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Geochemical Behaviour of the Tunisian Background Aerosols in Sirocco Wind Circulations

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ABSTRACT

This study examines spatial and time evolutions of the principal constituents of the Tunisian background aerosols under Sirocco wind circulations. Aerosols coming from the Sahara Desert were found to be loaded with particulate matter, especially silicon. The aerosols were shown to have varying geochemical behaviour along the "South-North" displacement of the Saharan plumes, depending on the wind flow characteristics, geomorphologic features and the nature of soils swept by the wind. In the south and the center part of the country, the transfer of aerosol constituents to the soil (by gravity and/or impaction) was probably predominated by localized enrichment phenomena. The latter are reinforced by the effect of turbulent winds over bare soils, wind wakes and probably selective disintegration, especially in the vicinity of the geomorphologic features of central Tunisia. These relatively high features, extending over important distances, appear to be of paramount importance for the phenomena of redistribution of aerosol constituents even during periods without Sirocco wind circulations. In the northern section of the country, aerosol constituent concentrations dropped to almost 50%, in spite of the abundance of localized turbulent winds. This may be explained by the effect of forests and the relatively dense vegetation cover, which clearly reinforces the transfer phenomena to the soil and the attenuate of dust entrainment.

Key words: aerosols, Sahara, Sirocco, Tunisia, turbulent winds

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1. Introduction

North Africa is widely regarded as the Earth's largest source of dust (N'Tchayi et al., 1997; Prospero et al., 2002; Washington et al., 2003). Dust emissions from this region stand out in terms of magnitude and spatial extent. The Saharan sources are considered by far the most active ones in the world, although their contribution is mainly confined to the northern hemisphere in the summer (Prospero, 1980; Prospero et al., 1981; Engelstaedter et al., 2006). Besides North Africa, major dust activity is evident in the Arabian Peninsula, in Iraq, Turkmenistan, Afghanistan, Northern India, the Namib and Kalahari Deserts and the Tarim Basin in China. Estimates of the annual Saharan dust are 1600 Tg yr^{-1} (Ozer, 2001), while those of the global dust emissions range from 1000 to 3000 Tg yr^{-1} (IPCC, 2001; Zender et al., 2004).

In the spring and summer, the air over North Africa is almost permanently loaded with significant amounts of dust, which are mobilised northward and eastward along the Mediterranean coast under low-pressure systems. Dust storm events first appear in the western Mediterranean Sea and move eastward to reach the eastern coast, where three periods of increased atmospheric dust are distinguished: (i) the spring (March–May), (ii) summer (July–August) and (iii) autumn (September–November) (Engelstaedter et al., 2006). The latter show three pathways of dust transport trajectories from North African sources. First, dust is transported over large distances across the Atlantic Ocean to the United States, the Caribbean and South America (Swap et al., 1992; Perry et al., 1997; Prospero and Lamb, 2003); secondly, dust is conveyed towards the Mediterranean Sea and Europe (Aviala and Penuelas, 1999; Borbely-Kiss et al., 2004); and third,

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dust moves towards the eastern Mediterranean and the Middle East (Ganor, 1994; Kubilay et al., 2000; Israelevich et al., 2003).

Erosion, transport and deposition of Aeolian dust have major environmental and economic consequences (Pye, 1987; Goudie and Middleton, 2001). Adverse effects include soil erosion, air pollution, vegetation damage, building corrosion and visibility reduction, which may lead to airport closures and road accidents. The resulting atmospheric aerosols can directly affect the climate by scattering and absorbing solar radiation (Charlson et al., 1992) and indirectly by their ability to nucleate cloud droplets (Twomey, 1977). Besides their climatic effects, light scattering and absorption by aerosols cause a reduction in solar radiation (Luria, 1996) and influence the visual air quality, thereby adversely affecting the visibility and the quality of the atmosphere (Seinfeld and Pandis, 1988).

Tunisia, like its North African neighbour, is fully subjected to the Saharan influences. On a seasonal basis, this Saharan wind named Sirocco (locally known as chehili or ghibli), is most frequent in the spring and summer (Bousnina, 1990). It blows from the south, southwest or deviates to the southeast and generally precedes a slightly active cold front. Its turbulent movement is frequently responsible for the localized lifts of fine sand particles in the atmosphere (Azri et al., 2002). It causes an increase in aerosol constituent concentrations, which may reach 10 times the average value (Azri et al., 2002). It is associated with a remarkable drop of relative humidity and an important elevation of temperature, especially in the summer period.

The present study examines the evolution in time and space of the principal constituents of the Tunisian background aerosols under Sirocco wind situations. The objective is to identify its geochemical behaviour along the "south-north" transfer of the Saharan plumes over Tunisia; a North African country characterized by an arid to semi-arid climate and a marked zoning of geomorphologic features.

2. Geomorphological context

The Tunisian geography is subdivided into the following three important geomorphologic units, from south to north (Fig. 1) (Ben Ayed, 1993):

(1) Southern Tunisia: lying next to the Sahara Desert, it contains vast plains characterized by very sparse vegetation and different soils rich in limestone, silica and gypsum (Watson, 1985; Mtimet, 1999). The southwest is a part of the low Sahara basin bordered on its northern side by the chotts line, and on the southern side by the desert, characterized by endless

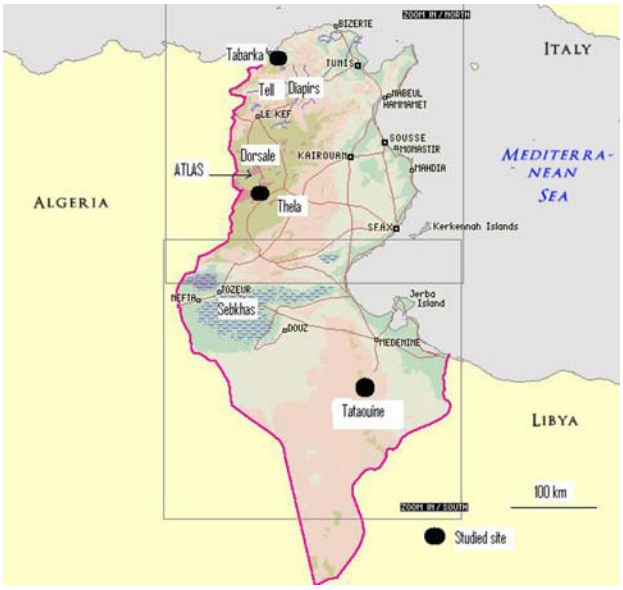

Fig. 1. Location map for aerosol sampling sites.

wind-swept dunes or sun-scorched fields of rock and stone. It represents the Dahr Plateau composed of discontinuous mountainous chains of modest altitudes, varying between 400 and 600 m, and the Jeffara Plains including Sebkhas of Chott Eljerid ($\approx 2 \times 10^6$ hm^2), Chott Elgharsa ($\approx 2 \times 10^5$ hm^2) and Chott Elfejej ($\approx 10^5$ hm^2). This region is characterized by limited rainfall, where the annual average does not exceed 150 mm yr^{-1}.

