Geochemical characteristics of particulate matter in the atmosphere surrounding a ceramic industrialized area

E. T. Gómez · T. Sanfeliu · M. M. Jordán · J. Rius · C. de la Fuente

Abstract Total suspended particulate samples (TSP) were collected and concentrations measured during seventeen months in the vicinity of a ceramic industrial area. A method of fractionating was applied to the samples in order to obtain two fractions corresponding to mineral particulate coming from dust emissions (Upper-F fraction) and to amorphous matter (carbon plus small amounts of S, Ca, Fe, etc) coming mainly from traffic and other combustion processes. Also for TSP samples several element concentrations were measured following two previous treatments: extraction of elements mainly associated with the soluble fraction of the samples (B, Fe, P, As, NO^{2-}, NH^{4+}, Cl^{-}, F) and acid digestion for trace metals and elements mainly associated with the non-soluble fractions of the samples (Fe, As, Cd, Ni, Pb, Zn Ca). Seasonal differences and the influence of meteorological parameters (temperature, relative humidity, pressure and wind conditions) on the air pollution levels, particles as well as ions, were studied.

Results show different seasonal and weekly evolution for mineral and amorphous carbonaceous particles because of the different origins in dust emissions or combustion processes respectively, and the different physical properties such as size grain. Of the ions analyzed Fe, Ca and Zn were clearly associated to mineral phases and consequently related to dust emissions, and NO^{2-}, NH4+, P, Cl^{-} were related to amorphous matter coming from combustion. Ni and Cd show lower levels than those reported as guideline values and the source is mainly related to the enrichment of these elements in clay materials. B and As content result in elevated concentrations, with the tendency to increase during cold months. The emission of these elements was associated with vaporization or volatilization during high temperature ceramic processes. The original gaseous state is influenced by temperature. In the winter the content for B and As is higher due to enhanced condensation of gas-phase boron onto particles, while in the summer the increase of air temperature results in elevated evaporation.

Keywords Air pollution · Particulate matter · Mineral matter · Amorphous matter · Spanish ceramic industry

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Introduction

The quality of air in ceramic industrial areas is mainly impoverished by particle emissions, (Boix and others 1995, Boix 1996). Previous studies in the ceramic area of La Plana in Castellón (NE Spain) have shown two effects associated with these atmospheric pollutants (Gómez and others 2001; Sanfeliu and others 2002; Jordan and others 2002): (1) daily and weekly periods with high concentrations of particles different grain size ranges and; (2) periods of high levels of B, As, F, Fe, etc., which seems not to be associated with the evolution of particle concentrations.

In this same area, the main source of particles are dust emissions coming from extraction, transportation, loading, unloading of mineral raw materials and combustion of

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fossil fuels from traffic. The origin of atmospheric pollution by trace metals or other elements is not so clear. Trace elements tend to distribute in atmospheric particles as ions adhered to particles, by adsorption to their structural surface or by forming the crystalline structure; in general, the process involved depends on the physical state at the moment of the atmospheric emission. Dust emissions mainly release to the atmosphere elements in the crystal structure but emissions produced in high temperature activities could be the source for ions with reference to the three distribution processes mentioned above.

Clay atomization, tile firing and frit-melting processes produce mainly the following emissions:

- Solid particles released by dragging or semi-molten from raw materials. These particles have compositions enriched with the elements Si, Ca, Mg, Zn, Zr (Vickery and others 1998; Jordan and others 2002).
- Sublimation and decomposition of raw materials produce vaporized or volatilized ions such as: B, Cl, Pb, F, SO_x (Lezaun Navarro and others 1998).
- Combustion of natural gas produces mainly emissions of NO_x and other gaseous compounds such as CO, CO_2, NH_4, etc. (Busani and Navarro 1994).

Despite the fact that traffic seems to release a high content of carbonaceous particles to the atmosphere in the area (Gómez 2002; Jordan and others 2002), it can also be the source of trace elements. Traditionally, Pb and Br were used as tracers of emissions coming from vehicles, but nowadays the usefulness of these tracers is not as clear because of the increase in the use of unleaded petrol and the high quantity of different elements emitted during fuel combustion. Xudong and others (1994) established Sb, Br and Zn as potential tracers from traffic emissions.

