# **Elemental deposition in the vicinity of a lignite power plant in Southern Greece**

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Bulk deposition samples were collected on a monthly basis in Megalopolis city, located near lignite power plants. A total of 23 elements (As, Br, Ca, Ce, Co, Cr, Cs, Eu, Fe, Hf, La, Mo, Na, Rb, Sb, Sc, Se, Ta, Tb, Th, U, Yb, Zn) were detected in the collected samples using INAA. The coal burning marker elements As and Se were found enriched in the collected samples together with Mo, Br, Sb, U and Zn. Factor analysis permitted the identification of four sources: re-suspended soil/road dust, lignite power plant, sea spray and traffic emissions. A wind direction effect on deposition rates of all elements (except rates of Na, Rb, Sb and Zn) was also found.

### **Introduction**

Trace elements are released into the atmosphere both from natural and anthropogenic sources and are removed by a variety of cleaning processes including dry and wet deposition mechanisms.1–3

Coal is a major anthropogenic pollution source, since during combustion for energy production a number of potentially toxic elements are released into the environment. These emissions are made up of primary (flue gas and fine fly ash particles) and secondary (gasto-particle conversion) particulate emissions.4,5 Although electrostatic precipitators have high collection efficiency  $(\sim 99.95\%)$ , considerable amounts of fly ash particles, enriched in several toxic elements, are discharged into the environment, due to the high rate of coal consumption. The fly ash particles, because of their high atmospheric mobilities, are transported from their source over a wide range of distances and deposited on the ground.<sup>5</sup> In order to assess the environmental impact of a particular power plant, specific studies should be carried out, since coal quality, combustion technology and local meteorological conditions are important factors controlling the emission, transportation and deposition of pollutants.4,5 Several studies in different countries have shown the environmental impact of coalfired power plants in the surroundings.  $6-12$ 

The current study has been focused on bulk deposition chemistry in the city of Megalopolis in southern Greece, which is located about 2.5 and 4 km NE to the lignite power plants A and B, respectively and far from other major pollution sources. For this purpose, bulk deposition samples were collected on a monthly basis, over a one-year period (February 1997–January 1998), at four sampling sites in Megalopolis city and one in the area of the lignite power plant A. The specific objectives of this study were the determination of elemental concentrations in the collected samples using INAA and the identification and apportionment of pollutants to their sources. Enrichment factor

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calculations for all the detected elements and factor analysis were used. Also, wind sectors supplying particulate matter to the area of Megalopolis city were examined. The natural radioactivity content and the dust and elemental deposition rates in comparison with samples collected simultaneously in an urban city (Patras) were studied earlier.13,14 Although a number of studies have been undertaken concerning radioactivity released from the power plants in Megalopolis basin this work constitutes the first attempt to evaluate the trace element impact from a lignite power plant in the area of Megalopolis city.<sup>15,16</sup>

#### **Experimental**

#### *Study area and sample collection*

Megalopolis is a small city in the centre of Peloponnese peninsula, with 10,000 inhabitants (Fig. 1). The city is located about 2.5 and 4 km NE to the lignite power plants A (550 MW<sub>e</sub>) and B (300 MW<sub>e</sub>), respectively. The lignite power plants are operated by the Public Power Corporation of Greece. Description of the power plants is given elsewhere.<sup>13</sup> The lignite deposit of Megalopolis basin is of very low calorific value  $(960-1385 \text{ kcal} \cdot \text{kg}^{-1})$  and of high water and ash consent (60% and 17%, respectively). There are no other major industrial activities in the immediate vicinity of the city.

Bulk (wet  $+$  dry) deposition sampling was carried out at five sites in Megalopolis. Four of them (M1–M4) were located in the city of Megalopolis, at different distances from the power plant A (M4, M3, M2, M1 in increasing distance), while M5 was located in the area of the power plant A, specifically on the roof of the Administration Building. The locations of all the sampling sites are shown in Fig. 1. Samples were collected from the 1st of February 1997 to 31th of January 1998, on a monthly basis. All 60 samples for bulk deposition were collected successfully.

