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Spatial distribution of heavy metals in urban soils of Naples city (Italy)

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"Capsule": *Cu, Cr, Pb and Zn in the surface and sub-surface soils of the Naples city urban area have been studied and compared with historical data.*

Abstract

Concentrations of surface and sub-surface soil Cu, Cr, Pb and Zn in the Naples city urban area were measured in 1999. Contour maps were constructed to describe the metals spatial distribution. In the most contaminated soil samples, metals were speciated by means of the European Commission sequential extraction procedure. At twelve sites, Cu, Pb and Zn levels in soil were compared with those from a 1974 sampling. Many surface soils from the urban area as well as from the eastern industrial district contained levels of Cu, Pb and Zn that largely exceeded the limits (120, 100 and 150 mg kg^{-1} for Cu, Pb and Zn, respectively) set for soils of public, residential and private areas by the Italian Ministry of Environment. Chromium values were never above regulatory limits (120 mg kg^{-1}). Copper apparently accumulates in soils contiguous to railway lines and tramway. Cu and Cr existed in soil mainly in organic forms (~68%), whereas Pb occurs essentially as residual mineral phases (77%). The considerable presence of Zn in the soluble, exchangeable and carbonate bound fraction (23%) suggests this element has high potential bioavailability and leachability through the soil. Concentrations of Cu, Pb and Zn have greatly increased since the 1974 sampling, with higher accumulation in soils from roadside fields.

Keywords: Urban soil; Heavy metals; Pollution; Speciation; Naples

1. Introduction

Human health in towns and cities is strongly dependent on the status of urban soils (Simpson, 1996). Strong compaction, contamination by wastes and atmospheric depositions, loss of organic matter, changes in soil reaction, structural degradation and infection by pathogenic microorganisms are only few of the many adverse processes that affect and modify the ecological functions of soils in urban areas (Bullock and Gregory, 1991; Jim, 1993; Beyer et al., 1995; Blum, 1998). Moreover, ground sealing, mainly deriving from expansion of residential and traffic areas, progressively reduces the surface under vegetation and soils (Oechtering, 2000).

Urban soil pollution may result from the accumulation of nonsoil origin pollutants as well, where little vegetation cover or dry conditions subsist, from re-entrainment of polluted soil particles. Heavy metals, radionuclides, chlororganic compounds and other toxicants are the most common contaminants, mainly due to emissions of industrial plants, thermal power stations, vehicular traffic and road infrastructures (Czarnowska et al., 1983; Piron-Frenet et al., 1994; Wu et al., 1998; Krauss et al., 2000; Kvasnikova et al., 2000). Heavy metals reaching the soil remain present in the pedosphere for many years even after removing of the pollution sources and increased amounts of heavy metals in soils of urban areas have been reported by many authors (Klein, 1972; Chen et al., 1997; Pichtel et al., 1997). However, the severity of pollution depends not only on total heavy metal content of the soil, but also on the proportion of their mobile and bioavailable

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forms, which are generally controlled by the texture and other physicochemical properties of soils (Brummer et al., 1986; Ma and Rao, 1997; Selim and Sparks, 2001). Synergistic effects among metallic pollutants may deteriorate the situation. Therefore, to define hazards and to propose treatments and eventually new, more appropriate utilisation of soils in urban areas the speciation, geochemistry and behaviour of heavy metals in soil have to be investigated.

There has been little research on urban soils in Italy. Naples is a very densely populated city, located near the sea and included in one of the largest urbanised and densely populated areas in the world. Vegetation and soil landscape mapping have been conducted in the urban environment by the Land Planning Service of the City Council, in collaboration with the University of Naples, providing reliable data for land use analysis and planning (Mazzoleni et al., 1999). These studies have shown that both physical and chemical degradation processes affect urban soils. The reasons behind this situation are associated to the "social development model" of the city of Naples, especially after the 1950s. An understanding of the problem may be inferred by taking into account the following: (1) uncontrolled urbanisation dynamics of the whole area in the last 30 years that has produced considerable loss in agricultural soils and urban greenspace, (2) the intense road traffic volume (265,000 vehicles cross the city per day, data from Municipality), being strongly influenced by the daily commutins of workers, (3) the existence of many industrial activities such steelworks, petrol refineries, and chemical industries in suburban areas that became included in Naples with the growth of the city.

In particular, the presence of various oil refineries (Q8, AGIP, ESSO) in the eastern and western districts of the city along with combustible deposits, steelworks (ILVA, Cementir) and industrial chemical plants (Eternit, Montedison) have produced widespread trace metal contamination of the surrounding green areas.

