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Chemical and Mineralogy Characteristics of Dust Collected Near the Phosphate Mining Basin of Gafsa (South-Western of Tunisia)

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Abstract

The study aimed at chemical and mineralogical characterization of whole particulate matter (PM) in the vicinity of a mining phosphate basin at urban area in Gafsa. Heavy metals concentrations in PM samples (Cd, Fe, Cu, Mn, Cr, Zn, Ni, Al, Pb), MgO and SiO₂ were determined by atomic absorption spectrometry. Calcium and phosphorus oxides (CaO, P_2O_5) were analyzed by Technicon Auto-analyzer. Potassium and sodium oxides (K₂O, Na₂O) were analyzed by flame photometer. Ionic species (S0₄²⁻, Cl⁻, NO₃⁻) were quantified by ion chromatography while organic carbon (OC) content was determined using carbon sulfur Analyzer. The mineral phase identified by X-Ray powder diffraction technique and SEM-EDX provides information about morphology and chemical composition of PM. As regards the chemical characterization it was found that samples were enriched predominantly in SiO₂, CaO and P_2O_5 which were detected only at mining area (S2 and S3) Weak concentrations of K₂O, Na₂O and MgO were also found. In addition the minerals phases identified in the samples were carbonate fluorapatite, Calcite, Heulandite, Gypsum and Dolomite. The values of ionic composition were in the flowing order SO₄²⁻>Cl->NO₃⁻. Results also showed that heavy metals pollution increased by the proximity to the mining area. SEM-EDX revealed that particles are almost spherical.

Keywords: Dust; Mining pollution; Metals; Mineralogy; Mining basin; Gafsa.

Introduction

Air pollution is a major problem nowadays. Exposure to ambient air pollution has been associated to several health outcomes, starting from modest transient changes in the respiratory tract and impaired pulmonary function, continuing to restricted activity, emergency room visits and hospital admissions and to mortality [1].

According the World Health Organisation (WHO), 4-8% of deaths occurring annually in the world are related to air pollution [2].

In recent decades, there has been a growing concern for the potential contribution of dust as one of major sources of atmospheric pollution. The interest in atmospheric particulate matter (PM) is mainly due to its effect on health and its role in climate change [3]. Dust can also affect biogeochemical cycling and air temperature because of the absorption and scattering of solar radiation and can influence sulphur dioxide levels in the atmosphere [4]. Fine particles in the air are most efficient in scattering light because they are similar in size to wavelength of visible light and have the largest surface area, so reducing visibility [5]. In daily life, dust presence in atmosphere dramatically reduces air quality leading to consequences such as respiratory and allergy diseases and aggravation in people suffering from heart diseases. Moreover, metals present in dust can accumulate in human body via directly inhalation, ingestion and dermal contact resulting in serious health problems [6]. In addition, the particulate material or its soluble components may be transported to organs some distance from the lungs and have a detrimental effect on these organs [7]. A large variety of dust is found in the atmosphere which originates from a variety of sources. Dust can have natural or anthropogenic origins. Maximal concentrations of PM₁₀ are found in mining area and the concentrations are gradually diminished with increase in distance due to transportation, deposition and dispersion of particles [8].

Currently, the knowledge of the chemical composition of particulate matter (PM) has gathered increasing importance in the scientific community as the necessity to differentiate PM components and their influence to both health effects and role in the climate change [9]. Also, research on the physic-chemistry properties of particulate matter is intense and toxicological studies attempt to identify which adverse biological responses (e.g. particle number, size, surface, chemical composition), and suggest that the chemical composition of PM (which reflects differences in source contributions) plays an important role in these responses [9]. In this framework, this study presents a comparative analysis of particulate matter at three sites located in Gafsa and identified minerals and trace metals, ionic components, and carbonaceous material.

The Study Area

Geology

Gafsa is the capital of Gafsa Governorate of Tunisia. It lies 369 Km by road southwest of Tunis. The geographical coordinates of the city are 34°25′N8°47′E. It is located in one perforated in the middle of an alignment mountainous, called "mounts of Gafsa" between DjbelBou-Ramli and DjbelOrbata which culminates to 1165 meters.