(2) Central Tunisia occupied by the steppes: These are divided into high and low steppes. The high steppes represent a region of lofty mountains continuously extending over a distance of more than 300 km. These chains, with a southwest to northeast orientation, represent a limit between central and north Tunisia. They extend from the north of Gafsa to the south of Dorsale, which is composed of chains of high altitudes, reaching 1544 m. The low steppes stretch over wide alluvial plains and hills cut across by large creeks. This region is also characterized by different soils more enriched in limestone compared to those of south Tunisia (Mtimet, 1999). The vegetation cover is made up of forests, often stunted, and fields of alfalfa grass. The continental climate contributes to the bareness of the region. This area has gradually developed into farming land, in which olive tree prevails. The average annual precipitation over this region varies between 200 and 450 mm yr^{-1}.

(3) North Tunisia occupied by the Tell and vast plains: The Tell is a region of relatively modest altitude (400 to 800 m) covered by forests. It is characterized by an important rainfall, reaching an annual aver-

age of 1000 mm. The plains are composed of different soils distinguished by the abundance of clay, enriched in Fe and organic matter with a dense vegetation cover (Mtimet, 1999). Furthermore, outcrops of evaporitic rocks (∼800 hm^2) enriched with potassium sulphates are also incorporated in these plains. They correspond to the diapirs zone.

The three aforementioned physiographic groups represent the oriental continuity of the Magrebine Ranges, which extend further west over Algeria and Morocco.

3. Meteorological context

The analysis of synoptic maps at the surface and at higher altitudes (at 500 hPa) during the four selected Sirocco periods, showed the following:

(1) During the first period (29 and 30 March 2003), the synoptic situation over Tunisia was characterized by the predominance of a current from the southwest supported by a thermal depression at the surface centred over Croatia (1010 hPa), and another high altitude current from the southwest causing sunny weather, with an increase of the maximum temperatures varying between 22°C and 32°C;

(2) During the second period (14 to 16 April 2003), the situation was characterized by the presence of a surface thermal depression (1000 hPa) over southern Italy and the predominance of a high altitude southwest current. This situation caused a southwest regime at the surface and also at higher altitudes (at 500 hPa) over Tunisia, resulting in hot Saharan air thrusts and high temperatures, varying between 26°C and 35°C;

(3) During the third period (13 to 15 July 2003), Tunisia was influenced by a thermal depression at the surface (990 hPa) centred over the east of Spain. This situation was associated with the predominance of a current at higher altitudes from the southeast related to the existence of a low geopotential area at 500 hPa (5380 gpm), which resulted in substantial increases of the temperatures over the Tunisian regions. Maximum temperatures reached 43°C in certain regions of Southern Tunisia.

(4) During the fourth period (28 to 31 August 2003), the synoptic situation was distinguished by the presence of a surface thermal depression (1000 hPa) centred over France, the predominance of a current from the southeast sector and another current at higher altitudes from the southwest, related to a low geopotential area at 500 hPa (5400 gpm). This resulted in very hot weather over Tunisia, where maximum temperatures varied between 36°C and 45°C.

During the four Sirocco periods, the wind rose (at 10 m above ground level) showed that the dominant wind direction (70% to 80% of total observations) fluctuated between the southeast and southwest sectors, for each selected site. Average wind velocities were shown to be moderate in the South and Center and relatively higher in the northern part of the country. They varied from 3 to 5 m s^{-1} for the "Tataouine" and "Thela" sites and reached values varying between 7 and 9 m s^{-1} in the vicinity of the "Tabarka" site. The remaining winds, representing only 20% to 30% of the total observations, have variable directions and magnitudes reaching 1.5 times those of the dominant winds. They are probably influenced by turbulent winds associated with the Sirocco blowing over the Tunisian regions. Under these situations, horizontal visibility was shown to be less than 5 km for each selected site.

At the aforementioned sites, back trajectories calculated at an altitude of 1000 m for each Sirocco period, showed the presence of four different paths (Fig. 2) originating from the Algerian Sahara for the first and second periods, and from the Libyan Desert for the others. Back trajectories have been calculated using the HYSPLIT 4 model (Draxler and Rolph, 2003; Rolph, 2003).

During the "post-Sirocco" days, when the depressions moved towards the centre (for the first two) and east of the Mediterranean Basin (for the last two), changes in the "southwest" and "southeast" wind regimes were registered. Predominant currents from the northwest, northeast and eastern sectors were established (between 85% to 95% of the observations). They resulted in a relative decrease of the air temperature, consisting of a drop of 2°C to 5°C of the maximum values all over the country. Slightly active disturbances (without storms) associated with relatively strong winds occurred especially in the North. Measured wind velocities varied between 15 and 19 m s^{-1} in "Tabarka", between 10 and 14 m s^{-1} in "Thela" and only from 7 to 9 m s^{-1} in "Tataouine".

4. Material and methods

The Tunisian aerosol quality under selected Sirocco wind situations is characterized by sampling particles present in the air at three distinct sites: "Tataouine", "Thela" and "Tabarka" (Fig. 1). Their corresponding characteristics are presented in Table 1.

Aerosol sampling at the selected sites was based on weather forecasts given by the Tunisian National Institute of Meteorology. Four periods influenced by Sirocco winds were selected:

First period: 29 to 30 March 2003; Second period: 14 to 16 April 2003; Third period: 13 to 15 July 2003; Fourth period: 28 to 31 August 2003.

