The relationship between indoor and outdoor levels of PM10 and its chemical composition at schools in a coastal region in Spain

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PM10 levels and its chemical composition were studied inside and outdoor of seven primary schools (3 in urban environment, 3 in industrial environment, 1 in rural environment) located in the Mediterranean coast in an area with an important industrial nucleus dedicated to the treatment of raw mineral materials. The main objective of this work is a comparison between these levels obtained inside and outside schools and also assess the influence of various natural and anthropogenic emission sources on particles concentrations found inside. The indoor airborne samples were collected using Respicon TM. In the three outdoor sampling stations was used a minivol air sampler type 3.1 LVS of Derenda. PM10 chemical composition was obtained by ICP-MS (elements) and ion chromatography. The ratio I/O (indoor/outdoor) has been calculated taking into account only the samples taken in the same conditions. In all schools the ratio I/O for PM10 was greater than unity (between 1.3 and 7.8), indicating that existed significant indoor sources of these particles. In the three schools located in the industrial environment were collected PM10 samples inside and outside in non-teaching periods. Comparing the values of I/O when the classrooms were unoccupied with respect to the average value of these same schools when the classrooms are occupied, the behaviour is different depending on the location. On the other hand, in an industrial school was obtained when some infrastructure works were being carried out outside of school. This caused a significant increase in the concentration of particles in the interior (I/O = 19.9). From the levels of As, Ni, Cd, Pb, Al, B, Zn, Mg, Sb, F−, ClO2−, NO3− and SO42− in PM10 inside and outside of each school, also the ratios I/O were calculated. These chemical ratios I/O were higher than unity in all cases and generally higher than those recorded in the case of PM10. Finally, Pearson correlation coefficients (r) between the elements and anions and the PM10, and between the different elements and anions were calculated for the purpose of establishing the existence of common emission sources.

1. Introduction

It is well known by epidemiological researches that there is a strong relationship between air pollution and morbidity and mortality, with air pollution potentiating a wide range of diseases and cardiovascular problems, especially in young, elderly and ill people. According to Xing et al. (2016), adults exposed to ambient air pollution, for example PM10 (particulate matter 10 micrometers or less in diameter) and coarse particulate, have shown increased prevalence of respiratory disease. Such health problems include cardiac arrhythmias, reduced lung function, asthma, chronic bronchitis, and increasing respiratory symptoms such as sinusitis, sore throat, dry and wet cough, and hay fever (Moreno et al., 2004). Recent researches also suggests that these problems can be related to specific components of particulate matter rather than mass (Ghio and Devlin, 2001; Khaniabadi et al., 2017). Children have certain characteristics that make them more susceptible to air pollution than adults. On the one hand, their breathing rate is higher (Faustman et al., 2000; Landrigan et al., 1999) and on the other hand respiratory systems of children under five are not fully developed and their immune system is not mature (Banerjee, 2000; Boy et al., 2000; Khalequzzaman et al., 2007).

However, people in developed countries are spending the majority of their time (more than 70%) in various indoor environments (Guo et al., 2019). This is an open access article under the CC BY-NC-ND license (http://creativecommons.org/licenses/by-nc-nd/4.0/).
most considerable portion of the day in, apart from at home (Silvers et al., 1994). Despite this evidence most air pollution studies focus on the general population (considering all age groups) and on the relationship with the pollutant levels outdoors.

Pallares et al. (2019) establish a catalogue that brings together the morphologic characteristics of the main types of particles found among the indoor airborne fine particles captured in the primary schools in the studied area and to assess the influence of outside emission sources, on the particles found inside of schools with natural ventilation.

It is generally accepted that indoor concentrations of particles derive from two sources: indoor and outdoor. However, the significance of both sources depends on a number of variables, the most important of which are the generation rate of particles indoor, the outdoor particle concentration, air exchange rate, particle penetration efficiency from the outdoor to the indoor environment, and the particle deposition rate in indoor surfaces (Kamens et al., 1991; Thatcher and Layton, 1995).