Sanfeliu and others (2002) have already identified the relationships of several elements with dust or high temperature process emissions in the ceramic area of Castellon. Al, Fe and Mg are associated with the transfer of clay raw materials, while the content of Ca may be from the exploitation of clays in the area. Finally, elements such as Zn, As and Zr seem to be mainly influenced by emissions from firing and melting kilns.

Three important relationships are considered: (1) the association of several ions with atmospheric particle levels as well as the distribution of the elements in these particles account for their behavior in the atmosphere; (2) the relationship of the same ions with traffic emissions as the possible source for them and; (3) the seasonal evolution of particles and atmospheric elements as indicators for the influence of the meteorological parameters in the area.

Materials and methods

Sampling

Samples of TSP were collected at the Escuela Taller site in Onda (Castellón, Spain). This sampling location is in the western portion of the ceramic industrial area. All of this area is within a coastal plain; therefore, atmospheric pollutants flow throughout the area according to meteorological parameters, mainly wind speed and direction (Boix 1995).

Seventy-five air samples of 24 hours duration were taken during ten periods from January 1999 to May 2000. The sampling programme obtained samples from different meteorological conditions and industrial activities (working and non-working days).

The sampler was a high-volume MCV sampler type CAV-A/HF, with an effective sampling area of 400 cm^2 and a pumping flow of 30 cm^3 h-1. Filters of cellulose membrane of 0.45-μm-pore size (Millipore HAWP142 50 20.5×25.4 cm) were used during sampling.

Fractionating of particle samples

Samples of TSP were split into two fractions according to the distribution of particles on the filter (Fig.1). From a quarter of each filter of TSP the following method was devised: (1) individual particulate material on the upper zone of the filter was mechanically extracted from the cellulose membrane, suspended with cyclo-hexane and deposited under vacuum on silver filters (Millipore 0.3 micrometers). This fraction of particles was denominated by *Upper*-F fraction 2) Particles embedded in the filter pores were mechanically extracted from the cellulose membrane with residues coming from the filter. Therefore, the residue and particulate material was suspended in cyclo-hexane, ultrasonically treated at 30 °C, sifted through a sieve (35 micrometers) and immediately deposited under vacuum on silver filters. This fraction of particulate material was denominated by the *Lower*-F fraction.

Each of these fractions was analyzed by means of X-ray diffraction (following the method reported by Jordan and others 2002; Goméz and others 2002) and by scanning

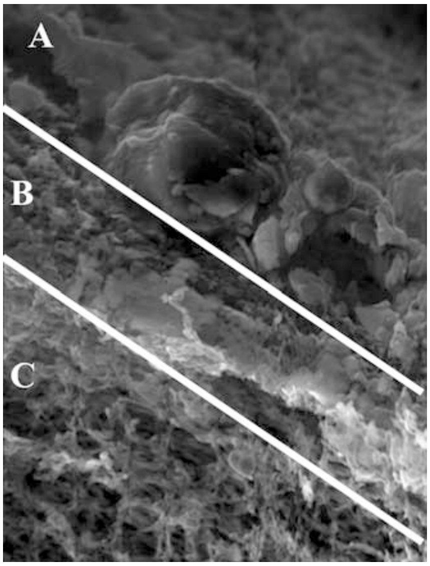


Fig. 1
Edge-on view of particles after collecting on permeable cellulose filter. Three zones can be distinguished: *upper right*, only particulate material; *bottom left*, membrane filter; and *between*, particles embedded in the filter pores

electron microscopy to evaluate their main composition (mineral or amorphous phase).

Chemical analysis

In the present work several elements mainly associated with different sources were selected for chemical analysis. Before analyzing, it was necessary to extract ions from atmospheric particles by following two methods:

1. *Extraction of elements mainly associated with the soluble fraction of the samples (ES)*; because of element-dependent differences in detection limits and techniques of analysis of each one of the selected ions in this group (B, Fe, P, As, NO_2^-, NH_4^+, Cl^-, F) a fraction of each filter was used and treated by liquid extractions. In general, the procedure was performed by mixing into a Teflon bomb a fixed volume of ultra pure water, a section of filter and different reagents. Each one of these mixtures was ultrasonically treated at 40 °C and filtered through cellulose membrane filters (Millipore AAWP004700). This liquid extraction without particles was used for chemical analysis.
2. *Acid digestion for trace metals and elements mainly associated with the non-soluble fraction of the samples (AD)*: Two fractions of 16 cm^2 for each filter was digested by means of acid attack following four steps: (1) 2 ml of concentrated supra-pure nitric acid were added to the filter and heated at 90 °C in a closed Teflon bomb for 24 hours; (2) once the mixture was cold, 2 ml of hydrogen peroxide and 2 ml of supra-pure citric acid were added and heated at 110 °C for 24 hours; (3) after cooling, 5 ml of ultra pure water was added and the solution was centrifuged (3000 rpm for 15 min twice) before separating the residue of particles (mainly graphite) from the solution; (4) the solution was transferred to a graduated volumetric flask to make a volume of 15 ml. Final acid concentration was 1% nitric acid. Blank filters digestion with and without NIST Standard 1665B Fly Ash, Fe, As, Cd, Ni, Pb, Zn and Ca was analyzed before treatment according to this method.

Both types of solutions were analyzed by inductively coupled plasma mass spectrometry (ICP-MS) or UV-Spectroscopy.

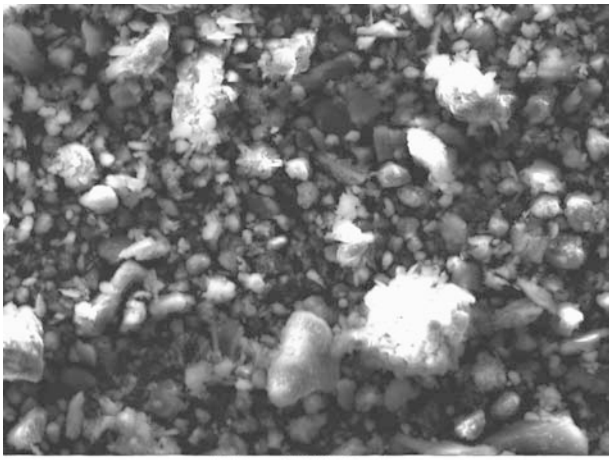
Meteorological parameters

Standard meteorological parameters were obtained by means of an automatic meteorological station near the sampling apparatus (200 m), which belonged to "Conselleria de Medio Ambiente de la Generalitat Valenciana". From these data a matrix was built for each sampling period with the following meteorological parameters: weekly mean temperature, weekly mean atmospheric pressure, weekly mean humidity and weekly total precipitation. For evaluating the influence of the wind, 4 day types were established according to the daily parameters of wind speed and direction: (1) day type 1A characterized by a sea breeze wind component with an average wind speed lower than 3 m s^{-1}; (2) day type 1B is defined by an intense local sea breeze with an average wind speed higher than 3 m s^{-1}; (3) day type 2A wind blows during periods of one or half an hour with variable direction and a speed lower than 2 m s^{-1} and; (4) day type 2B wind with wind direction between 250–290° W-NW and constant speed higher than 3 m s^{-1}.

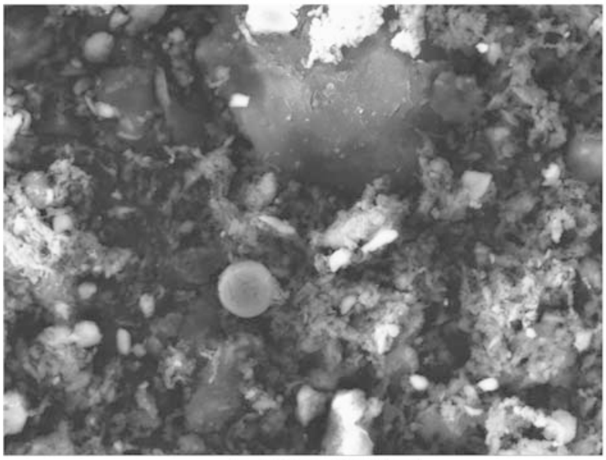
Results

Composition of TSP after fractionating

X-ray diffraction and SEM analysis of the *Upper*-F and *Lower*-F fractions coming from TSP showed important differences (Figs. 2 and 3). The *Upper*-F layer is mainly composed of individual particles of different irregular morphologies. The opposite is true in the *Lower*-F fraction, here three types of morphologies can be identified: (1) individual larger irregularly formed particles of non-spherical morphology; (2) perfect sphere-like particles; and (3) large masses with clot-like appearance of aggregate particles. In both fractions individual and larger irregularly formed particles corresponding to crystalline phases



a. (Mag. 3.50 K x)



b. (Mag. 8.5 K x)

