like Mn, Cr were higher at the Eastern part of the basin, due to emissions from smelters and metallurgical industries located at Georgia, Eastern part of Ukraine and central Russia. Besides the general evaluation of the elemental data also Factor Analysis (FA) was applied to the elemental data to characterize aerosols with respect to sources and to quantify the different sources and their importance for the composition of the aerosols at both Eastern and Western atmospheres.

KEYWORDS:
Eastern Black Sea, Western Black Sea, Trace elements, Aerosol, EDXRF

INTRODUCTION

There are variety of natural and anthropogenic sources of atmospheric particles. Since particles are released from different sources, they occur in different physical (particle diameter, surface area, density) and chemical states. Concentration and composition of the aerosols in the atmosphere depend on the meteorological factors that strongly affect atmospheric transport and transformation of particles. Since meteorology and demographic factors vary from one region to another on the Earth, characteristics of the aerosols are altered in a similar way.

Database on inorganic [1, 2] and organic [3, 4] composition of the atmosphere both in urban [5, 6] and rural [7, 8] regions of the Eastern Mediterranean increased significantly in last decades. These studies revealed demonstrated key issues about transport of pollutants to Eastern Mediterranean region and various atmospheric processes, such as anomalous SO_4^{2-} levels in the eastern Mediterranean atmosphere [9, 10] and how those levels developed in time [11]. Contribution of biogenic sources on unusually high SO_4^{2-} levels in the basin [12, 13], types and locations of source regions affecting chemical composition of particles and rain water [14, 15].

Studies that investigate existing aerosol and rain water composition in the Black Sea atmosphere is scarce. Most of the existing studies were conducted at urban areas surrounding the basin [16-19]. There is very limited number of rural studies, where source regions affecting composition of PM_{10} particles and rain water were investigated [20-23].

In this study aerosol samples were collected at two locations, one at the western and the other one at the eastern part of the Black Sea basin. Sampling station on the Eastern Black Sea is located at approximately 10 km from the Bulgarian border and approximately 50 km from the Black Sea coast. The station on the Western Black Sea is located at the Trabzon area. It is approximately 200 km from the Georgia Border and approximately 75 km from the coastline. Fine ($PM_{2.5}$) and coarse ($PM_{2.5-10}$) fraction particles were separately collected using a stack-filter unit. For both stations daily sampling was started in March 2011 and continued until December

2012. Collected samples were analyzed using an energy dispersive x-ray fluorescence spectrometry (EDXRF).

MATERIALS AND METHODS

Sampling sites and equipment. Western Black Sea station is located at a rural site (41°58'00"N 27°23'36"E) which is very close to the Bulgarian border of Turkey. Sampling station is approximately 30 km to Kırklareli city center, 50 km to the Black Sea coast and 10 km to the Bulgarian border. The altitude of the station is 574 meters. There are no pollution sources or large settlements within the 50 km radius of sampling station.

Eastern Black Sea station is located 1115 meters above sea level at a rural site (40°32'34"N 39°16'57"E) on the Eastern Black Sea coast of Turkey. The station is approximately 75 km to Black Sea coast and approximately 200 km from the Georgia border of Turkey. Settlement around the Eastern Black Sea station is similar with the Western Black Sea station, it is limited and there isn't any significant industrial activity around the station. Location of both stations is depicted in Figure 1.

At both stations, daily fine and coarse fraction particles were collected using a Gent Stack Filter Unit (SFU). Inlet of the sampler consists of a PM_{10} pre-impactor, which stops particles > 10 µm when the air is drawn through it. After the pre-impactor, air is directed to two 47 mm diameter polycarbonate filters, a coarse and a fine, placed in series. The coarse filter, which is the first filter in series, has a pore size of 8.0 whereas fine filter, which behind the coarse one, has 0.4 µm pore size. The sampling flow rate was maintained at 16.7 L min^{-1} using a flow controller. When the air is drawn at 16.7 L min^{-1} flow rate, coarse particles ($PM_{2.5-10}$) are collected on the top filter and fine particles ($PM_{2.5}$) are collected on fine filter. Operational characteristics of stack-filter-unit were discussed extensively in the literature [24, 25]. Filters were weighed before and after the sampling with a microbalance (Sartorius, Model MC-5) to determine the fine and coarse mass concentrations. Filters were conditioned in a constant temperature (25±5)ºC and humidity (26±5)% chamber for at least 24 hours.

At both stations sampling was daily and pumps were stopped only to change samples. For both stations the sampling was started at March 2011 and continued until December 2012. During the sampling period approximately 300 daily PM samples were collected at each station.