Ratios between the concentrations of particles and chemical elements found indoor and outdoor a building give an indication of the contribution of indoor sources. It is expected that in the absence of internal sources, the ratios I/O (indoor/outdoor) between the concentrations are less than or equal to 1 (Chao et al., 1998; Jones et al., 2000) and the ratios will be higher unit in the presence of significant indoor sources (Zuraimi et al., 2003; Stranger et al., 2007).

In places where there is no clear source of contamination in the interior, the occupant-related activities may represent the main source of particles. These are usually composed of clothing fibres, hair fragments, soil particles, skin cells, resuspension of particles of different nature due to the movement, and emissions of materials used, as well as paper, spores and fibres, etc. (Brans et al., 2005).

Moreover, the type of ventilation used is a very important factor that determines the level of particulate indoor and therefore the values of the ratio I/O (Parker et al., 2008). Numerous studies have compared different types of ventilation and determined its influence. Thompson et al. (1973) in his study found that the ratios I/O of the PST were less than 0.1 in air conditioned buildings with higher filtration systems while were much higher in buildings with natural ventilation where there was also a high movement of people. Similar events were observed by Zuraimi et al. (2007) with the PM2.5 particulate, with values of I/O close to unity with the natural ventilation and recording a significant decrease when using air conditioning systems with filters.

The highest concentration of PM10 and PM2.5 found by Gemenetzis et al. (2006) in Thessalonika indoors were associated with limited ventilation and high outdoor concentrations. The use of a receptor model indicated that emissions from traffic, resuspension of road dust and heating are the main sources of PM10 in the outside (Spamara et al., 2003).

The study area is located in the Mediterranean coastal basin, province of Castellón (Spain) with major problems of high concentration of particular matter in ambient air, and also is located next to an industrial centre based on the treatment of ceramic raw mineral materials as ceramic red clays and silicates (important particulate emissions). It is a strategic zone in the framework of EU pollution control. Around 80% of the European Union ceramic tile and ceramic frit manufacturers are situated in the mountainous interior (Fig. 1). These localities (Castellón, Alcora and Lucena) represent three different environments: urban, industrial and rural respectively.

In the city of Castellón (urban area) three schools were selected: S1, S2 and S3. As a fundamental characteristic, all are located near road networks with a high density of traffic. The three sampling sites were situated in different zones of the city, so as to analyse whether, the orientation with regards to the regimes of air flows, and the distinct urban morphologies influence the concentration levels of indoor particles. In addition, an outside sampling station was located on the terrace of a building in the centre of the city to obtain information of PM10 in ambient air.

The three schools in the town of Alcora share as a principle feature, their greater proximity to an important industrial belt. As in the case of the urban schools, three primary schools were chosen: S4, S5 and S6, in different points with differing orientations and surroundings. In addition, an outdoor sampling station was located on the terrace of a building in the centre of the town. In the municipality of Lucena (rural area), a village with a low density of traffic and a low concentration of industry, the school selected was S7. In this case, outdoor sampling point was located on the terrace of this school.

Table 1 summarizes main characteristics of the seven schools studied. In all schools, a natural ventilation of classrooms is performed.

### 2. Materials and methods

#### 2.1. Site description

This study concentrates on a strip of land in the province of Castellón (Spain) that ranges from its capital, located on the Plana de Castellón where there are no natural barriers, passing through the locality of Alcora situated on the slope of a mountain, and ending in the village of Lucena located in the mountainous interior (Fig. 1). These localities (Castellón, Alcora and Lucena) represent three different environments: urban, industrial and rural respectively.

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#### 2.2. Instruments and method

The sampling period spans from one year (July and August excluded). Samples were taken for duration of 5–8 hours depending on the school hours to obtain a monthly sample for each of the schools.

The indoor airborne samples were collected using RespiCon TM. This collector is a multi-stage, virtual impactor that traps airborne particles on three individual collection filters. At a flow rate of 3.11 L/min., the first stage separates out and collects the particles that are smaller than 2.5 μm. The second stage collects particles between 2.5 and 10 μm, while the third stage collects the remaining particles. Quartz fibre filters (37 mm diameter) were used to collect indoor particles.

