The Influence of Local Sources on the Mineral Content of Bulk Deposition Over an Altitudinal Gradient in the Filabres Range (SE Spain)

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Mineralogical characteristics of bulk deposition have been determined at seven sites along an altitudinal gradient in the southern face of Filabres Range from 1770 m altitude to sea level, over a period of 8 months, the rainiest in this semiarid area. X ray diffraction and scanning electron microscopy were used for identifying and semiquantifying minerals. The aim was twofold: to ascertain the contribution of mineral particles to both the high *pH* and alkalinity of bulk deposition, the highest in Europe, and to use mineralogy of bulk deposition as a possible fingerprinting technique to identify the source of insoluble particles. The *pH*, alkalinity, and electrical conductivity of rainwater increase down the altitudinal gradient as precipitation decreases, indicating a clear increase in alkaline particles, in agreement with the presence of calcite, aragonite, dolomite, and gypsum, which also explains high *pH* and alkalinity of the bulk deposition. Moreover, *pH* and conductivity of rainwater are somewhat more related to E-SE winds than to W-NW ones. In addition to carbonates and gypsum, seven silicate minerals were found. Some minerals, like feldspars, chlorite, and aragonite, are neither related to site nor to rain events. Some others, like quartz, high-crystalline illite, paragonite, and kaolinite, are only related to rain events. Finally, a few minerals, like low-crystalline illite, palygorskite, paragonite, and to some extent calcite and gypsum, are site related. Some minerals have been interpreted to come from local sources, e.g., high-crystalline illite, gypsum, and paragonite; others, like palygorskite and low-crystalline illite, smaller in size, may come from distant sources.

Introduction

There are no data for the mineralogical characteristics of bulk deposition in SE Spain, the driest area in Europe (containing the 200mm isohyet), where it is usually assumed that frequent muddy rains are influenced by aerosols from nearby northern Africa [*Loye-Pilot et al.*, 1986]. As southern Spain is frequently considered to be the transition zone between Africa and Europe, it is of great importance to know how atmospheric inputs influence geoecological variables in this part of the world in order to predict possible contamination or to simply ascertain the influence of African dust in southern Europe. Only *Barrios et al.* [1987] have shown the local importance of aeolian dust supplies to Spanish soils and the difficulty of identifying their sources because of the presence of ubiquitous wind-derived minerals like quartz, mica-illite, kaolinite, and smectite.

Previous work on bulk deposition in the Filabres Range, SE Spain, has shown that *pH* is from neutral to alkaline and HCO_3^- and Ca^{2+} constitute more than the 50% of total ions [*Domingo*, 1991] while in more industrial areas like those of Central Europe [*Matzner et al.*, 1982] and the United States [*Likens et al.*, 1977], acid *pH* and acidic ions dominate. An intermediate situation can be found in northern Spanish Mediterranean areas (*Bellot et al.* [1990] and *Lledó* [1990] in Prades Mountains, NE Spain; *Avila* [1987], *Belilla* [1989], and *Roda* [1983] for Montseny

Mountains, near Barcelona). *Loye-Pilot et al.* [1986] have documented the influence of Saharan dust on the acidity of rain and the atmospheric input to the Mediterranean as a counteracting effect to acidic rain.

Consequently, to document those differences it is of great interest (1) to see if the alkalinity of bulk deposition in the study area (SE Spain) is due to aerosols containing mineral particles; and (2) to ascertain the origin of these aerosols through the mineralogical characteristics of these particles. Mineralogy of dry deposition has been widely employed in some other locations [*Glaccum and Prospero*, 1980; *Coudé-Gaussen and Blanc*, 1985; *Uematsu and Duce*, 1985; *Bergametti et al.*, 1989] as a fingerprinting technique to determine the source of aerosols, which have been found to be of either local or remote origin.

Paquet et al. [1984] studied the mineralogical composition of about 30 aeolian dusts which were sampled in 1974 at 2 m above ground along a 5000-km N-S transect across the Sahara. They noted the abundance of the clay fraction (>25%) and identified a wide range of minerals: illite, chlorite, illite-smectite, and chlorite-smectite in mixed layers, and smectite, kaolinite, paligorskite, quartz, feldspars, calcite, gypsum, and traces of dense minerals. The minerals were distributed in such a way as to form main groups that indicate the importance of local sources and the erosion of surface soils and soft sediments.