Four other periods without Sirocco were also se-

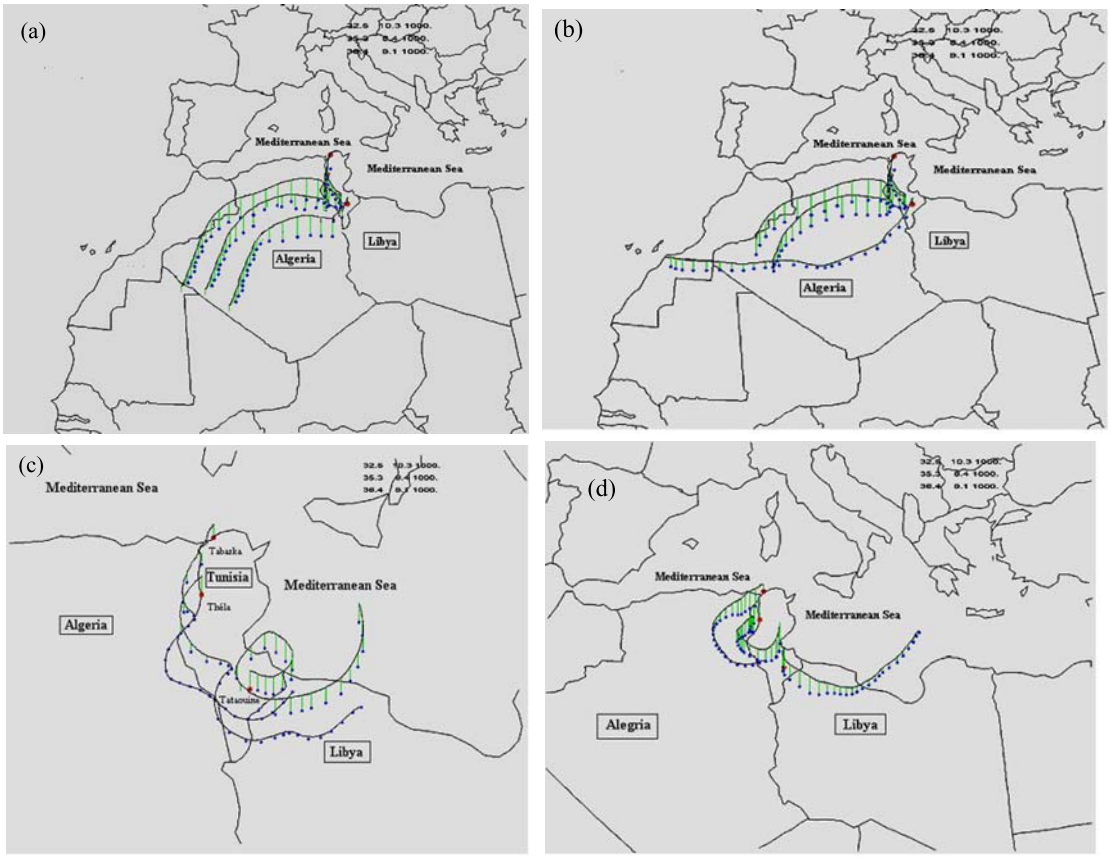

Fig. 2. Back trajectories calculated at an altitude of 1000 m for each Sirocco period: (a) 29 and 30 March 2003; (b) 14 to 16 April 2003; (c) 13 to 15 July 2003 and (d) 28 to 31 August 2003.

Table 1. Characteristics of the aerosol sampling sites.

Site	Type	Altitude above sea level (m)	Latitude	Longitude	Distance from the coast (km)
Tataouine	Rural region in southern Tunisia	242	32°55′N	10°27′E	100
Thela	Rural mountainous region in central Tunisia	1091	35°33′N	8°39′E	200
Tabarka	Coastal rural region in northern Tunisia	158	36°44′N	9°11′E	2

lected. They correspond to 31 March, 17 April, 16 July and 1 September 2003, representing the days that immediately follow the Sirocco periods.

Aerosol sampling was performed at 3 meters above the ground level by a total filtration of the air using nucleopore filters, characterized by a diameter and a porosity of 4.7 cm and 0.45 μm, respectively. The threshold aerodynamic diameter of the aerosol sampler nozzle was 10 μm. Aerosol sampling was performed on a daily basis at an hourly rate of 180 L. The collected samples were analysed by X-ray wavelength florescence spectrometry. Each sample was counted twice with K-alpha (or L-beta Pb lines) and PGFD or CS detectors with a 3 kW Siemens SRS 303 spectrometer and Rh tube.

5. Results and discussion

5.1 *Spatial and temporal evolution of the aerosol constituents*

Chemical analysis of the aerosol matter sampled during the considered Sirocco periods showed that the concentrations of Si, Ca, K, P, Al, Fe, Ti, Mg, Na, and Cl are characterized by a marked variability in time and space (Tables 2, 3, and 4). The total concentra-

Fig. 3. Spatial and temporal evolutions of the total concentration of aerosol constituents during Sirocco periods.

tions of the analyzed chemical elements were shown to vary between 56 and 180 μg m^{-3} (Fig. 3). The largest values were registered within the first two sites of "Tataouine" and "Thela". They fluctuated between 120 and 180 μg m^{-3}. Relatively low values varying between 56 and 80 μg m^{-3} were observed at the third site ("Tabarka"). In the vicinity of each site, the temporal evolution of the aforementioned elements is also shown to be characterized by very distinct values.

Examining Fig. 3, we can note that during the displacement of the Saharan plumes from "Tataouine" to "Thela", that is from Southern to Central Tunisia (Fig. 2), the evolution of the total concentrations of the analyzed elements show similar values, with particular specific maxima. The appearance of maxima in the vicinity of the "Thela" site, where concentrations exceed those registered at that of "Tataouine" (located upstream), may be explained by the marked effect of enriching sources. This may include the importance of the effect of turbulent winds in entraining dusts from poorly vegetated soils within regions of southern and central Tunisia and also eastern Algeria. Nevertheless, the blocking effect of the mountainous chains of central Tunisia, with a southwest to northeast orientation, cannot be ignored. They may reinforce particle concentrations by the phenomenon of wind wake created upstream and downstream of these lofty chains, characterized by a considerable spatial extension (300 km) facing the main Sirocco wind flow direction. These geomorphologic features were shown to cause wind wake cavities, where particle concentrations can be multiplied by a factor of 10 (Ryde, 1970; Bach, 1976).

The displacement of the Saharan plumes further north from "Thela" to the coastal site of "Tabarka" resulted in reduced concentrations of the analyzed elements, which decreased by almost 50% (Fig. 3). Such a drop may be attributed to the forest belt (with a relatively humid atmosphere) covering the Tell, which is located at a distance of about 30 km upstream of "Tabarka". This forest belt may reinforce the phenomena of the transfer of aerosol constituents to the soil (deposition by gravity and/or impaction). On the other hand, the abundance of a dense vegetative cover in the north may attenuate the phenomena of dust entrainment.

The aerosol elementary analysis at each studied site along the selected periods showed the abundance of silicon (Tables 2, 3 and 4). The corresponding rates fluctuate between 75% and 90% for both the "Tataouine" and "Thela" sites, with concentrations oscillating between 104 and 163 μg m^{-3}. At the "Tabarka" site, in spite of the remarkable decrease of the total concentration of the analyzed elements, silicon concentrations and the rates remained considerable in the aerosols (from 41 to 64 μg m^{-3}; cf. Table 4, with rates varying between 63% and 81%). The significant silicon concentrations in the aerosols at this

Table 2. Concentrations of the analyzed constituents of the aerosols sampled at the "Tataouine" site in periods of Sirocco (μg m^{-3}).