In the three outdoor sampling stations, the same data capture set was used; it is a minivol air sampler type 3.1 LVS of Derenda. This device is considered as a referent according to the European regulations on the particle sampling PM10 (UNE 12341:1999) and it allows the daily collection of the PM10 particle concentration. This set provides data on the aspirate volume and on the temperature, pressure and average relative humidity of those registered during the 24 hours of the sampling.
Fig. 1. Location of the studied area.
The aspirate air (2.3 m³/h) by using a pump goes through a cutting head which allows under 10 μm particles passing through. The particulate remains in quartz fibre filters with a diameter of 47 mm.

Indoor and outdoor samples from all locations were subjected to gravimetric analysis to determine the mass of PM10 analysis. Before the sampling and after being collected, the filters must be kept under conditions of a temperature of 20 ± 1 °C and controlled humidity (50 ± 5) % for 48 hours, after being weighted. Filters were weighed pre- and post-centrations are shown in.

Table 1
Main characteristics of the schools.

| School | Site | City zone | Traffic density | Classroom volume (m³) | Orientation of windows | Number of students |
|--------|------|-----------|----------------|-----------------------|-----------------------|--------------------|
| S1     | Urban | E         | High           | 268.03                | WNW                   | 40                 |
| S2     | Urban | NW        | Medium         | 159.56                | SIE                   | 21                 |
| S3     | Urban | W         | High           | 173.49                | ESE                   | 20                 |
| S4     | Industrial | SE       | Medium         | 36.17                 | WNW                   | 25-30              |
| S5     | Industrial | E       | Medium         | 109.09                | ENNE                  | 26                 |
| S6     | Industrial | SW      | Low-Medium     | 97.67                 | SIE                   | 60                 |
| S7     | Rural  | SE        | Low            | 182.60                | SE                    | 8-12               |

Table 2
Average of PM10 concentrations obtained and I/O ratios.

| School | Site | Indoor PM10 conc. calculated (μg/m³) | Outdoor PM10 conc. (μg/m³) | I/O ratio | I/O ratio unoccupied |
|--------|------|--------------------------------------|-----------------------------|-----------|----------------------|
|        |      | (min-max)                            |                             |           |                      |
| S1     | Urban | 132 [98-161]                         | 53 [36-77]                  | 2.6       | 2.6                  |
| S2     | Urban | 114 [65-140]                         | 54 [36-79]                  | 2.3       | 2.3                  |
| S3     | Urban | 141 [94-186]                         | 59 [31-100]                 | 2.6       | 2.6                  |
| S4     | Industrial | 69 [21-102]  | 53 [37-73]                  | 1.3       | 1.3                  |
| S5     | Industrial | 261 [205-322] | 36 [19-53]                  | 7.8       | 7.8                  |
| S6     | Industrial | 145 [97-183]  | 45 [37-65]                  | 3.5       | 3.5                  |
| S7     | Rural  | 71 [16-169]                          | 36 [16-55]                  | 2.1       | 2.1                  |

3. Results and discussion

3.1. Average concentrations and ratios I/O of PM10

A study of equivalence between the two samplers was carried out using the measurements (TM Respicom inside and INDO-1VS, outside), and were estimated the PM10 levels within the equivalence function that relates the values obtained both cases (UNE 12341-1999).

The averages of the ratios I/O obtained in each school were calculated (Table 2), for this purpose had not been taken into account the samples that had not been captured in the same conditions, as those which were collected in non-teaching periods and the sample that was taken during the construction works outside the classroom.

Table 2 shows that in all schools the ratio I/O for PM10 was greater than unity, indicating that there were significant indoor sources of these particles (Zhu et al., 2005). However, this ratio needs to be investigated further. The ratio I/O could be greater then unity also for a transport event from outside towards inside environment and not only for particle indoor sources.