In the framework of a national project on desertification to study both hydrological and geochemical balances in a semiarid catchment located between 1560-m and 1770-m altitude in the Filabres Range in SE Spain [*Domingo*, 1991], a sequence of bulk

deposition collectors were installed. The initial objective was to measure the variability of the volume of rainfall at different altitudes within the watershed, as well as to investigate the role of inputs of bulk open field precipitation with high ionic content in the total budget in the catchment. Also, to determine the origin of the chemical characteristics of bulk deposition in the studied catchment, additional bulk deposition collectors were installed in an altitudinal gradient between the catchment at 1770 m and at sea level, following an approximately 65-km-long N-S transect.

To achieve all the objectives, bulk deposition (wet and dry precipitation) was collected over a period of 8 months, from November 1989 to June 1990, from a series of bulk deposition collectors within the defined altitudinal gradient and analyzed for its pH, electrical conductivity, alkalinity, and mineralogy.

GEOGRAPHIC SETTING AND RAINFALL EVENTS

Bulk open field deposition was sampled by seven rain gauges (continuously open polyethylene funnel collectors with 490-cm² aperture) located along an altitudinal gradient between the top of Filabres Range at about 1770 m above sea level (asl) and the city of Almería at 20 m asl. Rain gauges were all placed 1.50 m above the ground and in clearings. The precise locations of the rain gauges, labeled A, B, C, D, E, F and G are shown in Figure 1. Sites A, B and C are located between 1770 m and 1300 m asl, within the above mentioned instrumented catchment, in a terraced slope covered by planted pine forest, litter, grasses, and perennial plants. The substrate, outcropping at the highest part of the catchment, consists of mica schists, in which quartz, muscovite, paragonite, feldspars, garnet, and graphite have been found. Site D is near the village of Senés, at 1010 m, in an area with the same lithological characteristics as the top of Filabres Range, with a cover of bushes and grasses. Site E is in the lower part of the southern alluvial fans of the Filabres Range, at 450 m with a very sparse cover of bushes and annual plants. Site F is in a badlands area developed on marine Miocene gypsiferous marls (with quartz, muscovite, paragonite, chlorite, kaolinite, calcite, dolomite and gypsum) within the Sorbas-Tabernas basin, near the junction of roads N-340 and N-324, at 300-m altitude, with a mixed ground cover: that is completely bare on south facing slopes and partially covered with plants on north facing slopes. Finally, site G is in Almería, at the top of a building, at 20 m asl.

All rainy events, seven in total, were sampled and listed from 1 to 7. The 8-month sampling period corresponded to the rainy season of the area. The dominant winds as well as the back trajectories of the 700- and 850-mbar air masses arriving at the area before and during all rainy events were identified by means of synoptic weather maps from the Servicio Meteorológico Nacional de España. Air masses came only from two directions, W-NW and E-SE. During the sampling period some rainy events in sites D, E, F, and G were muddy rains, while some rainy events in sites A, B, and C were snow (Table 1).

METHODS

An aliquot of the total bulk open field precipitation taken from every rain gauge after every rainy event was immediately filtered through a filter (0.45-μm-mesh diameter "Millipore HAWP"). Polyethylene collectors were replaced with clean ones after each event. In the filtrate, pH, electrical conductivity (EC), and alkalinity were measured by potentiometry, conductivity bridge,
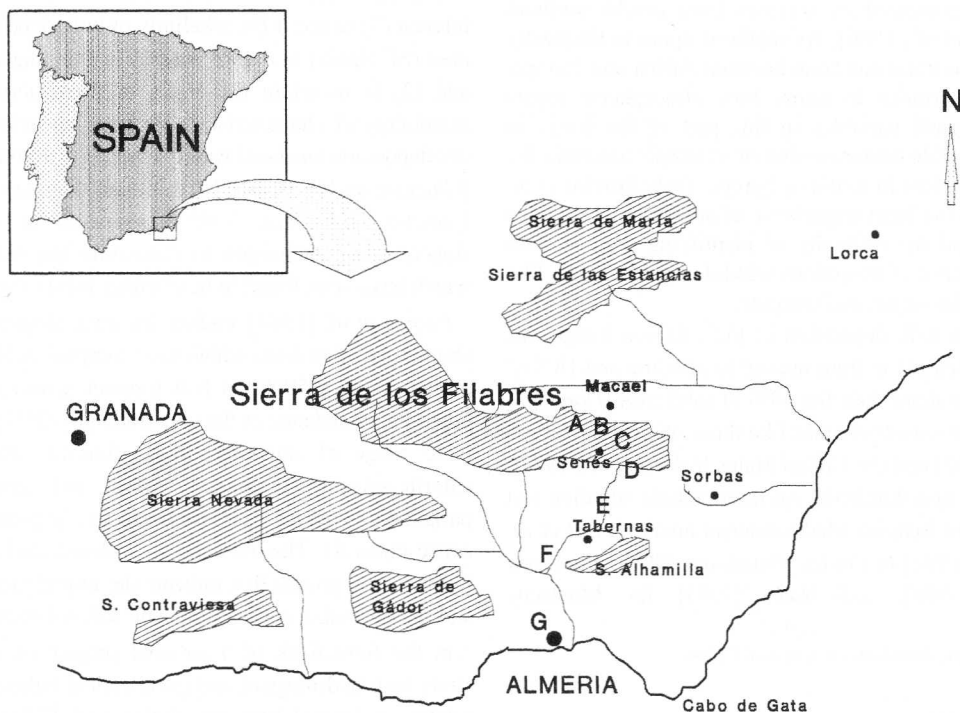