Data from the agencies for the control of air pollution in Campania (SCIA, Servizio Controllo Inquinamento Atmosferico; MARC, Monitoraggio Ambientale Regione Campania) and biomonitoring studies of air quality indicated that the urban area of Naples is seriously affected by nitrogen, sulphur and carbon oxides, ozone, dust, hydrocarbon and trace element from the atmosphere (Adamo et al., 2003; Alfani et al., 2000).

Early studies in the Naples urban area indicated elevated concentrations of Cu, Pb and Zn in soils surrounding industrial plants and streets (Basile et al., 1974). Accumulation of Pb, Zn and Cr in the upper part of the A horizon (0–5 cm) of the soil of a *Quercus ilex* L. wood inside the city influenced organic matter decomposition by strongly decreasing the rate of litter decay in the early stages of decomposition, via a reduction in soil of litter fungal biomass, and biological activity (Cotrufo et al., 1995). Limited information exists concerning the spatial distribution and availability of heavy metals in soil. Furthermore, there is no data regarding temporal changes of metal concentrations in contaminated soils.

The objective of this study was to assess spatial changes of metals in surface soils in the Naples urban area. Specifically, (1) the total content of Cu, Cr, Pb and Zn in surface and sub-surface soils of the city of Naples was measured, (2) the chemical and mineralogical forms of metals in soil were determined using a sequential fractionation procedure in order to assess mobility and availability to plants. In addition, some current data were compared with those of a 1974 sampling.

2. Materials and methods

2.1. The study area and soil sampling

The city of Naples is located in a coastal plain environment of south Italy where abundant falls of pyroclastic materials from the nearby major volcanic systems, the Phlegrean Fields and Vesuvio, have occurred. Consequently, the soil is derived from volcanic parent material. The city area occupies a surface of 2117 km^2, with 292 km^2 is classed as urban areas, and has approximately 1,200,000 inhabitants. The climate is typically Mediterranean with an average annual temperature of 15.4 °C and an average annual rainfall of 1026 mm (Ufficio Idrografico e Mareografico di Naples, 1960–1995).

This study concentrated on the urban core area, including the western and eastern districts of the city, where the motorway and the old oil refineries along with combustible deposits are located, respectively. For sampling, the study area was divided in regular grids of 500×500 m. One composite (10 subsamples), W-shaped surface soil sample (0–2 cm depth) was collected in 1999 in each grid in gardens, parks, roadside fields and industrial sites. A total of 173 surface soil samples were collected. In 36 of the selected sites soil samples at 10, 20 and 30 cm of depth were also collected.

Soils submitted to X-ray and electron microscopic analysis revealed a certain homogeneity in mineralogical assemblage consisting mainly of mica biotite, sanidine, leucite, garnet and pyroxenes. Leucite occurred only in the samples collected in the eastern and central areas of the city, which indicates the Somma-Vesuvius pyroclastic materials as main parent material of these soils (Beccaluva et al., 1991) (Table 1). The absence, with the exception of very small amounts of halloysite, of well crystalline clay minerals, suggested weak pedogenetic processes, likely due to continuous renewing of soils through addition of new primary products.

Table 1
Amounts of minerals occurring in soils from eastern and central (E–C) and western (W) Naples urban area[a]

Mineral	E–C	W
Biotite	12	19
Halloysite	1	2
Sanidine	19	27
Leucite	14	Not detected
Garnet	8	Not detected
Diopside	10	15
Augite	6	10
Glass	30	27

[a] % of soil mass (semiquantitative analysis according to Bish and Chipera, 1988; Chipera and Bish, 1995).

2.2. Chemical analysis

Soil samples were air dried, sieved to pass a 2 mm mesh and stored in polypropylene bottles. On the samples were determined: pH–H_2O (1:2.5 soil:water ratio), organic carbon (Walkley and Black method, 1934), carbonates (pressure Dietrich-Fruehling calcimeter method, Loeppert and Suarez, 1996), particle-size analysis after ultrasonic treatment at 20 kHz and 75 W for 15 min. Separation of the coarse sand (0.2–2 mm) and clay (<2 μm) fractions was achieved, respectively, by wet sieving and centrifugation.

Total heavy metal content was determined by treating 5 g of soil with HCl/HNO_3 (3:1 solution) at 100 °C for 1 h (Canepa et al., 1994). The four-step chemical extraction procedure (0.11 M HOAc, 0.1 M NH_2OH HCl, H_2O_2/1 M NH_4OAc, HF/HNO_3), developed by the Measurement and Testing Programme of the European Commission (Ure et al., 1993; Adamo et al., 1996; Singh et al., 1998), was used to fractionate heavy metals in easily reducible, reducible, oxidizable and residual forms in the 15 most contaminated soil samples.

To compare current data with those collected by Basile et al. (1974), extractions of Pb with Na_2EDTA (0.05 mol l^{-1}), at a ratio of sample:extractant of 1:10 with shaking for 2 h, and of Cu and Zn with $(NH_4)_2EDTA$ (0.05 mol l^{-1} at pH 9), at a ratio of sample:extractant of 1:10 with shaking for 24 h, were carried out on the soil samples from the same twelve locations used in the 1974 survey.