Fig. 2a, b
Micrographs of *Upper*-F (**a**) and *Lower*-F (**b**) fractions coming from TSP samples. **a** Mag 3.50 K x; **b** Mag 8.5 K x

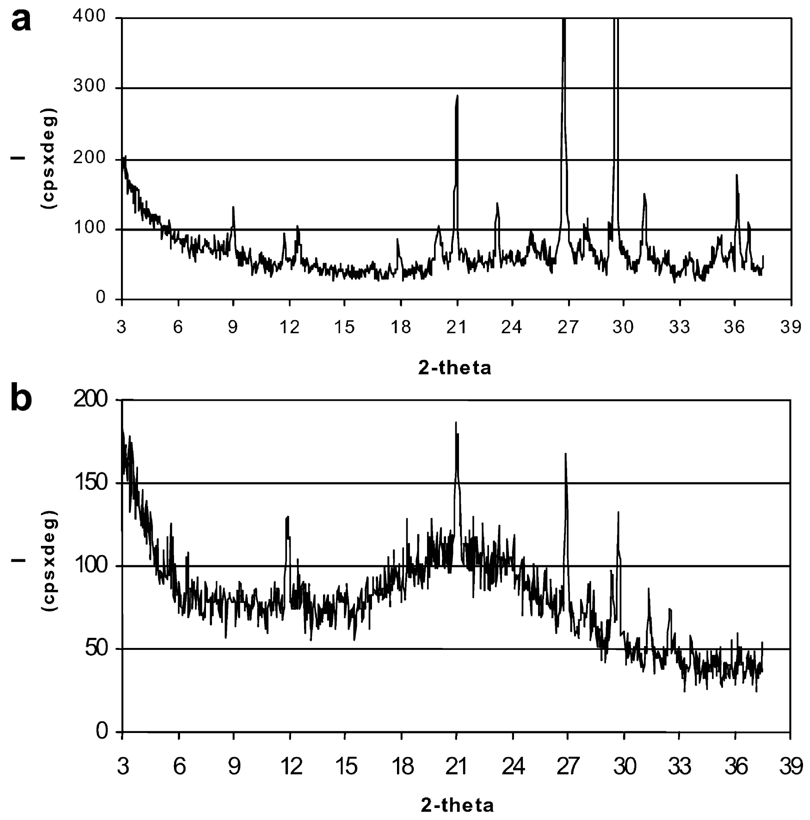


Fig. 3a, b
XRD diagrams corresponding to: **a** *Upper*-F and **b** *Lower*-F fractions coming from TSP samples

(different mineral particles as can be see in Gómez and others 2001; Gómez 2002) can be identified and studied by X-ray diffraction (Fig. 3).

The visual analysis by means of SEM in some samples, similar to Fig. 2, showed that masses of clot-like morphology of particulate aggregate dominates the *Lower*-F fraction with a proportion higher than 85% of the whole material. This type of morphology as clots of dendritic structure (Ortner 1999) is associated with amorphous matter of carbonaceous particles linked with organic compounds, both coming from the incomplete combustion of a long chain of hydrocarbon fuels, e.g., in diesel or fuel motors of cars. In the samples, the origin of this type of non-crystalline components in traffic emissions is due to chemical composition obtained by means of EDAX spot microanalysis on the *Lower*-F fraction material. Values with a high content of C and minor proportion of Si, S, K and Fe have been identified and these data agree with values reached by Ortner (1999) and Umbria and others (1999) for this type of anthropogenic components. Also, a minor content of Ca, Na, Zn and Cl has also been observed. Finally, the minor content in the *Lower-F* fraction correspond to sphere-like particles with grain size ranging from 10 μm to less than 1 μm, which are associated with atmospheric emissions from high temperature ceramic activities (Alvarez and others 1999).