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*Fig. 1.* Map of Greece showing the location of Megalopolis. The bulk deposition sampling sites and the location of power plants A and B are shown in the amplification

The sampling procedure is described in detail elsewhere.13 Briefly, bulk deposition samples were collected in high-walled plastic (PVC) pots with 32.5 cm opening in diameter, which were located on the flat roof of two-story buildings.17 The pot was placed in an open area on the first of the month and left for one month. In order to avoid removal of the deposited particles, mainly by wind, a layer of 2 cm in height ultra pure water (4D) was constantly maintained at the bottom of the pot. At the end of the month, the water was transferred into polyethylene bottles, which were transported, together with the pot to the Radiochemistry Laboratory at the University of Patras. All the content of the pot and bottle was transferred to a porcelain dish and was concentrated, by evaporation at 75 °C in a water bath. The final evaporation, till dryness, was conducted in a small porcelain dish of known weight. All materials used for collecting and concentrating the deposition samples were decontaminated by soaking in  $10\%$  HNO<sub>2</sub> for at least 48 hours, followed by rinses, first with 2D

and second with 4D water. Special precautions had been taken regarding the sampling, the transportation of the sampling device, the sample handling and analysis steps in order to avoid any contamination, damage or removal of the deposited particles.

#### *Analysis of elemental composition*

The analysis of the collected samples has been performed by instrumental neutron activation analysis  $(INAA)$  combined with high-resolution  $\gamma$ -ray spectrometry. INAA has been widely applied in traceelement pollution studies because of its sensitivity and its capability for multielemetal non-destructive analysis. The samples and appropriate standards were irradiated using the rotation system of the research reactor of National Research Center for Physical Sciences DEMOCRITOS (Athens, Greece) at a thermal neutron flux of  $(3-4) \cdot 10^{13}$  n·cm<sup>-2</sup>·s<sup>-1</sup>. After irradiation the samples and standards were counted using a  $\gamma$ -

spectrometry setup consisting of a HPGe detector (efficiency 20%, and FWHM 1.69 keV for the 1332 kV  ${}^{60}Co$   $\gamma$ -ray), connected to a Canberra 35+4Kmultichannel analyzer. The trace element concentrations were determined using the comparative method. For this reason, the IAEA standard reference material Soil-7 and the United States Governmental Geochemical Standard GXR-1 were used for determinations and quality control procedures. Peak areas were determined using Canberra's Spectran computer code. The countings were performed at the Nuclear Technology Laboratory of the National Research Center for Physical Sciences (NCRP) 'DEMOKRITOS' (Athens, Greece).

### *Meteorological data*

Meteorological data were obtained from the meteorological station of the Public Power Corporation, which is located between the power plant A and the city of Megalopolis at a distance of 1.2–2 km from the plant.

## *Statistical treatment*

Normality tests showed that the data distributions of the examined variables were not normal. Thus, the logtransformed data were used for all statistical treatments. One way analysis of variance (ANOVA) was used in order to compare the mean elemental concentrations between the different sampling sites. The wind direction effect was studied using Student's *t*-test. Factor analysis with varimax rotation was applied to the elemental concentrations and the dust amount. Since the number of samples in relation to the detected elements was relatively small, the elements Ce, Eu, Hf, Rb and Yb were not included in the factor analysis, as these elements were highly correlated with the crustal elements (e.g., Fe, Sc, Co, Cs, Th). The results in both cases were the same, but the total variance explained was improved. Possible sources were assigned to the principal components. All calculations were carried out using the SPSS v.12 software.