Copper, Cr, Pb and Zn concentration in the extracts was determined in acidic samples (pH below 2.0) using a Perkin Elmer 3030 B atomic absorption spectrometer (AA), equipped with deuterium-arc background correction.

2.3. Statistical analysis and spatial distribution maps

Data were submitted to descriptive statistical analysis to define their frequency distributions. Simple correlation analysis was used to examine the relationship between the analysed heavy metals. The evaluation of the degree of association among variables, assumed as normally distributed, was based on calculating the value of the correlation coefficient (r) and testing it $[|t\text{-test}| = |r|\sqrt{(n-2)/(1-r^2)} > t(n-2;\quad \alpha/2)]$ for 5% significance level. Scatter diagrams of all associated variables were drawn to check the uniform dispersion of plots and to help in the interpretation of significant and non-significant correlation coefficients.

The total concentrations of heavy metals in the surface soil samples were used to construct contour maps using the program SURFER (Golden Software Inc., Colorado).

3. Results and discussion

3.1. Soil properties

According to data in Table 2, the content of clay-size particles (<2μm) ranged from 2.6 to 8.6 g kg^{-1} with a mean value of 5.4 g kg^{-1}. Lower values characterized the samples from the eastern area of the city. The pH–H_2O value was generally neutral or sub alkaline, according with the diffuse presence of carbonates in soil. Only few samples from the central urban district showed pH values lower than 6.0. The content of total organic carbon in the soil ranged between 6.0 and 80.6 g kg^{-1} with higher mean values in the central area.

Table 2
Main physico-chemical properties of the studied surface soils from different districts of Naples city; the first line represents mean ± standard deviation, the second one minimum and maximum value[a]

Soil samples	Coarse sand (2–0.2 mm)	Fine sand (0.2–0.02 mm)	Lime (0.02–0.002 mm)	Clay (<2 μm)	pH (H_2O)	OC	$CaCO_3$
Western district	24.6±1.23	50.6±2.53	18.3±1.12	6.5±0.32	8.01±0.40	13.3±0.66	3.3±0.16
	(14.5–30.1)	(47.1–59.9)	(15.0–23.9)	(4.9–8.6)	(7.34–8.42)	(6.2–31.9)	(0–31.9)
Central district	30.1±1.74	41.0±2.05	22.8±1.14	6.1±0.24	6.81±0.24	30.3±1.51	4.7±0.23
	(20.8–41.8)	(35.3–49.1)	(17.1–26.4)	(3.7–8.1)	(4.31–8.10)	(12.6–80.6)	(0–19.1)
Eastern district	44.7±2.29	40.5±2.68	11.1±0.45	3.7±0.18	7.4±0.37	21.2±1.08	1.54±0.07
	(38.7–53.5)	(34.1–44.9)	(8.8–14.3)	(2.6–5.0)	(6.67–8.28)	(6.0–33.7)	(0–4.06)

[a] g kg^{-1}.

3.2. Heavy metal total content

The descriptive statistics of the Cu, Cr, Pb and Zn total data sets relative to surface soil samples are given (Table 3). The frequency histograms of the data are shown (Fig. 1).

Chromium, Pb and Zn total concentrations produce the same general unimodal distribution. The frequency histogram of Cu seems to indicate the presence of two overlapping populations. For all metals, total concentrations were not normally distributed, showing rather positively skewed data. Skewness, measured according to the formula 3×(sample mean−sample median)/sample standard deviation, ranged from 0.7 for Pb to 1.07 for Cu. Therefore, medians instead of means would describe such distributions more precisely.

Total copper content ranged from 6.2 to 286 mg kg^{-1}, with a median of 54 mg kg^{-1}. Approximately 85% of the analyzed soils contained from 6.2 to 120 mg kg^{-1} total Cu, and only 15% exceeded the maximum concentration (120 mg kg^{-1}) established for soils of public, residential and private areas by the Italian Ministry of Environment (Gazzetta Ufficiale della Repubblica Italiana,

Table 3
Heavy metal content of the Naples urban soils, 1999[a]

Number of samples	Element			
	Cu 173	Cr 173	Pb 173	Zn 173
Minimum value	6.2	1.7	4	30
Maximum value	286	73	3420	2550
Median	54	8.4	184	180
Mean	74	11	262	251
Standard deviation	56	9	337	253

[a] mg kg^{-1}.