Fig. 1. Location of rain gauges (sampling sites) within the study area.

TABLE 1. Meteorological Data Associated With Rainy Events

Site	P type	Rainfall, mm	EC, S/m	pH	Alk, mg/kg
		Event 1			
A	1	122.45	0.0015	5.60	3.00
B	1	102.86	0.0013	5.99	4.70
F	1	60.48	n.a.	n.a.	n.a.
G	1	95.31	0.0032	6.28	8.54
		Event 2			
E	1	33.01	0.0175	7.87	93.45
		Event 3			
A	3	35.31	0.0043	7.06	17.02
B	3	38.57	0.0039	6.72	11.29
C	3	36.27	0.0046	6.24	7.69
E	3	9.40	n.a.	n.a.	n.a.
		Event 4			
A	2	61.83	0.0024	6.40	6.00
G	1	8.06	0.0284	7.91	85.89
		Event 5			
A	3	81.17	0.0023	6.30	6.10
B	3	72.19	0.0023	6.40	5.02
C	3	73.25	0.0031	6.32	6.59
D	3	34.94	0.0080	6.62	23.18
G	3	14.49	0.0143	7.63	37.58
		Event 6			
B	2	55.51	0.0015	6.48	4.71
C	2	29.52	0.0024	5.93	6.71
D	1	21.81	0.0066	6.60	23.85
G	1	3.47	0.0297	7.71	41.79
		Event 7			
A	1	122.45	n.a.	n.a.	n.a.
B	1	122.45	n.a.	n.a.	n.a.
C	1	144.58	n.a.	n.a.	n.a.
D	3	47.72	n.a.	n.a.	n.a.
G	3	30.10	0.0082	7.55	28.18

Event 1, November 29, 1989; event 2, December 5, 1989; event 3, December 14, 1989; event 4, March 21, 1990; event 5, April 20, 1990; event 7, May 7, 1990. Mean dominant wind directions are N-NW for events 1 and 6 and E-SE for events 2, 3, 4, 5, and 7. Precipitation tipe 1, rain; type 2, snow; type 3, muddy rain. n.a., not available.

and titration with sulfuric acid to an end point of *pH* 4.5, respectively.

Solids retained on the filter were analyzed by X ray diffraction (XRD). Mineralogical analysis by XRD on filtration mountings is a well-known technique that has been largely used over the past decade and has even been recommended as a procedure for routine semiquantitative analysis of mineral phases [*Davis and Johnson*, 1982; *Davis*, 1984a].

The main drawback of filtration mountings comes when only small amounts of mineral samples are available, specially when they come from the filtration of very clean water, being necessary to filter large amounts to get enough residue, or when working with atmospheric samples. In both cases the analytical response is always reduced. Another drawback when working with filters is the considerable increase in background signal which hinders adequate identification of mineral species as well as their
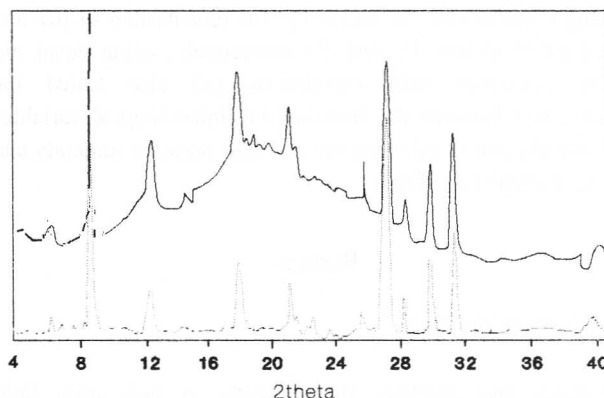