The lowest ratio I/O was obtained for the S4 school. This ratio close to 1 is because the location is constantly exchanging air from outside, which causes the concentrations obtained are similar inside and outside. If a window is open the ratio I/O is close to 1 for all particle sizes (Zhu et al., 2005). Perhaps, the fact that school facilities are situated on the first floor makes the access to the class is not straight from the street and thus the number of particles introduced by shoes is less. And finally it should be noted that this is a nursery school (not all children walk and do not present the same activity as students of an infant school) and thus the effect of resuspension of particles is minimized at this location.

On the other hand, must be stated that S5 school had the highest rate I/O for PM10 of all locations. These high concentrations were associated with school characteristics, such as the orientation and morphology of the streets surrounding it. This school is located next to an unpaved park that all students of children entering the centre have to cross, which significantly increases the contribution of particles due to transport through the shoes and clothing (Molnár et al., 2007). Then these particles are resuspended and an increase of the concentration is produced. Yang et al. (2009) in their study in different schools in Korea obtained average ratios I/O of 2.06 for PM10 in classrooms and Stranger et al. (2008), found ratios of up to 8.8 for PM2.5. The results are explained in both cases by the student activities that cause resuspension of particles. In S5 school also produces a large accumulation. In the study area there is a large effect of particle accumulation indoor and in ambient air mainly due to climatic characteristics (Hesak et al., 2001; Gómez et al., 2005). There are not many days during the year in which the wind speed is significant, i.e. days in which there is an effective exchange of indoor air by natural ventilation (type of ventilation used by all schools). The dominant regimes of winds in the study area are breezes which are insufficient at the
time for renewing the air inside a building.

The other schools had ratios I/O of PM10 from 2.1 (rural environment) and 3.5 (industrial environment, S6). In all three schools situated in Castellón (urban environment) similar ratios were obtained, but in the school S2 was slightly lower because this location is less influenced by traffic.

In some studies conducted in urban areas has been demonstrated the influence the orientation, location and configuration of the streets with respect to emission sources on the vertical dispersion of pollutants (Ruin et al., 1996). In confined spaces, a greater vertical drop in large-sized particles is produced which causes a progressive enrichment in fine particles (Zhu et al., 2005). In open spaces there is a greater variation in vertical profiles and concentration of particles and their distribution is highly influenced by winds as sea breezes, and the distance and intensity of anthropogenic activities (Ruin et al., 1996; Zhu et al., 2005).

### 3.2. Ratio I/O of PM10 unoccupied classroom

In the three schools located in the industrial environment (S4, S5 and S6) were collected PM10 samples inside and outside in non-teaching periods. The ratios I/O for PM10 collected in schools S4, S5 and S6 under these conditions were 3.4, 1.9 and 2.3 respectively.

Comparing the values of I/O when the classrooms were unoccupied with respect to the average value of these same schools when the classrooms were occupied (Table 2), a decrease in the ratio I/O was observed when there are not children in S5 and S6 schools, especially in S5. This highlights the importance of the activities of the occupants in indoor ambient. In various studies (Blondeau et al., 2005; Poupard et al., 2005) was found that concentrations of particles inside showed peak in the hours that rooms were occupied due to particle generation by occupants themselves, both by the contribution from the outside, caused by the activities of the school (such as dust from the blackboard) or by the resuspension of previously deposited particles.

However, in S4 school, the ratio I/O obtained in non-teaching periods was greater than the overall average obtained when the classrooms are occupied (Table 2). This is due to an important change in conditions, since in this case windows are completely closed, so there is no air exchange as occurs in other samples, resulting in an accumulation in the interior that causes an increase in the ratio I/O.

### 3.3. Average concentrations and indoor to outdoor ratios of PM10 chemical composition

Table 4 shows the descriptive statistics of the concentration values of the chemical elements and anions, analyzed in PM10, that were obtained inside the seven schools (S1, S2, S3, S4, S5, S6 and S7) and outdoor of the three types of location (urban, industrial and rural).