Chemical analysis and blank levels. Collected samples were analyzed using an energy-dispersive X-ray fluorescence (EDXRF) with ED2000 spectrometer at the Turkish Atomic Energy Authority, Sarayköy Nuclear Research and Training Center. Excitation source was an X-ray tube with Silver anode operated at a maximum current of 1000 µA with a maximum voltage of 50 kEV. A total of 18 elements including Na, Mg, Al, Si, S, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, As, Ba and Pb were detected with different frequencies. Detailed description of the analytical system was given elsewhere in [23].



FIGURE 1
Locations of the Western and Eastern Black Sea stations

Two different blanks, namely field blanks and laboratory blanks, were analyzed along with samples. Field blanks consisted of filters that were brought to the field like filters, loaded to the sampler like regular sample filters, but retained on the sampler for only one minute. After they were removed from the sampler they were treated and analyzed like sample filters. Laboratory blanks, on the other hand, consisted of filters taken from boxes and analyzed immediately. Laboratory blanks were not taken to the field. Generally one filter for laboratory blank was taken from each box. Blank levels and percent blank subtraction of the elements in the coarse and fine fractions represented in [23].

RESULTS AND DISCUSSION

Statistical summary of measured concentrations of the elements in fine and coarse fractions at the Eastern and Western Black Sea stations are given in Tables 1 and 2, respectively. Tables include average values, standard deviations and median values of 18 elements measured in this work.

Distribution characteristics of 18 elements were tested using chi-square test. Most of elements measured at both stations were log-normally distributed within a 95% confidence interval. Remaining elements which were not log-normally distributed, were also right skewed. As it can be seen from Table 1 and Table 2 average values of trace elements are twice as high as with the corresponding medians, indicating that the median is a better indicator of the data population.

Measured trace element concentrations at the Eastern Black Sea atmosphere varied between 0.46 ng m^{-3} for As and 2071 ng m^{-3} for SO_4^{2-}. For the Western Black Sea station concentrations varied between 0.6 ng m^{-3}- 3415 ng m^{-3} again for As and SO_4^{2-}, respectively.

Measured elements can be separated into three groups based on their coarse-to-fine concentration ratios. First group is consisted of crustal elements like Al and Si. These elements had higher concentrations in the coarse fraction. Coarse-to-fine concentration ratios varied between 1.03 for Ba at the Eastern Black Sea and 4.1 for Ca at the Western Black Sea. Typical C/F ratios for crustal elements is between 1.5 and 2.0. The ratios were > 1.0 at both Eastern and Western Black Sea. The only exception to this general pattern for soil related elements was Fe. Concentrations of Fe in coarse and fine fraction particles were comparable, indicating a fine fraction source for Fe. As crustal elements have mass median diameter between 3.0 – 3.5 µm [2], presence of crustal elements in coarse fraction is not surprising and reported frequently in literature [26, 27].

TABLE 1
Statistical summary of measured concentrations at Eastern Black Sea station (ng m^{-3})

	Coarse ($PM_{2.5-10}$)		Fine ($PM_{2.5}$)	
	Avg. ± SD	**Median**	**Avg. ± SD**	**Median**
Mass	11858 ± 24099	7041	10409 ± 21385	7795
B.C.	123 ± 291	86	294 ± 755	222
Na	67 ± 96	42	31 ± 39	25
Mg	139 ± 351	70	69 ± 144	39
Al	376 ± 867	210	178 ± 355	111
Si	1078 ± 3125	518	445 ± 855	300
SO_4^{2-}	308 ± 699	210	1763 ± 4858	1263
K	141 ± 375	94	90 ± 147	69
Ca	804 ± 1697	482	272 ± 526	153
Ti	196 ± 2455	13	22 ± 59	8
V	39 ± 194	1	3.89 ± 4.49	0.98
Cr	11 ± 40	6	7.06 ± 8.92	4.71
Mn	24 ± 71	11	14.74 ± 15.99	8.11
Fe	733 ± 3224	147	502 ± 1103	159
Ni	1.91 ± 3.60	0.88	3.70 ± 15.93	1.99
Cu	9.16 ± 19.72	3.27	6.92 ± 14.21	2.56
Zn	31 ± 44	22	29 ± 29	20
As	0.23 ± 0.69	0.16	0.23 ± 0.38	0.18
Ba	3.49 ± 2.02	3.27	3.90 ± 2.72	3.17
Pb	3.77 ± 5.81	2.35	4.10 ± 5.25	2.99

