Geoscience Frontiers 11 (2020) 783–792

Contents lists available at ScienceDirect

Geoscience Frontiers

journal homepage: www.elsevier.com/locate/gsf

Research Paper

Assessment of geochemical and sedimentological characteristics of atmospheric dust in Shiraz, southwest Iran

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ARTICLE INFO

Handling Editor: Vinod Oommen Samuel

Keywords:
Dust storm
Atmospheric dust
Heavy metals
Diatom
Palygorskite
Provenance

ABSTRACT

Geogenic dust is commonly believed to be one of the most important environmental problems in the Middle East. The present study investigated the geochemical characteristics of atmospheric dust particles in Shiraz City (south of Iran). Atmospheric dust samples were collected through a dry collector method by using glass trays at 10 location sites in May 2018. Elemental composition was analysed through inductively coupled plasma optical emission spectrometry. Meteorological data showed that the dustiest days were usually in spring and summer, particularly in April. X-ray diffraction analysis of atmospheric dust samples indicated that the mineralogical composition of atmospheric dust was calcite + dolomite (24%)>palygorskite (18%)>quartz (14%)>muscovite (13%)>albite (11%)>kaolinite (7%)>gypsum (7%)>zircon = anatase (3%). The high occurrence of palygorskite (16%–23%) could serve as a tracer of the source areas of dust storms from the desert of Iraq and Saudi Arabia to the South of Iran. Scanning electron microscopy indicated that the sizes of the collected dust varied from 50 μm to 0.8 μm, but 10 μm was the predominant size. The atmospheric dust collected had prismatic trigonal–rhombohedral crystals and semi-rounded irregular shapes. Moreover, diatoms were detected in several samples, suggesting that emissions from dry-bed lakes, such as Hoor Al-Azim Wetland (located in the southwest of Iran), also contributed to the dust load. Backward trajectory simulations were performed at the date of sampling by using the NOAA HYSPLIT model. Results showed that the sources of atmospheric dust in the study area were the eastern area of Iraq, eastern desert of Saudi Arabia, Kuwait and Khuzestan Province. The Ca/Al ratio of the collected samples (1.14) was different from the upper continental crust (UCC) value (UCC = 0.37), whereas Mg/Al (0.29), K/Al (0.22) and Ti/Al (0.07) ratios were close to the UCC value (0.04). This condition favours desert calcisols as the main mineral dust sources. Analysis of the crustal enrichment factor ($EF_{crustal}$) revealed geogenic sources for V, Mo, Pb, Sr, Cu and Zn (<2), whereas anthropogenic sources affected As, Cd, Cr and Ni.

1. Introduction

Geogenic dust is one of the most critical meteorological hazards in the Middle East; therefore, a thorough understanding of its physicochemical properties and mineralogy is crucial (Middleton, 2017; Karimian Torghabeh et al., 2019). Recent studies on dust storms have extended their scope to arid and semi-arid areas, and many surveys have been conducted on the importance of mineral dust to human life from different points of view (Middleton, 2017). This sedimentology phenomenon has wide-ranging effects on Earth's system, including oceanic and terrestrial ecosystems, global biogeochemical cycles, climate processes and air chemistry, atmospheric pollution and human health (Engelbrecht and Derbyshire, 2010).

Dust storm is different from air pollution caused by urban and industrial activities; such pollution is a temporary characteristic of the area and limited to industrial and urban environments (Žibret et al., 2013). Dust storm reduces air quality in certain conditions (e.g. wind speed, soil moisture, soil mineralogy, roughness, vegetation density and

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Peer-review under responsibility of China University of Geosciences (Beijing).

https://doi.org/10.1016/j.gsf.2019.08.004
Received 28 October 2018; Received in revised form 17 June 2019; Accepted 18 August 2019
Available online 7 September 2019

topography) in an area that is not necessarily completely dry. Wind erosion plays a central role in dust storm production and occurs when strong and persistent winds interact with dry, fine-grained, loose soil (Li et al., 2004; Yigiterhan et al., 2018a,b). When atmospheric dust particles are moved by wind, they are ceaselessly deposited by dry and/or wet deposition phenomena, such as leaching or diffusion (Bergametti and Forêt, 2014); they ooze into soils (Dehbandi and Aftabi, 2016), settle onto land or freshwater surfaces (Nickovic et al., 2001; Mahowald et al., 2005), are breathed in by humans and animals (Morman and Plumlee, 2014) or collected by plants (Norouzi et al., 2015).

Three important characteristics of dust storm events, namely, morphology, mineralogy and chemical properties, are typically investigated. These features provide valuable information on the positive and negative aspects of dust phenomena and help identify the sources of these particles.

Mineralogy studies are widely used methods of evaluating atmospheric mineral dust source contributions (Li et al., 2007; Ahmady-Birgani et al., 2018). Geochemical mass balance methods, such as enrichment factor, have also produced useful results in dust source identification (Weiss et al., 2002). For example, Schütz and Sebert (1987) proposed using the high frequency of calcite and palygorskite as a source mineral marker for dust particles originating from Northern Africa. In several atmospheric mineral dust studies, typical elemental ratios (e.g. Ca/Al, Mg/Al, Na/Al and K/Al) were used as markers for different mineral dust sources. Afro-Asian geochemical mineral dust studies accomplished by Chen and Li (2011) and Scheuvens et al. (2013) are successful examples of the application of elemental ratio techniques.

Several studies have been conducted on chemical and mineralogical properties to identify dust sources due to the high occurrence of dust storms, especially in western and eastern parts of Iran (Karimian Torghabeh et al., 2019) and increased public concern about the harmful effects of dust storms on human health. Three ideal provenances such as (I) desert areas in Iraq and Syria, (II) sedimentary basins in Saudi Arabia and Kuwait, and (III) dry lakebeds, such as the dry bed of Hoor al-Azim Wetland and Hamoun dry lakes for dust storms in Iran were reported by Zarasvandi et al. (2011); Rashki et al., (2013); Ahmady-Birgani et al., (2015) and Mojadam et al. (2018).

Hojati et al. (2012) investigated the characteristics of atmospheric dust in central parts of Iran and concluded that the palygorskite in dust particles probably originates from and within desert areas in Iraq, Saudi Arabia and Jordan, especially from Iraqi arid land to Zagros area and central Iran.

Iran lies along the world's dust belt and has an arid and semi-arid climate that has long been affected by dust events. The intensification of these events in recent decades in terms of severity, extent and frequency in the western, southern and central parts of Iran has resulted in a new phenomenon that needs to be addressed. Fars Province, due to its geographical location (located on the mid-latitude belt), has a low moisture content and is near the source of dust production in the deserts of Iraq and Saudi Arabia. This province is constantly exposed to the dust phenomenon. However, atmospheric dust reports on this area are unavailable, at least in literature. The first objective of this study is to investigate the bulk composition of atmospheric dust in Shiraz City. Given that this study provides the first reported measurements of atmospheric dust in the south of Iran, the second objective is to present baseline dust data that can be used to evaluate the characteristics and compositions of different fine fractions (e.g. PM10–2.5 μg/m^3 and PM2.5 μg/m^3).

