

Multivariate statistics and spatial interpretation of geochemical data for assessing soil contamination by potentially toxic elements in the mining area of Stratoni, north Greece

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ABSTRACT: A preliminary soil survey was undertaken in the mining region of Stratoni in Chalkidiki peninsula, north Greece. The objectives of the study were to assess the levels of soil contamination with respect to average concentrations of toxic elements in the region, to determine the associations between the different chemical elements and their spatial distribution, and to identify possible sources of contamination that can explain the spatial patterns of soil pollution in the area. Forty-nine surface soil samples were collected and analysed by inductively coupled plasma-atomic emission spectroscopy after digestion with a mixture of HClO₄-HNO₃-HCl-HF. The study focused on ten elements (Pb, Zn, Cu, As, Cd, Mn, Fe, Ni, Co, Cr), all of which were present in soil with concentrations well above the global soil means. Lead, As and Ni, with means of 895 µg g⁻¹, 364 µg g⁻¹ and 161 µg g⁻¹ respectively, exceeded the tentative trigger concentration set by the Netherlands and the UK Interdepartmental Committee on the Redevelopment of Contaminated Land (ICRCL) regulations. Factor analysis explained 89% of the total variance of the data through four factors. Combined with spatial interpretation of its output, the method successfully grouped the elements according to their sources and provided evidence about their geogenic or anthropogenic origin.

KEYWORDS: soil pollution, factor analysis, spatial interpretation, Stratoni, Greece

INTRODUCTION

Soil pollution by toxic chemical elements in mining areas is a widespread and significant problem. Soil is a medium of interaction between the atmosphere, the biosphere and the lithosphere, and the presence of toxic elements in soil can be harmful to plants, animals and humans, under certain circumstances. The magnitude of hazard caused by a certain chemical element depends on the concentration, the chemical form, the particle size, the soil or water pH and the extent of exposure to the element.

Mining areas are characterized by high concentrations of potentially harmful elements in soils, related not only to weathering of naturally enriched parent rocks and pedogenesis, but also to human activities of mining and metal processing. The identification of sources of soil pollution by toxic elements in such instances is important because the localized disturbance of the ground through human influence, for example at mine waste tips, shafts and metal processing plants, may cause increased dispersion and exposure of contaminants derived from natural sources (Appleton 1995). This in turn has financial, legal and health implications and affects decisions for remedial action.

A range of methods has been devised for discrimination between anthropogenic contamination and natural concentrations of toxic elements. Many of these methods employ detailed physico-chemical analysis such as partial and sequential

extractions (Li & Thornton 2001), study of variation of stable isotopes (Santos *et al.* 1993) and mineralogical studies (Boulet & Larocque 1998). Perhaps a more rapid and cheaper alternative for assessment of contamination sources can be based on multivariate statistical analysis of the data. Multivariate statistics have been used to assess soil contamination by toxic elements in areas of raised background concentrations (Kelepertsis *et al.* 1982, 2001). Davies (1997) used a combination of factor analysis and isoline plots to differentiate sources of pollution in an old industrial area of Wales. This approach has been further enhanced in recent years by the use of geographical information systems (GIS) that allow the integration of spatial databases. Examples of studies employing GIS for identifying metal pollution sources are available in the literature (Facchinelli *et al.* 2001; Ramsey *et al.* 1994).

This approach of combining multivariate statistical geochemical data with spatial information is adopted in the present study, which is based on a preliminary soil survey of the Stratoni area in north Greece. The long mining history of the area combined with the already raised natural geochemical background with respect to potentially harmful elemental concentrations and the variety of ore types that characterize the region, make it a very interesting case for environmental geochemistry studies that seek to identify sources of pollution. The objectives of this paper were: (1) to assess the levels of soil contamination in respect to average concentrations of toxic elements in the Stratoni mining area in North Greece; (2) to determine the