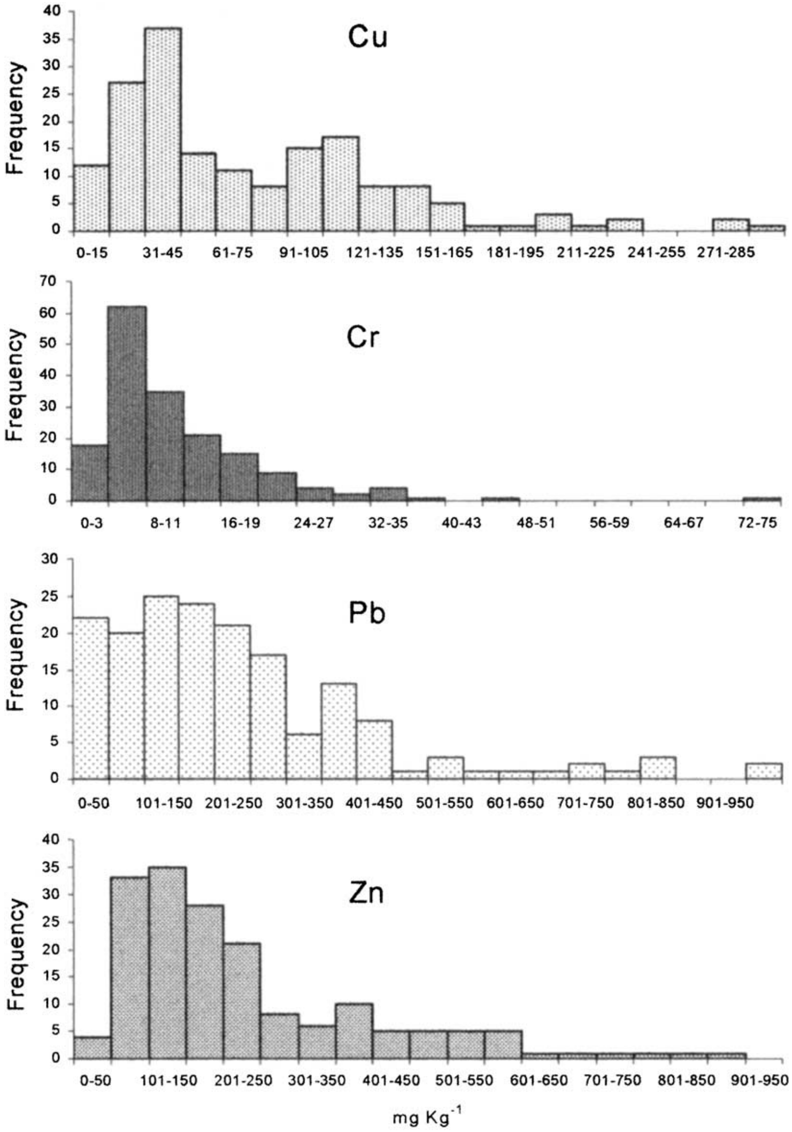

Fig. 1. Frequency histograms of the raw data set.

1999). Soil chromium content ranged from 1.7 to 73 mg kg^{-1} with a median of 8.4 mg kg^{-1}; the regulatory limit (120 mg kg^{-1}) was never exceeded.

Total soil Pb content ranged between 4 and 3420 mg kg^{-1}, with a median value of 184 mg kg^{-1}. A large number of soils (~76%) were polluted by Pb, with concentrations in excess of the 100 mg kg^{-1} limit set for public, residential and private areas.

Total soil Zn content ranged from 30 to 2550 mg kg^{-1} with a median of 180 mg kg^{-1} and a percentage of contaminated soil (Zn > 150 mg kg^{-1}) of 53%.

About 14% of the overall analyzed soils showed levels of Cu, Pb and Zn above the regulatory limits. In conclusion, the surface soils of the Naples urban area appeared to be polluted in the order by Pb > Zn > Cu≫Cr.

In all the soil samples collected at 10, 20 and 30 cm depths from 36 selected sites, total contents of Cu, Cr, Pb and Zn decrease with depth (Fig. 2). The trend was particularly steep for Zn (80–185 mg kg^{-1}) and Pb (170–278 mg kg^{-1}). Chromium, whilst not enriched in these soils, also shows a gentle decline from surface to 30 cm. Assuming that the studied soils were undisturbed, the depth of penetration may give some indication of the mobility of anthropogenic Pb in soil (Puchelt al., 1993), mainly by deposition from automobile exhausts. However, decrease of Pb levels with depth did not appear to be uniform. The discontinuity in the slope of the Pb and Cu lines, Fig. 2, from 0–20 cm to 20–30 cm, could be attributed to reduced metal inputs deriving, in the case of Pb, from recent increased utilization of unleaded fuels.

