CHARACTERIZATION OF URBAN SOIL POLLUTION

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Abstract
The objective of the project has been to provide a factual foundation describing the characteristics of diffuse soil pollution in urban areas. Investigation strategies to optimise data collection and predict soil contamination levels in relation to urban age, type of housing, soil fill etc. have been derived. Geostatistical tools and analysis of pollutant patterns have been used to improve the accuracy of these predictions. Over 900 soil samples have been analysed in 11 historical delineated areas in Copenhagen and a provincial town in Denmark. Point sources of pollution have been avoided. All soil samples are analysed for arsenic, copper, chromium, lead, nickel, zinc and PAH. A subset have been analysed for cadmium, mercury, total hydrocarbons (oil, tar), dioxins, PCB's, phthalates and pesticides. A close relationship between urban age and elevated levels of lead, zinc, cadmium, mercury, PAH and dioxins has been established. Impacts from the other parameters are negligible, but background levels are established. Impacts from traffic cause elevated levels of lead, copper, zinc and PAH in the upper soil layer within 10 - 20 m of the road. A geostatistical data treatment has been used to assist in mapping of diffuse soil pollution.

Introduction

The Agency of Environmental Protection in Copenhagen under the Danish Environmental Protection Agency's Technology Program has instigated a project concerning diffuse soil pollution in urban areas (1-6). The project is to provide background information concerning the characteristics of diffuse soil pollution in urban areas and on this basis to devise appropriate investigation strategies.

An important objective related to Danish legislative requirements is to describe the soil pollution with a sufficient degree of certainty, so that an overall investigation of diffuse pollution within a defined area can be assumed to represent each individual plot of land within the area. As land plots identified as polluted are subject to recommendations and restrictions concerning usage, knowledge of the accuracy of the predictions based on the soil investigation is of great importance. To assist in the process, systematic investigation strategies and planning processes have been developed and applied in an investigation of diffuse soil pollution in urban areas in Copenhagen and a provincial town in Denmark.

Methods
The basis for an investigation strategy is that the diffuse soil pollution via the historical description of activities for the area can be related to one or more events that caused the overall diffuse pollution of the area, and can be characterised by a conceptual model.

The planning of an investigation strategy for mapping of diffuse contaminated areas comprises the following systematic steps:

- Preparation of the historical description for the area.
- Drawing up of one or more conceptual pollution models to describe the diffuse soil pollution.
- Demarcation and definition of area(s) exposed to the same sources of pollution and similar historical development.
- Definition of hypotheses, which need to be clarified during the course of the investigation.
- Choice of suitable analytical parameters and measurement techniques.
- Drawing up of a sampling and analytical programme (one or more phases).

The Historical Description: The historical description needs to include identification of potential sources of diffuse soil contamination, the route(s) or events that led to contamination of the soil environment. Known point sources (hotspots) in the area of interest need to be identified.

The Conceptual Pollution Model: The conceptual model describes the way in which diffuse soil pollution can have occurred and takes account of the source, the emission, the spreading and expected loading of the soil environment. Based on our present knowledge concerning sources of pollution and experiences acquired by investigation of diffuse soil pollution, five pollution models have been defined, see figure 1.

Deposition model: A soil impact that originates from airborne emissions (dust, gases) from one or many point sources, e.g. incinerator stacks, coal fired power stations, crematories etc.
The diffuse soil pollution decreases in concentration with distance from the source and the area of deposition is dependent on wind conditions, topographical and physical aspects around the sources and the nature of the emission, concentration, chemical and physical form, particle size, temperature etc. The pollution is most evident in the topsoil and decreases with depth.