TABLE 2
Statistical summary of measured concentrations at Western Black Sea station (ng m^{-3})

	Coarse ($PM_{2.5-10}$)		Fine ($PM_{2.5}$)	
	Avg. ± SD	**Median**	**Avg. ± SD**	**Median**
Mass	9210 ± 10038	6923	10301 ± 7036	8923
B.C.	1837 ± 21997	72	2946 ± 22878	91
Na	170 ± 238	84	65 ± 85	41
Mg	261 ± 852	94	75 ± 122	30
Al	459 ± 1389	179	192 ± 339	82
Si	1107 ± 3037	438	460 ± 905	159
SO_4^{2-}	500 ± 530	354	2915 ± 1999	2593
K	149 ± 144	101	149 ± 132	108
Ca	700 ± 1382	322	198 ± 353	78.42
Ti	69 ± 443	16	19 ± 34	8.20
V	5.63 ± 23	1.48	1.58 ± 1.60	1.07
Cr	9.16 ± 11	6.42	6.91 ± 6.79	5.00
Mn	19 ± 35	11	9 ± 10	7.35
Fe	1000 ± 2459	311	514 ± 504	357
Ni	4.17 ± 7.19	1.90	1.84 ± 1.30	1.60
Cu	15 ± 34	5	5.44 ± 5.44	3.64
Zn	39 ± 85	25	23 ± 24	14
As	0.34 ± 0.82	0.19	0.26 ± 0.15	0.23
Ba	27 ± 89	5.66	7.71 ± 8.76	4.67
Pb	7.16 ± 12.27	4.35	5.46 ± 4.52	4.59

Concentrations of crustal elements were higher at the Western Black Sea atmosphere. In a previous study by [28] showed that Western Black Sea atmosphere region was under the influence of dust episodes origination from Northern Africa. In this case, effect of Saharan dust on this region is probably responsible for higher concentrations of crustal elements at the Western Black Sea atmosphere.

Second group of elements are consisted of Na and Mg, which are originated from sea salt. Also these elements were higher concentrations in the coarse fraction as expected. Concentrations of Na and Mg were observed at higher levels at the Western Black Sea atmosphere. Observed concentrations of these elements at a station are highly dependent with the wind regimes on the region and the distance of the station from the coastline. Therefore such sea salt elements like Na and Mg may vary greatly one station to another. Higher sea salt concentrations observed at the Western Black Sea region is due to closer proximity of the station to the sea.

The third group are consisted of elements with anthropogenic sources. Concentrations of anthropogenic species change significantly from one place to another, depending on the distribution of anthropogenic sources around the sampling point. Since these elements can be used as natural tracers of different compounds in receptor modeling studies, their concentrations at Eastern and Western Black Sea atmosphere is important as they provide preliminary information on source apportionment. At both stations anthropogenic elements, including Pb, As, SO_4^{2-}, and BC were higher in fine fraction. Association of these elements with the fine fraction is well documented in the literature [29, 30]. Copper was an exception. Copper concentration was approximately 25% higher in the coarse fraction. The reason is not clear.

Elements Ti, V, Cr, Ni, Mn had coarse-to-fine ratios > 1 in both stations. These elements have mixed sources, their measured concentrations are accounted for both crustal and anthropogenic sources. Coarse-to-fine ratios >1.0 for these elements indicate that contribution of coarse crustal particles on their measured concentrations both in Eastern and Western Black Sea are more important than contributions of anthropogenic sources.

Seasonal variations in concentration of measured elements at the Eastern and Western Black Sea are given in Table 3. Median values were used in the table, because most of these elements had lognormal distribution with 95% statistical confidence. Summer was the period between April and September, and winter was the rest of the year. This division was adopted from studies done in Mediterranean Region by our group [1, 11, 31]. The division in those studies was based on rainfall. In the Eastern Mediterranean region 80% of the rainfall occurs between September and April, which was our winter period. Although, rainfall pattern in the Black Sea region is not exactly the same with the pattern observed in the Eastern Mediterranean, we used this seasonal pattern in all studies in Turkey to be able to

compare results obtained in different parts of the country.

As it can be seen from Table 3 crustal elements, Al and Si, had 1.5 – 2 times higher concentrations in the summer season. This indicates the seasonality of the formation of soil particles. Soil particles in the atmosphere are formed with resuspension of surface soil under the influence of wind [32, 33]. Although wind speed is lower at summer season compared to winter season, formation of the particles is more likely as the surface is dry. This formation mechanism results in higher crustal element concentration in the summer season. Higher concentrations of soil-related elements in summer period is observed, not only at the Black Sea, but also in most of the studies performed in Turkey [12, 31, 34].

