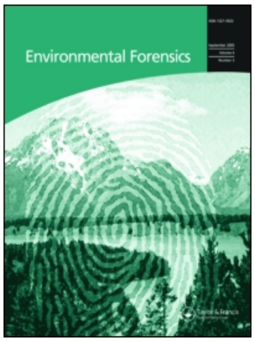

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Chemical and Mineralogical Characteristics of Dry Deposition in the Surrounding of a Cement Factory in Jordan

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Dry deposition samples were collected from 28 residential rooftops in Fuhais, Jordan, during the dry seasons of the year 2004. The samples were analyzed for trace and heavy metal concentrations to investigate the impact of total suspended particles emitted from the cement industry in the city of Fuhais. The cement factory has been operational for the past 50 years, and the management was evaluating the use of petcoke blending to reduce fuel costs. No baseline data on heavy metals due to the current operations existed prior to this study. The present study showed that lead (Pb) and copper (Cu) concentrations were statistically significant in the northwest sector of the city compared with the other quadrants. This significance was attributed to the heavy traffic of trucks carrying raw materials and cement in and out of the cement factory as the main route of transportation to the factory runs through this quadrant of the city. Cadmium (Cd), aluminum (Al), iron (Fe), zinc (Zn), manganese (Mn), nickel (Ni), molybdenum (Mo), and chromium (Cr) concentrations were high in all sampled areas. The overall average concentrations of all elements except Mo were found to be higher on the rooftops of the residential areas in Fuhais City in comparison with a remote reference site near Amman where no anthropogenic activity exists. The trace and heavy metal concentrations and the mineralogical composition of dry deposition samples collected from residential rooftops are representative of the current cement industry operations, which dominate the air quality of Fuhais City.

Keywords: heavy metals, cement industry, dry deposition, Jordan

Introduction

The Jordan Cement Factory (JCF) has been operational in the city of Fuhais about 5 km outside the great municipality of Amman, Jordan, since 1954. The JCF plant is located in the center of the city of approximately 12,000 inhabitants. There are schools, shops, a nursing home, and a hospital located within 1 km of the JCF site. Fuhais is located in a valley surrounded by different land uses consisting of residential, commercial, and agricultural activities.

The JCF consists of two cement kilns/coolers with clinker production capacities of 1 million tons per year each. The kilns are equipped with pre-heaters and electrostatic precipitators (ESP). One of the clinker coolers has an ESP, and one is equipped with a gravel filter bed. The facility has a raw mill, crushing operation, and cement mill that are potential air emission sources. Storage silos, conveyors, vehicle travel, and other unquantified fugitive dust sources are also considered significant contributors to particulate emissions. The local air quality is dominated by the JCF operations (Senes Consultants and Al Shamil Engineering, 2003b).

An environmental audit and air-quality impact assessment conducted in Fuhais concluded that total suspended particulate (TSP) concentrations downstream of the pollution-control devices exceed design specifications and that the monitoring programs for air quality around JCF need to consider more parameters (e.g., heavy metals) and need to be more extensive and frequent (Senes Consultants and Al Shamil Engineering, 2003a, 2003b). The current JCF operation has exceeded the Jordanian air quality standard for particulate matter less than 10 μm in size (PM_{10}), and the facility is one of the major contributors to overall atmospheric particulate loading in the vicinity of the plant (Senes Consultants and Al Shamil Engineering, 2003c).

Although the Jordanian air quality standards limit particulate matter in the air space, there is no current limitation on concentration or mass loading of heavy metals due to particulate emissions. The purpose of this study was to quantify the metal concentrations and the distribution of those concentrations in the dry deposition in areas surrounding the cement plant in the city of Fuhais, Jordan, and to compare those results with those found in a similar dry deposition samples collected from an unaffected reference area. Momani et al. (2000) and Jiries et al. (2002) have documented heavy metal content in rooftop dust in Amman, Jordan.