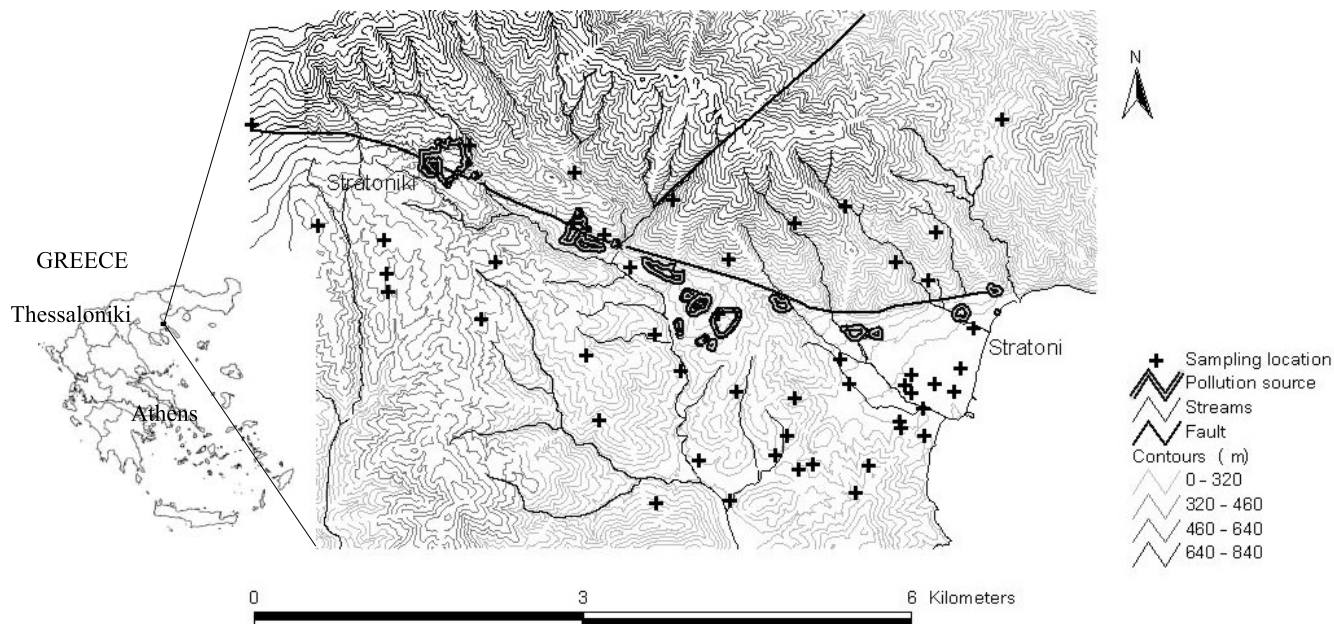


Fig. 1. Topographic map of the Stratoni area with pollution sources and sample locations.

associations between the different toxic elements and their spatial distribution; and (3) to identify possible sources of contamination that can explain the spatial patterns of soil pollution in the area.

STUDY AREA

Geographical location and land use

The Stratoni area is situated in the Chalkidiki Peninsula, northern Greece (Fig. 1). The study area lies between latitudes $40^{\circ}30'$ and $40^{\circ}32.5'$ and longitudes $23^{\circ}45'$ and $23^{\circ}32.5'$. It is characterized by mountainous relief with 92% of the surface covered by forest. Although the total area is classified as mining concessions it is adjacent in the north to a European Union protected area of NATURA 2000. The three main towns of the area (Stratoni on the coast, Stratoniki and Stagira further inland to the west) have a history closely linked to mining activities, the consequences of which in terms of environmental pollution have raised public concern in recent years.

Geology and mineralization

The rocks of the area belong to the Serbo-Macedonian massif which is composed of metamorphic rocks of the Kerdilion and the Vertiskos tectonic units (Pe-Piper & Piper 2002). The two units are in contact through the Stratoni–Varvara fault. Rocks of the Kerdilion sequence comprise variable proportions of biotite gneiss, biotite–hornblende gneiss, microcline–plagioclase gneiss, marbles and amphibolites, and locally show migmatitic textures. The overlying Vertiskos sequence consists largely of biotite–muscovite gneiss (Pe-Piper & Piper 2002; Barbieri *et al.* 1995; Kalogeropoulos & Economou 1987). Between the two sequences are lenses of amphibolite–metagabbro complexes interpreted as ophiolites. Sapountzis *et al.* (1990) have described small amphibolites of mafic protolith within the Kerdilion sequence at Stratoni.

The metamorphic rocks of the Vertiskos sequence have been intruded by calc-alkaline post-orogenic granites of Tertiary age: the Stratoni granodiorite and the quartz-diorite porphyry of Fisoka. These intrusive bodies are related to several ore deposits

in the area (Arvanitidis *et al.* 1994). Starting *c.* 10 km west of Stratoni village, a Pb–Zn–Mn deposit is present and exploited in the Varvara Mine. Moving eastwards along the Stratoni–Varvara fault, Pb–Zn sulphide ore forms within the marble as well as along the contact between the gneiss and the lower marble horizon. Major deposits of this type have been exploited at Madem Lakkos, Olympias and Mavres Petres mines. The major minerals present in the Stratoni deposit are pyrite, arsenopyrite, galena and rhodochrosite (Nicolau & Kokonis 1980). Copperiferous pyrite has also been extracted at Madem Lakkos during the 1970s. Another ore type of the area is the porphyric Cu–Au deposits in Skouries and Fisoka, south of Stratoni village. Major minerals present in this ore are pyrite, chalcopyrite, bornite and magnetite.