At both stations anthropogenic elements showed similar trends in terms of their winter and summer season concentrations except Cu and As. Anthropogenic elements, Cr, Ni, Zn, Pb had higher concentrations in the summer season at both stations whereas Cu and As had higher concentrations in the winter season. Higher winter concentrations of As was also observed at the Mediterranean atmosphere [32] and attributed to local sources of this element. Higher concentrations of As and Cu is probably due to same reason, because pollutants transported from distant source regions experience more extensive wet scavenging during winter and thus have higher concentrations during summer. However, elements from local sources can have higher concentrations in winter, or comparable concentrations in both seasons, because they will not be scavenged as efficiently during shorter transport distances. SO_4^{2-} showed more significant seasonality relative to other anthropogenic elements. This is partly due to less effective scavenging in summer, like other elements, and partly due to more extensive photochemistry in summer, with increased solar flux.

Contribution of sea salt to observed Na levels is highly dependent on the distance between sampling point and coast. Since sea salt particles are coarse and they settle down with increasing distance with coast. As it can be seen from Table 3, observed Na levels at both stations show a seasonality that is higher at the summer season. The similar seasonality at this region was also confirmed by [23]. This indicates observed Na levels at both stations come from soil rather than sea salt.

Factor analysis. In this study, factor analysis (FA) was applied to the elemental data sets generated at both stations to identify aerosol sources affecting the two parts of the Black Sea basin. Calculated factor loadings for Western Black Sea and Eastern Black Sea atmospheres are given in Table 4 and Table 5, respectively. Only factors with eigenvalues > 1 were retained (Kaizer criteria). Elements with missing data > 20% were not included in the FA

TABLE 3
Seasonal median values of elements (ng m^{-3})

	Eastern Black Sea					Western Black Sea			
	Coarse		Fine			Coarse		Fine	
	Summer	Winter	Summer	Winter		Summer	Winter	Summer	Winter
Mass	7072.8	6929.1	8138.0	6898.9	**Mass**	6124	7828	7649	9704
BC	89.0	72.7	223.8	214.5	**BC**	86	52	108	86
Na	48.1	27.7	29.1	17.5	**Na**	82	86	56	33
Mg	72.1	58.0	37.4	41.9	**Mg**	115	69	57	15
Al	239.5	165.7	117.0	63.4	**Al**	240	103	153	21
Si	566.3	458.0	308.8	187.6	**Si**	684	200	380	43
SO_4^{2-}	228.5	190.2	1450.8	929.7	**SO_4^{2-}**	292	466	2726	2498
K	98.1	77.3	74.1	60.2	**K**	139	79	129	90
Ca	498.1	433.3	158.8	124.0	**Ca**	375	277	146	35
Ti	13.5	11.6	8.6	7.8	**Ti**	22	10	12	3
V	0.9	1.7	1.1	1.0	**V**	1.7	1.3	1.2	0.9
Cr	6.4	5.8	4.8	4.6	**Cr**	6.1	6.6	4.8	5.4
Mn	11.1	9.3	9.2	5.9	**Mn**	15.6	10.5	9.3	7.5
Fe	156.6	135.4	138.6	266.5	**Fe**	322	300	324	387
Ni	1.0	0.5	2.3	1.4	**Ni**	1.4	2.4	1.9	1.5
Cu	3.9	2.7	2.2	2.9	**Cu**	4.3	5.8	3.5	4.0
Zn	21.9	20.8	25.1	8.6	**Zn**	18	32	11	17
As	0.2	0.2	0.2	0.2	**As**	0.19	0.18	0.22	0.25
Ba	2.7	4.3	2.7	4.6	**Ba**	5.7	5.7	4.7	4.6
Pb	3.0	2.1	3.8	2.1	**Pb**	4.4	4.2	5.2	4.1

TABLE 4
Western Black Sea atmosphere FA analysis – factor loadings

	Factor 1	Factor 2	Factor 3	Factor 4	Estimated Community
Mass	0.79				0.68
Na	0.40		0.66		0.63
SO_4^{2-}	0.76				0.76
K	0.80				0.74
Cr		0.89			0.86
Cu		0.85		0.34	0.83
Zn				0.95	0.92
Si			0.91		0.84

exercise. Aluminum, which is a very important crustal marker was not included due to this reason. Lack of Al is not a serious drawback in this case, because Si, which is detected in almost all of the samples is equally good crustal marker.