The spatial distributions of Cu, Cr, Pb and Zn are shown in the maps (Fig. 3a, b, c, d, respectively). For Cu, Cr and Zn all of the data are included; for Pb, the two largest values measured in soil samples collected on the edges of the city motorway are excluded to improve the visualization of the geochemical anomalies. High levels of all metals are located in sites of the eastern part of the city, corresponding with areas of heavy industry and where various oil refineries (Q8, AGIP, ESSO) along with combustible deposits occur. Copper apparently accumulates in soils contiguous to railway lines and tramways mainly along the south eastern coastal line.

Lead concentrations fluctuate throughout the city. However, from the city centre and in industrial eastern and western areas, there is substantial Pb contamination of soils. The most contaminated soils are in the proximity of the motorway and streets with high traffic flows. Only soils on the northwest part of the city, which is characterised by greater elevation (~150 m above sea level), contain low Pb levels. Despite the sharp increase of unleaded fuel utilization, followed by a rapid decline of Pb levels in the atmosphere, the content of Pb in urban soil still remains high with a consequent associated risk for children via the soil–hand–mouth pathway (Bargagli, 1998).

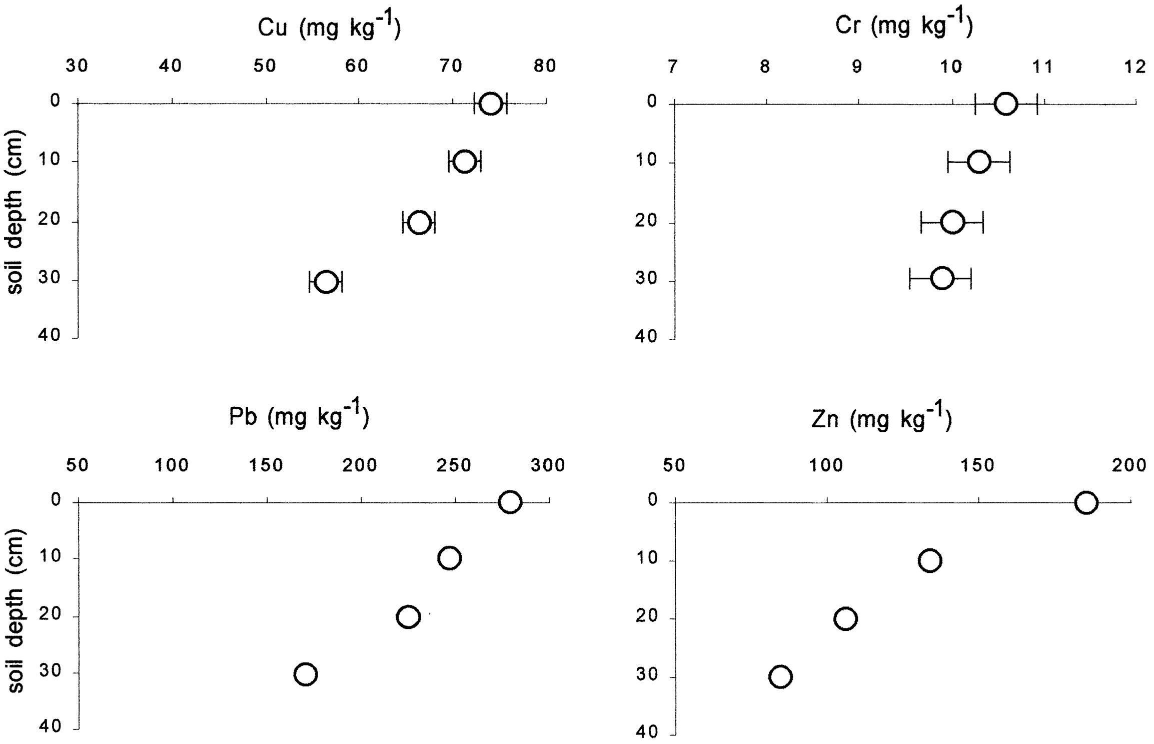

Fig. 2. Vertical distribution of Cu, Cr, Pb and Zn in soils from Naples urban area (values are means of 36 data).