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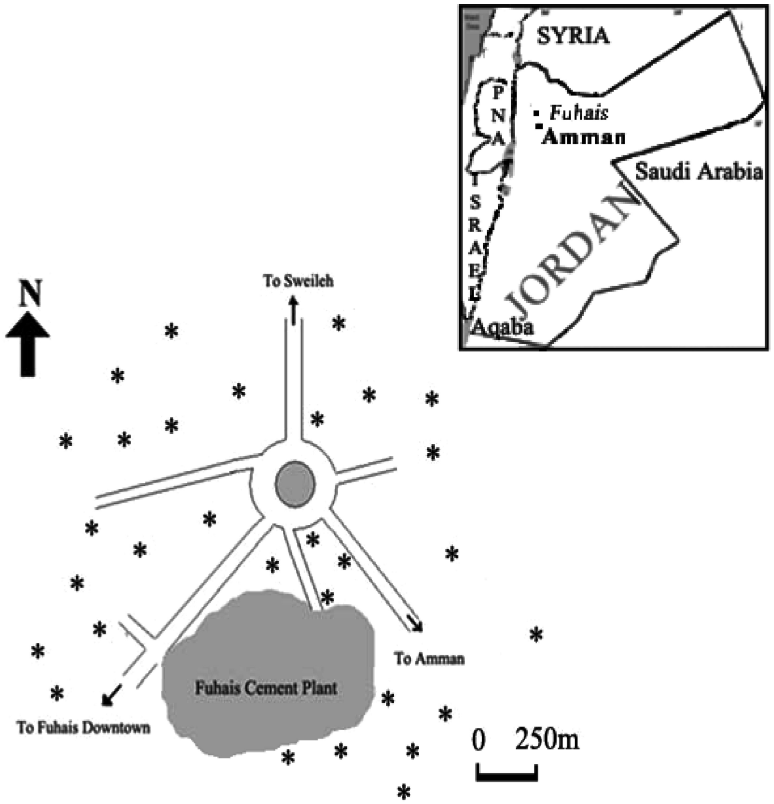

Figure 1. Map of Jordan showing cities of Fuhais and Amman and map of sampling sites (*) surrounding the cement factory in Fuhais.

Heavy metals are contained in trace concentrations in the raw materials used to manufacture cement (Serclerat et al., 2000). The concentrations of heavy metals in the resultant ash from cement manufacturing increase depending on the fuel source (Kikuchi, 2001). Abdel Razaq et al. (1999) investigated the effect of cement-industry dust emissions on the nitrogen and phosphorus content of vegetation in Al-Rashadia, Jordan. Fatima et al. (2001) investigated the chromosomal aberrations in men in India and correlated the results to exposure to cement dust. Legator et al. (1998) found a high statistical significance in the relationship between symptoms of respiratory effects in humans and proximity to cement kilns. Al and silica are the prime components of cement dust and several investigators have reported the adverse effects of Al in living systems (Ajoy et al., 1990; Ballal et al., 2004; Garruto et al., 1984). Nickiforov et al. (1979) measure the accumulation of heavy metals in the body tissues of rabbits in the vicinity of cement plant discharges, and the negative impact of toxic metals in cement dust on the abundance and size characteristics of plants has been investigated (Iqbal and Shafig, 2001).

Materials and Methods

Dry deposition samples were collected from the rooftops of houses in Fuhais in a distributed systematic pattern from the four directional quadrants around the cement factory during the summer of 2004 (as shown in Figure 1) to investigate the impact of the cement plant TSP discharge on the surrounding city. The sampling strategy took into consideration the dominant wind direction (northwest) in addition to the heavy transportation movement in the investigated area. Therefore, sites that are located south and east of the point-source pollution (the JCF) are the primary targets of airborne pollutant emissions. For comparison, samples were collected from a remote reference site, where absence of cement industry influences and limited anthropogenic activities are the dominant conditions. Roof dust deposits were sampled from each station (represented by asterisks in Figure 1) by using a clean plastic bucket to capture the dust deposition. A clean plastic dustpan, brush, and scoop were used to remove the samples from the buckets. Samples were then dried in an oven at 110°C, sifted through a 2-mm sieve to remove extraneous materials, and retained for further analysis. Two grams of the sample were accurately weighed and digested for total heavy metal analysis using the wet digestion procedure. The sample was transferred to a polyethylene test tube and digested with 15 mL of a 1:1 mixture of nitric acid (HNO_3):perchloric acid ($HClO_4$). The mixture was heated at 80°C for 2–3 hours using a heating block. The contents were filtrated and transferred through a simple filtration apparatus into a 25-mL polyethylene volumetric flask. The filter paper was washed several times with 1% HNO_3, and the volume was completed to 25 mL with 1% HNO_3. The solutions were stored in sealed plastic bottles prior to analysis. The heavy metals (Pb, Cu, Zn, Ni, Fe, Al, Cr, Co, Cd, Mo, and Mn) were determined in dust samples using a Perkin Elmer Flame Atomic Absorption Spectrophotometer model Analyst 300 (Perkin Elmer, Wellesley, MA, USA). Standard stock solutions of each metal (1,000 µg/mL) in 1% HNO_3 were used to prepare working standards. Calibration curves were built up for each metal where $r > 0.995$. Reagents blanks (two per eight samples) were prepared by carrying out the whole extraction procedure. All materials used for analysis (e.g., bottles, glassware) were washed thoroughly, rinsed with 1% HNO_3, and finally rinsed with distilled water.