The governorate of Gafsa is composed of different towns such as: El Guettar, El Kaser, Gafsa, Mdhila, Métlaoui, Moularès, Rdyef and Sned. The city has 90.000 inhabitants (2005 estimate), the governorate has 340.000 inhabitants (2005 estimate) and an area of 8990 Km².

Mining basin of Gafsa is a part of the south-western dry area of

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Tunisia. It covers 325 Km², representing 42% of the entire governorate of Gafsa area and 38% of its total population [6].

Measurements were conducted at urban area of south Gafsa (S1) and (S2, S3) were chosen at Metlaoui as shown in (Figure 1) because Metlaoui is considered as a predominantly mining zone. Phosphate is the main wealth of the region. Metlaoui is located in the southwestern part of Tunisia (34°18'/34 °19'N; 8°22'/8°25'E), at 250 m above sea level. There are less than 40.000 inhabitants in the province of Metlaoui. The regional landscapes are mountains, basins and plains. It is a transition zone between steppes and desert [10].

Climate

Gafsa has a dry continental climate. It is characterized by important annual variation in temperature. Winters are cold and dry; with a mean annual rainfall of 137 mm and monthly mean temperatures ranging from 40°C in July to 7°C in January. Summers are very hot and dry because of dry sirocco blowing from the Sahara desert. Wind speed varies from 7 to 10 knots throughout the year and winds are predominately from the west or northwest [11].

Soil type

Soil in Gafsa is characterized by high salinity. The soil is stony and contains gypsums crusts and limestone. Mobility of sand is the limiting factor for the development of soil and rain-fed agriculture in general [12].

Vegetation

Vegetation includes steppe of this climate; Thymeleahirusta

(menten), Artemisia eampestris (tgouft), Artemisia bleached on grass Alba (chir), Diplotaxix (harra) and Peganumharmala (harmel). Oasis of south-western Gafsa, El-Kasba, El ksar, Lala and Guettar cover about 3200 hectares. The oasis counts 32500 palm trees belonging to seven different variets; "Degletnour", "Allique ", "Kenta","BesserHlou"," Quabrichou" and "Hammouri" [12].

Industry

Gafsa also specialized in the Craft industry of wool carpets, in particular the Klim, the margoum intended for exportation. Gafsa has developed thanks to the mining of the phosphate whose layer was discovered in 1886 as one of the most important in the world .It extracts from its mines nearly eight millions tones phosphate in 2005 which make Tunisia the fifth world producer [13].

Materials and Methods

Sample collection

Aerosols were collected using air blower (Black and Decker, serial number (49613) Characteristics:

Frequency=50/60 Hz

Power =335 W

Capacity =3.2 m²/min

These samples were collected during July 2011

Details of sampling are given in Table 1.



Chemical analysis

Malvern Mastersizer 2000 was used for determining particle size distribution. Dust is introduced into a tank with circulation containing a dispersing liquid. The suspension is then transported towards a laser cell measurement. Themastersizer has a fully optimized optical design which allows characterizing particle in the size range 0.02-2000 micron. The results are given in function of volume of the size of particles.

Metals concentration (Cd, Fe, Cu, Mn, C r, Zn, Ni, Al, Pb) and magnesium oxide MgO, SiO₂, were determined using the Atomic spectrometer spectra AA 220. CaO and P_2O_5 were analyzed by with Technicon Autoanlyzer. K₂O, Na₂O were characterized by using flame photometer 410 model.

Mineral phases identified by X-RAY powder diffraction technique. In this technique peaks occur when the path of the diffracted X-rays is equal to an integer multiple of the path difference expressed by Bragg's equation which is given by: $N\lambda = 2d \sin\theta$

Station code	Location	Period sampling
S1	Urban area	17/7/2011 - 22/7/2011
S2	Near Landry of phosphate	22/7/2011 - 26/7/2011
S3	Near Breakers of phosphate	26/7/2011 - 30/7/2011

Table 1: Details of sampling collection.

Where n is an integer, λ is the wavelength, d is the inter-atomic spacing and θ is the diffraction angle.