Fig. 2. X ray diffraction spectra. Top curve is uncorrected, and the bottom curve is corrected for background.

quantification [*Sturges et al.* 1989, *Davis*, 1984b]. Mineralogical quantitative analysis by XRD of samples on filtration mountings has been discussed by *O'Connor et al.* [1980], *Davis et al.* [1982a, b] and *Sturges et al.* [1989], who all consider the results to be only semiquantitative. In order to overcome all the drawbacks and difficulties, it was necessary to enhance the analytical response to the samples by two procedures:

1. The spectral response of mineral phases was enhanced by increasing the counting time at every step; instead of the usual 2 s, a 6-s period was used.

2. The computer-based spectral record was interpreted after correction of the background to enhance the discrimination of peak maxima. Examples of an uncorrected spectrum and a corrected spectrum are shown in Figure 2.

For the semiquantitative analysis, the method of *Chung* [1974], which integrates peaks, was used. A Siemens D500 X ray diffractometer, with a graphite monochromator, set at 40 kV and 30 mA, was used. Spectra were registered and stored in a computer data base for further analysis. Samples were analyzed within the range 4°-70°, taking data every 0.05° during the 6-s period.

To evaluate the particle size of dust grains on filter mountings, as well as to get a morphological appraisal of minerals identified by XRD, selected samples from every sampling site were observed with a scanning electron microscope (SEM) (JEOL JSM 6400), with a coupled microanalysis system of energy-dispersive X ray (EDX) type (LINK LZ-5). Drawbacks similar to those for XRD results affect the SEM observations. Images are not very clear because of high electrostatic charges associated with the filters supporting the mineral particles, even when they are covered with graphite. All chemical and mineralogical data were statistically analyzed.

Dependence or independence of samples with respect to both sites and events was investigated by the Kruskal-Wallis test. Sites and precipitation events were arranged in two ways: (1) seven classes of sites and seven classes of events were considered, taking into account all sites and events; and (2) sites and events were grouped into two classes of sites and events (one for the upper part of the gradient, comprising sites A, B, and C, and one for the lower part of the gradient, comprising sites D, E, F, and G; and two classes of events, one related to air masses coming from the west and NW, and one related to air masses

coming from the east, SE and SW). The probabilities of two and seven group means, P_2 and P_7 respectively, being equal are given. Spearman rank correlations (R) also tested the relationships between the chemical and mineralogical variables and altitude and wind direction and also between minerals and chemical properties of rain.

RESULTS

Chemistry of the Rain

Volumes and chemical characteristics of bulk open field precipitation are summarized in Table 1. Being most frequently near neutrality, pH ranges from 5.6 to 7.9. There is an inverse relationship between pH and altitude (Spearman correlation R=-0.52, n=19), with electrical conductivity (R=-0.76, n=19) and alkalinity (R=-0.76, n=19). These relationships agree with precipitation volumes (Spearmam correlations between rainfall and chemical variables are: R_{CE} = -0.86, R_{pH} = -0.67, and R_{alk} = -0.85).

The Kruskal-Wallis test has also confirmed these relationships: EC, pH, and alkalinity are dependent on sampling sites (probabilities of these variables being equal are P_7=0.0265, P_7=0.1709, and P_7=0.1018 respectively, and also P_2=0.0007, P_2=0.0318, and P_2=0.005 respectively).

Electrical conductivity and alkalinity have different values according to the type of precipitation, the highest being for both rains and muddy rains and the lowest being for snow. Some dependence is also found between EC and pH on particular rain events (probabilities of these variables being equal are P_7=0.09 and P_7=0.17, respectively, and also P_2=0.15 and P_2=0.23, respectively). Slightly higher pH and conductivities are associated with E-SE winds. Also pH is correlated with EC (R = 0.81) and with alkalinity (R = 0.83), indicating that basic ions which contribute to high pH dominate in the soluble fraction.

Mineralogy

The results of the mineralogical analyses of the solid residues are presented in Table 2. Eleven mineral species have been identified: seven are silicates (quartz, feldspars, mica-illite, paragonite, chlorite, kaolinite and palygorskite), three are carbonates (calcite, dolomite, and aragonite), and one is a sulfate (gypsum). Most mineral species in all samples are phyllosilicates, with the mica family (represented by both mica-illite and paragonite) dominating.