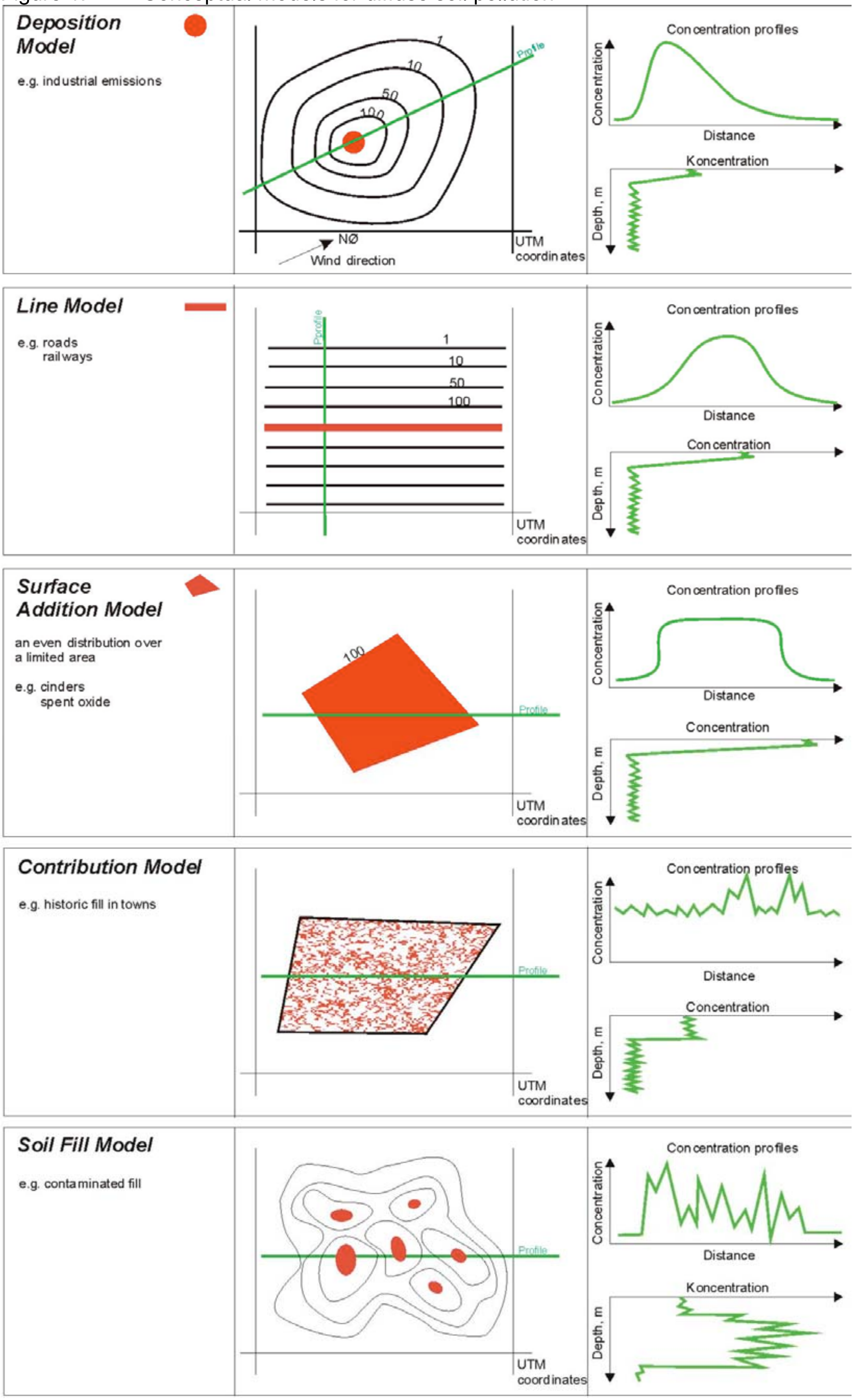
Line model: A soil impact that originates from moving point sources aligned along a long linear element in the landscape, e.g. a road, railway etc.
The diffuse soil pollution decreases with distance for the line source and with depth in the soil profile.

Surface addition: A soil impact that originates from the spreading of a contaminating material, e.g. former uncontrolled spreading of cinders, sewerage sludge, sediment, spent oxide from gasworks etc.
The diffuse soil pollution constitutes a uniform load in the upper topsoil.

Contribution model: A soil impact that originates from small random contributions through the years (10 – 200 years), e.g. the historic fill on top of which a town develops.
The diffuse soil pollution constitutes a variable and random load to the historic fill and the depth is dependent on urban age. Urban age, dumping of waste, domestic heating, housing, demolition and town fires are thought to play a major role.

Soil fill model: A soil impact that originates from the systematic dumping or filling with soil, waste or other material of unknown origin, e.g. land reclamation, backfill, of low lying areas and coast regions, construction of harbours, construction activities, earthworks, etc.
The diffuse soil pollution constitutes a variable and random load to the soil fill and can extend to greater depth.

Figure 1: Conceptual models for diffuse soil pollution



Investigation sites: Three types of diffuse soil pollution have been investigated in Copenhagen city (established 1100 AD inner city population 500,000, Greater Copenhagen population 1,200,000) and in a provincial historic town (Ringsted, established 1000 AD, population 67,000) in 2002/2003.

- Diffuse soil pollution caused by urban sprawl is related to the type of housing, housing materials, construction age, sources of domestic heating, soil fill etc. - contribution model
- Diffuse soil pollution caused by emissions from traffic – line model
- Diffuse soil pollution caused by atmospheric deposition from industrial sources – deposition model.

The investigation sites are as follows:

- 5 housing areas (terraced housing from 1600, 1890, 1930 and modern housing estates from the 1950´s and 1960´s) in Copenhagen.
- A park (formerly a historic defensive earthwork) in Copenhagen.
- 5 housing areas (terraced housing from 1880, detached housing (villa) from 1915-1920, 1940´s, 1950´s and 1980´s) in a provincial town - Ringsted
- A 1.6 km section of main road in the city of Copenhagen (established 1920, 4 lanes, 90 km /h, 50,000 cars / day).
- A 0.7 km section of road in the city of Copenhagen (established in 1900, 2 lanes, 60 km /h, 20,000 cars / day).
- A housing estate in an industrial suburb of Copenhagen from the 1930´s close to an industrial metal processing facility.