Ion chromatography (Shimadzu Degasser SCL.10ASP) was used to analyze the three anions $(SO_4^{-2}, CI^-, NO_3^-)$.

Scanning Electron Microscopy (SEM) coupled with energy dispersive X-ray spectrometry (EDX) (FEI Quanta 200) provides information on morphology and chemical composition of individual particles down to sub-micrometer and nano-meter size. Organic carbon (OC) content was determined using Horiba EMIA-220V series carbon sulfur analyzer.

Results and Discussion

Particle size distribution

The particle size of dust is a very important parameter. Particle size is often described by the diameter of the particle. It has already been proved that the degree of harmful effect of inhaled dust is closely related with dust particle size. The results of particle size distributions are summarized in Tables 2-4 and their particle size distribution curves (S1, S2, S3) are shown respectively in Figure 2. The particle size distribution patterns marked by overlapping of the curves.

% volume	0.21	1.38	1.03	1.43	5.41	6.62	10.12	21.67	15.95	6.64	12.1	9.36	4.87	3.18	0.02
					Table	2: Particle	size dist	ribution o	fS.						
			-											-	

Particle size (µm)	0 .010	1	6	10	20	32	40	50	71	90	100	125
% volume 1.21 3.9 3.06 16.67 23.1 12.55 12.04 15.48 7 2.12 2.71 0.16												
Table 3. Particle size distribution of S2												

% volume 0.89 5.28 3.87 10.75 15.6 10.57 11.85 18.14 9.71 3.34 5.03 2.86 1.04 0.6 0.07 0.26	Particle size (µm)	0 .010	1	6	10	20	32	40	50	71	90	100	125	160	200	315	340
	% volume	0.89	5.28	3.87	10.75	15.6	10.57	11.85	18.14	9.71	3.34	5.03	2.86	1.04	0.6	0.07	0.26







S1

S2

S3

Percentage of particles between 0.01 and 10 µm: The mesh that contains very fine particles between 0.01 and 10 microns is represented on Figure 3. The samples obtained (S1 S2 S3) contain (2.62 %, 8.17%, 10.04 %) of total sample volume respectively. Fraction of coarse mining dust superior 10 µm around mining area is considered to be nonbreathable, that is why is considered with less attention than breathable dust. Breathable fractions of airborne ambient particulate matter (PM_{10}) specifically those fractions that are less than 2.5 μ m (PM_{25}) in size. Fine particles have been identified as potential risk of general public. It has been one the largest occupational problem. They are small enough to penetrate into lungs, where they may exacerbate conditions such as bronchitis and asthma and have also been associated with visibility degradation and climate change. It causes lung damage such as pneumoconiosis and in particular silicosis, asbestosis, damage to the noise; throat and eyes and damage to the skin; it may cause various types of dermatitis, which are a widespread and often serious problem or even skin cancer [14].

Mineralogical and chemical composition

Mineralogical composition: Based on XRD analysis, samples of dusts collected at mentioned sites (S1, S2, S3) are composed of minerals listed in Table 5 with their formula.

The characterization in XRD, S_1 shows that this sample mainly consists of quartz (78%), this sample is very rich in breathable silica crystalline in the form of quartz.

For S2 and S3 silica is not detected by diffraction on the X-ray but this phase appears later on compared to the results obtained by MEB-EDX.

The other phases identified by this sample are calcite (15%), gypsum (1%) and dolomite (6%).

The S3 sample is characterized by a major phase of heulandites 65% and a minor gypsum phase 4%. The S2 sample is less rich in heulandites (37%) and presents calcite 10%.

As it is shown in Table 6, Carbonatefluorapatite is not uniform in

Name	Formula
Carbonate fluorapatite	Ca9.55 (PO ₄)4.96F1.96 (CO ₃)1.283
Calcite	CaCO ₃
Heulandite	((C_2H_5) NH ₃)7.85 ((Al 8.7 Si 27.3) O72)(H ₂ O)6.92
Quartz	SiO ₂
Gypsum	Ca(SO4) (H ₂ O)2
Dolomite	Ca Mg (CO ₃)2

 Table 5: Formula of investigated minerals.