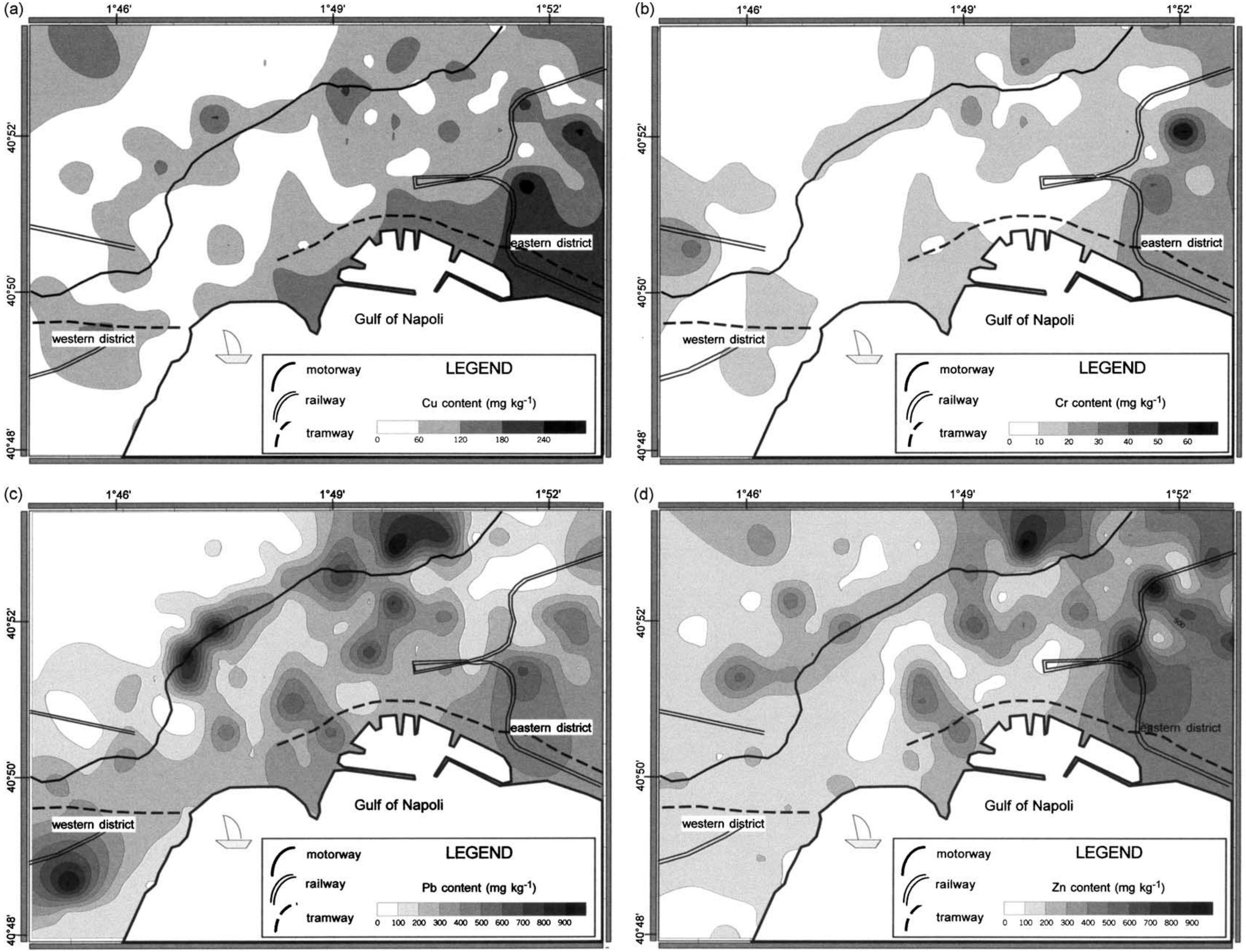

Fig. 3. Kriged maps of (a) Cu, (b) Cr, (c) Pb and (d) Zn content in surface soils of Naples urban area.