2. Materials and methods

2.1. Description of the study area

Shiraz is the largest city in southern Iran (52°54′E, 29°62′N) and the capital of Fars Province (Fig. 1). The city has an area of 1268 km^2; it is 28 km wide on its northwest to southeast axis and approximately 9 km wide on its east to west axis. Its population is above 1,500,000, and it is known as the sixth most populated city in Iran. Geologically, Shiraz is a syncline valley city (NW–SE elongated) that formed between the Baba Kohi and Derak anticlines (Jahandari, 2015), and it is located in the Zagros Fold–Thrust Belt Zone (Alavi, 1994). Shiraz is mainly characterised by sedimentary rocks, such as conglomerate, limestone, marl and shale (Ghorbani, 2013).

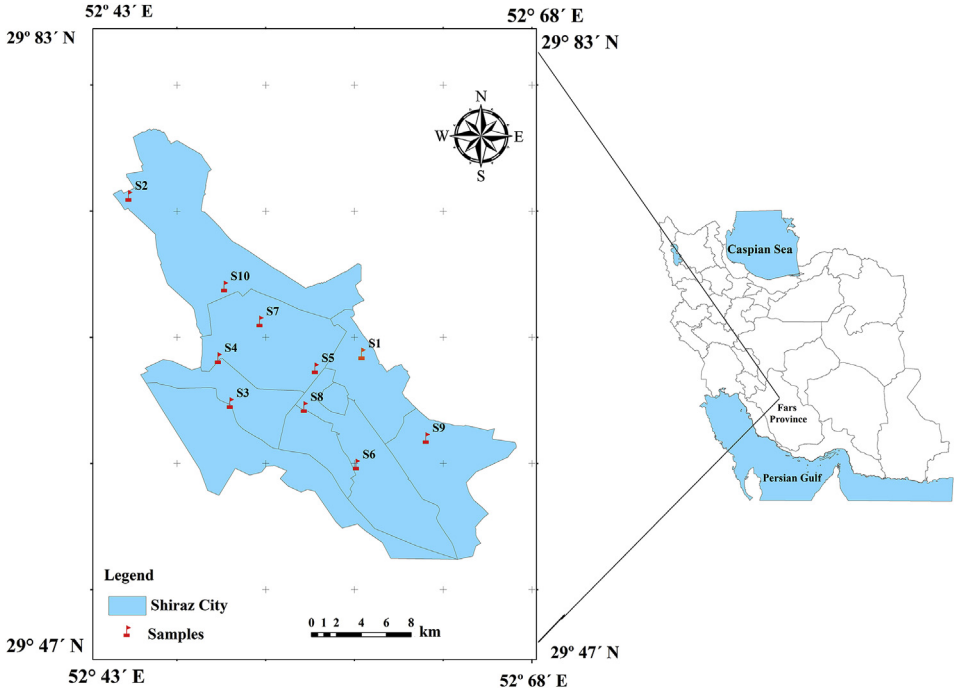

Fig. 1. Map of Iran showing Fars Province and the locations of the atmospheric dust sampling sites in Shiraz City.

2.2. Dust sampling

Atmospheric dust samples were collected using a dry method. The dust collectors had a dry glass surface (Menendez et al., 2007) with an area of 1 m^2. Each tray contained a flat glass-coated plastic net (2 mm mesh openings) on its upper part to form a roughness plate, which is for trapping atmospheric dust and preventing dust from being transported (Fig. 2). Each sampling tray was installed 3–5 m from the ground level on flat-roofed buildings in 10 different locations (Fig. 1). In addition to reducing the possibility of vandalism at the sampling site, the placement of traps at this altitude also eliminates the effects of low-altitude wind that contains local dust, such as urban dust. Samples were gathered weekly by using a small plastic brush to scrape off materials deposited on the glass trays. All sampling tools (plastic brushes and glass trays) were washed with distilled water after sampling. The dust samples were stored in a polyethylene micro-tube, numbered and transported to the laboratory. Given that the highest incidence of dusty day in the study area occurs in spring, 10 samples were collected from the sites in May 2018.

2.3. Chemical and mineralogical analysis

We combined several geochemical methods, including X-ray diffraction (XRD) and inductively coupled plasma-optical emission spectrometry (ICP-OES), to understand the dust geochemistry of bulk samples, particularly with respect to the mineral assemblage of particles and trace elements, and determine their geological sources.

The minerals of atmospheric samples were identified through XRD analysis. ATESCAN-Vega 3 scanning electron microscope at the central laboratory of Shiraz University was used to identify the morphology and textural features of the dust samples.

The elemental concentrations of dust samples were determined using a strong acid (HNO_3–HCl_4) pseudo-total digestion method via ICP-OES (Agilent model 735, United States). Quality assurance and control included duplicate analyses, certification of materials and analysis of blanks. The relative standard deviations were 1.8% for As, 9% for Cd, 3.8% for Co, 1.4% for Cr, 5.1% for Cu, 1.6% for Mo, 3% for Ni, 8.3% for Pb, 4.6% for Sr and 0.69% for Zn.

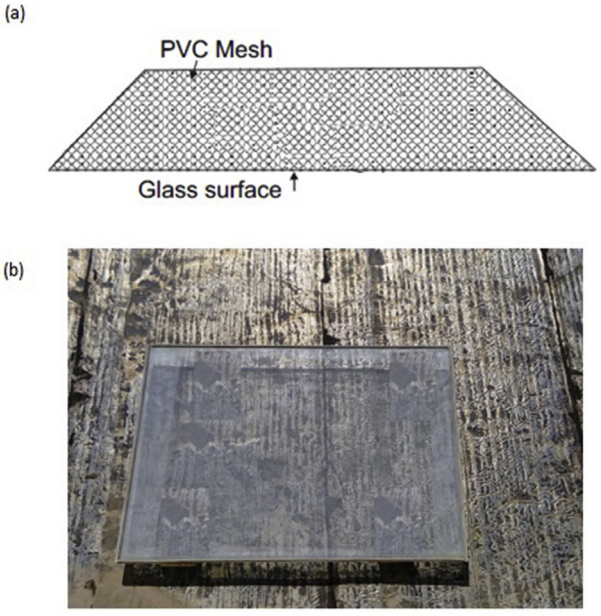

Fig. 2. Schematic of atmospheric dust samplers that consist of a glass tray (1 m × 1 m) covered with a 2 mm mesh PVC net modified by Menéndez et al. (2007).