Mining activity

The deposits of the area have a long exploitation history that started *c.* 600 BC and continues today, mainly because of their Au potential. It is believed that the mining activity in Chalkidiki during ancient times was the major source of gold during the era of Phillip II and Alexander the Great. It is estimated that the exposed smelting material of this period's mining activities reaches 350 000 tonnes (Arvanitidis *et al.* 1994).

During the last century, several mining and metallurgical companies exploited the sulphide deposits in the area. Between 1970 and 1990 *c.* 10 million tonnes of mixed sulphides and 18 million tonnes of pyrite were extracted from the mines at Madem Lakkos, Mavres Petres and Olympias (Papakonstantinou *et al.* 1996). Apart from the mines, other sources of pollution associated with the mining activities include the metal processing factory located by the coast at the north end of Stratoni town and the loading pier located in the port of the town. Solid wastes from recent mining activity include mine tailings, flotation tailings, metallurgical slags, waste rock and pyrite concentrate stockpiles. All of the above are deposited at various locations in the wider area parallel to Stratoni–Varvara fault between the villages of Stratoniki and Stratoni (Fig. 1), mainly along the streams running west of Stratoni town (Papakonstantinou *et al.* 1996). Measures to

Table 1. Summary statistics for ten elements ($\mu\text{g g}^{-1}$) extracted by $\text{HClO}_4\text{--HNO}_3\text{--HCl--HF}$ (number of soil samples = 49). Global soils mean concentrations after Levinson (1980).

	Mean	Median	St.Dev.	Min.	Max.	Geom. mean (\log_{10})	Geom. median (\log_{10})	Global soils mean
Pb	895	325	1663	11	10 996	2.5	2.5	17
Zn	654	240	1149	62	6830	2.5	2.4	50
Cu	150	104	142	26	694	2.1	2.0	20
As	364	119	656	5	3690	2.1	2.1	5
Cd	3.3	1.3	6.1	0.4	37	0.2	0.1	1
Mn	3037	1501	5209	132	24 101	3.2	3.2	530
Fe	61 160	59 500	22 480	2 0600	141 100	4.8	4.8	35 000
Ni	161	69	228	7	1287	1.9	1.9	30
Co	33	32	30	4	169	1.4	1.4	10
Cr	395	178	540	24	2824	2.3	2.3	50

prevent acid mine drainage have been devised only since 1986 and include covering the pyrite stockpiles with PVC, building cement canals to control the flow of rainwater draining the mines, and backfilling the mine shafts with waste material mixed with cement (Adam & Gazea 1994). Mines in Stratonis ceased to operate in 2003 but there are plans for further exploitation in the near future.

METHODOLOGY

Field sampling and chemical analysis

A total of 49 soil samples were collected over an area of $\approx 28 \text{ km}^2$. It was not possible to follow accurately the initial regular sampling grid of $500 \times 500 \text{ m}$ because of the mountainous terrain; however, care was taken to preserve a uniform distribution of sampling sites over the study area. The sampling locations of the study are presented in Figure 1. At each sampling location, the surface of the soil was cleared of superficial debris, vegetation and the O-soil horizon material before digging to a depth of 10 cm. Soil type is similar over the whole area with well-developed soil profiles. Thus the sampled 0–10 cm of topsoil has been judged to be an appropriately homogeneous soil horizon to sample.

After collection, the samples were dried in an oven at a temperature of 40°C , disaggregated in an agate mortar and sieved through nylon screens of 2 mm. The finer fraction ($<2 \text{ mm}$) was homogenized and pulverized to $<0.075 \text{ mm}$ in an agate mill. Then the soil samples were digested with a mixture of $\text{HClO}_4\text{--HNO}_3\text{--HCl--HF}$ and analysed for the following elements: Sb, Ba, Be, B, Cd, Cr, Co, Cu, Pb, Mn, Mo, N, Ag, Sn, V, Zn, and Sr. A portion of each soil sample was digested with a mixture of $\text{HCl--HNO}_3\text{--H}_2\text{O}_2$ and analysed for As and Hg. All samples were analysed by the inductively coupled plasma-atomic emission spectroscopy (ICP-AES) method at the Section of Economic Geology and Geochemistry, National and Kapodistrian University of Athens.

Analytical data quality was assured by introduction of internal reference samples in randomized positions within the analytical batch and by duplicate analysis of a proportion of the samples (Ramsey *et al.* 1987). Analytical precision was calculated from these duplicate samples and was found to be within international standards.