## **Results and discussion**

## *Elemental concentrations*

Arithmetic mean, arithmetic standard deviation and geometric mean of the monthly measurements of dust amount and elemental concentrations deposited at all the sampling sites in Megalopolis during the one-year sampling period are presented in Table 1. A total of 23 major, minor and trace elements were detected. As can be seen from this table, the concentrations of all elements vary over relatively large ranges. This is

attributed mainly to weather conditions that strongly affect the dispersion and deposition of air dust, as well as to the fluctuation of emission sources over the seasons. The differences in mean values (GM) of dust amount between the five sampling sites were statistically significant (*F* = 7.032 on 4 and 59 *df*, *P*<0.001). On the other hand, the differences in mean elemental concentrations were not statistically significant for most of the analyzed elements, except for As  $(F=8.629)$  on 4 and 59 *df*, *P*<0.001), Ca (*F* = 7.829 on 4 and 59 *df*, *P*<0.001), Na (*F* = 2.656 on 4 and 59 *df*, *P* = 0.042), Mo  $(F=4.346$  on 4 and 59 *df*,  $P=0.004$ ), Se  $(F=4.077$  on 4 and 59 *df*, *P* = 0.006) and U (*F* = 5.294 on 4 and 59 *df*,  $P=0.001$ ). After multiple comparisons tests, all significant effects were attributed to the difference of site M5, from the sites M1–M4. The location of site M5 in the area of power plant A explains this difference. Summary statistics of the dust  $(g·m<sup>-2</sup>)$  and elemental deposition rates ( $\mu$ g·m<sup>-2</sup>) are presented elsewhere.<sup>14</sup> An important feature of the deposition data is the consistency with which they change with the distance from the power plant A and the increasing trend in the monthly deposition rate for dust and most elements with decreasing distance from power plant A. Moreover, the differences in mean elemental deposition rates between site M5 and sites M1–M4 were statistically significant for most of the analyzed elements. It should be also noted that the deposition rates of most elements detected in Megalopolis samples were significantly higher (except Br, Sb, Ca and Zn) in comparison with those measured in Patras samples during the same period.14

### *Influence of wind direction*

The consideration of elemental deposition rates in relation to wind direction may be helpful in the identification of emission sources of the detected elements. For this purpose, the monthly deposition values were classified according to the prevailing wind direction during the sampling period. As mentioned above, the city of Megalopolis is located NE to the lignite power plants. This means that the northwesterly flows in the region favor transport of pollutants emitted from plant A and plant B. According to the meteorological data (Table 2), the prevailing wind direction was NW during 2/97 and 4–8/97. A comparison between dust and elemental deposition rates coming from NW directions and all the other ones was done by Student's t-test (site M5 was not included in the analysis). The results showed a wind direction effect on the calculated deposition rates. The differences in mean values (GM) of NW directed elemental deposition rates were significantly higher than those from the other directions, except that of Na, Rb, Sb and Zn.

Table 1. Summary statistics for major, minor and trace element concentrations and dust amount in bulk deposition samples from Megalopolis during February 1997–January 1998



This means that the contribution of the lignite power plants to these species was higher than the average level and the relation of this source to the species was strong in this direction. On the contrary, no wind direction effect was found for site M5. This could be explained by the fact that there are open fly ash deposits around the power plant A. Consequently, winds from any direction transfer fly ash particles to site M5, which was located in the area of power plant A. Another important meteorological feature is the average low wind velocity during the sampling period. This feature might be also responsible for the accumulation of pollutants over the plants' area resulting in high concentrations of elements in bulk deposition.

#### *Source identification*

*Enrichment factors:* One way of establishing the principal sources of elements in bulk deposition samples is via the use of enrichment factor (*EF*). The concept of enrichment factor is based on the fact that some elements originating from well-defined sources can be distinguished from other elements derived from different sources. The *EF* value was calculated according to the following equation:

$$
EF = \frac{(X/C)_{sample}}{(X/C)_{reference}}
$$

where  $X$  is the concentration of the element of interest and *C* is the concentration of a reference element, which is predominately of natural origin and non-volatile. In this study, Sc was used as the reference crustal element for the derivation of enrichment factor. The *EF*s were calculated on the basis of Earth's crust mean abundance of the elements given by TAYLOR.<sup>18</sup> By convention, an *EF* of ten or greater is indicative that the source of the element is anthropogenic and not from the natural environment. Elements present in the atmosphere primarily due to natural processes, such as soil resuspension, generally have an *EF* close to one.19