As for the concentration values obtained outside the locations, the highest outside levels of B were obtained in the rural location, and the higher Pb concentrations in industrial one. The urban station reported the highest concentrations of Ni, Zn, Mg, Sb, ClO2 and Cl−. The rest of elements and anions analyzed showed similar values in at least two types of location. Similar Cd concentrations were obtained in the rural and urban environment, of Al and NO3 in the industrial and urban areas and similar levels of As, F− and SO4 in three types of location. However, the concentrations recorded inside the schools did not follow the same trend than those obtained outdoor.

The values of ratio I/O obtained for each of the elements and anions (analyzed in PM10) of the seven schools are shown in Table 5. The levels found in the interior were higher in all cases. The ratios I/O allow to have...
an idea of enrichment due to both processes of accumulation as generation by indoor sources.

Besides, Table 5 shows that the ratios I/O calculated for the chemical elements and anions, analyzed in the PM10, were higher than unity and generally higher than those recorded in the case of PM10 (Table 2). PM10 particles generally have high concentrations of some trace elements, however, some emissions sources are highly enriched in toxic metals without causing thereby an increase in the levels of PM10 (Bilos et al., 2001). The ratios I/O of metals and anions were varied, but highlighted the high values obtained in the case of two of the elements, Cd and B. The metal concentrations in the interior are often greater than the outside due to selective enrichment of metal-rich fine particles during cleaning processes or due to an internal source (Paustenbach et al., 1997). In their work, Rasmussen et al. (2001) found that concentrations of Pb varied with the type of heating. These results suggest that the method of home heating has an effect of accumulation of metals in the particulate, possibly because of differences in air circulation and the use of particulate filters.

Table 5 shows that the ratios I/O of Zn and fluoride were similar in the seven schools studied. In the S7 school, located in rural area, were registered the highest ratios of chloride and nitrate anions. In S5 and S6 schools (industrial area) were obtained the highest ratios I/O for chloride and sulphate registered outside schools. The values obtained were 0.02 and 0.09 respectively. These indicated that the levels of these elements and anions registered in ambient air were not related, i.e. do not exhibit the same behaviour. Therefore, the high correlation that is registered inside the schools is because the most important sources of these elements and anions are indoor sources.

Moreover, Pearson correlation coefficients were calculated for these same elements and anions obtained in outside locations to see if they have the same behaviour that inside of schools. Table 6 presents a summary of the results obtained from applying this correlation coefficient for concentrations of elements and anions analyzed in the PM10 captured inside and outside schools.

The highest correlations obtained inside schools correspond to those recorded between the concentrations of Cd and Pb (r = 0.88), Mg and Al (r = 0.89) and those of chloride and sulphate (r = 0.72). These high and positive correlations indicate the existence of common emission sources among them, which are of great importance in the concentration levels obtained indoor.

The Pearson correlation coefficients were calculated for the concentrations of Cd and Pb, and of chloride and sulphate registered outside schools. The values obtained were 0.02 and 0.09 respectively. These indicated that the levels of these elements and anions registered in ambient air were not related, i.e. do not exhibit the same behaviour. Therefore, the high correlation that is registered inside the schools is because the most important sources of these elements and anions are indoor sources.

A potential source of cadmium and lead in the interior is the gas or coal heating (Meyer et al., 1999; Komarnicki, 2005), although these elements can be part of painting in old buildings (Meyer et al., 1999), and other classroom materials. The origin of chloride and sulphate within schools is mainly associated with the cleanup of the classroom, as these anions are part of the composition of some of the cleaning products used in these tasks.

On the other hand, Pearson correlation coefficient obtained between the Mg and Al in outdoor locations was 0.79 (Table 6). This showed that both inside and outside are related positively and have a common emission source of importance. Mg and Al have a mineral origin, both natural and anthropogenic. These are all part of the composition of mineral raw materials used in the ceramic industry, such as clays and dolomites (Gómez et al., 2001; Sanfelìu et al., 2002) but in turn have a natural origin in the study area as part of the soil that may be resuspended by wind or other factors such as vehicle traffic.