	Date (day/month/year)	P	Si	Ca	K	Al	Fe	Ti	Mg	Na	Cl	Sum
First	29/03/2003	0.015	120.212	12.162	0.947	0.505	0.107	0.031	0.328	2.631	1.040	137.978
	30/03/2003	0.016	118.918	12.394	1.721	1.929	0.113	0.095	0.678	3.362	0.190	139.416
Second	14/04/2003	0.011	162.521	11.582	0.760	0.534	0.091	0.031	0.251	1.479	0.340	177.600
	15/04/2003	0.015	114.857	15.682	1.609	1.359	0.130	0.080	0.582	2.227	0.110	136.651
	16/04/2003	0.014	111.519	11.499	1.342	0.183	0.123	0.085	0.427	1.168	1.640	128.000
Third	13/07/2003	0.004	118.610	10.928	0.275	1.622	0.101	0.011	0.095	0.271	1.150	133.067
	14/07/2003	0.012	113.869	14.526	1.282	1.083	0.097	0.031	0.611	3.860	6.230	141.601
	15/07/2003	0.017	115.953	12.062	0.809	0.913	0.113	0.051	0.280	1.023	7.320	138.541
Fourth	28/08/2003	0.029	109.603	14.977	1.658	1.431	0.084	0.078	0.577	2.003	1.970	132.410
	29/08/2003	0.024	105.578	13.156	0.935	0.845	0.111	0.040	0.403	1.927	2.279	125.298
	30/08/2003	0.022	102.815	12.290	0.974	0.713	0.134	0.051	0.268	1.004	1.918	120.189
	31/08/2003	0.093	136.448	12.654	5.335	5.514	0.197	0.308	1.336	0.549	0.612	163.046

Table 3. Concentrations of the analyzed constituents of the aerosols sampled at the "Thela" site in periods of Sirocco (μg m^{-3}).

	Date (day/month/year)	P	Si	Ca	K	Al	Fe	Ti	Mg	Na	Cl	Sum
First	29/03/2003	0.016	110.164	12.114	1.060	0.007	0.134	0.087	1.040	0.208	0.191	125.021
	30/03/2003	0.014	117.958	17.125	3.604	2.686	0.161	0.153	0.065	7.070	6.325	155.161
Second	14/04/2003	0.012	116.256	12.021	0.761	0.930	0.143	0.052	1.610	5.171	1.123	138.079
	15/04/2003	0.029	113.338	11.297	0.390	0.456	0.145	0.024	0.880	0.297	0.978	127.834
	16/04/2003	0.042	128.691	12.132	1.127	0.735	0.155	0.063	2.310	0.217	1.739	147.211
Third	13/07/2003	0.014	111.522	11.657	0.308	0.224	0.140	0.012	0.600	1.588	1.022	127.087
	14/07/2003	0.027	110.086	12.979	1.899	0.194	0.142	0.155	1.600	0.246	1.248	128.576
	15/07/2003	0.022	105.433	11.756	0.781	0.853	0.176	0.042	1.410	0.594	0.927	121.994
Fourth	28/08/2003	0.012	105.238	17.342	0.865	0.775	0.139	0.076	0.876	0.532	0.894	126.749
	29/08/2003	0.023	104.133	11.486	0.754	0.556	0.136	0.300	0.042	0.367	1.938	119.735
	30/08/2003	0.018	115.728	17.166	0.878	0.544	0.161	0.420	0.030	0.432	1.719	137.096
	31/08/2003	0.066	112.243	14.774	4.484	1.899	0.221	0.075	0.117	2.585	2.153	138.617

Table 4. Concentrations of the analyzed constituents of the aerosols sampled at the "Tabarka" site in periods of Sirocco (μg m^{-3}).

	Date (day/month/year)	P	Si	Ca	K	Al	Fe	Ti	Mg	Na	Cl	Sum
First	29/03/2003	0.012	55.393	11.243	1.063	0.210	0.296	0.016	0.171	1.545	1.723	71.672
	30/03/2003	0.004	57.211	10.916	0.537	0.192	0.958	0.015	0.113	0.610	0.835	71.391
Second	14/04/2003	0.008	53.719	11.053	0.462	0.120	0.338	0.009	0.121	0.872	1.312	68.014
	15/04/2003	0.010	41.063	11.544	0.706	0.029	0.752	0.005	0.124	0.783	1.107	56.123
	16/04/2003	0.023	42.520	11.980	1.212	0.411	0.885	0.010	0.119	0.678	0.932	58.770
Third	13/07/2003	0.021	51.182	11.847	0.910	0.053	0.331	0.008	0.127	1.672	2.025	68.176
	14/07/2003	0.050	55.190	13.638	3.142	0.080	1.334	0.012	0.201	1.932	2.475	78.054
	15/07/2003	0.024	41.346	14.359	6.154	0.094	0.700	0.017	0.241	1.321	2.310	66.566
Fourth	28/08/2003	0.023	41.317	10.091	0.048	0.039	0.842	0.010	0.110	2.624	4.638	59.742
	29/08/2003	0.019	63.546	10.303	0.189	0.080	0.501	0.020	0.102	1.963	3.119	79.842
	30/08/2003	0.044	52.396	12.773	3.497	0.090	0.452	0.025	0.132	3.315	5.232	77.956
	31/08/2003	0.027	54.833	13.574	1.071	0.704	3.173	0.075	0.110	1.382	2.633	77.582

particular site may be related to the additional local enrichment from the sand dunes bordering the coast. This phenomenon may also be enhanced by the characteristics of the local air flow, which include:

(1) The abundance of localized turbulent winds, which improve the rise of dust, especially silica, highly influenced by the saltation phenomena (Pye, 1987).

(2) The relatively high wind velocity (averages are between 7 and 9 m s^{-1}), which was shown to reinforce earth dust entrainment in the air when velocities exceed 7 m s^{-1} (Reydet, 1984; Belghith, 1999).

The concentrations of the aerosol constituents at all of the studied sites were found to be very important during the selected Sirocco periods. They exceed by almost 2.5 to 8 times those of the aerosol constituents sampled in situations without Sirocco (Fig. 4). Increases of approximately 10 times were also found in other periods characterized by Saharan influences (Azri et al., 2002). This clearly shows the effect of Sirocco winds in enriching the Tunisian background aerosol with particulate matter, especially silicon. It is important to note at this stage that during periods without Sirocco, silicon concentrations did not exceed 5 μg m^{-3}, (Table 5), with rates rarely exceeding 10% of the total concentrations of the analyzed elements.

Different approaches were adopted to better understand the phenomena of enrichment of the sampled aerosols in the Sirocco periods and to determine the origin of its constituents. These approaches include enrichment factors (EF), contribution rates of feeding potential sources and intra-sites enrichment rates.

5.2 *Enrichment factors*

Different studies performed on continental aerosols showed that aluminium (Al) might be considered as the most representative indicator of the crustal source. It is very abundant in the earth's crust (81300 ppm) and slightly emitted by the marine source (0.002 ppm). Its anthropogenic emissions are also negligible. Therefore, this element was considered as a crustal reference to study the enrichment of the aerosol particles with respect to the earth's crust.

Sodium (Na) and chlorine (Cl) may be selected as marine reference elements because of their abundance in the marine source. Nevertheless, the presence of crustal sodium with non-negligible quantities

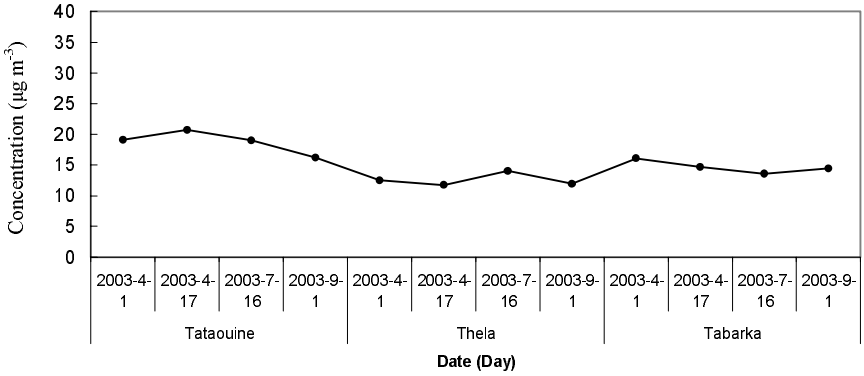

Fig. 4. Spatial and temporal evolutions of the total concentration of aerosol constituents during "no Sirocco" periods.