2.4. Enrichment factor

The crustal enrichment factor is commonly used to study the elemental composition of atmospheric dust, assess crustal/non-crustal sources of elements and evaluate the degree of anthropogenic influence (Lammel et al., 2002; Khillare et al., 2004). The formula to calculate the $EF_{crustal}$ value is expressed as:

$$EF_{crustal} = \frac{\left[\frac{C_x}{C_r}\right]_{sample}}{\left[\frac{C_x}{C_r}\right]_{crustal}} \quad (1)$$

where C_x refers to the concentration of target elements and C_r is the content of the reference element in the samples and upper continental crust (UCC). In the present study, aluminum (Al) was employed as the lithogenic tracer due to its abundance in the Earth's crust, lack of anthropogenic origin and non-mobility in weathering processes (Taylor and McLennan, 1995). According to Chester et al. (1984), the $EF_{crustal}$ value of 1 pertain to a crustal origin, whereas ratios greater than 10 denote a non-crustal source. Moreover, $EF_{crustal}$ ratios in the range of 1–5 indicate that anthropogenic processes significantly affect the elemental compositions of atmospheric dust. In general, increasing values of the crustal enrichment factor indicate an increase in the contribution of non-crustal processes to the chemical composition of atmospheric dust.

2.5. Annual occurrence of dusty days and meteorological parameters

The meteorological condition of the sampling period was established using 42-year archives from 1971 to the end of 2013. Fars Province has been affected by drought in the past few years (Moradi et al., 2011). This situation suggests that due to the reduction of the moisture content of soil, the frequency of dust events in the region has increased in recent years. In this regard, Figs. 3 and 4 show diagrams of long-term monthly precipitation and wind speed in the study area, respectively. The mean maximum long-term monthly precipitation occurred in January (77.6%), and the mean maximum long-term wind speed jointly occurred in April, May and June. Omidvar and Omidi (2013) analysed dust phenomena in southern and central Fars Province from 1992 to 2008 and argued that most stormy days in this province occur in spring and summer, with Shiraz and Fasa stations having the highest monthly frequency. The results of previous studies that analysed dusty days in Shiraz City show that spring (44.31%) and autumn (6.1%) have the highest and lowest seasonal frequency of dust phenomena, respectively. Meteorological data recorded in Fars Province from 1992 to 2008 show that Shiraz experiences the largest number of monthly dust storms in May, with a total of 24 days, and the smallest number monthly dust storms in January, with a total of nine days (Omidvar and Omidi, 2013). The authors suggested that because the average annual wind speed of dust in this region is

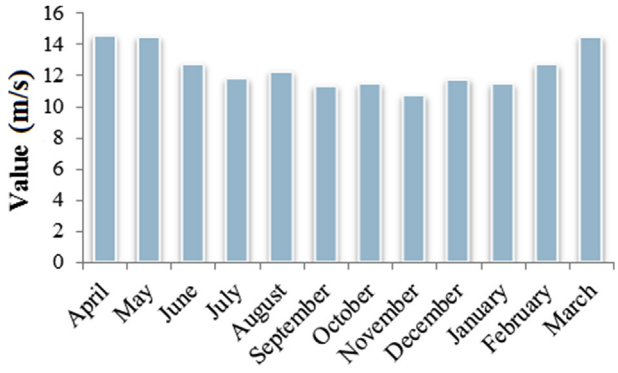

Fig. 3. Long-term mean maximum wind speed (m/s) in Shiraz meteorological station during the 29-year study period.

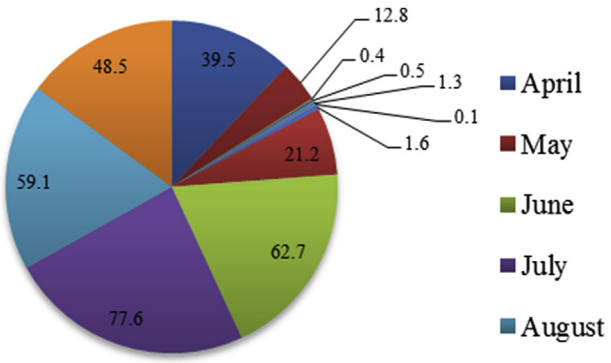

Fig. 4. Long-term monthly precipitation in Shiraz meteorological station during the 29-year study period (unit: %).

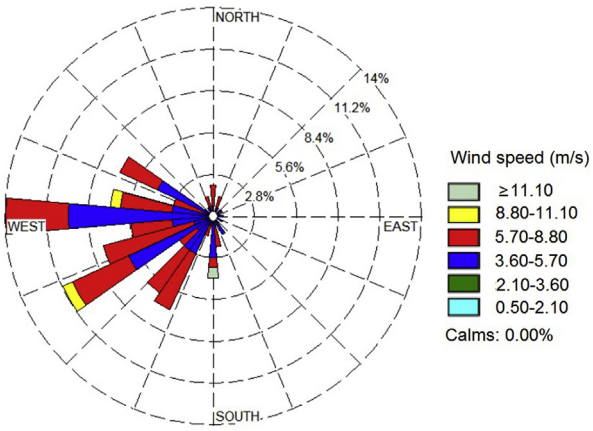

Fig. 5. Rose plot of wind speed direction from 2016 to 2017 in the study area.

almost 1 m/s, the dust phenomenon in Fars Province does not have a local source. Fig. 5 shows the dominant wind direction (rose plot) from 1993 to 2012 in Shiraz City obtained using archived data on recorded meteorological parameters. Similar to the situation in other southern and western cities of Iran, such as Ahwaz, western and southwestern winds are the most frequent winds in Shiraz. Broomandi et al. (2017) suggested that Arabian countries around the south of the Persian Gulf, particularly Kuwait, Iraq, and the Arabian Peninsula, can act as dominant origins of dust storms in west and southwest areas of Iran.

3. Results

3.1. Morphology of atmospheric dust samples

Several high-resolution SEM images with different magnifications of selected samples revealed different ranges in the size of particles and surface coatings of calcite, quartz and other minerals. The images also showed that calcite, silica (quartz and diatoms), other silicates and gypsum to some extent were commonly covered with hydrous aluminium phyllosilicates and different-sized particles, particularly clay-sized particles that are visible as small aggregates (Fig. 6).

Quartz particles displayed typically sub-angular and sharp edges and were mostly larger in size (50 μm) than the other mineral particles. Calcite particles appeared as trigonal–rhombohedral crystals with semi-rounded and irregular shapes of different sizes, whereas the large crystals of 10–20 μm were identified as gypsum. Calcite and quartz were the dominant minerals in all of the atmospheric dust samples. Rock-forming minerals with diatomaceous materials (hydrated amorphous silica) and clay minerals were prevalently observed in the dust samples, and their sizes ranged from 2 μm to 10 μm. Diatom was detected in Sample 1, which was collected at a higher altitude compared with the other samples. This amorphous silica had microorganisms that are generally found in dry lake beds (Chou et al., 2008).