These results show that mineral phases dominate the *Upper*-F layer composition, and represent the influence of dust emissions. However, the main component of the *Lower-F* layer is amorphous matter (carbon plus small amounts of S, Ca, Fe) coming from traffic.

Seasonal evolution of atmospheric particles

High values of particulate matter in the air as TSP can be attributed to three situations (or facts): high values of the *Lower*-F fraction (September 1999), high values of the *Upper*-F fraction (May 2000) or high values for both fractions (July 1999).

In general, particulate matter concentration of the *Lower*-F fraction, 16–91 μg/m^3 (an average of 59 μg/m^3) were significantly higher than aerosol concentrations of the *Upper*-F fraction, 0–80 μg/m^3 (an average of 33 μg/m^3). The *Lower*-F particulate matter accounts for 33–100% of the TSP, while the *Upper*-F fraction accounted for 0–67% of the TSP. Generally, the levels of particulate matter in the air for TSP and the *Upper*-F fraction increased with similar weather conditions. The higher values for the *Upper*-F fraction are identified during long periods with sea-earth breeze winds type 1B, with the additional presence at this time of a high-pressure trough situated over the eastern portion of Spain and non-precipitation. The amount of particulate matter shows wide variations between periods and even between days in the same period (note the higher values for standard deviation in the *Upper*-F fraction). Levels for the *Upper*-F fraction considerably decreased in the weeks with high values of total precipitation. Because of such a trend, a relationship between ambient dryness and the input of mineral particulate matter is clear.

However, the *Lower*-F fraction levels showed homogeneous mass concentrations. The evolution of the *Lower*-F fraction is not associated with the same weather conditions as that of the U*pper*-F and TSP particulate matter. In general, the levels of particulate in the air for the *Lower*-F fraction increased during the warmer months (July, August, September). During August the industrial activity decreased in the area, so the mass concentration of 62 μg/m^3 for the *Lower*-F fraction in this month is a high value.

From these general trends, the highest values of TSP in July 1999 are due to the union of weather conditions that lead to the increase of both mass concentration levels, the *Upper*-F and the *Lower*-F. Similarly, the weather conditions during January 2000 contributed to lower values for particulate in the air for the *Upper*-F as well as for the *Lower*-F fraction, producing the lowest mass concentration levels for TSP.

Seasonal and weekly evolution of elemental composition

The mean atmospheric concentrations of the elements can be divided into a total of five types of situation as follows:

1. The contents of B, Cland As (ES) tend to increase during the winter months.
2. The contents of F, NH_4^+ and As (AD) tend to decrease during periods with alternation or predominance of sea-breeze winds of low speed.
3. Atmospheric concentrations of Ca, Zn and Fe (AD) tend to increase with increasing the content of the *Upper*-F fraction from TSP, therefore increasing with mineral phases coming from dust emissions.
4. The content of NO_2^- seems to be associated with particulate matter of the *Lower-F* fraction from TSP.
5. The content of Ni, Cd and Pb cannot be associated with any parameter (seasonal, meteorological or particles concentration).

To evaluate weekly evolution, the fall (or increase) in the mean element concentrations during non-working days (Saturdays, Sundays) for nine sampling periods was calculated and analyzed (Fig. 4). From this figure the content of several elements during non-working days can be divided into four groups:

1. Largest fallouts corresponding to the content of Fe (AD), Ca, P, Ni and F, and these fallouts are identified in all the periods analyzed.
2. The content of NO_2^- and Cl^- tends to decrease during non-working days but the fallout is located around a low percentage.
3. The content of Cd, Pb, As (AD) and B can slightly increase or decrease during non-working days.
4. During several sampling periods the content of Fe (S), NH_4^+ and As (S) is increased on non-working days.

Relationship between element content and particles

To evaluate the relationship between particles and element content, PST samples were gathered according to the *Upper*-F and *Lower*-F mean concentration of particles. For these groups of samples mean element content was calculated and performed. From the *Upper-F* fraction four groups were established (Fig. 5) and three groups for the *Lower-F* fraction (Fig. 6).

As can be seen in Fig. 6 the content of Fe (AD), Ni, Cd, Ca, F and Zn clearly tends to increase with increasing particles concentration of the *Upper*-F fraction. The contents of B, Fe (ES), NH_4^+, As (ES), As (AD) and Pb are identical in all the groups established according to the *Upper*-F concentration.