Table 5. Concentrations of the analyzed constituents of the aerosols sampled at the "Tabarka" site in periods without Sirocco (μg m^{-3}).

	Date (day/month/year)	P	Si	Ca	K	Al	Fe	Ti	Mg	Na	Cl	Sum
Tataouine	01/04/2003	0.007	3.625	1.702	1.075	0.192	0.032	0.004	0.046	5.296	7.120	19.099
	17/04/2003	0.005	4.524	1.665	1.061	0.123	0.029	0.006	0.036	6.256	7.020	20.725
	16/07/2003	0.008	3.732	1.712	2.089	0.132	0.088	0.009	0.039	4.352	6.830	18.991
	01/09/2003	0.005	2.435	1.662	1.083	0.099	0.090	0.009	0.036	4.185	6.618	16.222
Thela	01/04/2003	0.002	0.443	0.534	0.013	0.082	0.017	0.003	0.025	4.376	7.018	12.513
	17/04/2003	0.002	0.376	0.456	0.022	0.085	0.021	0.003	0.011	4.231	6.536	11.743
	16/07/2003	0.003	0.356	0.235	0.013	0.072	0.023	0.005	0.021	6.321	6.987	14.036
	01/09/2003	0.003	0.267	0.424	0.021	0.062	0.016	0.003	0.018	4.543	6.579	11.936
Tabarka	01/04/2003	0.002	1.224	0.174	0.014	0.049	0.024	0.003	0.007	6.435	8.153	16.085
	17/04/2003	0.001	1.141	0.219	0.023	0.036	0.023	0.003	0.004	5.235	8.024	14.709
	16/07/2003	0.001	1.177	0.289	0.022	0.033	0.023	0.002	0.005	4.919	7.113	13.584
	01/09/2003	0.002	1.132	0.165	0.025	0.023	0.019	0.001	0.004	5.459	7.627	14.457

(Na/Al=0.34 according to Mason, 1966) resulted in selecting chlorine as a reference element. Mason (1966) and Brewer (1975) showed that this element is scarce in the earth's crust (approximately 130 ppm) but very abundant in marine waters (18 800 ppm). Its selection in this study was also based on the analysis of its particular temporal evolution, which is different from that of the other elements supposed to be mostly of crustal origin (Tables 3, 4, and 5). Thus, with respect to the selected aluminium and chlorine references, enrichment factors of a selected element (X) are determined by formulae (1) and (2), and may be subdivided into three classes:

$$\mathrm{EF}_{\mathrm{crust}}(X) = \frac{([X] \times [\mathrm{Al}]^{-1})_{\mathrm{aerosol}}}{([X] \times [\mathrm{Al}]^{-1})_{\mathrm{crust}}}\,, \qquad (1)$$

$$\mathrm{EF}_{\mathrm{sea\ water}}(X) = \frac{([X] \times [\mathrm{Cl}]^{-1})_{\mathrm{aerosol}}}{([X] \times [\mathrm{Cl}]^{-1})_{\mathrm{sea\ water}}}\,. \qquad (2)$$

$\mathrm{EF}_{\mathrm{crust}}(X)$: Enrichment factor of the compound X with respect to the crust;

$\mathrm{EF}_{\mathrm{sea\ water}}(X)$: Enrichment factor of the compound X with respect to the sea water;

$[X]$: Concentration of the compound X;

[Al]: Concentration of Al;

$([X] \times [\mathrm{Al}]^{-1})_{\mathrm{aerosol}}$: Concentration of the compound X with respect to that of Al in the aerosol;

$([X] \times [\mathrm{Al}]^{-1})_{\mathrm{crust}}$: Concentration of the compound X with respect to that of Al in the crust;

[Cl]: Concentration of Cl;

$([X] \times [\mathrm{Cl}]^{-1})_{\mathrm{aerosol}}$: Concentration of the compound X with respect to that of Cl in the aerosol;

$([X] \times [\mathrm{Cl}]^{-1})_{\mathrm{sea\ water}}$: Concentration of the compound X with respect to that of Cl in the sea water.

Slightly enriched: EF<10;

Enriched: 10<EF<1000;

Highly enriched: EF>1000.

Because of the lack of models, particularly appropriate to North African soils, those of Mason and Brewer were used to determine the enrichment factors of the different analyzed elements. Data corresponding to these factors are illustrated in Fig. 5. Based on this data, the following points can be noted:

(1) The elements Al, Si, Ca, K, P, Fe, and Ti are essentially of a crustal origin. The aerosol is slightly enriched and enriched in terms of these elements, with respect to the crust (EF<10) and sea water ($10<\mathrm{EF}< 10^9$), respectively;

(2) The chlorine is typically marine for both the

"Tataouine" and "Tabarka" sites because of its high enrichment factor with respect to the crust ($10<EF<10^4$). The aerosol enrichment in terms of this element for both sites is attributed to the maritime influence. In the vicinity of the "Tataouine" site, which is at a distance of 100 km from the coast, the already felt maritime influence is probably attributed to the significant effect of deviated Saharan winds over the Mediterranean Sea. At the "Thela" site, which is at a distance of more than 200 km from the coast, the continental influence is non-negligible. The aerosol is non-enriched to slightly enriched in terms of chlorine, compared to the seawater and to the crust, respectively ($EF<10$). The chlorine is then of a mixed origin;

(3) The sodium has a mixed origin (crustal and marine) for both the "Tataouine" and "Tabarka" sites; the aerosol is slightly enriched in terms of this element compared to both the crust and the seawater. It is of a dominant crustal origin at the "Thela" site.

During the "south-north" displacement of the Saharan plumes, the evolution behaviour of chlorine and sodium in the vicinity of the central "Thela" site may be explained by the phenomena of dust entrainment from the localized Sebkhas in Southern Tunisia, which extends over large areas (Fig. 1), namely Chott Eljerid, Chott Elgharsa, and Chott Elfejej. These Sebkhas were shown to be non-negligible feeding sources of aerosols in terms of chlorine and sodium (Blum et al., 1998).