The SEM imaging measurements indicated that the sizes of dust particles in Shiraz varied between 50 μm and 0.8 μm. Fine (<10 μm) and ultrafine particles (<2.5 μm) can penetrate into the respiratory system and cause various illnesses (WHO, 2006). SEM studies showed that <10 μm particle size is the most frequent size of Shiraz atmospheric dusts. This situation can exert adverse effects on the health of citizens because disposable dust masks cannot prevent the entry of these microscopic particles into human lungs (Mazzoli and Favoni, 2012). These observations are consistent with the results of Mohammadi et al. (2015), who concluded that 25.3% of the total mortality that occurred in 2012 was attributed to PM10 with concentrations > 40 $\mu g/m^3$.

3.2. Element composition

The elemental composition of atmospheric dust reflects mineral-bearing soil from dust provenance. Most studies interpreted Ca–Mg rich as carbonate phases and Si–Na–Mg–K–Al rich as silicate phases. Hence, evaluation of the relationship between these elements is helpful in clarifying the geochemical nature of dust storms.

3.2.1. Major elements

The statistical characteristics of the studied elements of the Shiraz atmospheric dust samples are presented in Table 1. The total major element contents in the collected samples decreased in the order of Ca > Al > Mg > K > Na > Ti. According to the mineralogical results (Section 3.3), Al–Mg enriched represented palygorskite, whereas high contents of Ca were related to carbonate particles. High Ti, Na and K contents reflected the presence of anatase, albitic plagioclase and muscovite in the samples, respectively. The concentrations of these elements should be compared with UCC (Table 1) to understand the status of the major elements in atmospheric dust. The concentration of Al in all samples was significantly lower than the UCC value. The concentrations of Ca and Mg in the samples exceeded the corresponding values of UCC, whereas the mean concentrations of the other elements were lower than the UCC average. Compared with Saharan dust (North African dust), Shiraz atmospheric dust is richer in Ca and Mg (Moreno et al., 2006; Chou et al., 2008). These results confirm that Middle East atmospheric dust has high percentages of Ca and Mg (Engelbrecht et al., 2009b; Yigiterhan et al., 2018a,b). The main reason for these observations is the geology of the region, which is mainly characterised by sedimentary rocks (Engelbrecht et al., 2009b).

Elemental ratio tracers make up a practical means of determining the origin of dust phenomena, and scientists have used them extensively. This method is commonly used in geochemical studies of atmospheric dust (Yigiterhan et al., 2018a,b). Elemental ratio tracer assists to detect depletions and enrichment of reactive elements with respect to Al and to clarify the probable provenance areas. In geochemical studies, Al is considered as an immobile element in the weathering process having a high abundance of Al_2O_3 (Chen and Li, 2011; Scheuvens et al., 2013; Yigiterhan et al., 2018a,b).

Table 2 compares the elemental ratios between the published data (UCC) and Ca, Mg, K and Ti to Al in the Middle East atmospheric dust used in this study. There was no significant abnormal enrichment of Ti/Al, Mg/Al and K/Al ratios found in the studied samples relative to UCC ratios. The Ti/Al and K/Al ratios in our samples are close to the UCC ratios. The ratio of Ca/Al in Shiraz atmospheric dust samples was 1.14, which is similar to Iraq, Kuwait and the Arabian Peninsula, having higher than the ratio of UCC (0.37). Ca/Al ratio shows that Ca content, which is one of the major elements for Shiraz atmospheric dust, can be explained by more carbonate fraction contribution in the Middle East atmospheric dust (Engelbrecht et al., 2009a, b; Yigiterhan et al., 2018a,b). The Mg/Al ratio of studied dust samples was significantly lower than those from Kuwait and Iraq. Meanwhile, The Mg/Al ratio in this study is close to the

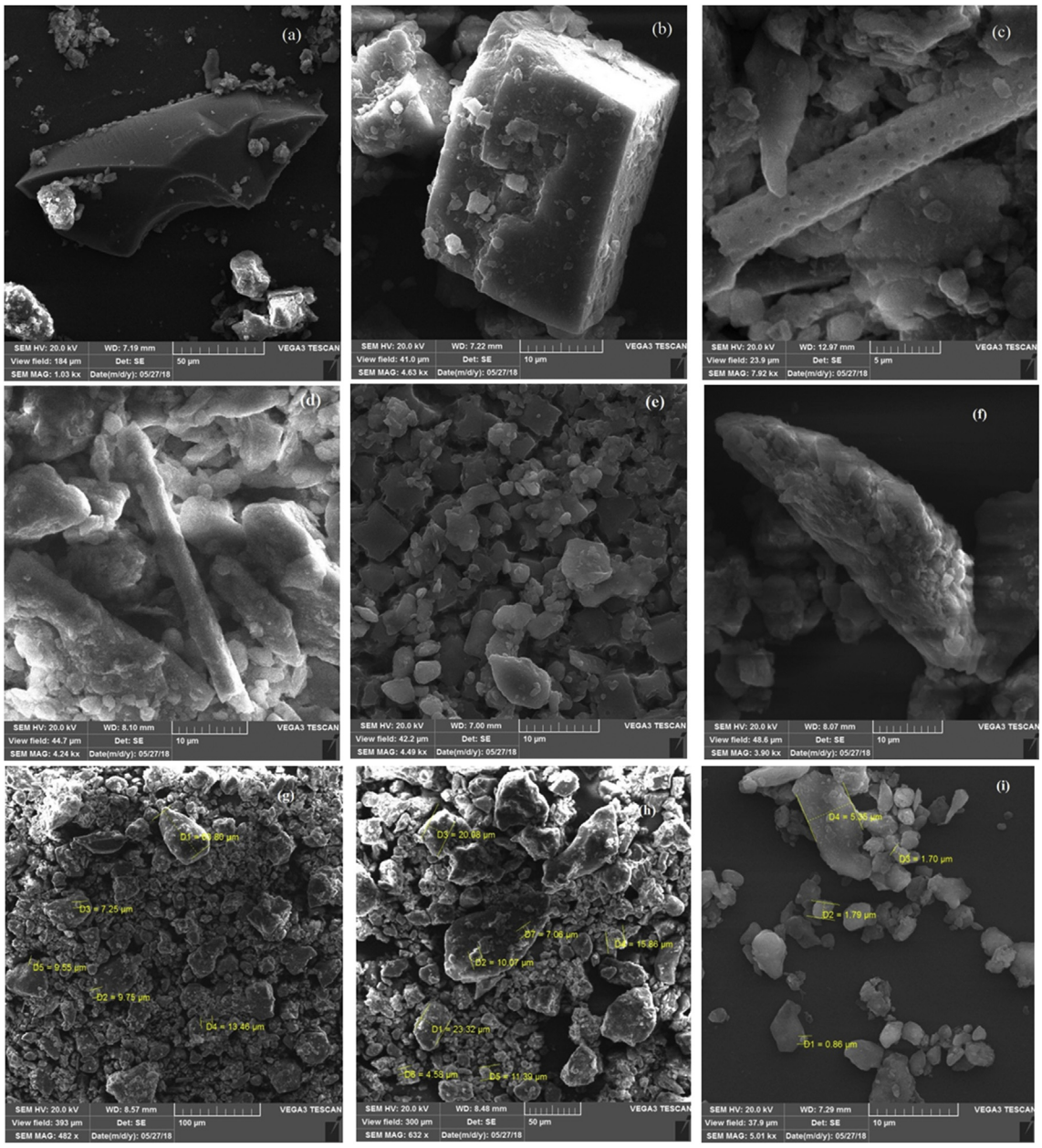

Fig. 6. SEM images showing the morphology and particle size of the atmospheric dust samples: (a) quartz, (b) calcite, (c) diatom, (d) clay aggregates, (e) gypsum and (f) particles agglomerated by fibrous palygorskite; (g), (h) and (i) reveal the presence of various particle sizes in several samples. The scale bar is defined for each image.

ratio of the Arabian Peninsula (Table 2). This trend is due to the large contribution of Al–Mg-silicate (palygorskite) minerals relative to Mg–Ca-carbonate minerals such as dolomite.