Statistical analysis and use of GIS

The study focused on ten elements, namely Pb, Zn, Cu, As, Cd, Mn, Fe, Ni, Co, and Cr, on the basis of specific interest related to the ore composition and lithology of the area. The computer program MINITAB® 12 for Windows was used for data analysis. Factor analysis (Howarth & Govett 1983) was the main technique used for statistical analysis. This technique can

simplify a complex data set by identifying one or more underlying 'factors' that might explain the dimensions associated with data variability. The 'loading' of each factor, i.e. the degree of association between each variable and each factor, allows the recognition of clusters. A varimax rotation was applied to the initial factor loadings in order to maximize the variance of the squared loadings (Harmon 1976). One-way analysis of variance has also been employed in order to examine whether the map units are different from one another with respect to the soil factor score values.

ARC VIEW® 3.1 for Windows GIS was used to create a simplified digital geological map showing the main lithological types, faults and streams that appear in the study area. This map was created by digitizing a published geological map (IGME 1978) of scale 1:50 000. Additional layers of spatial information, including the topography of the area and mapped sources of pollution, were superimposed to the geological map within the GIS. The source of this information was a digital map created by the mining company Hellas Gold as part of their environmental monitoring programme (V. Gazea, pers. comm.). The spatial database developed in ARC VIEW® was completed by plotting the sampling points of the survey, which enabled the interpretation of the factor analysis scores in association with the spatial features of the area.

RESULTS

The summary of the results of chemical analyses is shown in Table 1. All elements demonstrated positively skewed distributions, indicated in the summary statistics by the arithmetic mean $>$ the median, and were approaching normal distribution after the arithmetic values were transformed (\log_{10}). This transformation was necessary to fulfil the assumption of normality in the application of parametric statistical tests on the data.

The correlation matrix was then computed for 49 pairs of variables after normalization. Pearson correlation coefficients and levels of statistical significance are presented in Table 2. Principal component factor analysis with a varimax rotation was subsequently applied to the data in order to create factors, each representing a cluster of interrelated variables within the data set. The rotated factor loadings, communalities, and the proportion of the variance explained by four factors are presented in Table 3.

Graduated symbol plots of factor scores were created in ARC VIEW®. The symbol plots for each factor were classed according to its score data quartiles and were projected on the digital map in order to visualize the spatial relationship between the factor scores and the geological features as well as various sources of pollution in the study area. These maps are presented in Figures 2 to 5.

Table 2. Pearson correlation coefficient matrix for soil data (49 samples) after \log_{10} normalization. For each element, the upper value is the correlation and the lower value is the p -value at the 95% confidence level.

	Pb	Zn	Cu	As	Cd	Mn	Fe	Ni	Co
Zn	0.77 0.000								
Cu	0.33 0.019	0.50 0.000							
As	0.87 0.000	0.77 0.000	0.38 0.000						
Cd	0.66 0.000	0.86 0.000	0.54 0.000	0.71 0.000					
Mn	0.51 0.000	0.79 0.000	0.32 0.023	0.58 0.000	0.75 0.000				
Fe	0.08 0.592	0.23 0.118	0.22 0.135	0.21 0.142	0.29 0.044	0.36 0.010			
Ni	-0.10 0.480	-0.16 0.279	-0.28 0.055	-0.14 0.355	-0.04 0.781	-0.04 0.799	0.22 0.123		
Co	-0.33 0.020	-0.22 0.122	-0.23 0.112	-0.29 0.045	-0.10 0.538	-0.10 0.481	0.46 0.001	0.80 0.000	
Cr	-0.11 0.471	-0.22 0.137	-0.26 0.072	-0.15 0.308	-0.10 0.591	-0.14 0.339	0.24 0.096	0.97 0.000	0.75 0.000

Table 3. Varimax component loadings of four factors, communality and percentage of variance explained for ten soil variables.

Variable	Factor 1	Factor 2	Factor 3	Factor 4	Communality
Pb	0.917	0.028	0.189	0.103	0.888
Zn	0.897	0.154	-0.210	0.173	0.902
Cu	0.304	0.196	-0.145	0.902	0.965
As	0.906	0.074	0.020	0.125	0.843
Cd	0.824	0.020	-0.276	0.288	0.838
Mn	0.730	0.102	-0.570	-0.075	0.874
Fe	0.121	-0.231	-0.805	0.201	0.756
Ni	-0.025	-0.978	-0.074	-0.111	0.977
Co	-0.216	-0.761	-0.518	-0.138	0.913
Cr	-0.070	-0.984	-0.015	-0.042	0.976
%Variance	0.384	0.264	0.142	0.103	0.893

DISCUSSION

Levels of soil pollution

All ten studied elements are present in soil at high concentrations, well above the global soil mean values (Table 1). Also, many of the elements have mean concentrations above the

tentative trigger concentrations in polluted soils according to the Netherlands and the UK regulations (Table 4). Specifically, the measured mean concentration of Pb in soil ($895 \mu\text{g g}^{-1}$) exceeds the action limit of $600 \mu\text{g g}^{-1}$ set by the Netherlands regulations and approaches the limit of $1500 \mu\text{g g}^{-1}$ for open space set by the UK regulations. The mean concentration of Cu ($150 \mu\text{g g}^{-1}$) is very close the Interdepartmental Committee on the Redevelopment of Contaminated Land (ICRCL) trigger concentration of $140 \mu\text{g g}^{-1}$. Arsenic, with a measured mean of $363 \mu\text{g g}^{-1}$, is about nine times above the regulated limits and Ni, with a measured mean of $161 \mu\text{g g}^{-1}$, exceeds the ICRCL limit of $35\text{--}70 \mu\text{g g}^{-1}$.