In addition, the As showed significant correlations (r > 0.60) with the Ni, Al and Mg, and Ni also showed it with Al. Good correlations between these elements were also obtained outdoor. This is due to these elements, although are parts of the composition of different type of raw materials, are all used in the ceramics sector, and therefore have a common origin. Also, as previously said in this subsection, these elements were those with a higher correlation with PM10, indicating the existence of potential sources of common importance among these elements. Their origin (elements and PM10) was associated atmospheric particulate emissions related to the use of raw materials ceramics.

On the other hand, a significant correlation between the anions nitrate and sulphate was detected, both inside and outside locations. This relationship is associated with an important common origin, combustion processes of fossil fuels, such as traffic and natural gas combustion in industrial skills or heating. In this case these anions are not related to concentrations of PM10 in the interior.

### 4. Conclusions

PM10 concentrations and the levels of the chemical elements and anions analyzed in this fraction of particulate were greater in all cases inside schools than outside. This indicates the presence of indoor emission sources of importance.

Chemical elements and anions, in general, were more enriched in the interior of the buildings than the PM10 fraction. This is associated with selective increase of metal-rich fine particles during the cleaning process or due to an internal source and the existence of some emissions sources that have highly enriched in toxic metals without causing thereby an increase in the levels of PM10.

The calculation of Pearson correlation coefficients between the

| Ratio I/O | S1  | S2  | S3  | S4  | S5  | S6  | S7  |
|----------|-----|-----|-----|-----|-----|-----|-----|
| As       | 4.9 | 7.7 | 4.6 | 3.0 | 4.9 | 2.6 | 4.3 |
| Ni       | 10.7| 13.5| 11.6| 10.9| 16.0| 10.8| 13.8|
| Cd       | 245.1| 217.1| 171.3| 190.0| 232.1| 252.3| 167.1|
| Pb       | 23.3| 20.2| 12.0| 5.4 | 6.8 | 7.2 | 18.1|
| Al       | 8.4 | 14.2| 11.3| 11.0| 12.4| 7.0 | 9.9 |
| B        | 161.4| 125.9| 148.5| 120.9| 144.9| 116.1| 125.7|
| Zn       | 10.7| 9.9 | 11.8| 13.2| 12.4| 10.1| 9.6 |
| Mg       | 4.2 | 5.1 | 5.3 | 6.6 | 11.7| 4.3 | 6.0 |
| Sb       | 1.9 | 2.1 | 1.7 | 4.9 | 3.3 | 3.5 | 3.0 |
| Fluoride | 19.6| 19.8| 19.7| 18.5| 21.1| 20.8| 21.7|
| Chloride | 23.1| 24.6| 27.4| 48.5| 67.4| 65.7| 49.8|
| Chloride | 8.3 | 10.0| 15.1| 14.4| 16.9| 19.5| 22.0|
| Nitrate  | 6.8 | 6.7 | 9.3 | 6.2 | 11.2| 10.0| 13.9|
| Sulphate | 4.7 | 6.8 | 8.4 | 7.6 | 11.6| 11.5| 8.9 |
Table 6
Correlations (Pearson coefficient) between the concentrations of elements and anions analyzed in the PM10 captured indoor and outdoor.