5.3 *Elementary abundances of collected particles*

The following formulae (3 to 6) were used to estimate the relative contributions of the earth's crust and seawater (Belghith, 1999):

For a given element X with abundance $[X]_{\text{aerosol}}$ in the aerosol mainly originating from the crust $[X]^{\text{crust}}_{\text{aerosol}}$:

$$\frac{([X]\times[\text{Al}]^{-1})^{\text{crust}}_{\text{aerosol}}}{([X]\times[\text{Al}]^{-1})_{\text{crust}}}=1\,. \tag{3}$$

$([X]\times[\text{Al}]^{-1})^{\text{crust}}_{\text{aerosol}}$: Concentration of the compound X with respect to that of Al in the crustal component of the aerosol;

$([X]\times[\text{Al}]^{-1})_{\text{crust}}$: Concentration of the compound X with respect to that of Al in the crust.

Based on the assumption that 100% of aluminium originates from the crust; $[X]^{\text{crust}}_{\text{aerosol}}$ is determined as:

$$[X]^{\text{crust}}_{\text{aerosol}}=([X]\times[\text{Al}]^{-1})_{\text{crust}}\times[\text{Al}]_{\text{aerosol}}\,. \tag{4}$$

$[X]^{\text{crust}}_{\text{aerosol}}$: Concentration of the compound X in the crustal component of the aerosol (its crustal concentration).

$[\text{Cl}]^{\text{crust}}_{\text{aerosol}}$ may therefore be estimated by Eq. (4) and the marine chlorine abundance is given as:

$$[\text{Cl}]^{\text{marine}}_{\text{aerosol}}=[\text{Cl}]_{\text{measured}}-[\text{Cl}]_{\text{crust}}\,. \tag{5}$$

$[\text{Cl}]^{\text{marine}}_{\text{aerosol}}$: Concentration of marine chlorine in the aerosol;

$[\text{Cl}]_{\text{measured}}$: Concentration of total Cl in the aerosol;

$[\text{Cl}]_{\text{crust}}$: Concentration of crustal chlorine in the aerosol.

For a given element X, with abundance mainly originating from the sea $[X]^{\text{marine}}_{\text{aerosol}}$ is given by:

$$[X]^{\text{marine}}_{\text{aerosol}}=([X]\times[\text{Cl}]^{-1})_{\text{marine}}\times[\text{Cl}]^{\text{marine}}_{\text{aerosol}}\,. \tag{6}$$

$[X]^{\text{marine}}_{\text{aerosol}}$: Concentration of the marine component of the compound X in the aerosol;

$([X]\times[\text{Cl}]^{-1})_{\text{marine}}$: Concentration of the compound X with respect to that of Cl in the sea water;

$[\text{Cl}]^{\text{marine}}_{\text{aerosol}}$: Concentration of marine chlorine in the aerosol.

Compared to the crustal source contribution rate, results presented in Fig. 6 show that the rate of contribution of the marine source is very small for the three studied sites. Their cumulative contribution rates, fluctuating between 5% and 75%, may not exclusively explain the enrichment of the studied aerosols. This would imply the contribution of another source, with relatively important rates varying between 25% and 95%. Indeed, in the absence of anthropogenic sources emitting elements of a crustal and/or marine origin, the aforementioned source may be related to the effect of the Sirocco wind flow characteristics and landscape. Such an effect is attributed to:

(i) The abundance of localized turbulent winds all over the country; it is a frequent phenomenon associated with Sirocco that is responsible for dust entrainment;

(ii) The mountainous chains of Central Tunisia, facing the main Sirocco wind flow direction; they reinforce the phenomena of wind wake and probably selective disintegration of aerosol constituents.

Nevertheless, one cannot neglect the effect attributed to the nature of Tunisian and Algerian soils, which appear to be different from the chosen models of Mason and Brewer. This difference is confirmed by the computations performed in this study, which show enrichment factors less than 1 (Fig. 5), proving thereby that even the aluminium, chosen as a crustal reference element, is partly enriched.

5.4 *Intra sites enrichment of studied aerosols*

Intra-sites enrichment rates of studied aerosols were computed as following (Yoboue, 1991).

$$T(\%)=\frac{[X]_{\text{site1}}-[X]_{\text{site2}}}{[X]_{\text{site1}}}\times 100\%\,, \tag{7}$$

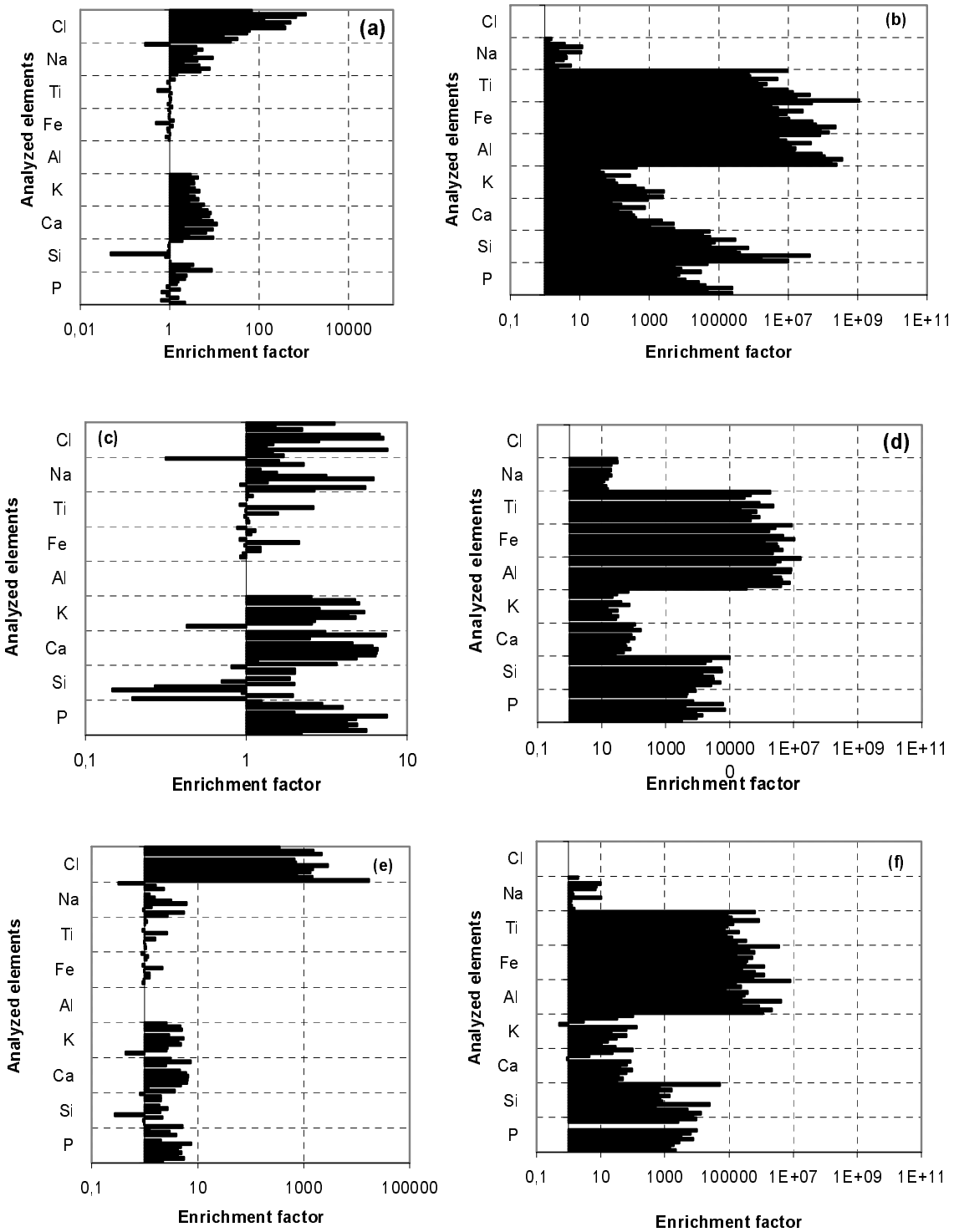

Fig. 5. Enrichment factors of the different constituents of the aerosols referred to aluminium (a, c and e) and chlorine (b, d and f) in Tataouine, Thela and Tabarka, respectively.