3.2.2. Trace elements

The trace metal contents in the Shiraz atmospheric dust samples and their descriptive statistics are summarised in Table 1. The average levels of the trace metals decreased in the order of Sr > Zn > V > Cr > Ni > Cu > Pb > Co > As > Cd. The mean concentrations of Mo, Sr and Co were close to their UCC values, whereas the mean concentrations of As (4.51 mg kg^{-1}), Cd (0.28 mg kg^{-1}), Cr (95.8 mg kg^{-1}), Ni (68.1 mg kg^{-1}), Pb (30.6 mg kg^{-1}), Cu (40.5 mg kg^{-1}), V (102.7 mg kg^{-1}) and Zn (10.6.5 mg kg^{-1}) were approximately 3, 2, 2.8, 3.4, 1.5, 1.6, 1.7 and 1.5 times higher than the corresponding UCC values, respectively. The concentrations of these elements are compared with those of atmospheric dust from other parts of Iran in Table 1. The atmospheric dust in Shiraz showed higher average Co, Cr, Cu, Ni, Pb and V concentrations compared with dust from southwest and southeast parts of Iran, but the Zn level in the dust was lower than that of the southwest region. These differences are probably related to sampling sites, times, methods and analyses. However, the enrichment of these trace elements has been reported in atmospheric dust samples from Iraq and western Iran (Table 1).

To improve our evaluation of whether the trace elements were extracted from geogenic or anthropogenic sources, we implemented the enrichment factor and hierarchical cluster analysis on the investigated elements (Karimian Torghabeh et al., 2018). The calculated crustal EF values of the studied elements in atmospheric dust are presented in Fig. 8. The mean values of crustal EF computed for the elements increased in the order of Na < K < Mo < Sr < Mg < Zn < Pb < V < Co, Ca < Cr < Cd < As < Ni, with values of 0.3, 0.6, 0.62, 0.98, 1.77, 1.79, 1.8, 2.04, 2.23, 3.08, 3.36, 3.45, 3.59 and 4.07, respectively. The $EF_{crustal}$ values of all studied elements are less than 10, indicating a predominantly geogenic source.

Table 1
Summary statistics of concentration levels trace elements (mg/kg) and major elements (%) in the atmospheric dust of Shiraz during May 2018.

Element	Minimum	Maximum	Mean	S.D.	CV	UCC[a]	Southwest of Iran[b]	Southeast of Iran[c]	Western Iran[d]	Iraq[e]
Al (%)	6.48	7.02	6.72	0.18	0.02	8.04	–	–	–	–
Ca (%)	7.12	9.46	7.72	0.71	0.09	3.0	–	–	–	–
Mg (%)	1.91	2.0	1.96	0.02	0.01	1.33	–	–	–	–
Na (%)	0.91	0.79	0.86	0.03	0.03	2.89	–	–	–	–
K (%)	1.4	1.5	1.5	0.03	0.02	2.80	–	–	–	–
Ti (%)	0.48	0.53	0.51	0.01	0.03	0.30	–	–	–	–
As (mg/kg)	3.2	5.9	4.5	0.93	0.2	1.5	1.48	–	–	–
Cd (mg/kg)	0.25	0.31	0.28	0.01	0.06	0.098	0.41	–	10.4	27
Co (mg/kg)	18	20	18.7	0.67	0.03	10	3.1	–	21	49
Cu (mg/kg)	40	43	40.5	0.97	0.02	25	10.05	11	50	53
Cr (mg/kg)	93	118	98.4	7.5	0.07	35	15.92	77	128.4	–
Mo (mg/kg)	0.68	0.88	0.79	0.05	0.07	1.5	0.84	–	–	–
Ni (mg/kg)	67	70	68.1	1.4	0.02	20	21.68	17	111.2	131
Pb (mg/kg)	23	57	30.6	9.96	0.3	20	10	10	109.2	182
Sr (mg/kg)	279	299	287	7.4	0.02	350	54.76	139	–	–
V (mg/kg)	99	106	102.7	2.3	0.02	60	10	69	–	–
Zn (mg/kg)	101	130	106.6	8.6	0.08	71	810.96	54	472.2	154

[a] Taylor and McLennan (1995).
[b] Zarasvandi et al. (2011).
[c] Rashki et al. (2013).
[d] Al-Dabbas et al. (2012); Najafi et al. (2014).
[e] Al-Dabbas et al. (2012).

Table 2
Elemental ratios for atmospheric dust samples from current study, compared to those of bulk dust samples from other countries of the Middle East.

	Ti/Al	Mg/Al	Ca/Al	K/Al	Analysis	Reference
Shiraz atmospheric dust	0.07	0.29	1.14	0.22	ICP-OES	This study
Kuwait	0.65	1.25	1.23	0.94	XRF	Engelbrecht et al. (2017)
Iraq	0.72	1.11	1.31	1.04	XRF	Engelbrecht et al. (2017)
Arabian Peninsula	0.14	0.11	2.17	0.34	XRF	Engelbrecht et al. (2017)
UCC	0.04	0.16	0.37	0.34	–	Taylor and McLennan (1995)

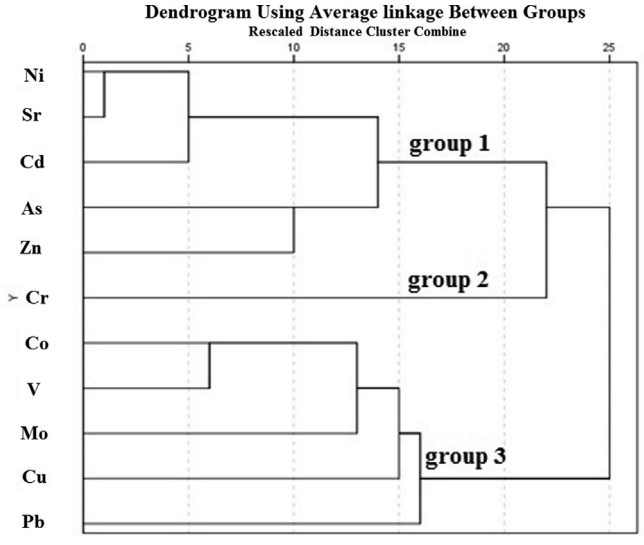

Fig. 7. Dendrogram of trace element concentrations of atmospheric dust in Shiraz.