As first-order mica-illite peaks from the studied samples have appeared under two different widths (Figure 3), the Kubler or crystallinity index was calculated for every sample containing this species. In Figure 4 two well-differentiated mica-illite families can be seen: in the higher part of the gradient, most mica-illites show a low crystallinity (Kubler index = 2); on the contrary, in the lower part their crystallinity is higher (Kubler index = 1) (Kruskal-Wallis test of probability of being from the same population P=0.035). Lower crystallinity is usually attributed either to some degradation of the mineral or to small particle sizes of the order of 0.2 µm or less. Crystallinity has

TABLE 2. Semiquantitative analysis of Mineral Species in Bulk Deposition

Site and Event	Q	I	Chl	K	F	CA	DO	AR	GY	PL	PG	KI
A1	5	42	35	2	0	0	0	0	0	15	0	2
A3	6	36	29	5	4	3	0	0	0	17	0	2
A4	2	64	11	6	3	0	0	0	0	0	14	1
A5	3	47	8	29	4	0	2	0	0	0	6	1
A7	15	35.5	11.5	12.5	8	1.5	2	1.5	0	12	0	2
B1	5	52.5	21	7.5	1	1.5	1	0.5	0.5	9.5	0	2
B3	8	38.5	27.5	2	4.5	1.5	2	2.5	0	14	0	2
B5	2	61	6	20	4	0	0	0	0	0	7	1
B6	2	70	2.5	4	3	0.5	0	0	2.5	0	15	1
B7	11	46.5	12	6.5	5.5	3	2	1	0	7.5	5	2
C3	5	46	30	3	4	0	0	0	0	11	0	2
C5	4	60	11	10	5	0	0	0	8	0	2	1
C6	3	88	3	0	0	0	0	0	0	6	0	1
C7	4	51	7	32	6	0	0	0	0	0	0	1
D5	3	47	14	7	4	1	1	0	4	0	21	1
D6	4	59	16	2	8	0	0	0	0	0	11	1
D7	5	45	22	10	6	0	0	0	0	0	12	1
E2	7	83	0	0	0	0	0	0	0	0	10	1
E3	4	54	5	7	4	0	0	0	0	0	27	1
F1	2	56	9	21	6	0	0	0	6	0	0	1
G1	5	57	5	20	6	0	0	0	7	0	0	1
G4	4.5	58	10.5	5	4.5	2.5	4	1	0.5	0	10	1
G5	5	51	16	6	4	0	3	0	0	0	15	1
G6	3	63	13	5	4	0	3	0	0	0	9	1
G7	5.5	54	13	6.5	2.5	0	3.5	0	4.5	0	9	1

In percent, Q, quartz; I, mica-illite; Chl, chlorite, K, kaolinite; F, feldspars; CA, calcite, DO, dolomite; AR, aragonite; GY gypsum; PL, palygorskite; PG, paragonite; along with the KI, Kubler index for mica-illite.

been used as a tracer to define the origin and processes in Neogene sedimentation in the Betic Ranges [Nieto et al., 1989]. In further analyses, mica-illite will be considered not as a single variable but as two variables: low-crystalline mica-illite (LCI) and high-crystalline mica-illite (HCI).