here, $T(\%)$: Enrichment rate of aerosols for site 1 with respect to that of site 2, fixed as a reference;

$[X]_{\text{site1}}$: Concentration of the compound X in the aerosol sampled at site 1;

$[X]_{\text{site2}}$: Concentration of the compound X in the aerosol sampled at site 2.

5.4.1 *Intra-sites enrichment in Sirocco periods*

Compared to the "Tataouine" site, the "Thela" site is enriched in terms of Ca, Ti, Mg, P, and Fe (Fig. 7). This is expected to be explained by the influence of soils and rocks of southern and central Tunisia and eastern Algeria swept by Sirocco winds. Nevertheless, the lack of enrichment in terms of Cl, Na, and Si at this site is unexpected. In fact, Cl and Na were proved to be highly emitted by the Sebkhas of southern Tunisia, while Si is abundant in the silica soils of southern Tunisia and Algeria. The lack of enrichment in terms of the aforementioned constituents at this mountainous site may be attributed to the importance of the redistribution of localized phenomena of the aerosol constituents, especially in the vicinity of the mountains of central Tunisia.

The work elaborated by Masmoudi et al. (2003) for the "Thela" site, in periods of Sirocco, showed a bimodal distribution of the different constituents of the aerosols, with an important enrichment of fine fractions. The average diameter of most constituents of the aerosols fluctuates between 0.1 and 3.6 μm. This size distribution is different from that observed at the "Tatouine" site, which is characterized by the dominance of coarse constituents with an average diameter varying between 0.1 and 10 μm (Masmoudi et al., 2003). This variability of the size distribution of aerosol constituents in the vicinity of the "Thela" site may be explained by the importance of the phenomena of gravity and/or impaction deposition:

(i) It was shown that gravitational settling causes the deposition of particles, especially the larger ones (diameter>1 μm) (Sehmel, 1980). Furthermore, Belghith (1999) showed that the gravity deposition is active for low wind velocities less than 5 m s^{-1}. Such a condition was fully fulfilled at the "Thela" site and even at the "Tataouine" site, located upstream;

(ii) The mountainous chains localized in central Tunisia enhanced impaction deposition, especially at the exposed sides.

In this study, the absence of a net decrease of the total concentration of the aerosol constituents at the "Thela" site on one hand, and the appearance of individualized maxima at this particular site on the other, may be explained by either the limitation of the aforementioned phenomena (gravity and/or impaction deposition) or their camouflage by other processes with a marked local influence. The latter are probably attributed to the phenomena of wind wakes. It can be concluded, therefore, that the aerosol enrichment in terms of the aforementioned elements (Ca, Ti, Mg, P, and Fe) may not reflect the representation of the different sources swept by the Sirocco wind. It rather results from the influence of local airflow characteristics associated with the geomorphologic features of central Tunisia.

Compared to that of the "Thela" site, the coastal site of "Tabarka" was shown to be enriched in terms of Fe, Na, Cl, and K due to the contribution of the soils of northern Tunisia enriched in Fe (Mtimet, 1999) and the marine source (for the case of Na and Cl). At this site, the particular enrichment in terms of potassium, shown as a crustal element ($10<\text{EF}_{\text{sea water}}<100$; $\text{EF}_{\text{crust}}<10$), is probably attributed to the effect of diapirs (Fig. 1). These are outcrops of evaporitic rocks

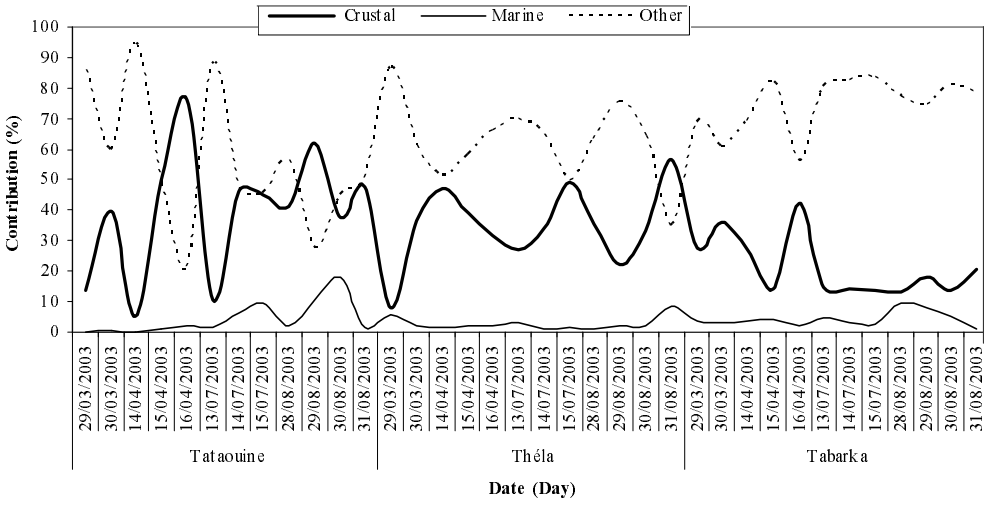

Fig. 6. Rate of contribution of enriching sources.

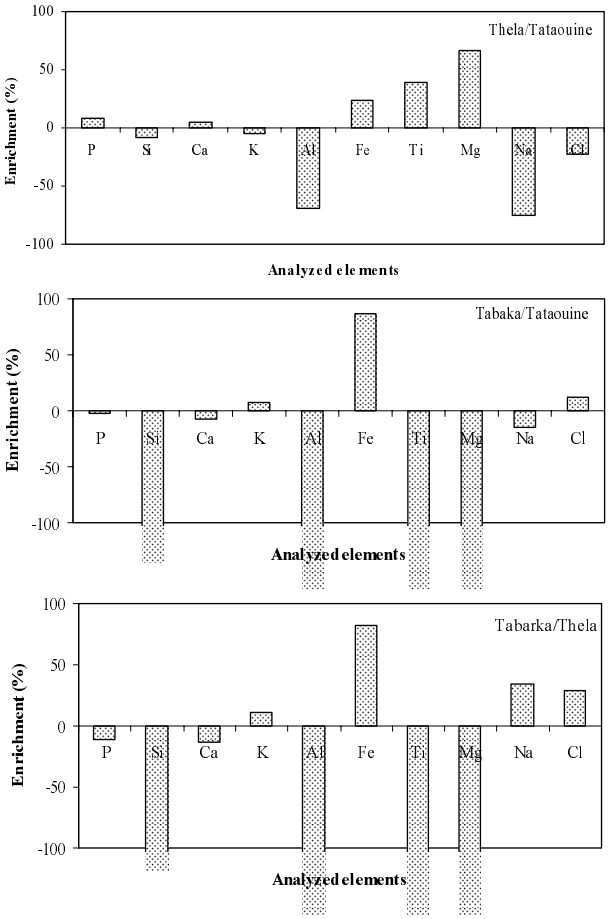

Fig. 7. Intra-site enrichment of the compounds of the aerosols sampled during Sirocco periods.