The relationship between element content and the *Lower*-F fraction have shown much variability. The content of P, NO_2^-, NH_4^+ and Cl^- clearly increase with the increase of the *Lower*-F particles concentration. The contents of Fe (AD), As (ES), As (AD), Ni, Cd and Pb exhibit exactly the opposite trend decreasing with increasing *Lower*-F content. The contents of F, Fe (ES), Ca and Zn are identical in all the groups. Finally, the content of B is increased in samples with a middle mean concentration of the *Lower*-F fraction.

Discussion

Of the ions analyzed and studied, several are clearly associated with the different fractions (*Upper*-F and *Lower*-F) coming from TSP samples.

The atmospheric contents of Fe, Ca and Zn are strongly associated with the *Upper*-F particles, and consequently depend on the dust emissions in the ceramic industrial area. The observed concentrations of Fe and Ca were higher than those reported by several authors around the world (Bergametti and others 1989; Maenhaut and Cornille 1989; Pacyna 1998; Lee and others 1994). Therefore, the concentrations of Fe and Ca relate to the high content of these elements in the raw clay materials used in ceramic industry.

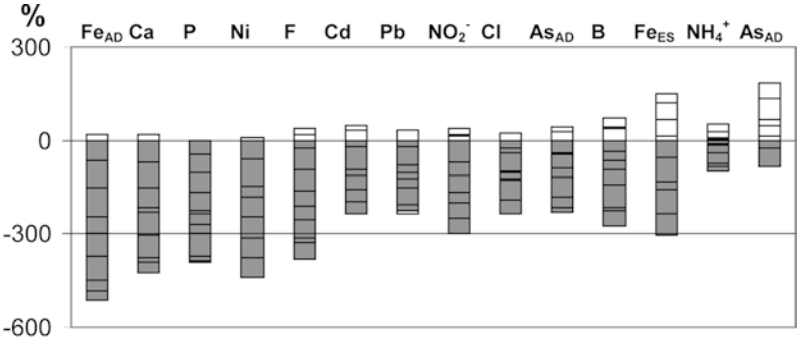


Fig. 4
Mean element content fall (*grease*) or increase (*white*) during non-working days in ten sampling periods

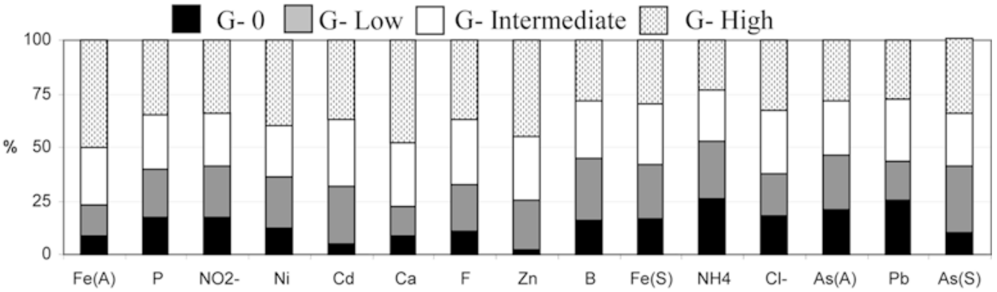


Fig. 5
Element content distribution (%) according to particles concentration of *Upper-F* fraction from TSP. *G-O*, TSP samples without *Upper-F* fraction; *G-low*, TSP samples with *Upper-F* concentrations lower than 20 µg/m^3; *G-intermediate*, TSP samples with *Upper-F* concentrations between 20 and 40 µg/m^3; *G-high*, TSP samples with *Upper-F* concentrations higher than 40 µg/m^3

However, the source of Zn is not so clear. Traditionally, the main source of Zn was from traffic, but in this work the use of Zn as a tracer of emissions from vehicles is dismissed. The range and the average of Zn concentration (182 ng/m^3) agreed with values established by Lee and others (1994) in urban locations, only during some working days are these values exceeded. This last fact and the association of Zn with the *Upper*-F fraction from TSP led to the association of atmospheric Zn content with emissions coming from mechanical activities of raw materials used in frit and enamel and tile production.