Moreover, the EF values of As, Ni, Cd and Cr are between 2 and 5, indicating that the concentrations of these elements are affected by the

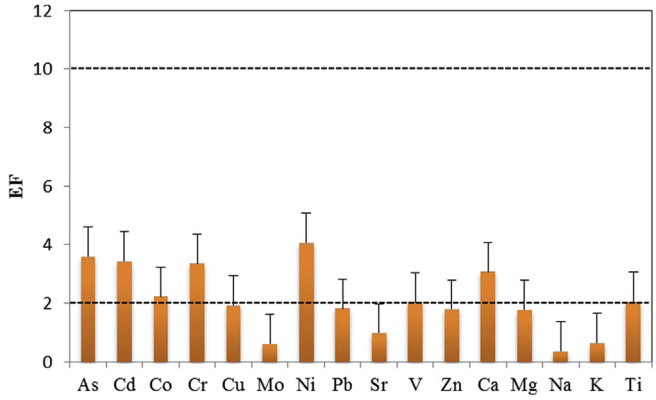

Fig. 8. Elemental crustal enrichment factors of atmospheric dust samples; the error bars display standard deviations.

anthropogenic processes.

The results of cluster analysis (Fig. 7) showed that the trace elements in Shiraz atmospheric dust can be classified into three groups: Group 1 consisting of Ni, Sr, As, Cd and Zn; Group 2 consisting of Cr; and Group 3 consisting of Co, V, Mo, Cu and Pb. Group 1 included typical pollution-derived heavy metals that were realised by anthropogenic sources. All elements of Group 3 had crustal EFs values less than 2 in the samples, demonstrating that these elements were mainly derived from natural sources. However, this group could be minimally influenced by human activities. In the second group, because Cr exhibits a distinct geochemical behaviour, our argument is that Cr has a mixed source (crustal and anthropogenic source). Nevertheless, based on the crustal EF value of Cr > 3, we posit that atmospheric dust Cr concentrations were affected by human activities.

3.3. Mineralogy of atmospheric dust samples

The mineralogical compositions of bulk dust can be a useful tool to recognise the source of dust provenance and/or determine the health effects of the particles on human society. Generally, the major mineralogical phases of global dust are silicates, quartz, feldspars (albite, orthoclase, anorthite) and parts of the group of clay minerals (illite, kaolinite, smectite and chlorite), carbonates (calcite, dolomite), sulphates (gypsum) and iron oxides (haematite, goethite) (Scheuvens and

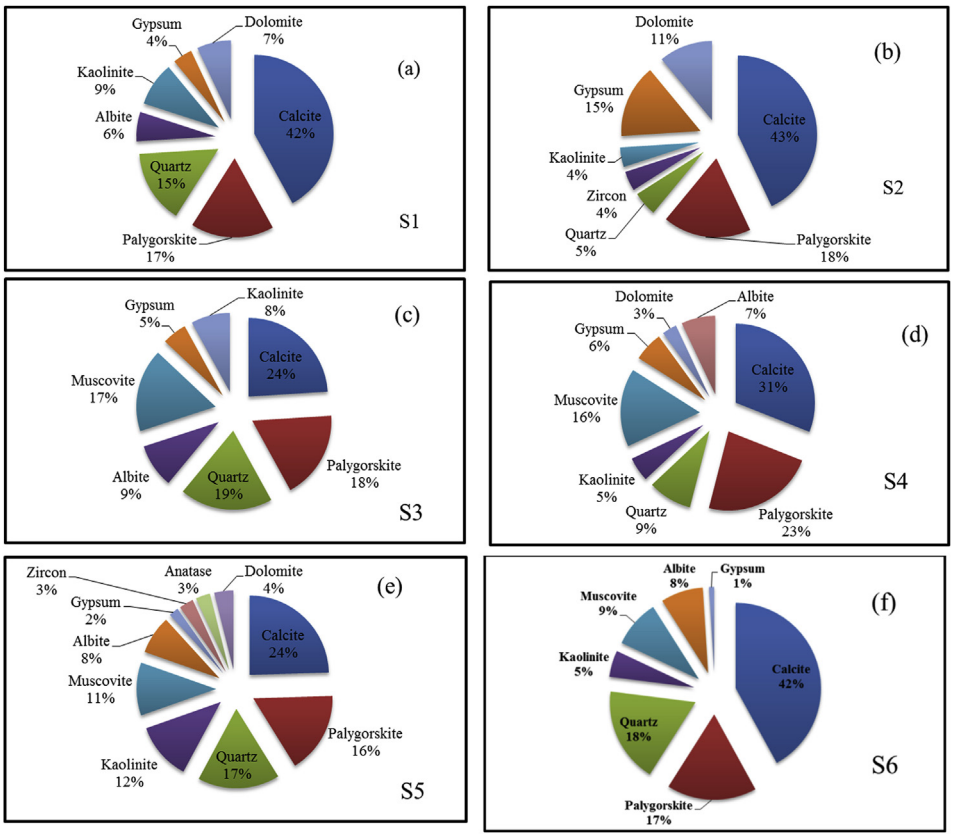

Fig. 9. Mineralogical composition by XRD analysis of collected atmospheric dust samples at various locations in Shiraz.

Kandler, 2014). A semi-quantitative analysis (wt.%) result of six atmospheric dust samples analysed via XRD isillustrated in Fig. 9. Notably, the values calculated by XRD analysis should be interpreted carefully because this method does not provide an accurate quantitative analysis. These values only present an overview of the approximate magnitude order of crystallographic composition. Nevertheless, the analysis of XRD data showed that the Shiraz atmospheric dust samples were predominantly composed of calcite ($CaCO_3$), quartz (SiO_2) and palygorskite ($(Mg, Al)_2Si_4O_{10}(OH)4H_2O$). The two calcareous particles, consisting mainly of calcite and dolomite ($CaMg(CO_3)_2$) (24%–42%) and palygorskite (16%–23%) compounds, accounted for more than 40% of the dust particle contents in all of the collected samples. The third dominant mineral in the six samples was quartz with a mass percentage between 19% and 5%, whereas muscovite ($KAl_2(AlSi_3O_{10})(FOH)_2$), albite ($NaAlSi_3O_8$) and kaolinite ($Al_2Si_2O_5(OH)_4$) contributed approximately 14%, 11% and 7%, respectively. The remaining compounds contained a small proportion of the total mass of dust, and gypsum ($CaSO_4{\cdot}2(H_2O)$) did not exceed 15% in all the studied samples. The other primary silicate minerals, namely, zircon ($ZrSiO_4$) and anatase (TiO_2) as heavy minerals, were observed only in samples with low average mass percentages (3%–4%). Notably, palygorskite and quartz were more dominant in Shiraz atmospheric dust than other silicates (muscovite, albite and kaolinite). The presence of heavy minerals in several samples (Fig. 9) indicates that mineral dust originating from dry lands of Iraq, Saudi Arabia, Syria and Jordan were probably the major source of the dust events that occurred in the Middle East and Iran (Awadh, 2012; Najafi et al., 2014).