Taking into account the mining history of the area, perhaps a more appropriate comparison can be attempted with the ICRCL guidelines for toxic element trigger concentrations in mine spoil-contaminated soils (Table 5). It is noteworthy that these concentrations are acceptable only when soil contamination is derived from mine spoil. In other situations, the elements may be present in forms which are more available to plants and animals when lower trigger concentrations will be appropriate (ICRCL 1990). According to these values, Pb, As and Cd exceed the threshold trigger concentration but only the mean concentration of Pb approaches the limit of $1000 \mu\text{g g}^{-1}$ for grazing livestock.

Factor analysis and spatial geochemical interpretation

Examining the correlation matrix of the elements (Table 2), many of the correlation coefficients are significant and some are very strong, e.g. Pb–Zn, Pb–As, Zn–Cd, Ni–Cr. This observation indicates common sources for the elements. The factor analysis yielded four factors, explaining 89.3% of the total variance in the data (Table 3). Factor 1 comprises Pb, Zn, As, Cd and Mn which show high positive loadings with a small contribution from Cu. Factor 2 contains Ni, Co and Cr with similar high negative loadings, Factor 3 is dominated by Fe and Factor 4 by Cu.

The variability of the elements in the first factor appears to be controlled by the mixed sulphide mineralization as all elements present in this factor are characteristic of the sulphide ore chemistry in the area. This factor accounts for 38% of the total variance. When the factor scores are projected on the map (Fig. 2), sampling locations with the highest positive values, indicating geochemical anomalies, lie within the Neogene deposits, whereas locations with the lowest scores lie within the

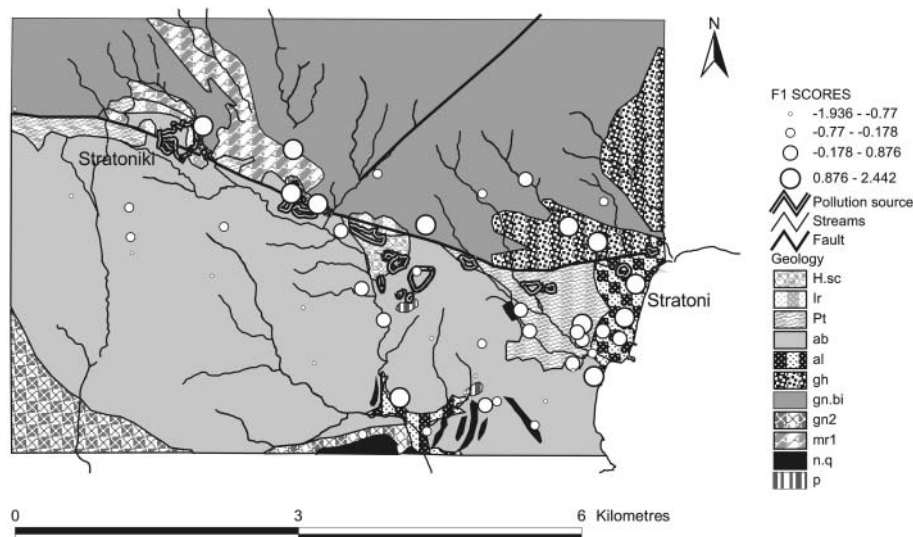


Fig. 2. Graduated symbol plot of Factor 1 scores projected on the simplified geological map of the area. High positive scores indicate geochemical anomaly for Pb, Zn, As, Cd and Mn. al, Alluvial deposits; Pt, Pleistocene deposits; H.sc, mine tailings; Ir, area of important gossan; mr1, marble; gn.bi, biotite gneiss; ab, amphibolite; gh, granodiorite; gn2, two-mica gneiss; n.q., diorite porphyry; p, peridotite and dunite (ultramafics). Source map from IGME (1978).