Summary statistics of the calculated *EF* values of all elements detected in the collected samples at each site in Megalopolis during the sampling period are presented in Table 3. The *EF*s of all elements (except Zn) were of the same order of magnitude at all sites. All elements also follow exactly the same pattern. According to Table 3, the elements As, Br, Mo, Sb, Se and U behave as enriched at all five sites, which indicates that a

significant proportion of these elements has non-crustal source. Zinc behaves as enriched only at sites M1, M2 and M4. Site M5 presents the higher *EF* values for U, As, Mo and Se and the lower one for Sb. Selenium shows the highest *EF*s, ranging from 403 to 607 with the highest value at site M5. It is worth to note that Se and As are considered as coal burning marker elements.<sup>19</sup> All the other elements behave as non-enriched (*EF*<10), which means that they have a significant crustal source. Samples of lignite, bottom ash and fly ash from the lignite power plant A were also collected on May 1997. These samples were subjected to elemental analysis using INAA.<sup>20</sup> It should be noted that As, Br, Mo, Sb, Se and U were also enriched in lignite samples, while As, Mo, Sb, Se and U in fly ash samples. Selenium also presented higher *EF*s in lignite and fly ash samples (443 and 352, respectively). It is also worth to note that As, Se, Sb and U are between the 189 substances cited as potentially hazardous air pollutants.21

*Factor analysis:* In a further attempt to assess the major sources affecting the elemental composition of the bulk deposition samples, factor analysis with Varimax rotation was applied to our data. Sixteen elements (As, Br, Ca, Co, Cr, Cs, Fe, La, Mo, Na, Sb, Sc, Se, Th, U, Zn) and the dust amount were considered for the construction of the matrix (17×60). Four factors were retained, which were also the only ones with eigenvalues higher than unity. They account for 82.0% of the variance in the data set. The values of Varimax rotated loadings for the 4 factors, as well as the percentage of the explained variance are given in Table 4. Factor loadings higher than 0.350 were used in order to interpret the sources associated with each factor.

*Table 2*. Meteorological data concerning the sampling sites of Megalopolis

Month	Mean velocity, $m s^{-1}$	Prevailing wind direction
2/97	1.6	NW
3/97	2.1	E
4/97	2.2	NW
5/97	1.8	NW
6/97	1.8	NW
7/97	2.3	<b>NW</b>
8/97	2.1	NW
9/97	1.6	<b>ESE</b>
10/97	1.6	Е
11/97	1.4	<b>SE</b>
12/97	1.6	<b>ESE</b>
1/98	1.4	<b>ESE</b>