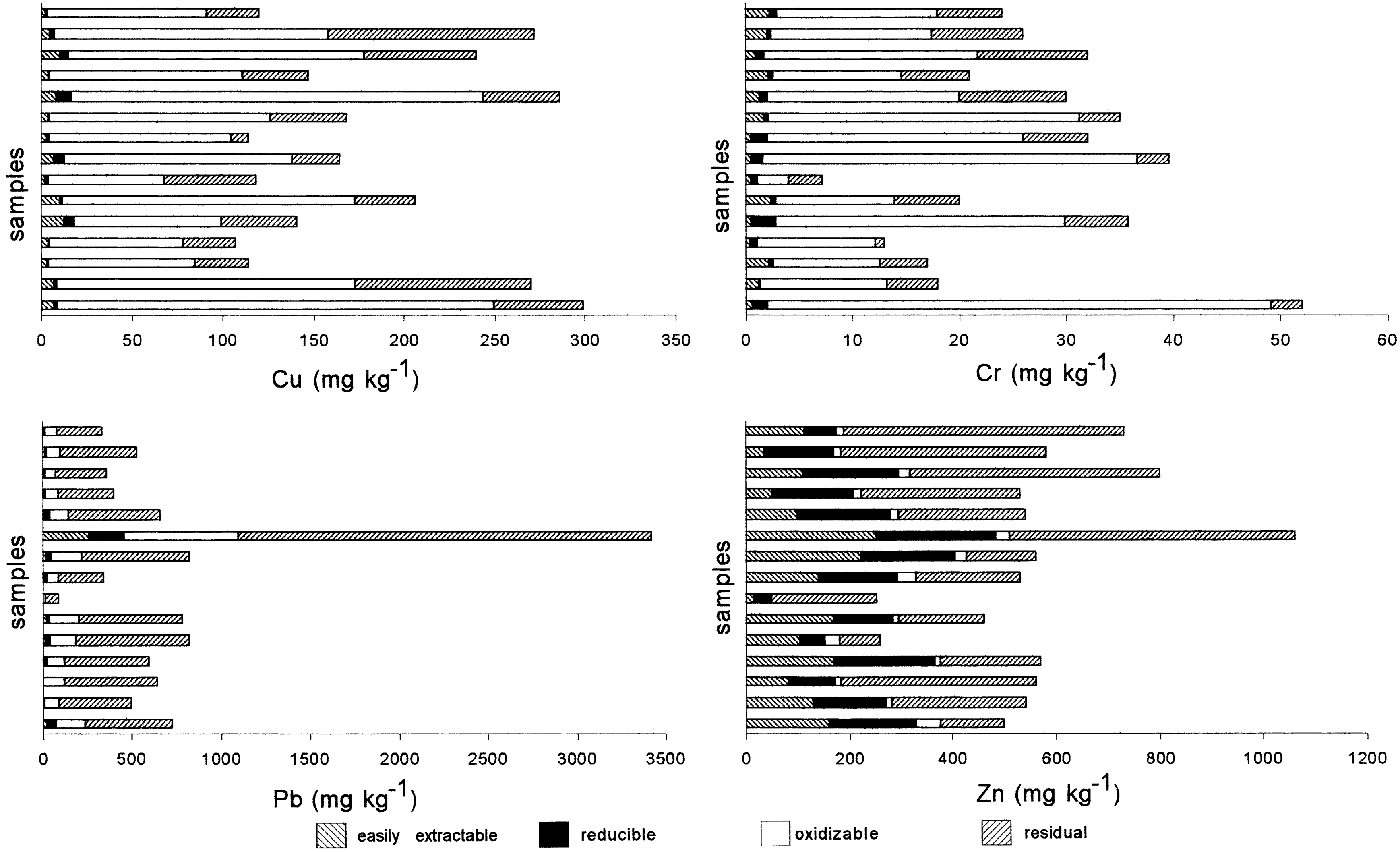

Fig. 4. Contribution of Cu, Cr, Pb and Zn in the sequentially extracted fractions.

3.3. Heavy metal speciation

The contribution of the easily extractable, reducible, oxidizable and residual pools of Cu, Cr, Pb and Zn from the selected fifteen soil samples are given (Fig. 4).

Results indicated that Cu and Cr were mainly held in the oxidizable (~68%) and residual (~24%) fractions, with other forms making up much less than 10% of the total. Lead was primarily held in the residual mineral fraction (77%) with smaller amounts held in oxidizable (18%) and reducible (3%) forms. Zinc was uniformly distributed between easily extractable (23%), reducible (24%) and residual (49%) fractions, with a very small percentage occurring in oxidizable forms (4%).

Thus, among the studied metals only Zn appeared to occur in consistently bioavailable and leachable forms in these soils. The same pools were lower for Cu and Cr, which were associated mainly with organic matter, for which both metals have high affinity (Kabata-Pendias and Pendias, 1992; Stevenson, 1982). Oxidation of organic matter, which is typical of Mediterranean environments, can make these metals more potentially available.

In the case of Pb, and to a lesser extent of Cu and Cr, the residual pool was prevalent, indicating scant relatively mobility.

3.4. Comparison between present and historical data

For comparison between present and historical data, emphasis has been placed on selecting soil sampling locations and methodologies of metal extraction that had been previously used. Twelve sites were selected for comparison: three urban parks (P), five square gardens (G) and four roadside fields (R).

The 1974 results showed soils from roadside fields and square gardens had higher contents of heavy metals when compared with soils from parks, which are more protected from pollution. Comparison between the 1999 data and those of 1974, made by subtracting the 1974 from the 1999 value and expressing the difference as a percentage of the 1974 value, revealed significant increases in Cu, Pb and Zn contents of the surface layer of all the Naples urban soils (Fig. 5). In comparison to 1974, Cu content in soil increased from 41% (site G5) to 878% (site R3), with mean increase of 134%. Lead content increased from 17% (site G1) to 876% (site R1), with mean increase of 182%. This trend was observed also for Zn, whose levels in soil at sites G1 and R1 were characterised, respectively, by the lowest (22) and highest (765) percentages of increase. Zinc levels in urban soils showed the higher percentage increases from 1974 to 1999 data due to possible accumulation from garden fertilizing activities, traffic and industry inputs. Presumably, the significant increases in Zn content of urban soils can be related to a continuous input of the metal in the urban environment principally as a consequence of vehicle emissions and tyre and brake abrasion (Garty et al., 1985; Ward, 1989). This would confirm that urban traffic is the main anthropogenic source of airborne trace elements in the Naples urban area. In contrast, the utilization of unleaded fuel could explain the lesser increases in Pb content of urban soils over this period.