Most of the mineral species identified by XRD have been identified further by their morphology (by SEM) together with EDX analysis. The following morphologies have been recognized: (1) small fibrous particles (about 5 µm long and less than 1 µm wide with high contents of Si, Al, and Mg and some Ca determined when there are high concentrations of particles, as analysis of individual particles is not possible because of their small size) have been attibuted to palygorskite; (2) acicular particles (averaging 10-15 µm long but reaching up to 20 µm and containing mostly Ca and C) identified as aragonite (Figure 5); (3) rhomboidal particles (from 10 to 20 µm, formed by Ca and C) recognized as calcite (Figure 5); (4) pseudohexagonal particles (3-8 µm in diameter with high contents of Si and Al and the presence of a little K) identified as kaolinite; the presence of K might be attributed to the association of kaolinite with illite; (5) quite large, well-exfoliated minerals (up to 150 µm long, containing Si, Al, Fe, Mg, and some Ca or Si, Al, and K) which
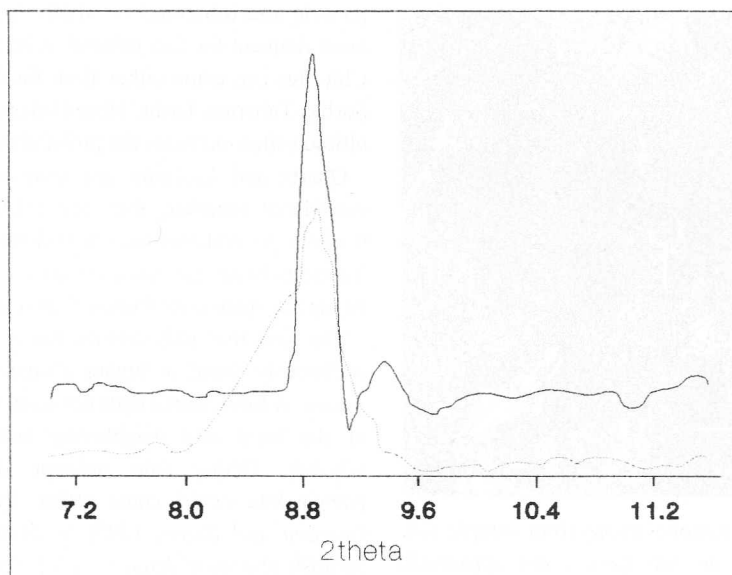
Fig. 3. Mica-illite content and the corresponding crystallinity index in the topographic gradient for all rainfall events.

have been identified as chlorites (Figure 5) and micas, respectively; and (6) equant, sometimes rounded particles of Si generally smaller than 10 μm, of Si, identified as quartz.

Moreover, other less frequent particles have been identified in most sites: rounded particles of C, 5 μm in diameter with vacuolar pores, attributed to residues of combustion; diatoms; plant residues; and perfect siliceous spheres, 7-8 μm in diameter, of unknown origin.

SEM observations have not been suitable to detect clear differences on the average size of mineral grains from the different sites (Figures 5 and 6).

Statistical analyses were performed by the Kruskal-Wallis test to establish the relationships of mineral species with sites, events, and bulk deposition chemistry (Table 3). Some minerals such as quartz, feldspars, chlorite, kaolinite, aragonite, and dolomite are present in most samples and in proportions not related to the altitudinal gradient. Some others, like HCI, calcite,

and gypsum, are somewhat more abundant at lower altitudes. Paligorskite, paragonite, and LCI are significantly related to the site: paligorskite and LCI are more abundant at higher altitudes and paragonite is more abundant at lower altitudes.

Palygorskite increases with altitude ($R=0.59$, $n=25$) and is positively correlated with calcite ($R=0.51$, $n=25$), which also increases with altitude and seldom appears at lower altitudes. Some degree of dependence on events or dominant winds has also been found for some minerals: quartz, kaolinite, and paragonite are somewhat preferently transported by E-SE winds. HCI is somewhat transported by W-NW winds. All the other minerals have been transported by both W-NW and E-SE winds.

Fig. 5. Scanning electron microscope image showing heterogeneous mineral grains from sample B3. The largest particle on the left in the photo, with Si, Al, Fe, and Mg, might be chlorite. The rhombohedral crystal above the latter, with Ca and C, is calcite. Acicular particles on the right, with Ca and C, are aragonite. Particles smaller than 5 μm, with Si, Al, and K, are attributed to mica-illite.
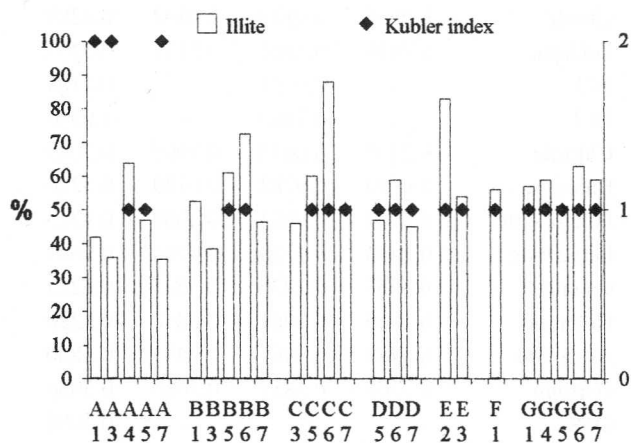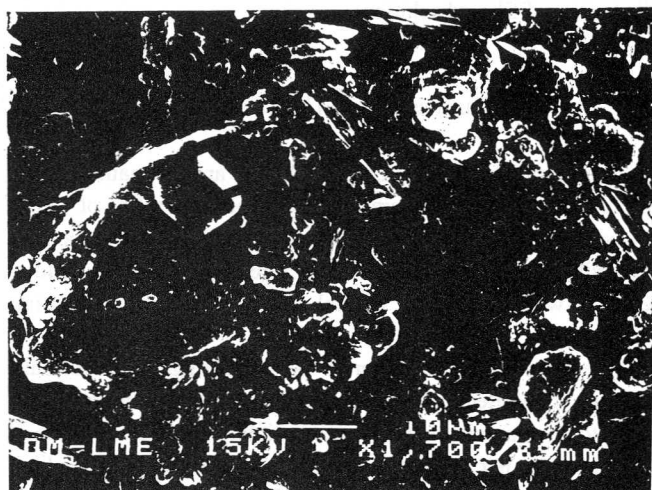