FA identified 4 factors at the Western Black Sea Atmosphere. The first factor identified at the Western Black Sea atmosphere has almost 80% of the loadings of SO_4^{2-} and K. Also, $PM_{2.5}$ mass has its 79% of the loading in this factor. SO_4^{2-} is generally a tracer of aerosols arising from fossil fuels, particularly SO_4^{2-} is a well-defined tracer for thermal power plants. Potassium, beside the well-known crustal and sea salt origins, is also emitted from biomass burning. A previous study conducted at this region by [28] showed that this region was under influence of forest fires especially in Ukraine. Having potassium and SO_4^{2-} in the same factor may be indicating SO_4^{2-} is originating from biomass burning rather than fossil fuels.

Second factor has high loadings of Cr and Cu. These two elements are the important tracers of smelters. Therefore, this factor is considered to represent particles formed during the process of obtaining metals from ores. There are a number of smelters located on Turkish coast (Samsun), Ukraine and Georgia, which may be the reason for appearance of a clear smelter factor.

Third factor has high loadings of Si. Therefore third factor is identified as crustal factor. The last and fourth factor identified in the Western Black Sea atmosphere is an industrial factor, because it has high loadings of Zn and Cu.

Factor analysis again identified 4 factors for the Eastern Black Sea atmosphere. Factor 1 is identified as an anthropogenic factor because, it has high loadings of SO_4^{2-}, V, Mn, Ni and Cd. SO_4^{2-} has 77% of its loading in this factor. This indicates that factor 1 is a combustion factor. Also, owing to presence of majority of loadings of Ni and V in this factor, factor 1 represents petrol combustion rather than coal combustion.

Fe, a well-known crustal tracer, has its 94% loading in Factor 2. Also, 96% of the loading of the $PM_{2.5}$ mass is explained by this factor. Therefore this factor is considered to represent a crustal factor.

Factor 3 has high loading of Na, which is a sea salt constituent. This factor is identified as a sea salt factor owing to presence of Na. The fourth factor identified in the Eastern Black Sea atmosphere has high loadings of

TABLE 5
Eastern Black Sea atmosphere FA analysis – factor loadings

	Factor 1	Factor 2	Factor 3	Factor 4	Estimated Community
Na			0.67		0.49
Mass		0.96			0.96
SO_4^{2-}	0.77				0.69
Ca	0.37		0.71		0.65
Ti			0.77		0.60
V	0.67				0.57
Cr				0.52	0.30
Mn	0.58			0.45	0.58
Fe		0.94			0.91
Ni	0.50			0.51	0.51
Zn					0.50
Cd	0.65				0.56

Cr, Mn and Ni. As mentioned before, these tracers released during the process of obtaining metals from ores. Therefore, this factor is considered to represent these particles.

CONCLUSIONS

Daily fine ($PM_{2.5}$) and coarse ($PM_{2.5-10}$) particulate matter samples were collected 2 different stations, Western and Eastern Black Sea, at the Black Sea basin of Turkey. Sampling was performed between March 2011 and December 2012 and collected samples at both stations analyzed using EDXRF for their trace element compositions.

Crustal elements observed at higher levels at the Western Black Sea atmosphere. Previous studies showed the influence of dust episodes to this region. Sea salt elements had comparable concentrations or slightly higher at Western Black Sea. This is due to closer proximity of the Western Black station to the sea.

At the Western Black Sea atmosphere most of anthropogenic trace elements had higher concentrations, which is not surprising when the distribution of industrial sources around sampling location is considered. But Mn, Cr were higher on the Eastern part of the basin.

According to FA results, it can be concluded there are some similarities and differences between the components of aerosol population of Eastern Black Sea and Western Black Sea atmospheres. In both atmospheres there are sea salt particles, crustal particles and particles formed during the separation of metal sprouts. Combustion factor is also common for both atmospheres. But combustion components are not same in the regions. There are some symptoms indicating that the combustion particles at the Western Black Sea originating from biomass burning whereas at the Eastern Black Sea combustion particles originating from petrol combustion. In both regions combustion originated particles may be also representing a mixture of particles released from multiple sources. For example, at the Western Black Sea factor analysis identified SO_4^{2-} and K in the same factor. During the transport of particles containing high amounts of SO_4^{2-} released from thermal power plants might have been mixed with the particles containing high concentrations of K released from forest fires. Due to lacking of sufficient tracers in the factor analysis, it may not possible to distinguish these sources.

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