The mineral constituents of the dry deposition samples were determined using x-ray diffraction system (Philips-X` pert MpD, PANalytical B.V., Almelo, The Netherlands). The fine powder samples were randomly mounted on special slides and then scanned between 2° and 65° 2 θ, using Ni-filtered Co Kα radiation, 40 kv/40mA, divergent and scattering slits of 0.02° mm, a receiving slit of 0.15 mm, with stepping of 0.01° and scanning speed of 3°/min.

Results and Discussion

Heavy metals are naturally occurring constituents in the environment in low concentrations. Anthropogenic activities can cause elevated levels of metal concentration in various parts of the ecosystem. Heavy metals in river sediments, for instance, are considered more sensitive than dissolved contamination as indicators of contamination (Gaiero et al., 1997). The anthropogenic sources of heavy metals in an urban environment could result from multiple sources. Cu can result from electronic and metallurgical industries, whereas Ni, Pb, Cd, and Zn can result from fossil fuel combustion and manufacturing industries such as that for batteries, paints, printing and graphics, medicine, and metallurgy (Alloway and Ayres, 1997).

A statistical summary of the chemical analysis for dry deposition samples are presented in Table 1. The concentration of heavy metals showed a wide range in concentrations within the same quadrant due to the fact that, within the same quadrant, the sampling sites were distributed over a wide area—some close to the source of pollution showing high concentrations and others in the same quadrant but further away from the source showing low concentrations. The average concentration levels of selected heavy metals (Mn, Cu, Ni, Co, Pb, Cd, Zn, Cr, Al, and Fe) within the four directional quadrants are shown in Figure 2. Initial qualitative observation of the average concentration results for each quadrant indicates that Ni and Fe were higher in the southwest quadrant; Pb, Cu, Co, and Cd were higher in the northwest quadrant; Zn and Mn were higher in the southeast quadrant; and Al and Cr were higher in the northeast quadrant. Mo was low and approximately equal in concentration throughout the study area.

Because the mean concentration of heavy metals appeared unequal in most of the investigated sites, one-way analysis of variance (ANOVA) was carried out to test the significant differences between mean concentrations of heavy metals with respect to the sampling quadrants. The results of the test showed that significant statistical differences (at a level of $\alpha = 0.10$) were found for Pb ($P = 0.082$) and Cu ($P = 0.081$) in the northwest quadrant. All other differences in mean concentrations of heavy metals were found to be not statistically significant by quadrant within the range of sampling distances used from the cement plant. This lack of statistical significance is considered to be due to the influence of the cement industry operations on the local air quality throughout Fuhais City. Cu and Pb were significantly correlated in the northwest, where they are considered to be directly related to the heavy traffic pattern for trucking and automobiles transporting raw and finished materials in and out of the cement factory.