Analytical parameters: The choice of analytical parameters is related to the conceptual model, but all soil samples are analysed for arsenic, copper, chromium, lead, nickel and zinc by X-ray fluorescence and for 7 PAH´s (fluoranthene, benzo(b)fluoranthene, benzo(j)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene (BaP), dibenz(a,h)anthracen and indeno(1,2,3-cd)pyrene) by GC-MS. A portion of the samples are also analysed by ICP-AES for the above mentioned metals as well as cadmium and mercury. Some of the samples are analysed for composition of the PAH profile PAH´s (about 30 PAH´s including alkyl derivatives) to evaluate patterns of composition according to source of pollution in diffuse urban soil pollution. Furthermore, a number of samples are analysed for dioxins, PCB's, phthalates and pesticides.

Data collection: Soil samples have been taken from sub sampling areas of 4-10 m², randomly distributed across the area under investigation. In each sub sampling area up to five samples are taken for evaluation of the sampling heterogeneity. For a few samples, 2-6 replicate analyses are carried out. No composite samples are used, and samples are taken from different soil profiles in the topsoil; 0-5, 2-10, 20-30, 45-55 and 95-105 cm´s depth. Most samples are taken in 2-10 cm´s depth. The sub sampling areas are randomly allocated across the area under investigation, and these areas are then physically inspected to ensure suitability. Any sites with clear evidence of newly turned soil, recently established gardens or buildings, soil excavation or new backfill are rejected for the purposes of this study. Any site with registered or potential sources of point pollution is avoided, as the purpose of the investigation is to establish the level for diffuse soil pollution arising from a common source in the defined area. Hotspots are therefore avoided. The purpose is to characterise diffuse soil pollution and provide reference levels for the urban diffuse pollution in general and not to identify point sources of pollution.

Data treatment: The results are characterised by simple statistical parameters. The data from many of the areas investigated does not appear to be normally distributed. For some substances, the data is skewed to the right (many low values especially common around the detection limits for metals such as chromium and arsenic) and for other substances skewed to the left (many elevated values). For nearly all sites, a few very high values are present. To describe the data, the mean, median, minimum, maximum and selected quantiles are calculated. Quantile plots, which makes no assumptions about the data distribution and show all data points giving a graphical representation of the data are used for a visual presentation of the data. The geostatistical data treatment requires comparison of variance for pairs of data at different distances. To estimate the nugget effect which describes the combined sampling and analytical variation for samples taken from the same position, it is necessary to have sufficient data points for samples at small distances from each other otherwise this part of the semivariogram (interception on the Y-axis) is poorly estimated. Diffuse soil pollution in the same depth in each sub sampling area is assumed to be homogenous or to exhibit a relationship with distance from the source. The measured analytical variation in each sub sampling area is therefore due to the micro/macro scale heterogeneity in the soil and to the mechanism for deposition/distribution of pollution in the soil.

Results

I figures 2 - 8 the results are illustrated with the median values for Pb, Cu, Cd, Hg Zn, BaP and dioxin. As can be seen, the diffuse soil pollution can be related to the age of development since this determines the type of housing, housing materials, construction age, sources of domestic heating and duration of soil exposure to pollutant loads. Generally the soil contamination decreases with depth in the soil profile, but extends deeper in the soil profile in the older areas. Contamination levels for heavy metals are lower in the provincial town, Ringsted, than in Copenhagen, but the PAH levels (as illustrated by BaP) are comparable for areas of similar age in both Ringsted and in Copenhagen.

The influence of the rolling mill at the metal processing plant is clearly seen for parameters such as lead, copper, cadmium and zinc, since levels are higher than expected according to the age of development for the area. A higher natural background level for cadmium in the Ringsted area is also clearly seen at all depths in the soil profile.

Figure 2: Median for lead content in soils with different ages for housing development