The contents of Ni, Cd and F show a certain relationship with the *Upper*-F fraction but it is not as strong as the above mention elements. Therefore, besides dust emissions, there are other sources. A proportion of F content is associated with vaporization and volatilization during tile firing which is a continuous activity without stopping on the weekend.

Results show relatively low concentrations of Cd and Ni with levels lower than those recommended by the European Commission. Both elements are associated with the *Upper*-F fraction of TSP, so the contents of Cd and Ni are mainly related to the enrichment of these elements in raw clay materials.

Of the ions associated with the *Lower*- F fraction of TSP, NO$_2$$^-$ shows a strong relationship while NH$_4$$^+$, P and Cl$^-$ are also related but with lower intensity. Traditionally, in the ceramic area the main source for these ions was gaseous emissions from gas combustion and traffic. Atmospheric levels of NO$_2^-$ are clearly associated with these sources, while the content of NH$_4^+$, P and Cl$^-$ originated from other types of emissions. Atmospheric levels of Cl$^-$ can be associated with three sources: vaporization or volatilization during high temperature ceramic processes, traffic and marine aerosol. Zhuang and others (1999) obtained Cl$^-$ concentrations around 0.14 µg/m^3 and 0.13 to 2.2 µg/m^3, respectively, for parking and coastal urban sites. Clarke and others (1999) measured Cl$^-$ concentrations that ranged between 0.5 to 2.89 and 0 to 1.94 µg/m^3, respectively, for urban and rural sites in the UK. On the other hand, if a natural source for Cl$^-$ content was assumed, the ratio between Na and Cl concentration (Na/Cl) would be around 1 (Ohta and Okita 1990). The average for Na concentrations measured by Boix (1995) in the area was 2.26 µg/m^3 while the average of Cl$^-$ concentrations in the present study ranged between 0.18 to 8.20 µg/m^3. If these values are considered, there is an excess of Cl$^-$ content for natural origin, besides values of Cl$^-$ that exceeded data reported for areas with the direct influence of traffic. Therefore, processes of vaporization and volatilization of Cl$^-$ are affecting the atmospheric levels of Cl$^-$ in the area.

In the area, concentrations of NH$_4^+$ were slightly higher (0.74 to 4.05 µg/m^3) than those measured by Clarke and others (1999) at urban and rural sites in the UK, respectively 0.67 to 4.89 and 0.36 to 2.35 µg/m^3. Consequently, atmospheric levels of NH$_4$$^+$ are similar to those reported for coastal urban areas where the NH$_4$$^+$ source is associated with combustion processes and the content of this ion in marine superficial waters. This last fact is reinforced because of the decrease of NH$_4$$^+$ content during periods of lower wind speed for the sea-breeze wind.

In spite of the relationship between the content of P and the *Lower-F* fraction of TSP, the same as Cl$^-$ and NH$_4$$^+$, the reduction of P content during non-working days is lower than those ions. This fact can be related to the weekly

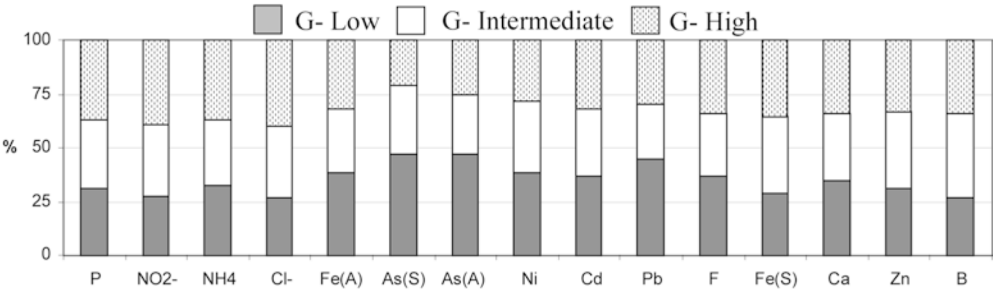


Fig. 6
Element content distribution (%) according to particles concentration of *Lower-F* fraction from TSP. *G-Low*, TSP samples with *Lower-F* concentrations lower than 30 µg/m^3; *G-intermediate*, TSP samples with *Lower-F* concentrations between 30 and 60 µg/m^3; *G-high*, TSP samples with *Lower-F* concentrations higher than 60 µg/m^3

continuous emissions of P during clay atomization where phosphates are used for treating clays.