| Indoor Correlations | As | Cd | Pb | Al | B | Zn | Mg | Sb | Fluoride | Chlorite | Chloride | Nitrate | Sulphate |
|---------------------|----|----|----|----|---|----|----|----|-----------|-----------|-----------|---------|----------|
| As                  | 0.68** | -0.18 | -0.25 | 0.64** | -0.14 | 0.17 | 0.66** | 0.13 | 0.20 | 0.20 | 0.34** | 0.15 | 0.18 |
| Cd                  | 0.52** | -0.37 | -0.37** | 0.63** | -0.50** | -0.02 | 0.55** | -0.07 | 0.29* | 0.34* | 0.22 | 0.27* | 0.17 |
| Pb                  | 0.29 | 0.12 | 0.88** | -0.27* | 0.52** | -0.19 | 0.05 | 0.06 | 0.20 | 0.07 | -0.21 | 0.20 | -0.14 |
| Al                  | 0.36** | 0.37** | 0.14 | 0.23* | -0.30* | 0.36** | 0.89** | -0.28** | 0.06 | 0.20 | -0.05 | -0.21 | 0.40 |
| B                   | 0.27* | 0.01 | 0.22 | 0.20 | 0.46** | 0.53** | -0.18 | 0.36** | 0.20 | -0.15 | 0.17 | -0.12 | 0.10 |
| Zn                  | 0.66** | 0.25* | 0.44** | 0.24* | 0.29* | 0.59** | 0.35** | 0.08 | 0.19 | 0.09 | -0.29* | -0.18 |
| Mg                  | 0.45** | 0.36** | 0.45** | 0.20 | 0.79** | 0.26* | 0.47** | 0.33* | 0.28* | 0.32** | 0.72** |
| Chloride            | 0.12 | 0.24* | 0.36* | 0.14 | 0.02 | -0.31** | -0.11 | 0.37** | 0.04 | 0.36** | 0.41** | 0.47** |
| Nitrate             | 0.04 | 0.18 | -0.06 | 0.02 | -0.06 | -0.02 | 0.07 | 0.01 | 0.18 | -0.17 | 0.15 | 0.66** |
| Sulphate            | 0.32* | 0.45** | 0.10 | 0.19 | -0.02 | 0.22 | 0.23* | 0.03 | 0.24* | -0.12 | 0.09 | 0.52** |

| Outdoor Correlations | As | Cd | Pb | Al | B | Zn | Mg | Sb | Fluoride | Chlorite | Chloride | Nitrate | Sulphate |
|----------------------|----|----|----|----|---|----|----|----|-----------|-----------|-----------|---------|----------|
| As                   | 0.52 | -0.18 | -0.25 | 0.64 | -0.14 | 0.17 | 0.66 | 0.13 | 0.20 | 0.20 | 0.34 | 0.15 | 0.18 |
| Cd                   | 0.52 | 0.01 | 0.22 | 0.20 | 0.46 | 0.53 | -0.18 | 0.36 | 0.20 | -0.15 | 0.17 | -0.12 | 0.10 |
| Pb                   | 0.29 | 0.12 | 0.88 | -0.27 | 0.52 | -0.19 | 0.05 | 0.06 | 0.20 | 0.07 | -0.21 | 0.20 | -0.14 |
| Al                   | 0.36 | 0.37 | 0.14 | 0.23 | -0.30 | 0.36 | 0.89 | -0.28 | 0.06 | 0.20 | -0.05 | -0.21 | 0.40 |
| B                    | 0.27 | 0.01 | 0.22 | 0.20 | 0.46 | 0.53 | -0.18 | 0.36 | 0.20 | -0.15 | 0.17 | -0.12 | 0.10 |
| Zn                   | 0.66 | 0.25 | 0.44 | 0.24 | 0.29 | 0.59 | 0.35 | 0.08 | 0.19 | 0.09 | -0.29 | -0.18 |
| Mg                   | 0.45 | 0.36 | 0.45 | 0.20 | 0.79 | 0.26 | 0.47 | 0.33 | 0.28 | 0.32 | 0.72 | 0.66 |
| Chloride             | 0.12 | 0.24 | -0.06 | 0.02 | -0.06 | -0.02 | 0.07 | 0.01 | 0.18 | -0.17 | 0.15 | 0.66 |
| Nitrate              | 0.04 | 0.18 | -0.06 | 0.02 | -0.06 | -0.02 | 0.07 | 0.01 | 0.18 | -0.17 | 0.15 | 0.66 |
| Sulphate             | 0.32 | 0.45 | 0.10 | 0.19 | -0.02 | 0.22 | 0.23 | 0.03 | 0.24 | -0.12 | 0.09 | 0.52 |

* Significant correlation; ** Good correlation.

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