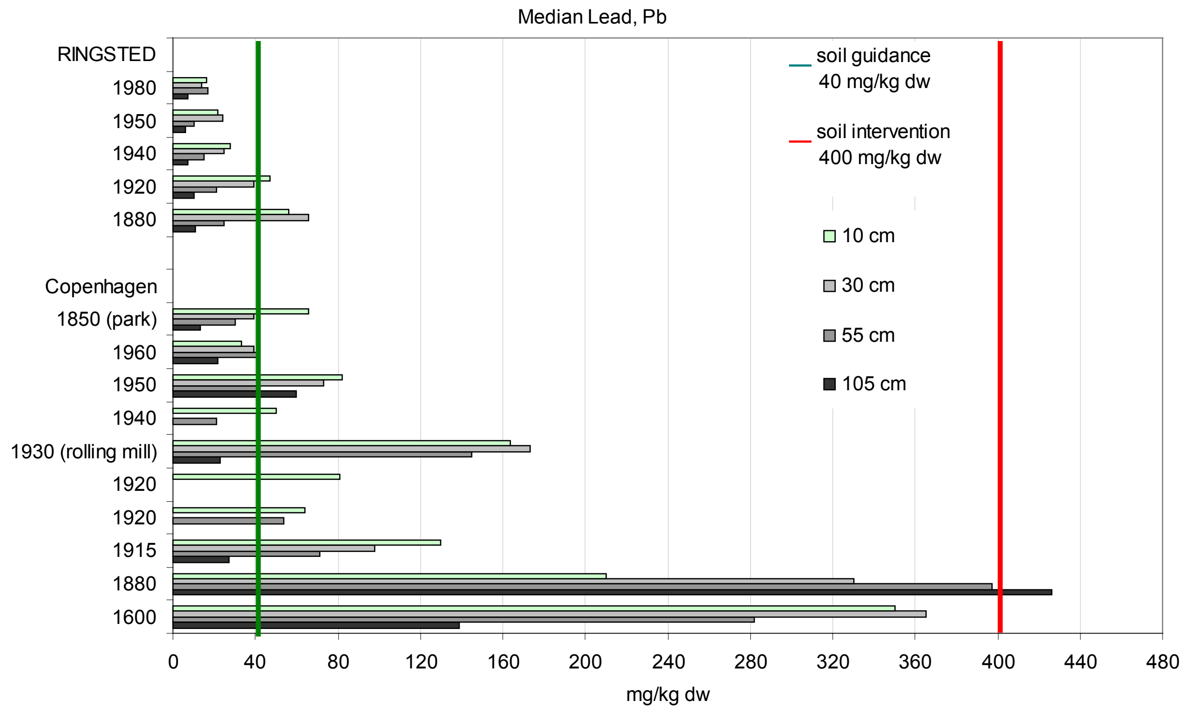


Figure 3: Median for copper content in soils with different ages for housing development

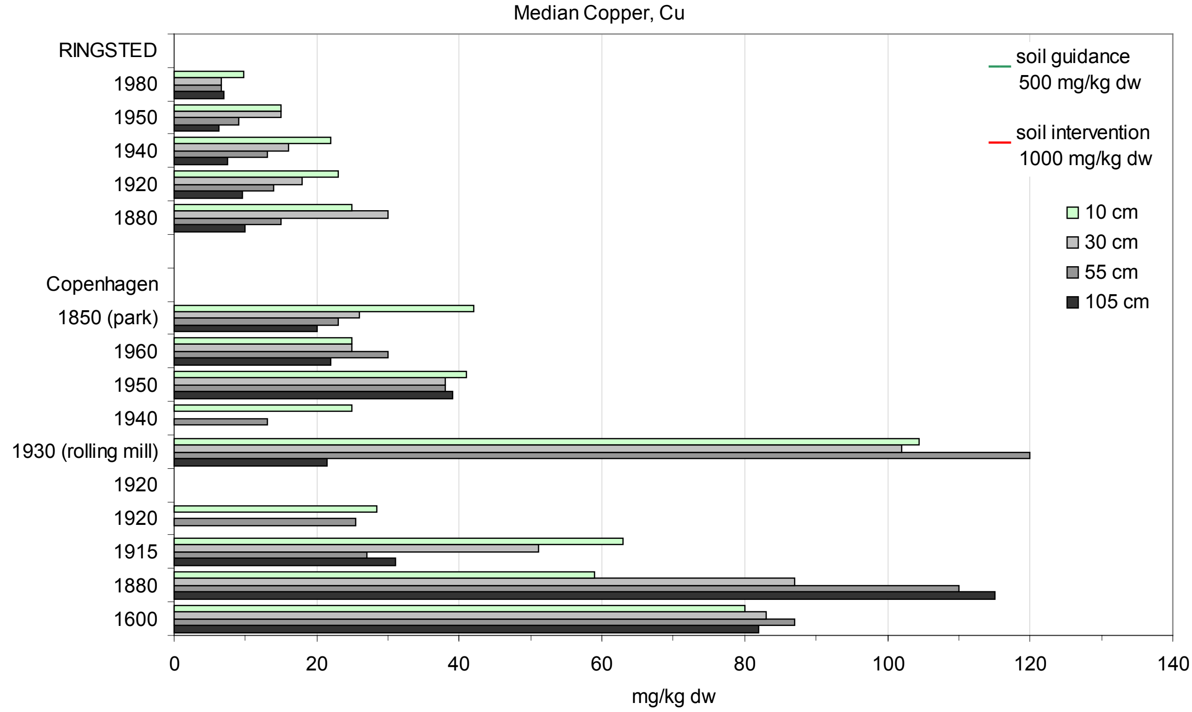


Figure 4: Median for cadmium content in soils with different ages for housing development