The content of several elements B, As, Fe (ES) and Pb are not associated with any fraction (*Upper*-F and *Lower*-F) coming from TSP.

Atmospheric concentrations of B and As show strong relationships with weather conditions, which results in the trend to increase during the cold periods. Besides, both elements are associated with vaporization or volatilization during high temperature ceramic processes.

The range of B content in the present study (6 to 173 ng/m^3, average 65 ng/m^3) can be compared to the data reported by Anderson and others (1994) for continental sites with coal-fired power plants, municipal incinerators and intensity traffic (10 to 26 ng/m^3), coastal areas (2.4 to 82.8 ng/m^3), remote marine sites (0.6 to 13.4 μg/m^3) and direct emissions in coal-fired power plants (29 to 162 μg/m^3). Therefore, it is considered that the experimental concentration data of B in the area is associated with a coastal area where some human activity is increasing the natural atmospheric B content.

The average of the As (AD) concentration (111 ng/m^3) agreed with values established by Bretschneider and Kurfüst (1987) corresponding to different sites around the world (3 to 190 ng/m^3), but it is higher compared to the guideline values of the European Commission (4 to 13 ng/m^3). Therefore, there is an anthropogenic contribution of As in the ceramic industrial area.

The identification of higher concentrations of B and As during cold periods is probably due to enhanced condensation of these elements in the gaseous phase after coming out of the industrial ceramic funnel onto atmospheric particles. Cold weather would enhance or aid the deposition of these elements during transport. Other authors have identified similar situations. Hrsak and others (2001) identified a strong positive relationship between Hg atmospheric concentrations and air temperature, which results in the strong influence of elevated temperature to the evaporation of mercury. Also, Anderson and others (1994) identified higher total B concentrations during wintertime; however, the individual behavior of concentrations of gas-phase boron and particulate-boron was opposed. Concentrations of particulate boron were higher during wintertime while levels of gas-phase boron were higher during the warmer period. These facts were associated with the enhanced condensation of gas-phase boron onto particles during cold weather. In the ceramic industrial area a similar phenomenon can occur, after vaporization or volatilization of B and As in high temperature ceramic processes, these elements can continue in gas-phase or condense onto particles according to temperature.

The atmospheric Pb content in the area can be associated with several sources. However, results show relatively low concentrations compared to the guideline values and data reported by other authors such as Lee and others (1994). According to this author the average Pb concentrations in the area is lower than the range of concentration values established for urban areas. Besides, the Pb content cannot be associated with traffic because it cannot be related to amorphous carbon particles of the *Lower-F* fraction of TPS.

Finally, the authors would like to emphasize that all the ions associated with combustion processes and/or high temperature ceramic activities (B, F, NH_4^+, Cl^- and As), except NO_2^-, trend to suffer lower reductions on non-working days during the periods with the smallest concentrations of TSP. This fact is associated with the continuity during weekend of these activities. On the other hand, the behavior of these elements after emission is strongly related to the gas-phase state, which is conditional on weather conditions.

Conclusions

Mineral content is increased over long periods during which high speed sea-earth breeze winds are prevalent, in addition to the presence at this time of a high-pressure trough situated over the eastern portion of Spain combined with non-precipitation. During such weather conditions there is no washout in the atmosphere and the specific meteorological conditions (winds and atmospheric pressure) help to increase the processes of accumulation of atmospheric particles, mainly mineral phases with a grain size higher than amorphous matter coming from combustion.

Clearly Fe, Ca and Zn are associated with dust emissions in the ceramic industrial area, and NO_2^-, NH_4^+, P and Cl^- are related to amorphous carbonaceous matter coming from combustion processes, mainly traffic.

B and As are related to gaseous emissions after vaporization or volatilization during high temperature ceramic processes. Seasonal evolutions of B and As concentrations depend on this original gaseous state. The content of these elements tends to decrease during warmer months as a consequence of the influence of elevated temperature to the evaporation. In the winter the content for B and As is increased in the area due to enhanced condensation of gas-phase boron onto particles.

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