Because ANOVA showed no statistical significance in the variation of the means of the heavy metal concentrations by quadrant concentrations at the $\alpha = 0.01$ and 0.05 level, the overall mean concentration for each metal from this study in Fuhais was compared with the average concentration found in dry dust deposition, which was measured at a remote reference site. The concentrations of all of the heavy metals were found to be higher

Table 1 *Statistical summary of measured heavy metal concentrations in mg/kg^{-1} from the four quadrants of the investigated area*

	Northwest			Northeast			Southwest			Southeast		
	Min.	Max.	Mean ± SD	Min.	Max.	Mean ± SD	Min.	Max.	Mean ± SD	Min.	Max.	Mean ± SD
Pb	17	117	57 ± 35	28	73	39 ± 16	15	32	25 ± 6	30	40	35 ± 5
Cu	12	38	24 ± 9	13	29	17 ± 6	13	21	17 ± 3	12	18	16 ± 2
Zn	56	222	141 ± 52	80	245	158 ± 75	48	209	108 ± 76	72	239	167 ± 64
Ni	16	24	21 ± 3	15	33	22 ± 6	18	25	23 ± 3	16	26	21 ± 4
Fe	4295	11396	6081 ± 2161	4030	7766	6354 ± 1339	4328	11896	7960 ± 3237	4405	7161	5606 ± 1098
Al	3386	8875	6123 ± 1687	4204	12085	7030 ± 2621	3944	8109	5984 ± 1508	3456	6568	5367 ± 1149
Cr	11	25	15 ± 5	10	55	20 ± 16	12	26	19 ± 6	11	17	13 ± 2
Co	7	39	15 ± 9	8	14	12 ± 2	10	14	12 ± 2	8	14	12 ± 2
Cd	1	28	5 ± 8	2	11	3 ± 3	2	2	2 ± 0	2	4	2 ± 1
Mo	ND*	4	2 ± 2	ND	4	1 ± 2	ND	3	1 ± 1	ND	3	1 ± 1
Mn	90	179	141 ± 31	107	175	144 ± 26	99	170	146 ± 27	130	189	161 ± 34

* Not detected.