3.4. Backward trajectory simulation

In this study, backward trajectory simulations were performed to identify the source of dust events and motion direction of dust plume over Middle East and the study area in May 2018. Generally, backward trajectory simulations are helpful in tracking the air mass path and aerosols at the receptor site. Fig. 10 shows the backward trajectory modelling plots of Shiraz for three initial evaluations of 50, 500 and 1000 m (at 0 UTC on May 8 (Fig. 10a) and at 0 UTC on May 15 (Fig. 10b)). On May 8, 2018, the air mass affecting Shiraz was mainly passing through the eastern desert of Saudi Arabia, Kuwait and Khuzestan Province.

4. Discussion

The SEM analysis for dust particles showed a range of different morphologies including rhombic, sub-angular, semi-rounded and irregular shapes with sizes varying between 2 μm and 50 μm. The shape of dust particles is closely related to the geological nature of dust provenance, meteorological conditions and transport distance among dust provenance and deposited region (Al-Dabbas et al., 2012; Ahmady-Birgani et al., 2015). Calcite was the dominant mineral in all of the studied samples with different shapes and sizes. The variation in the size and shape of the calcite particles may be due to their low hardness and high solubility. Different morphology of calcite can be related to the dust sources. For example, calcite particles with trigonal–rhombohedral crystals could have originated from the nearest area compared to the irregular shapes that might have transported a long-distance to the study area. Also, diatom detected in SEM analysis as a unique feature. Several scholars hypothesised that diatom is a biological marker to identify atmospheric dust sources lifted from African dry lakebeds (Muhs et al., 2014). In separate papers, Moreno et al. (2006) and Chou et al. (2008) reported that the Bodélé Depression in the southern edge of the Sahara Desert in Chad is the main geogenic source of silicate dust, which commonly originates from dehumidified diatomaceous lake deposits. Thus, the presence of diatom as a unique feature in the atmospheric dust samples in Shiraz is possibly associated with a specific source region (e.g. originated from dry lakebeds), despite the fact that in Iran and neighbouring countries (Iraq and Arabian Peninsula), the existence of diatoms

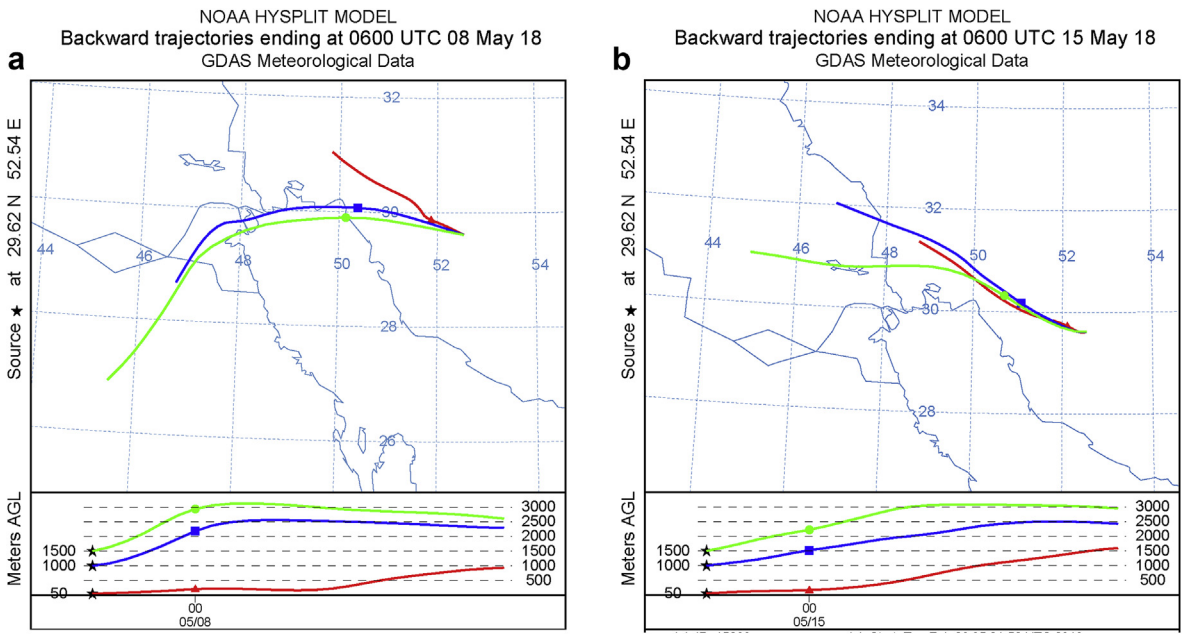

Fig. 10. HYSPLIT back trajectory simulations (a) at 0 UTC on May 8 and (b) at 0 UTC on May 15. The blue, red and green colour lines denote isobaric contours beginning at 50, 500 and 1000 m, respectively. These plots are calculated by NOAA's HYSPLIT trajectory model.

in dust storms has not been reported. Moreover, Dehghan Madiseh (2013) identified 85 species of diatoms in Hoor al-Azim Wetland. Our argument is that the presence of diatoms in the collected samples is probably related to the dry bed of Hoor al-Azim Wetland that is located in the western borders of Iran and Iraq and known as the main local source of a dust storm in western Iran (Mojadam et al., 2018). Notably, the appearance of diatom in the studied samples is not enough to support this theory. Therefore, further research is recommended.

The Ca/Al ratio in the studied samples (1.14) was greater than the UCC average (0.37). By contrast, Ca and Al yielded similar elemental ratios for atmospheric dust samples of Iraq (1.31), Kuwait (1.23) and the Arabian Peninsula (2.17), but a significant difference was observed in the Ca/Al ratios (studied samples = 1.14, UCC = 0.37). This difference is attributed to the regional distribution of calcisols in the mentioned countries. Meanwhile, the Mg/Al ratio of the studied samples was lower than those from Iraq and Kuwait. This result reflects the larger contribution of Al–Mg-bearing minerals to the chemical composition of Shiraz atmospheric dust compared to dolomite. The Mg/Al ratio in the current study is close to the ratio for Saudi Arabia due to the high abundance of palygorskite in the sediments and soils of the Eastern Arabian Peninsula (Shadfan et al., 1985).