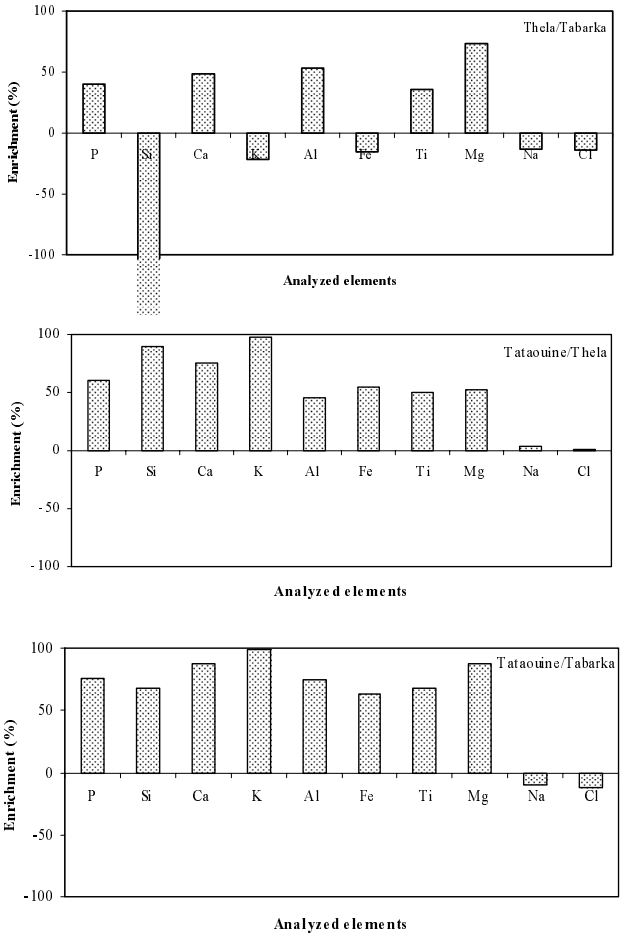

Fig. 8. Intra-site enrichment of the compounds of the aerosols sampled during periods without Sirocco.

(∼800 hm^2) enriched in terms of potassium sulphates.

5.4.2 *Intra-sites enrichment in periods without Sirocco*

Intra-sites enrichment in periods without Sirocco, under the predominance of northwestern, northeastern and eastern currents, was also shown to be quite important, but different from that of the Sirocco periods. Figure 8 shows that the "Tabarka" site corresponds to the lowest enrichment in terms of the analyzed elements. Compared to this coastal site, the "Thela" site, which is enriched in terms of Al, Ca, Mg, P, and Ti may neither reflect the effect of soils nor that of the diapirs of northern Tunisia, enriched in Fe and K. Alternatively, the obtained enrichment results was the effect of the mountainous chains of central Tunisia in flow redistribution, as was proved by the study of Sirocco winds.

Compared to that of "Thela", the "Tataouine" site is enriched in terms of all of the analyzed elements (Si, Al, Ca, Mg, P, K, Fe, Ti, Na, and Cl), implying the influence of multiple sources: soils and rocks of central Tunisia, Sebkhas, and the soils of southern Tunisia and the Mediterranean Sea. These multiple enriching sources are related to both the large field of wind action over vast areas characterized by its sparse vegetation covers and the mild and gentle topography upstream of the "Tataouine" site.

During the periods without Sirocco, the entrainment phenomena were certainly enhanced by the effect of the increase of the wind velocity beyond 7 m s^{-1}, a value judged to be of a remarkable action with respect to the phenomena of dust entrainment (Reydet, 1984; Belghith, 1999). In spite of the high enrichment rates registered during these periods, aerosol concentrations, which are usually subjected to the phenomena of wind wake, impaction and selective disintegration (especially in central Tunisia), remained lower than those of the aerosols affected by the Sirocco wind. This clearly proves the high rate of contribution of the Sahara in the enrichment of the studied Tunisian background aerosols.

6. Conclusions

The geochemical behaviour of the Tunisian background aerosols under Sirocco wind circulations was characterized. Spatial and time evolutions of the principal constituents of the aerosols (Si, Ca, K, P, Al, Fe, Ti, Mg, Na, and Cl) during four selected Sirocco periods were monitored at the "Tataouine", "Thela" and "Tabarka" sites, located in southern, central, and northern Tunisia, respectively. High constituent concentrations, exceeding 2.5 to 8 times those of aerosols sampled during the periods without Sirocco, were registered. These high concentrations are influenced by those of silicon, which represent rates exceeding 63%. Rates obtained are much more important than those registered during the periods of "no Sirocco", which rarely exceed 5%. This clearly shows the contribution of the Sahara in terms of loading aerosols with particulate matter, especially silica.

The study of the constituent enrichment factors showed the following:

(1) The elements Al, Si, Ca, K, P, Fe, and Ti are mainly of crustal origin;

(2) Chlorine is typically of a marine origin for both the "Tataouine" and "Tabarka" sites, while it is of a mixed origin for the "Thela" site;

(3) Sodium is characterized by a mixed origin for the "Tataouine" and "Tabarka" sites and mainly of a crustal origin for the "Thela" site.

Analysis of the elementary abundance of aerosol constituents proved that the examined aerosol enrichment cannot be explained exclusively by the aforementioned crustal and marine sources. In fact, it was also shown to be affected by the Sirocco wind flow characteristics (abundance of localized turbulent winds) and topography (especially the mountainous chain of central Tunisia).

Examining the intra-site enrichments of the studied aerosols confirmed the influence of such characteristics, especially those attributed to the phenomena of wind wake and the selective disintegration in the vicinity of the geomorphologic features of central Tunisia. The latter, characterized by relatively high altitudes and considerable spatial extensions, appear to accentuate the phenomena of the redistribution of aerosol constituents even under "no Sirocco" situations. They mask both processes of entrainment and deposition.

Future recommended research steps may include:

(1) The study of the dry deposition and aerosol size distribution in Sirocco wind circulations;

(2) Extended monitoring (under different Sirocco situations) of the aerosols is also required to ensure a better understanding of the geochemical behaviour of the Tunisian background aerosols.

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