Carbonate fluorapatite	Calcite	Heulandite	Quartz	Gypsum	Dolomite
-	15	-	78	1	6
53	10	37	-	-	-
31	-	65	-	4	-

Table 6: Major minerals determined by XRF (%).

different sites. Very high percentage of Carbonate fluorapatite at S2 and S3 and do not have in the Gafsa urban area could be explained from the distance of mining activities (landry and Breakers of phosphate) because Carbonate fluorapatite (francolite) is the principal mineral of phosphorites, the sedimentary rock phosphates which is an combination of phosphate radical PO₄ with water, calcium and the trace element fluoride [15].

Inhalation of fluoride containing phosphate dust can damage the tooth forming cells, leading to a defect in the enamel known as dental fluorosis.

Fluorosis is a serious health problem .White and yellow glistening patches on the teeth are seen which may turn brown. The brown streaks may turn black and affect the whole tooth and may get pitted perforated and chipped. Dental fluorosis not only poses cosmetic problems but has also social problems [16].

Contents of heavy metals in dusts

Particulate samples collected from 3 sites located in Metlaoui around mining phosphate area and urban area Gafsa were analyzed for (Cd, Fe, Cu, Mn, Cr, Zn, Ni, Al, Pb).

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Table 7 shows the results of the heavy metals. Graphs obtained using concentration of the heavy metals in 3 samples is given in Figures 4-12.

The Ni concentrations measured at 3 sample points were in the range 57-224 ppm .The highest concentration of nickel was found at site S1 (224 ppm) flowed by S2 (60 ppm) ,but the lowest value was found in S3.The data are represented in Figure 4.

Ni concentrations measured in the present study were higher than concentration at the SelebiPhikwe nickel-copper plan area in Botswana (41 ppm) [17].

The high concentrations of Ni were attributed to the pollution originating from traffic in Work place and anthropogenic activities (landry and Breakers of phosphate).

This element is mainly associated with emissions from stationary and industrial sources; it can also be emitted from vehicle exhaust as it is used as an additive in fuels. Health hazards associated with the exposure to Ni the occupational environment have resulted primarily from inhalation.

About 10% of women and 2% of men in the population are highly sensitive to Ni. A portion of these sensitive people can develop skin rash called nickel dermatitis if they are exposed to Ni through direct contact [18].

Cadmium concentration measured in all samples, presented the highest value at S2 site (67 ppm), but the lowest value was found at S1 site (4 ppm).Concentration values of cadmium in those three sites were significantly higher than this measured at SelebiPhikwe nickel-copper plan area in Botswana (0,03 ppm) [17] (Figure 5). A high concentration of Cd in S1and S2 may be due to phosphate activities because Cd can be found in phosphate rocks as can be seen in Table 8 [19].

People living near mine sites, in our study especially site S2 and S3

Sites	Cd	Fe	Ni	Cu	Zn	Mn	Cr	Pb	AI
S1	4	1500	224	-	80	177	71	15.8	1345.33
S2	67	2400	60	47	367	30	183	6.22	9953.23
S3	49	2940	57	170	300	35	277	13.7	3848.72







Phosphate rock origin	Cd concentration (ppm)
USA	3-186
Morocco	3-165
Peru	2-186
Russia	0.1-<13
North Africa	60
South Africa	2-<13
Brazil	4
Jordan Aman	6<13
Тодо	44-179
Tunisia ,Gafsa	38-173
Algeria	22.5-62,6
Syria	6.1-52
China(Yunam)	4
Mexico	8
Egypt (Quseir)	Aug-74

Table 8: Global pattern of cadmium in contents in phosphate rocks.

try to keep their windows and doors closed, because after inhalation, the absorption of Cd compounds may vary greatly depending upon the particles sizes and their solubility. Cadmium is a metal, which can cause severe toxicity in humans. Prolonged exposure to Cd can affect a variety of organs with the kidney being the principal target [20]. Also it induces chlorosis and necrosis in plants and exhibits mutagenic and carcinogenic effects in animals 33%-72% of the local Cd is supplied by air and airborne cadmium is transferred predominately by large scale atmospheric transport [21].