The lithogenic elements (Ti, K, Na, Mg, and V) and several trace elements (Co, Sr, Zn, Pb, Mo, and Cu) showed crustal EFs less than 2 with respect to Al (UCC) in the samples from the study area, thereby demonstrating a predominantly geogenic source. The crustal EFs for Ca ranged between 2.82 and 3.35. The high value of EFs for Ca suggests the widespread distribution of carbonate rocks in the Middle East. The results indicate that the crustal EF values of As, Cd, Cr and Ni ($2 < EFs < 5$) exhibit a modest enrichment. Hence, atmospheric dusts are indeed influenced by anthropogenic sources when they are transported to Shiraz. This influence is likely due to increasing industrial activities near Fars Province, particularly in oil and gas industries. The most probable reason for these observations is the petroleum industries in Khuzestan and neighbouring countries (Zarasvandi et al., 2011; Tsiouri et al., 2015), which are rich in dust-borne heavy metals, resulting in fly ash with a relatively high abundance of Ni, Cr, Cd and As that can be absorbed by particulate matter, such as clay minerals.

The present mineralogy results are consistent with those of Engelbrecht et al. (2009), Zarasvandi et al. (2011), Al-Dabbas et al. (2012), Hojati et al. (2012), Modaihsh and Mahjou (2013), Najafi et al. (2014), Ahmady-Birgani et al. (2015) and Javed et al. (2017), who conducted studies in the Middle East region. Hence, dust episodes in the southern and western parts of the Middle East are expected to have a similar mineral composition. The high contents of quartz and calcareous particles, the low content of gypsum and the presence of kaolinite, albite and muscovite in the dust were mainly due to a detrital sedimentary basin (Zarasvandi et al., 2011). Engelbrecht et al. (2009a), Modaihsh and Mahjou (2013) and Najafi et al. (2014) argued that middle and eastern regions of Iraq, in addition to the western parts of Saudi Arabia, are possibly the primary source of carbonatic phases during dust storms in western and southern Iran. Also, Modaihsh (1997) stated that the high value of calcite (32%) in the atmospheric dust of western parts of Saudi Arabia originate from calcisols, which is distributed in the Arabian Shelf, because the amount of rainfall is insufficient for leaching out carbonates. Similar findings on mineralogy with dominant carbonates and quartz have been reported for dust episodes in Iraq and Kuwait (Al-Dabbas et al., 2012; Al-Awadhi and Al-Shuaibi, 2013).

In addition, palygorskite occurrences have been recognised in dust samples in the central and western regions of Iran (Hojati et al., 2012; Najafi et al., 2014). Palygorskite as a hydrous Mg–Al silicate is a rare clay mineral that is unsteady in the regions with precipitation levels of more than 300 mm. Thus, arid climates are a favourable environment for the formation of palygorskite in soil. Researchers have argued that palygorskite is the main clay mineral in potential source soils and sediments of Iraq (Dhannoun and Al-Dabbagh, 1988; Al-Juboury et al., 2009), Saudi Arabia (Shadfan et al., 1985) and Jordan (Shadfan and Dixon, 1984). Hence, the occurrence of palygorskite in dust samples can be described by dust events uplifted from dry and semi-arid regions in Iraq, Saudi Arabia and Jordan. The high palygorskite frequency in all of the current samples can thus be considered a useful indicator for identifying the source of dust storms in southern Iran. The dominant clay minerals, namely, palygorskite and kaolinite, are attributed to certain climatic environments because these minerals reflect arid and semi-arid climatic conditions (Al-Dabbas et al., 2012). These source areas are in accordance by backward trajectory simulations of dust plume trails (Fig. 10).

5. Conclusion

To determine the characteristics of atmospheric dust and identify the potential provenance of dust storms, dust mineralogy and geochemistry

characteristics in the area of Shiraz, southern Iran, were investigated by gathering atmospheric dust samples at 10 sites.

The results of meteorological data indicated that spring has the largest dust phenomenon, and most dusty days occurred in May. Prevailing winds were from western and south western to easterly directions.

SEM analyses of dust samples showed that atmospheric dust has semi-rounded irregular, sub-angular, prismatic and rhombic shapes, with clay-sized particles as small visible aggregates. Moreover, Shiraz atmospheric dusts vary in size from 50 μm to 0.8 μm. However, the dust size is smaller than 10 μm in most cases. The diatom detected in the samples of the study area is a noticeable feature. The diatom present in the collected dusts may have originated from dried-up beds of Hoor al-Azim Wetland in the southwest of Iran.

Shiraz atmospheric dust has higher mean Ca and Mg content than UCC, whereas other major elements in the samples are similar to the UCC value. For trace elements, the mean concentrations of As, Cd, Co, Cr, Cu, Ni, Pb, V and Zn in the dust are higher than those in UCC. The studied elements in Shiraz atmospheric dust can be divided into two groups based on cluster analysis and crustal enrichment factor: (1) Ni, Sr, As, Cr, Cd and Zn represent affected by anthropogenic sources, and (2) Co, V, Mo, Cu and Pb likely originate from a geogenic source. The EF values of As, Cd, Ni and Cr ($2 < EFs < 5$) are indicative of modest enrichment. This enrichment suggests that the atmospheric dust transported to Shiraz is affected by human activities, especially petroleum industries.

On the basis of the results of XRD analyses, six mineral groups were identified in Shiraz atmospheric dust as follows: (1) carbonates (calcite and dolomite), (2) silica (quartz), (3) clay minerals (palygorskite and kaolinite), (4) primary silicates (muscovite and albite), (5) evaporates (gypsum) and (6) heavy minerals (zircon and anatase). Despite the similarities in mineralogy of the studied atmospheric dust samples and dust from other regions of Iran, the relative abundance of palygorskite is different. The high presence of palygorskite in all samples could be a useful tracer for detecting the provenance of atmospheric dust episodes in southern Iran. For instance, Khalaf et al. (1985) showed that the dominant clay-mineral fraction of Kuwait dust storms is characterised by a high relative frequency of palygorskite (40%–50%). The authors posited that palygorskite originated from ancient Mesopotamian floodplain deposits in southern Iraq and Sabkha sediments in the southwest area of the Persian Gulf. If the source of dust storms had been principally from desert areas bordering the Arabian Peninsula and Iraq, then palygorskite would have been much more abundant than it is at present. Therefore, the high occurrence of palygorskite in the collected dust samples is not surprising and indicates that these areas are the likely sources of dust events in southern Iran. Nevertheless, dust storms occur because of multiple environmental factors, and any comprehensive study necessitates the incorporation of interdisciplinary science to thoroughly understand the nature of this phenomenon. However, in accordance with the aforementioned discussions and the results of Khalaf et al. (1985), Shadfan and Mashhady (1985) and El-Sayed (2001), we posit that aridisols from the Mesopotamian floodbasin (in Iraq), the desert area located in the southwest of the Persian Gulf and the eastern parts of Saudi Arabia and Kuwait are three probable sources of dust emission dominated by palygorskite in the south of Iran.

Acknowledgments

This study was financially supported by the Shiraz University and INSF (Iran National Science Foundation, Project No. 97002616). Thanks to two anonymous reviewers and the editorial comments of Prof. Vinod O. Samuel for their critical comments, which helped us to improve the quality of the manuscript.

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