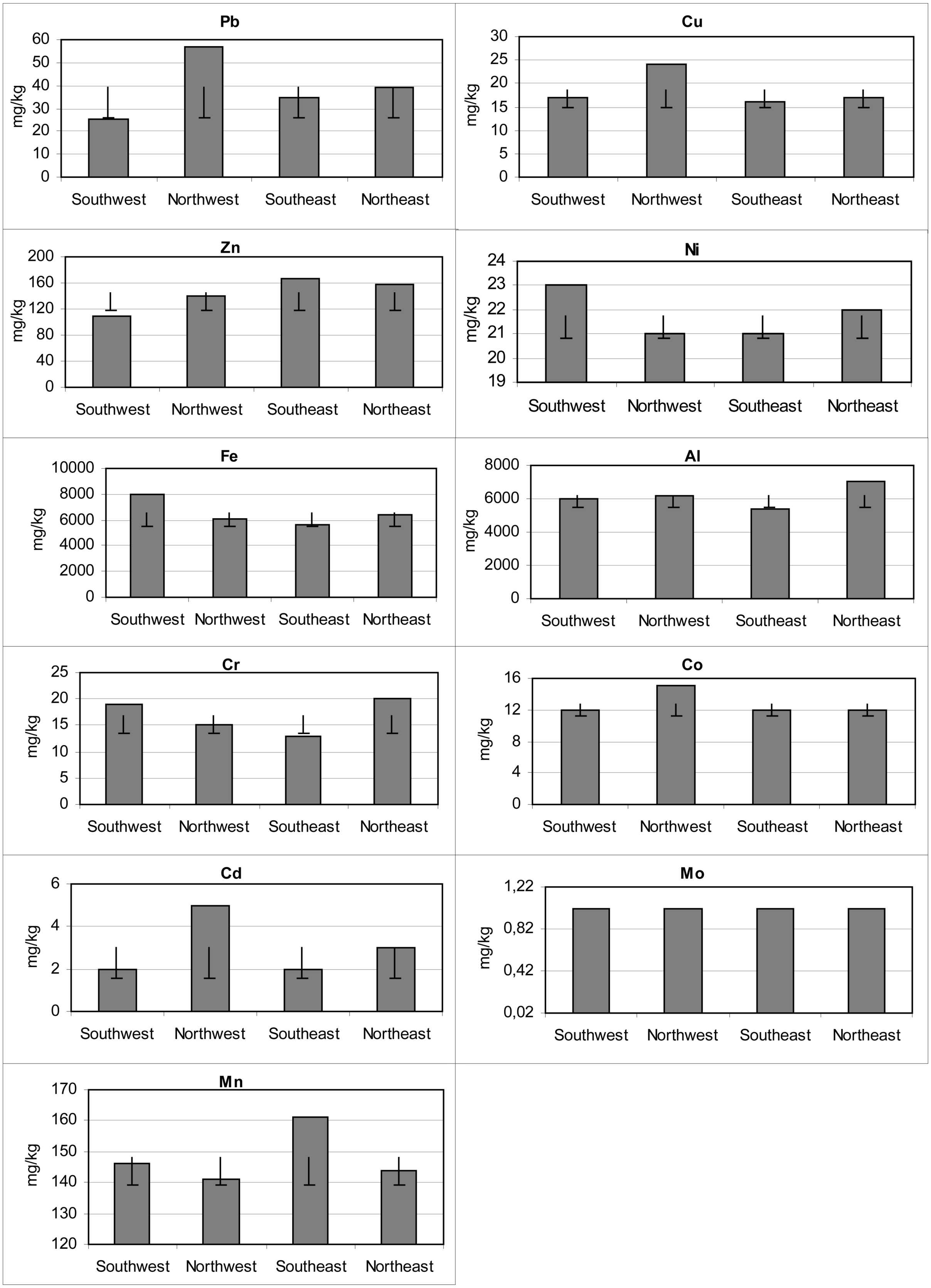

Figure 2. Mean concentration of heavy metals (mg/kg) and Standard Deviation (STD) for all sampling quadrants.

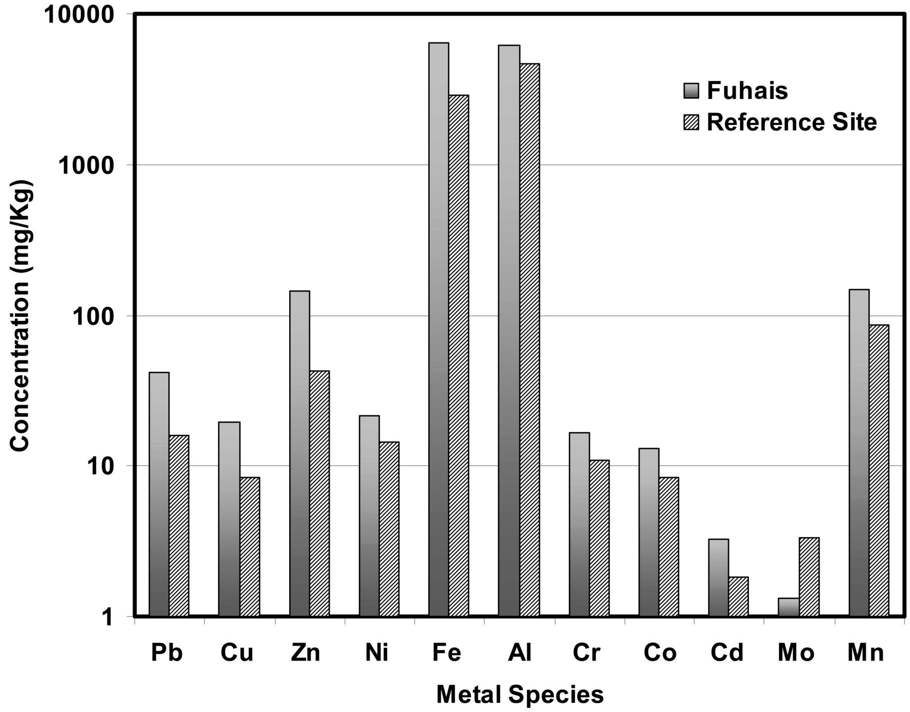

Figure 3. Comparison of the composition and concentration of dust from rooftops in Fuhais and the reference site.

on the rooftops in Fuhais than in the reference site as shown in Figure 3. The variations in the mean values of the concentrations of the metals (Pb, Cu, Zn, Ni, Fe, and Mn) in the dust between Fuhais and the remote reference site locations were found to be statistically significant at a level of $\alpha = 0.05$ ($P = 0.017$, Pb; 0.015, Cu; 0.011, Zn; 0.006, Ni; 0.010, Fe; 0.003, Mn), and differences for Al, Cr, and Co were significant at a level of $\alpha = 0.10$ (0.06, 0.10 and 0.09, respectively).