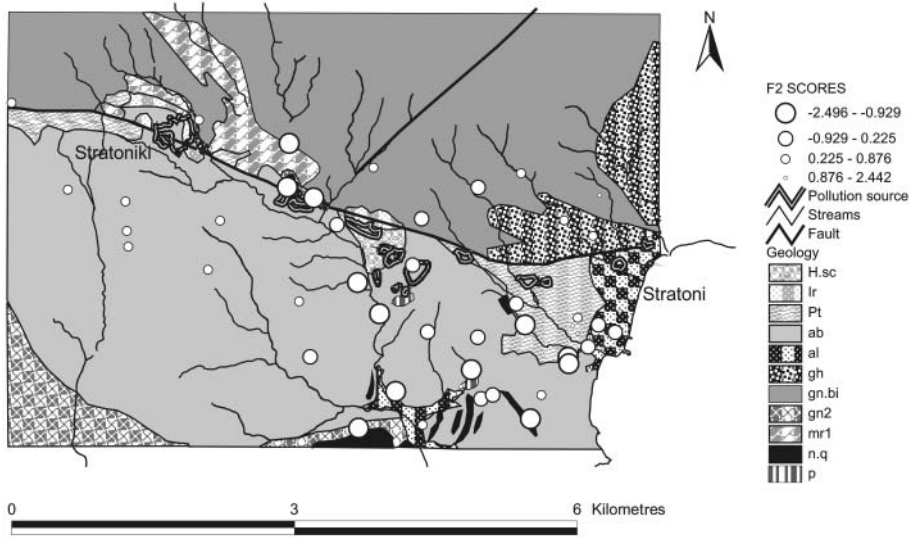


Fig. 3. Graduated symbol plot of Factor 2 scores projected on the simplified geological map of the area. High negative scores indicate geochemical anomaly for Cr, Co, Ni.

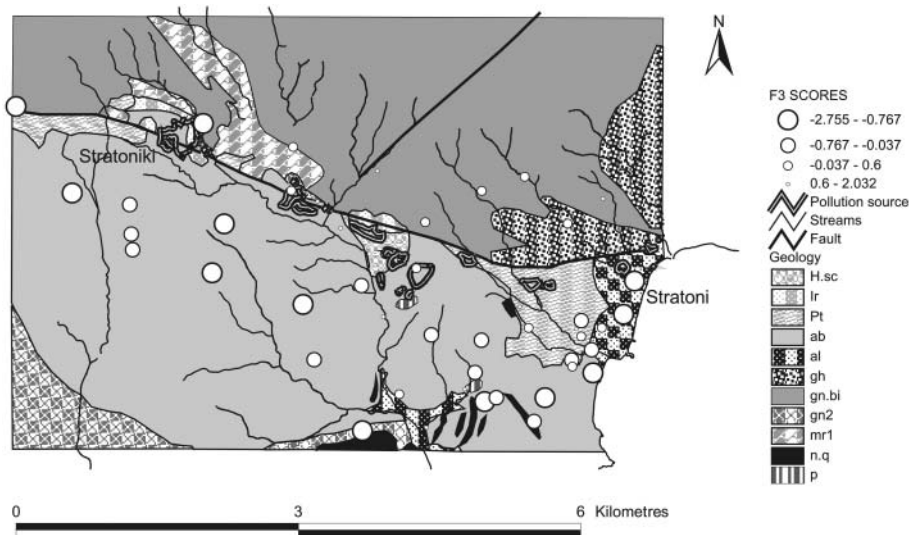


Fig. 4. Graduated symbol plot of Factor 3 scores projected on the simplified geological map of the area. High negative scores correspond to locations with high Fe content.

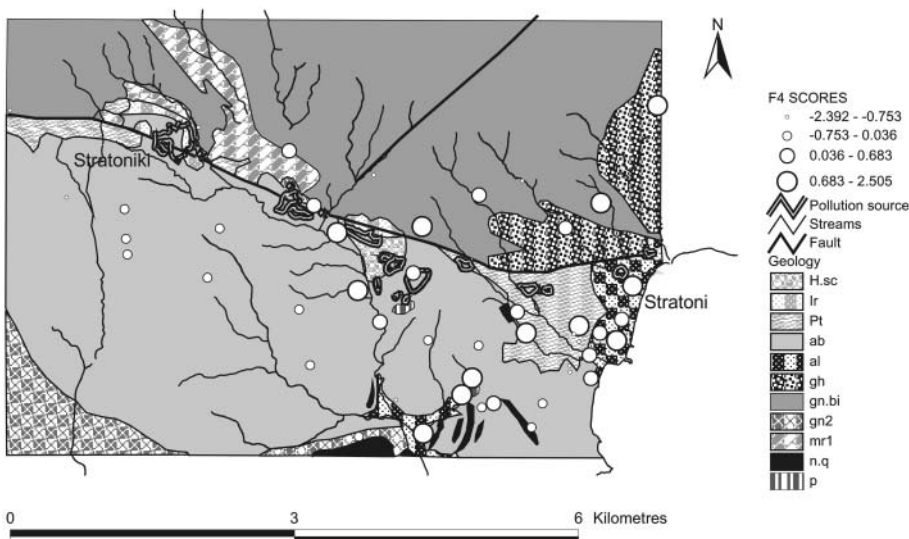


Fig. 5. Graduated symbol plot of Factor 4 scores projected on the simplified geological map of the area. High positive scores indicate locations with increased Cu content.