Concerning Cu, the high concentrations of copper found at S3 may be associated with electrical and chemical working; also Cu may be derived from the mechanical abrasion of vehicles and may be associated with diesel engine and wearing of break vehicles [22].

Lead is the most significant toxin of the heavy metals and affects are of toxicological and neurotoxic in nature, which include irreversible brain damage. From Table 7 and Figure 8 it can be seen that the Pb concentration in the range of 6 to 78 ppm. The highest lead concentration has been found in sample collected in urban environment S1, and the lowest at S2 sample. These may be due to the heavy traffic density that used lead such as deterioration of lead paint, home age, smoking and cooking activities.

It is noticed that important concentration of lead was found near Work place S3 can be associated to cars services site. The contents of Zinc in the dust samples are in the range of 50-367 ppm .the highest Zn concentrations were respectively in S2 and S1 as given in (Table 7 and Figure 9). This could be attributed to the use of zinc in accumulators of motor vehicles or in carburetors. Moreover, zinc may come from lubricating oils and tires of motor vehicles.

To better understand the pollution level of elements associated on dust in 3 sites, the concentrations of (Ni, Cd, Cu, Pb, Zn) measured at various places around the world are shown in Table 9 [20]. With regard to Fe, when comparing Fe concentrations with those measured at SelebiPhikwe nickel-copper plan area in Botswana (91 ppm) [17]





0:4 /		Element	s			
City /cou	ntry	Cd	Cu	Ni	Pb	Zn
Africa	Land /Angola	11.5	41.78	10	351.3	316.6
A	Amman/Jordan	3.1-11.2	66.5-350	43-88	210-1131	116-410
Asia	Bahrain	72	-	126	697.2	151.8
	Madrid/spain	-	188	44	1927	476
Europe	Palermo/Italy	1.1	98	14	544	207
	Paris/France	1.7	1075	25	1450	840
North	Hermosillo/ Mexico	4.25	26.34	4.7	36.15	387.98
America	Ottawa/ Canada	0.37	65.84	15.2	39.05	12.5

Table 9: Global comparison of heavy metals with present study data (ppm).

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, the levels recorded at sites in this study presented very high Fe concentrations (Figure 6).

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Al was the most abundant metal element varying from 9953, 2 to 1345, 3 ppm. The ranges obtained are comparable though lower those presented by Chandima Gunawardana et al. which range from 1.40 to 5.88 ppm [23]. Cr and Mn exhibit levels varying from 71 to 277 ppm and from 30 to 177 ppm respectively.

The level (Mn, Cr) in this work are compared with data reported from other cities in the world in Table 10. The Cr concentration in S1 (71 ppm) is higher than other cities except for Baoji, Xi'an and Guangzhou. For S2 (183 ppm) and S3 (277 ppm) the concentration of Cr is higher than all cities. The Mn concentration in 3 sample points in this work is lower than other cities considered.

Major oxides

The main oxides analyzed in this study are MgO, CaO, P_2O_5 , K_2O , Na₂O and SiO₂ (Table 11). The dominant major oxides in three samples are SiO₂, CaO and P_2O_5 . The abundance of SiO₂ in S₁ is mainly due to quartz as can be seen in Table 6.

Ionic composition

The analytical results of the major components (Cl⁻, NO₃⁻, SO₄²⁻) are presented in Table 12. The SO₄²⁻ was the most abundant ionic species, ranging in concentration from 5. 4 to 17.7 g/kg. The Cl⁻ is the second ionic species in samples ranging in concentration from 1. 4 to 2.6 g/Kg. Chloride may be originated from transporting of dusts from desert and industrial activities around the city by wind. The obtained results show that the Cl⁻ content in the samples is higher than the concentration of Cl⁻ (1.17 g/kg) in atmospheric aerosols in the coastal region of Mumbai [24,25]. The NO₃⁻ concentration ranged from 1.108 to 1.227 g/kg.



Figure 8: SEM images of dusts.