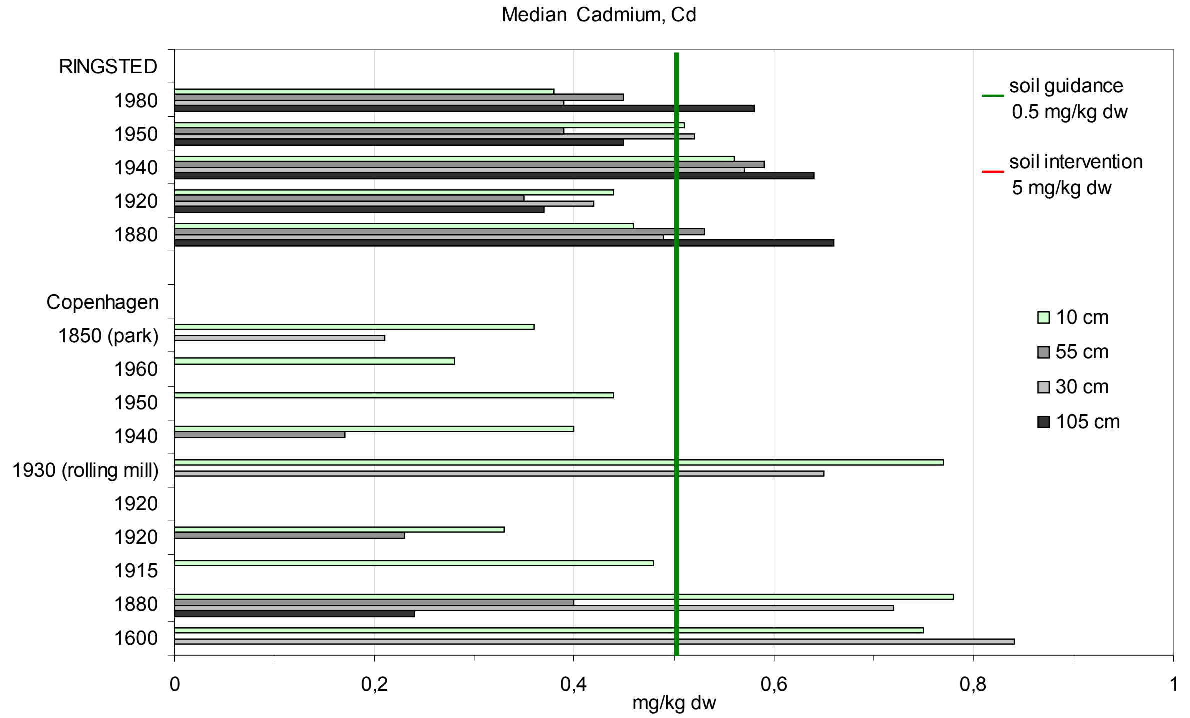


Figure 5: Median for zinc content in soils with different ages for housing development