amphibolite of the Vertiskos unit. High positive scores are also plotted along a zone that runs parallel to the Varvara–Stratoni fault, within the Kerdilion sequence. One-way analysis of

variance applied on scores of Factor 1 indeed revealed a statistically significant difference between the three map units with respect to the soil factor score values at the 95%

Table 4. Trigger concentrations (in $\mu\text{g g}^{-1}$) of potentially toxic elements in polluted soils (Alloway 1995).

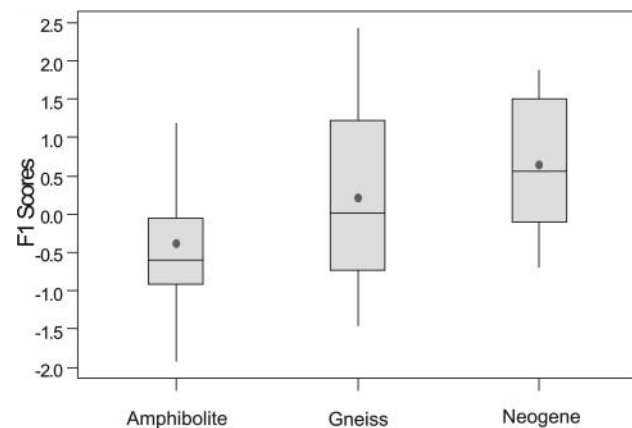
Element	Netherlands ^a			UK (DOE-ICRCL) ^b			
	A	B	C	Small garden	Large garden	Amenity grass	Open space
Pb	50	150	600	550	550	1500	1500
Zn	200	500	3000	280	280	280–560	280–560
Cu	50	100	500	140	140	140–280	140–280
As	20	30	50	20	10	40	40
Cd	1	5	20	5	3	12	15
Ni	50	100	500	35	35	35–70	35–70
Co	20	50	300				
Cr	100	250	800	600	600	1000	1000

^aIn the Netherlands system, A is the background value, B indicates need for further investigation and C means clean-up definitely required.

^bDepartment of the Environment, Interdepartmental Committee on the Redevelopment of Contaminated Land (ICRCL), Consultation Paper.

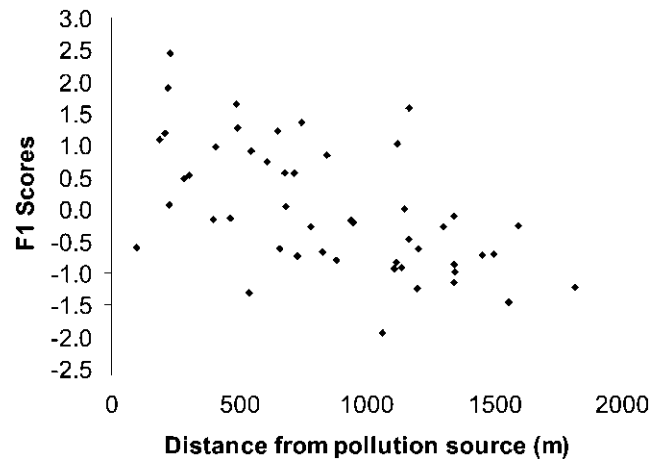
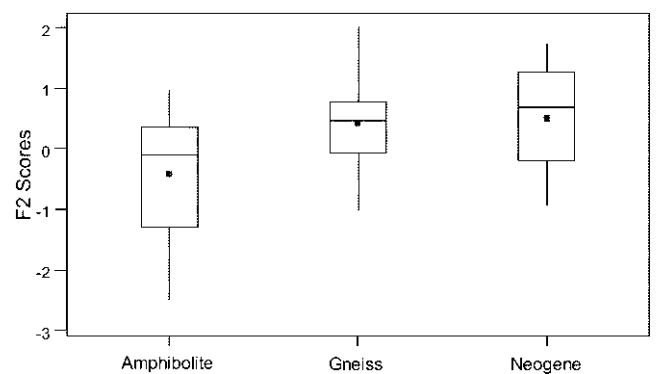
Table 5. Guidelines for toxic element trigger concentrations (in $\mu\text{g g}^{-1}$) in mine spoil-contaminated soils (ICRCL 1990).