Element	M1	M <sub>2</sub>	M <sub>3</sub>	M <sub>4</sub>	M <sub>5</sub>
	$AM \pm SD$	$AM \pm SD$	$AM \pm SD$	$AM \pm SD$	$AM \pm SD$
As	$12 \pm 4$	$11 \pm 3$	$11 \pm 3$	$10 \pm 3$	$16 \pm 4$
Br	$54 \pm 35$	$44 \pm 24$	$46 \pm 29$	$43 \pm 28$	$50 \pm 34$
Ca	$4.5 \pm 1.9$	$3.3 \pm 0.8$	$3.8 \pm 1.3$	$3.2 \pm 0.6$	$2.5 \pm 0.4$
Ce	$1.7 \pm 0.3$	$1.7 \pm 0.4$	$1.7 \pm 0.3$	$1.7 \pm 0.4$	$1.7 \pm 0.4$
Co	$1.5 \pm 0.1$	$1.5 \pm 0.1$	$1.5 \pm 0.1$	$1.5 \pm 0.1$	$1.5 \pm 0.1$
Cr	$3.8 \pm 0.8$	$3.6 \pm 0.8$	$3.6 + 1.1$	$3.4 \pm 0.6$	$3.2 \pm 0.9$
Cs	$2.7 \pm 0.1$	$2.8 \pm 0.1$	$2.7 \pm 0.2$	$2.7 \pm 0.1$	$2.7 \pm 0.1$
Eu	$1.5 \pm 0.3$	$1.6 \pm 0.2$	$1.5 \pm 0.1$	$1.6 \pm 0.1$	$1.6 \pm 0.2$
Fe	$1.6 \pm 0.4$	$1.3 \pm 0.1$	$1.3 \pm 0.1$	$1.3 \pm 0.1$	$1.5 \pm 0.1$
Hf	$2.1 \pm 0.2$	$2.2 \pm 0.4$	$2.3 \pm 0.3$	$2.1 \pm 0.3$	$1.9 \pm 0.2$
La	$1.8 \pm 0.1$	$1.8 \pm 0.1$	$1.8 \pm 0.1$	$1.7 + 0.1$	$1.7 \pm 0.1$
Mo	$46 \pm 18$	$53 \pm 17$	$52 \pm 18$	$53 \pm 19$	$67 \pm 22$
Na	$1.1 \pm 1.0$	$0.7 \pm 0.3$	$0.6 + 0.4$	$0.7 \pm 0.4$	$0.4 + 0.2$
Rb	$1.6 \pm 0.4$	$1.4 \pm 0.2$	$1.4 \pm 0.2$	$1.4 \pm 0.2$	$1.4 \pm 0.4$
Sb	$96 \pm 106$	$56 \pm 48$	$97 \pm 151$	$39 \pm 38$	$35 \pm 24$
Se	$433 \pm 159$	$438 \pm 99$	$437 \pm 195$	$403 \pm 141$	$607 \pm 233$
Ta	$0.7 \pm 0.2$	$0.7 \pm 0.2$	$0.7 \pm 0.2$	$0.6 \pm 0.2$	$0.6 \pm 0.2$
Th	$1.6 \pm 0.1$	$1.6 \pm 0.1$	$1.6 + 0.1$	$1.6 \pm 0.1$	$1.6 \pm 0.1$
U	$22 \pm 4$	$22 \pm 3$	$23 \pm 3$	$23 \pm 4$	$30 \pm 3$
Yb	$1.3 \pm 0.2$	$1.4 \pm 0.2$	$1.4 \pm 0.2$	$1.3 \pm 0.2$	$1.3 \pm 0.1$
Zn	$10 \pm 5$	$10 \pm 4$	$7.8 \pm 3.8$	$11 \pm 9$	$7.5 \pm 3.1$

*Table 3*. Summary statistics of the *EF* values calculated for all the detected elements in deposition samples at the five sampling sites in Megalopolis during February 1997–January 1998

*Table 4*. Varimax rotated factor loading matrix for bulk deposition samples collected at the five sampling sites in Megalopolis area, from February 1997 to January 2008

Variables	Factor 1	Factor 2	Factor 3	Factor 4
Dust	$-0.065$	0.369	$-0.348$	$-0.689$
As	0.315	0.842	$-0.113$	$-0.158$
Br	$-0.150$	0.723	0.008	0.548
Ca	$-0.185$	$-0.290$	0.192	0.192
Co	0.968	0.148	0.051	$-0.038$
Cr	0.547	0.110	0.735	$-0.081$
Cs	0.968	0.031	$-0.054$	$-0.016$
Fe	0.867	0.233	0.144	0.022
La	0.955	$-0.035$	$-0.054$	$-0.100$
Mo	0.257	0.561	$-0.569$	0.036
Na	$-0.143$	0.147	0.071	0.887
Sb	$-0.297$	0.442	0.655	0.258
Sc	0.989	0.028	$-0.066$	$-0.056$
Se	0.083	0.917	0.034	$-0.025$
Th	0.960	$-0.070$	$-0.002$	$-0.143$
U	0.844	0.244	$-0.328$	$-0.056$
Zn	$-0.037$	$-0.011$	0.440	0.122
Variance, %:	42.2	18.1	11.0	10.7

Factor 1 explains about 42.2% of the total variance and is dominated by crustal components, since it shows high loadings for the crustal elements Co, Cs, Fe, La, Sc, Th and U and may suggest the dominant contribution of re-suspended soil/road dust. This statement is also supported by the results of Table 3 where the mean *EF*s for Co, Cs, Fe, La, Sc, Th (except U) show crustal origin. Associated with this factor is Cr, which has also *EF*<10. This factor has been identified as " re-suspended soil/road dust factor".