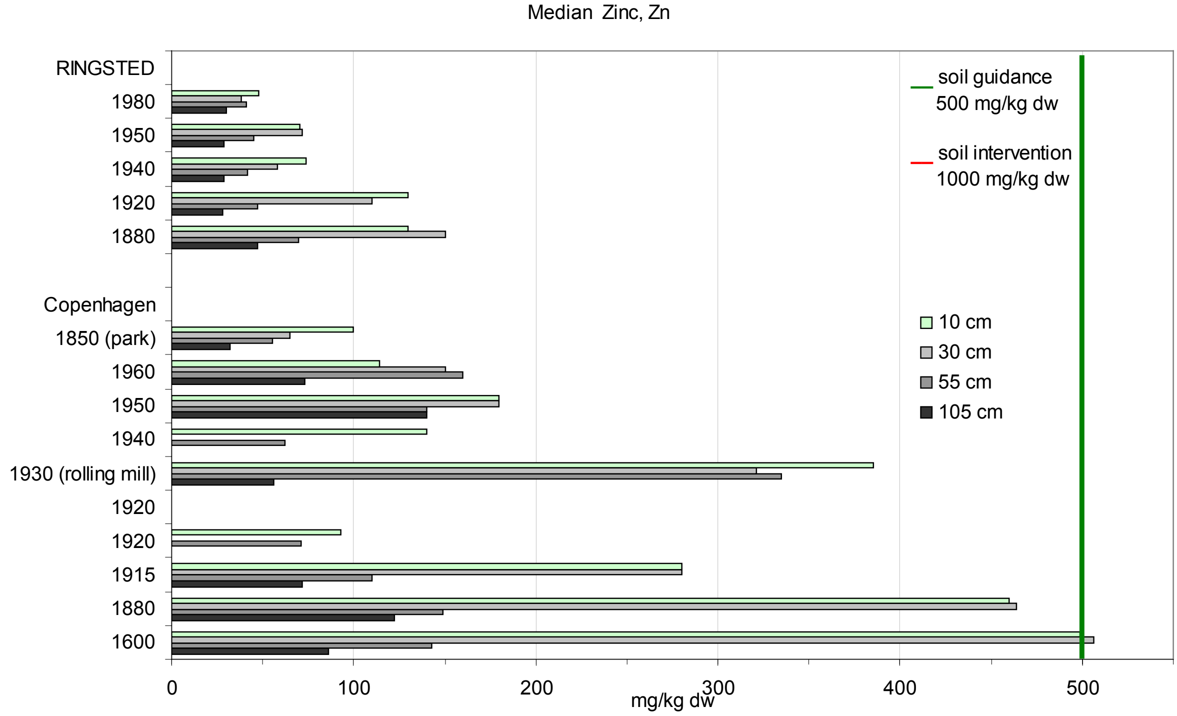


Figure 6: Median for mercury content in soils with different ages for housing

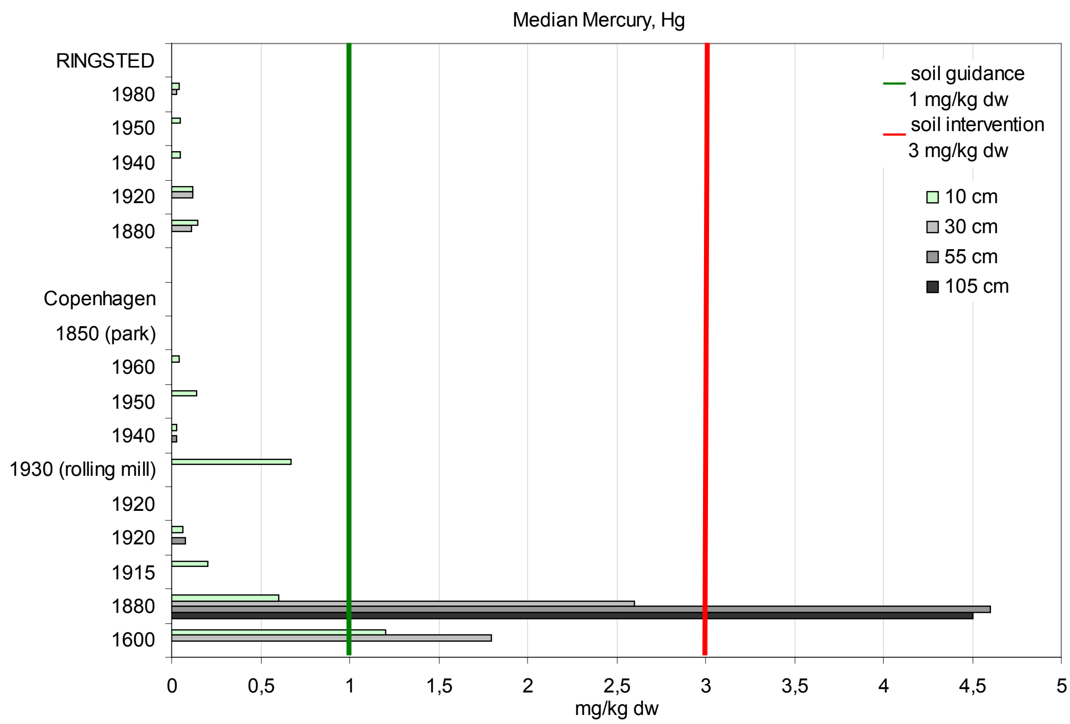


Figure 7: Median for BaP content in soils with different ages for housing development