Fig. 4. X-ray diffraction peaks of mica-illite from sites A to G. The highest peak A has a much higher crystallinity than the widest peak G. Peaks measured 1 nm.
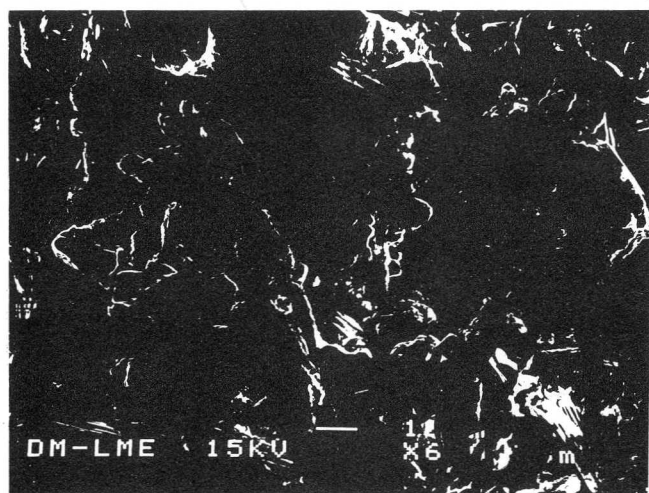
Fig. 6. Scanning electron microscope image from sample G4, showing how mineral grains do not have sizes apparently different from those shown in Figure 5.

Relationships Between Chemistry and Mineralogy of Rain

No correlations were found between contents of calcite and aragonite and the chemical variables, pH and alkalinity. There are, however, some correlations between dolomite and chemical variables ($R_{pH} = 0.53$, $R_{alk} = 0.47$, $n=19$). Calcite and aragonite may have been partly dissolved in rainwater so that no relationship has been found between calcite remaining in the filter and the chemical variables of the corresponding filtered solution. However, dolomite, as a less soluble mineral and, consequently, only partly dissolved, does show a positive correlation with all chemical variables ($R_{pH} = 0.53$, $R_{alk} = 0.47$, and $R_{EC} = 0.46$). Calcite and aragonite, upon dissolving, may have contributed considerably to the high pH, EC, and alkalinities of filtered solutions.

DISCUSSION AND CONCLUSIONS

Gypsum, calcite, aragonite, and dolomite are the main mineral species which can explain the alkaline character of bulk deposition in the study area. Abundant anions from carbonate and sulfate have a clear buffering effect upon usually acidic aerosols which may be natural or produced by industrial contaminants.

Three main groups of minerals can be distinguished from all minerals deposited in the studied altitudinal gradient: (1) ubiquitous minerals that are very slightly related or neither related to site nor to particular events or dominant winds, like feldspars, chlorite, calcite, and aragonite; (2) minerals preferably associated with some sites, like dolomite, or minerals related to either higher or lower altitudes, like HCI and LCI, paligorskite, and paragonite; and (3) minerals related to either precipitation events or to dominant winds, like quartz and kaolinite and, less significantly, HCI and paragonite.

No source area could be ascertained for feldspars, calcite, and aragonite. The only significant result for chlorite is its large

particle size (observed by SEM). If this size is accepted as the most frequent for this mineral, a local source must be admitted. Chlorites can come either from the Filabres Range or from the Sorbas-Tabernas basin. Nevertheless, low plant cover at lower-altitude sites increases the probability of the latter.