Factor 2 has high loadings for As and Se together with moderate loadings for Br and Mo. This factor also appears to have weak loadings for Sb and dust. It explains about 18.1% of the total variance. It should be pointed out that the profile of this factor has chemical composition quite similar with particles of fly ash resulted from the lignite combustion. This is true with regard to the *EF*s of fly ash and deposition samples. All these elements have *EF*s>10 both in deposition and fly ash samples and are of anthropogenic origin.<sup>21</sup> Bromine is enriched in lignite and not in fly ash samples because it goes off as a gas, but it is deposited as secondary pollutant (gas-to-particle conversion).4,21 In addition, Se and As are considered as coal burning marker elements.19 The weak loading for dust of this factor means that a considerable amount of the deposited dust is dominated by fly ash particles escaping from the stacks of the lignite power plants transported to Megalopolis city. This trend is also supported by the wind direction effect mentioned above. This factor was attributed to the lignite combustion products and has been identified as "fly ash factor".

Factor 3 accounts for 11% of the total variance and is dominated by anthropogenic emissions other than lignite power plants. It is a bipolar factor. It presents relatively high positive loading for Cr and weak for Sb and Zn. It also presents moderate negative loadings for Mo and dust. The *EF*s of Sb and Zn are >10 in deposition samples while that of Cr<10. As non-catalytic cars are not used any more, Zn and Sb are considered as marker

elements of motor vehicle emissions. Zn is the predominant element emitted from car tires and Sb from break abrasion.22 The correlation of this factor with Cr is not easily explained, as Cr is mainly product of metallurgical processes. Since Factor 1 is also correlated with Cr, it seems that Cr is contributed both by crustal and anthropogenic sources. This factor mainly represents a "vehicle emissions factor".

Factor 4 is also characterized as a bipolar factor and explains 10.7% of the total variance. The positive pole has high loading for Na and moderate for Br. The negative pole has a moderate loading for dust. Sodium has *EF*<10, while Br>10. As stated above, Br is a coalburning product. Bromine is also a marker element of marine aerosols. The strong correlation of Br with Na (Pearson  $r = 0.558$ , significant at the 0.01 level) and their consistent temporal pattern in our data show that probably a part of Br originates from the same source of Na, that of sea spray. The marine contribution to this factor might be explained by the long distance atmospheric transport of sea spray to the sampling sites, since Megalopolis is located about 40 and 60 km from the Ionian and Aegean Sea, respectively.<sup>9</sup> This could also explain the negative loading of this factor for dust amount. This factor has been identified as "sea spray factor".

It is apparent that the results of factor analysis were consistent with the results of *EF* calculations. These combined results suggest that the lignite power plants' emissions, together with re-suspended soil/dust, vehicle emissions and sea spray may control the elemental concentration levels in bulk deposition samples collected in the studied area.

## **Conclusions**

INAA technique was used in the elemental characterization of bulk deposition samples collected in Megalopolis city located NE to the lignite power plants.

After enrichment factor calculations the elements As, Br, Mo, Se, Sb, U and Zn were identified as enriched elements in the collected samples. Selenium, which is considered as coal combustion marker element, had very high values of *EF* (403–607). The elements As, Mo, Sb, Se and U were also found to be enriched in fly ash samples collected from the power plant A.

An evaluation of the elemental deposition rates with respect to meteorological interactions showed all elemental deposition rates at Megalopolis city sites (except rates of Na, Rb, Sb and Zn), directly influenced by NW directed emissions.

The results of factor analysis further indicated that there are four factors with statistical significance that explain 82% of the total variance: re-suspended soil/road dust, burning of lignite, vehicular emissions and sea salt.

Further studies should be undertaken, especially on PM10 and PM2.5 particulate matter, for assessing the potential hazard of anthropogenic sources mainly that of lignite power plant to the local population.

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