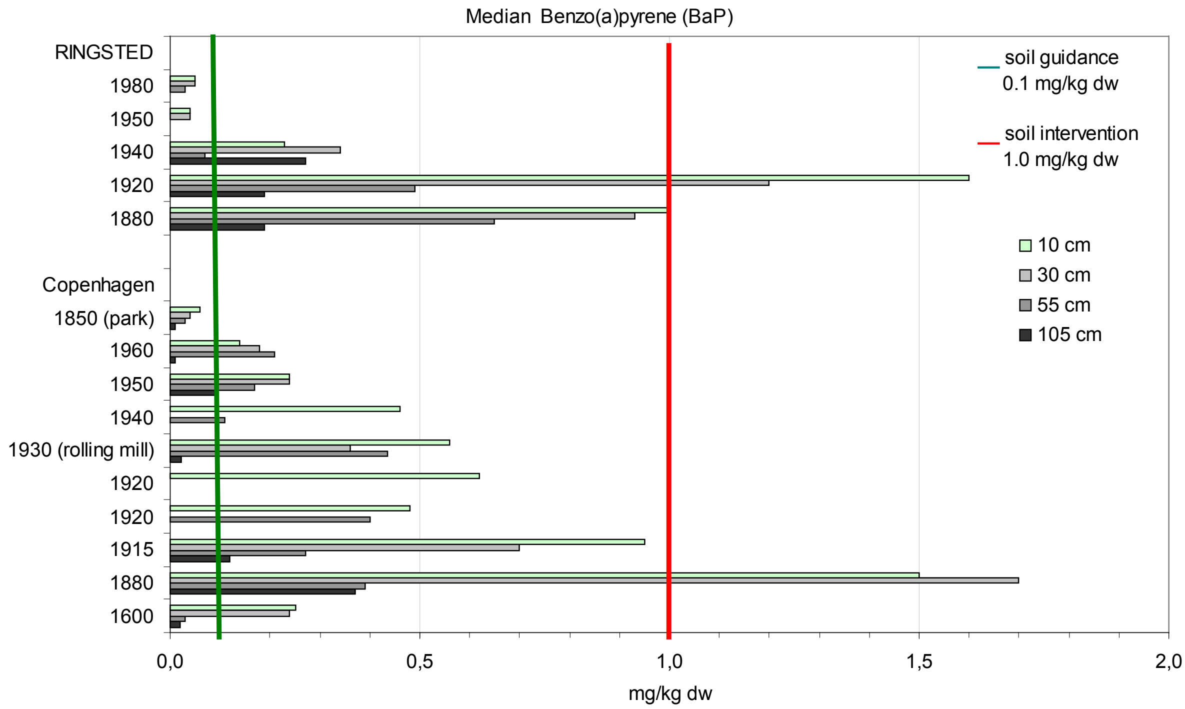
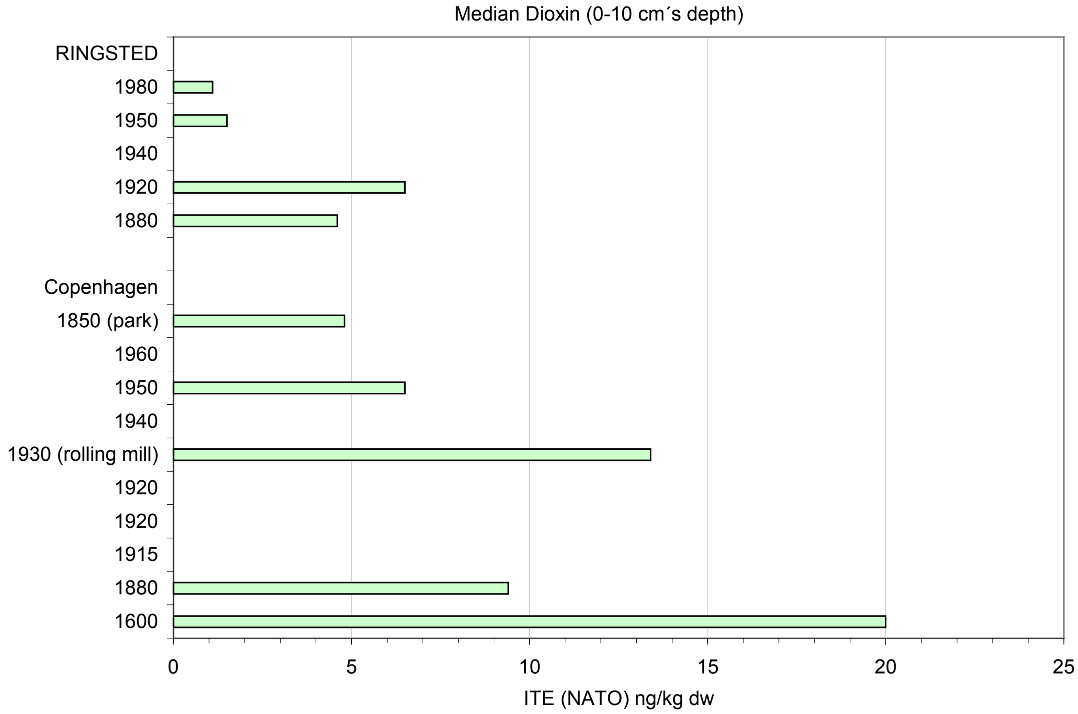


Figure 8: Median for dioxin content in soils with different ages for housing development