Quartz and kaolinite are ubiquitous minerals; however, if considered together, they are related to E-SE winds ($P_2 = 0.1092$). As both minerals are relatively abundant in the Sorbas-Tabernas basin, we could invoke a local source, but not without ruling out some contribution from a more distant one.

The fact that palygorskite has a small particle size and is exclusively found at higher altitudes might indicate a distant source. A local source does not seem to be likely given the nature of the local and neighboring substrate (granatiferous mica schists). Taking into account data for wind direction, palygorskite could come either from North Africa [*Coudé-Gaussen and Blanc*, 1985] or from the southern part of the Spanish plateau [*Vázquez*, 1978]. As the statistical analysis did not show any significant relation between palygorskite and dominant winds, both sources may have contributed to its deposition.

Dolomite is somewhat related to sites A, B, and G and is missing in the intermediate part of the topographic gradient. E-SE winds have contributed slightly more than W-NW winds to deposition of dolomite. At site G, dolomite might come from aerosols generated by the nearby cement industry for which this mineral is the main raw material.

Gypsum is a mineral species mostly found at lower altitudes, which surprisingly, is, somewhat, but not exclusively, related to N-NW winds. As a consequence, two different and

TABLE 3. Probabilities of the Differences Among Two or More Group Means Being Equal (Kruskal-Wallis Test)

| Variables | Probabilities of Group Means Being Equal | | | |
| | Sites | | Events | |
	7 Groups	2 Groups	7 Groups	2 Groups
EC	0.0265	0.0007	0.2865	0.1508
pH	0.1709	0.0318	0.1777	0.2367
Alkalinity	0.1018	0.0050	0.6086	0.7999
Quartz	0.9635	0.9128	0.0392	0.1299
Feldspar	0.7431	0.3961	0.5111	0.5407
LCI	-	0.0351	-	0.2104
HCI	-	0.1889	-	1.0000
Chlorite	0.2465	0.6614	0.3903	0.7049
Kaolinite	0.9999	0.9782	0.0189	0.3220
Paligorskite	0.2761	0.0061	0.4007	0.8313
Paragonite	0.1493	0.0160	0.1704	0.2684
Calcite	0.3655	0.2736	0.9523	0.5217
Dolomite	0.1209	0.3961	0.6842	0.3220
Aragonite	0.6424	0.3961	0.9393	0.5800
Gypsum	0.7563	0.2857	0.5680	0.3080
Q+K	-	-	-	0.1092
C+A+D	-	0.8911	-	0.2944

Abbreviations: EC, electrical conductivity; LCI, low-crystalline mica-illite; HCI, high-crystalline mica-illite; Q+K, quartz + kaolinite; C+A+D, calcite + aragonite + dolomite.

geographically opposite origins may be invoked for this mineral. First, the gypsiferous marls of Sorbas-Tabernas basin are probably a nearby and important source for gypsum aerosols in the area. Second, justifying a N-NW source, to the north and west of the Filabres Range, outcrops of Eocene and Miocene sediments contain gypsum. In fact, gypsum is almost ubiquitous in all tertiary basins to the north and west of the studied area.

Paragonite is found in significantly higher proportions at the lower part of the gradient and is somewhat related to E-SE winds. Assuming that paragonite is found as relatively large particles (larger than several microns), its principal source area must be the Sorbas-Tabernas basin, where this mineral has been detected. The presence of relatively abundant paragonite within the Betic Ranges has been known since the beginning of the 1970s. Although paragonite has also been identified in other locations in the Iberian Peninsula, it is always scarce [*García-González and Aragoneses*, 1990].

Taking into consideration the lithological differences between high and low altitudes, it can be inferred that local factors are quite important in the formation of aerosols precipitating near their source. Large particle sizes in the collected minerals, like HCI, chlorites, calcite, aragonite, and dolomite, clearly indicate the influence of local sources. Other minerals, like LCI, paligorskite, and possibly some of the carbonates which have contributed to high pH, EC, and alkalinities with sometimes smaller particles, might have come from distant sources that cannot be inferred at present. We can only observe that air masses from the east and SE contributed more significantly to higher pH and EC than air masses from the west and NW. Unfortunately, no significant relationship has been found between dominant winds and alkalinity, so the main source of calcite and aragonite cannot be inferred. S-SE winds contribute somewhat more to dolomite deposition than N-NW winds.

Mineralogical and chemical characteristics of bulk open field precipitation in SE Spain have proved to be useful in understanding the geochemical characteristics of atmospheric dust and in fingerprinting, in a few cases, the source areas of some minerals.

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