	S1	S2	S3	Baoji	Xi'an	Guangzhou	Hong Kong	Ottawa	Avilés	Calcutta	Madrid
Cr	71	183	277	126.7	167.3	78.8	124	43.3	41.6	54	61
Mn	177	30	35	804	687	481	594	432	1661	619	362

Table 10: Comparison of the heavy metal (Cr, Mn) concentration (ppm) of the three sample points and other selected cites.

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Oxides Sites	P ₂ O ₅	MgO	CaO	K₂O	Na ₂ O	SiO ₂
S1	0	8.49	15.48	4.9	5.52	65.61
S2	20.5	4.04	38.51	1.16	2.36	31.98
S3	17.48	6.4	32.06	4.48	3.86	35.72

Table 11: Major oxides (%).

lons sites	CI.	NO ₃ -	SO4 2-
S1	2.633	1.23	5.351
S2	1.578	1.11	17.73
S3	1.43	-	10.14

Table 12: Ions concentration in dust (g/Kg).

Sites	S1	S2	S3
OC (%)	2.26	4.69	2.84
Table 12: OC in comple points			







Organic Carbone content

Measurement results of OC content are given in Table 13. The % of OC ranged between 2.26% and 4.69%.

Scanning electron Microscopy (SEM) and Electron dispersive x-ray (EDX) data

Morphology of dusts: SEM can provide size and morphology information of particles on the submicron scale. The size and morphological characteristics of dust particles observed by ESM are illustrated in Figure. The SEM studies indicate that samples are characterized by multi-modal particle-size distributions; spherical, irregular, long and prismatic, crystalline are the most common shapes of dusts in the samples (Figure 13).

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EDX analysis

S1 sample: The three spectra obtained are shown as (P1), (P2) and (P3) in Figure 9. It must be noted that chemical composition of the particle is mainly Si and O with minor Mg, Al, S and K as expected the atomic percentages of these elements determined by analyzing the three EDX spectra. Figure shows that spherical grains (P4) have very high iron (Fe) content with small amounts of S, K and Al.

S2 sample: EDX spectrum P1 (Figure 9) indicated that the sample







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are predominantly composed of Ca (32.101%), O (35.29%) and P (12.12%).

The two spectra obtained are shown as (P2) and (P3) in Figure 9. It must be noted that both spectra exhibit characteristic peaks at the same element (C, O, Ca) although there are difference in the intensity.

The elements detected at spectrum P4 (Figure 9) are (C, O, Na, Mg, Al, Si, P, S, K, Ca, Fe, Sr, Ba), where O, C, Ca have the largest contribution as is presented in the atomic percentages of these elements.

Spectrum P5 Figure 9 clearly shows that this sample have very high carbon content (76.07%) with small amounts of (O, Mg, Al, Si, P, S, Ca).

The high fraction of C was possibly due to the mixture carbonaceous aerosols during the dust event. Figure 9 SEM micrographs of representative S_2 sample showing the morphologies of the main constituents and their corresponding EDS spectrum of element.

S3 sample: Measurement in P1 (Figure 9) indicated that the dominant elements were O (33.55%), Ca (33.42%) and P (12.69%).

As shown in Figure 9 the particle P2 is rich in O (36.83%) and Si (28.02%), other elements are present with a smaller percentage. From Figure 9 we can see that the main elements of P3 are Fe (42.01%), O (15.11%), Si (13.47%) and Ca (11.49%). As it can be seen in P4 (Figure 9), sample shows the highest content in Zn (37. 21%), other element (C, O, Al, Si, P, S, Cd, K, Ca, Fe, Zn, Se) are detected with small amounts.

Conclusions

This paper provides a detailed update of the chemical and mineralogical composition of PM at different site types in Gafsa. There is a clear evidence of metallic trace elements contamination in the city; this contamination is much higher around mines areas. It is highly recommended to keep a healthy distance between the mining and urban areas to reduce the exposure of population to contaminants. More guidelines and standards need to be produced to specify the maximum allowable level of every mineral substance without endangering the health of the population, to put these standards in place, the cooperation of Hygienists with people from the mining sector are paramount. The hygienist can provide people with more accurate information on exposure potential, dust sources, existent prevention methodology and others.

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