The dry deposition samples were analyzed for crystalline minerals content using x-ray diffraction. The analyses of these minerals were estimated from the height of the peaks. Dolomite, calcite, quartz, and gypsum were the predominant minerals as

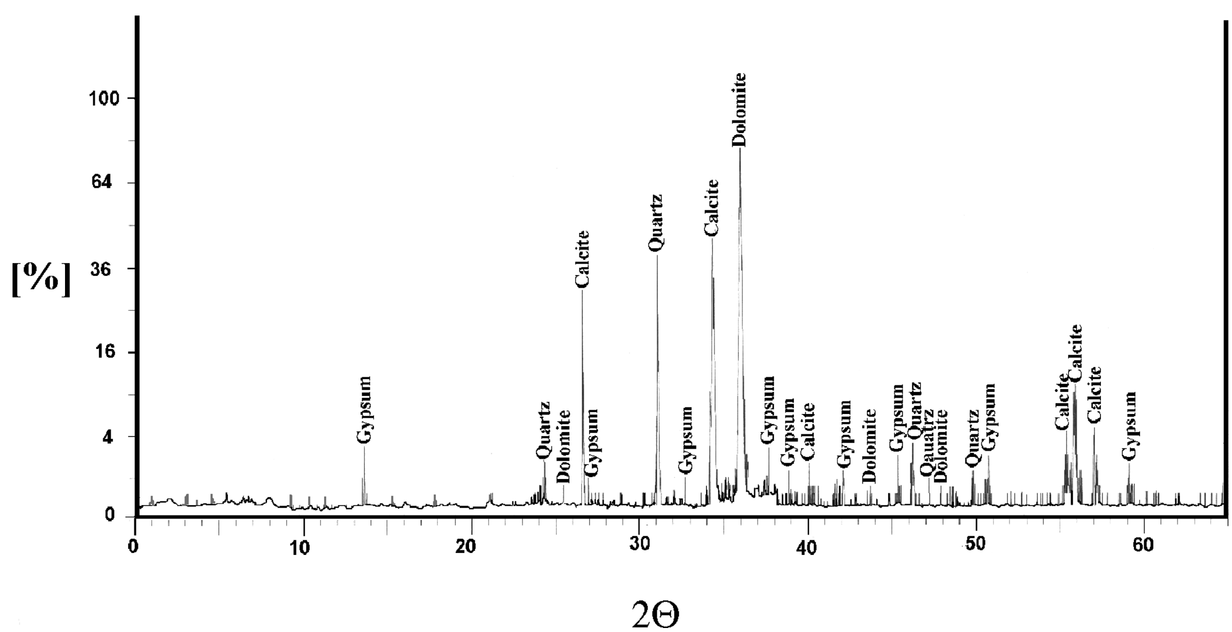

Figure 4. X-ray diffractogram of Fuhais dry deposition sample.

shown in the difractogram (Figure 4). All of the previously mentioned minerals can be components of the cement factory emission rather than natural sources as these minerals are used for cement production.

Conclusions

The results of this study indicate that the concentration of the heavy metals in the dust collected from residential rooftops in the city of Fuhais is higher (for all metals measured except Mo) than the heavy metal concentrations found in a remote reference site. The overall high concentrations of the heavy metals on the rooftop dust in Fuhais are representative of the current cement plant operations that dominate the air quality in the city. A significant difference in Pb and Cu concentrations were found in the northwest quadrant of the city, are representative of the traffic pattern from and to the cement industry. The mineral constituents of some dry deposition samples showed the presence of gypsum, dolomite, calcite, and quartz, which indicates the contribution of cement factory on the dry deposition of the area. All these results of the present study suggested that dry deposition samples from Fuhais city are strongly influenced by emission from the cement factory rather than natural sources.

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