Element	Threshold trigger concentration	Maximum (action trigger) concentrations (values not to be exceeded for use as specified)	
		For grazing live-stock	For crop growth (risk of phytotoxicity)
Pb	300	1000	–
Zn	1000	3000	1000
Cu	250	500	250
As	50	500	1000
Cd	3	30	50

**Fig. 6.** Box-plots comparing the map units with respect to soil Factor 1 score values. Solid circles indicate mean values.

confidence level. This comparison is presented graphically by means of side-by-side box-plots in Figure 6.

When interpreting these patterns, although the geogenic nature of the factor is profound, the anthropogenic influence must also be noticed referring to the high values noted in the Neogene deposits at Stratoni village as well as those in proximal distance to the pollution sources. It is possible that human activities, including the operation of the metal processing factory and the loading pier in Stratoni harbour, have influenced the metal contents of the soil. To demonstrate the effect of the pollution sources, scores of Factor 1 were plotted against the

**Fig. 7.** Soil Factor 1 scores as a function of proximity to mapped pollution sources.**Fig. 8.** Box-plots comparing the map units with respect to soil Factor 2 score values. Solid circles indicate mean values.

distance from the nearest pollution source (Fig. 7). A significant trend is shown on this graph with higher score values corresponding to shorter distances from pollution sources. However, this relationship is probably also affected by other factors, such as the prevailing wind direction and the surface slope, that hinder the extraction of a quantitative equation between the plotted variables.

The second factor, accounting for 26.5% of the total variance, contains Co, Ni and Cr. These elements are present with high concentrations in ophiolitic rocks, the protolith of the amphibolite present in the study area; therefore the factor can be characterized as 'lithogenic'. By examining the plotted factor scores on the geological map, anomalous values for Co, Ni and Cr in soil are represented by the highest negative values (Fig. 3). Comparison of the three main geological units by means of box-plots with respect to Factor 2 scores shows significantly lower values within the amphibolite at the 95% confidence interval (Fig. 8). This observation supports the argument of a 'lithogenic' control for the factor.

The third factor, explaining 15% of the total variance, is dominated by Fe with a smaller contribution from Mn and Co. The spatial distribution of the score values indicates increased elemental concentrations at sample locations of high negative scores (Fig. 4). Although there is no significant pattern observed with respect to the geological units, the presence of Fe and Mn oxides in the area could explain the high concentration of these elements in the sampled soil. Finally, Cu is the dominant element of the fourth factor, which explains 10% of the total variance. This element participates in the chemistry of the

mixed sulphide mineralization; however, it presents its own geochemical signature in the factor analysis. Again, there is no significant spatial pattern observed. However, like Factor 1, high positive score values in the coastal area might signify some anthropogenic contamination of the soil whereas high positive values in the centre of the map can be associated with the copperiferous pyrite, characteristic of the mineralogy of the deposit at the mining site of Madem Lakkos.

CONCLUSIONS

The results of soils analyses from the mining region of Stratoni, north Greece, show elevated concentrations of metals. All ten elements studied are present in soil with concentrations well above the mean values of global soils, and some elements (Pb, As, Ni) have concentrations exceeding the tentative guidelines set by the UK and the Netherlands regulations.

Factor analysis combined with spatial interpretation of its output, enabled by the use of GIS, proved a successful method for grouping the elements according to their sources, and provided evidence of their geogenic or anthropogenic origin. The factor analysis applied to the data explained 89% of the total variance through four factors, each corresponding to common sources of elements.

The most significant factor comprises the elements participating in the chemistry of the mixed sulphides mineralization, i.e. Pb, Zn, As, Cd, Mn and to a lesser extent Cu. Spatial interpretation of the high scores of this factor signified both geogenic and anthropogenic origin of pollution in the area. Of course, mine wastes will probably show similar multi-element geochemical signatures to soils developed over lithologic units containing the mineralized rocks being mined. Given the long mining history of the area and the lack of a pre-mining survey, we cannot be certain about the origin of pollution; however, the metal-processing plant at Stratoni village near the coast is a very suspect source for elevated elemental concentrations in that particular area.

The second factor accommodated elements that relate to the amphibolite metamorphic rocks of the area (Co, Ni, Cr) and has a lithogenic character. Factor 3 comprises Fe and to a lesser extent Mn and represents the existence of oxides. Finally, the fourth factor is dominated by Cu that corresponds to the copperiferous pyrite of Madem Lakkos and possibly to the anthropogenic pollution in the coastal zone.

The results of this study, although they provide only some evidence on the apportionment of soil pollution sources, can form the basis for more detailed research utilizing comparative geochemical and mineralogical data, metal speciation or stable isotope analysis in order to further clarify the portions of lithogenic and anthropogenic inputs to pollution. Such studies are necessary for devising the most appropriate methodology for remedial action after closure of the mines in the area.

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