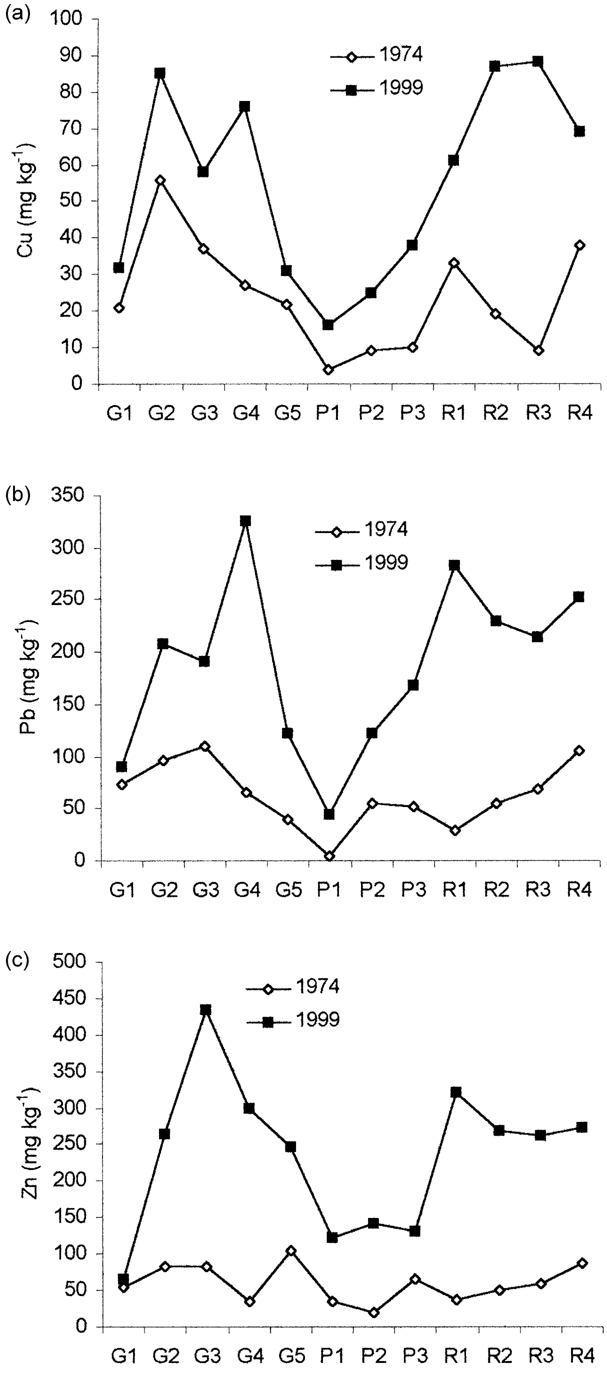

Fig. 5. Comparison of Cu (a), Pb (b) and Zn (c) amounts extracted by Na_2EDTA (0.05 mol l^{-1}) and $(NH_4)_2EDTA$ (0.05 mol l^{-1} at pH 9) for the 1974–1999 sampling sites in the Naples urban area.

4. Conclusions

The Naples urban area has been affected by human activity leading to a high accumulation of heavy metals Cu, Pb and Zn. The topsoil enrichment clearly revealed an anthropogenic origin of the pollution. The distribution of the metal concentration of the soil in the city indicated that the industry together with the traffic were mainly responsible for metal pollution, as the highest metal concentrations were found in the industrial eastern district, which always increased with respect to 1974, and close to the motorway and high traffic volume-roads.

Automobile emissions are probably the major source of the elevated Pb content of Naples urban soils. Highest Pb concentrations were detected in soil samples collected from the border of the city motorway and from streets areas with high traffic flows. It is probable that automobile exhausts are also a source of Zn and Cu. Although the industrial activity in the east area has drastically declined it might have contributed to most Zn accumulation in soil over the past 20 years. Tyre and line abrasion processes in the rail and tram-ways may be also responsible for the soil Cu pollution. The predominant fraction of Cu and Cr remained mainly linked with the organic matter, which can become available after organic matter mineralization. However, most of the Pb remained in insoluble and stable mineral forms of low availability. In respect to 1974, the 1999 soil metals assay of the urban area revealed an increasing level of Cu, Pb and Zn in the soil surface layer. The trend was particularly evident for Zn in the proximity of the roadside fields, indicating increased atmospheric deposition from road traffic.

Among all metals the more readily available pool was particularly consistent for Zn ($\sim$23% of the total), indicating this element is potentially bioavailable and may leach through the soil.

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