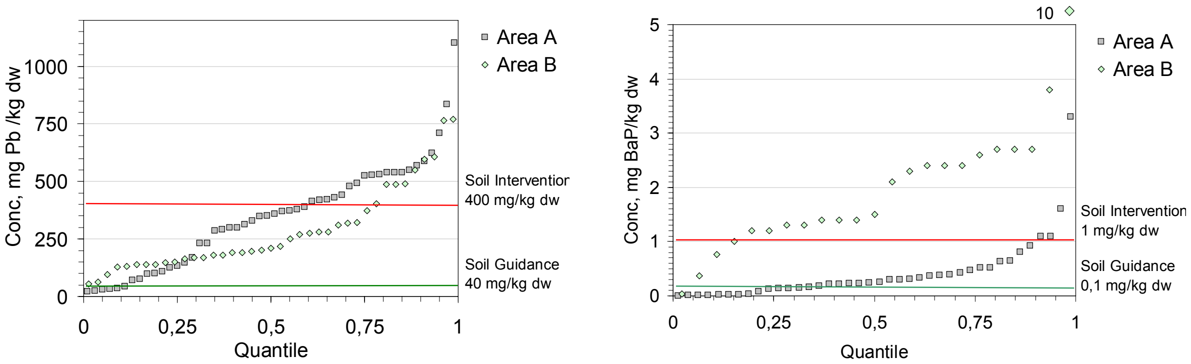


Generally some correlation was found between the content of lead, copper, zinc and cadmium (R^2 >0.65), but there was no correlation between the content of metals and PAH (R^2 <0.5).

The concentration levels of arsenic, nickel and chromium are not elevated in urban areas. Polychlorinated biphenyls (PCB) are generally not found in soil samples from areas with urban housing (PCB is not detected in 90% of samples). Low content of phthalates are measured, but levels are greatly below the Danish soil quality criteria of 250 mg/kg dw (Phthalates are not detected in 30% of samples, and only one sample had a content of more than 1 mg/kg dw). All the analysed soil samples had a low content of dioxins (1 - 20 ng international toxic equivalents (ITE)/kg dw), see figure 8. These values can be compared with the background level in rural and urban areas in Germany of 1- 5 ng ITE/kg dw and 10 – 30 ng ITE/kg dw respectively. The content of persistent pesticides was analysed in 10 soil samples, but only low levels of DDT and parathion were found in four of the 10 samples.

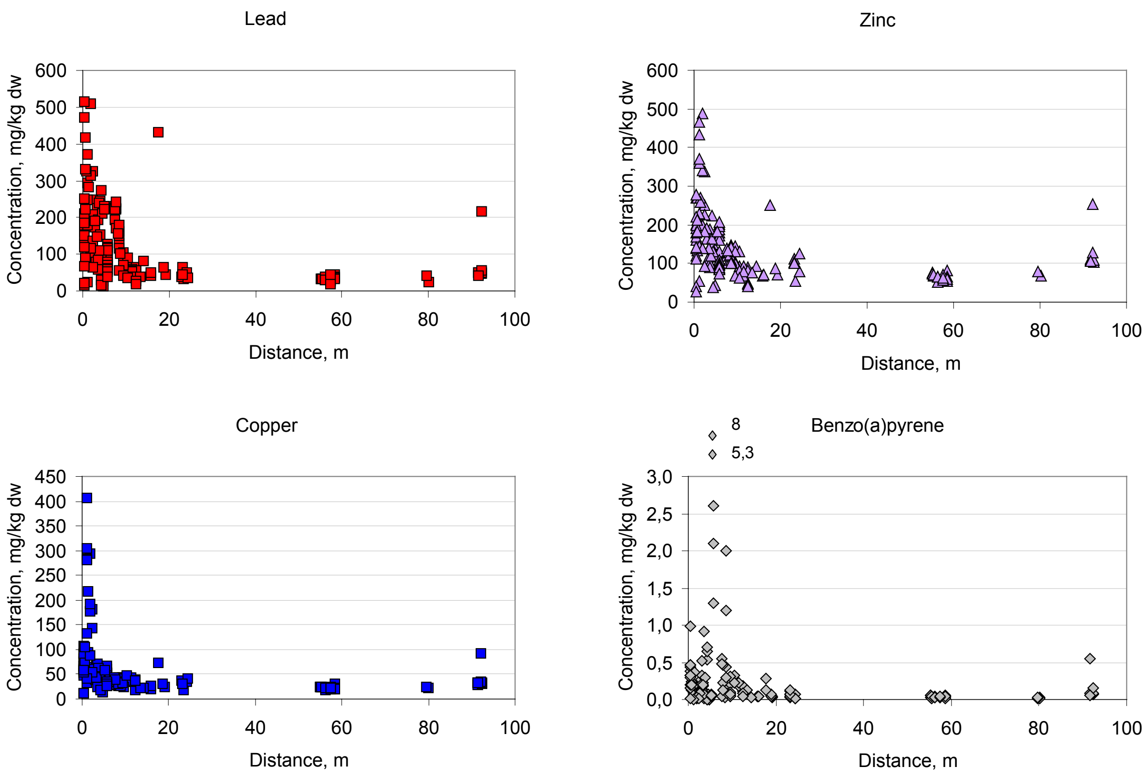
During the project, various forms for data presentation have been compared and in figure 9, a comparison of results for lead and BaP in two different areas is illustrated using a quantile plot, which shows all data measurements – the 0.5 quantile is the median value. Figure 9 illustrates the fact that high soil levels of BaP are not necessarily correlated with high soil levels of lead.

Figure 9: Quantile plot for lead and BaP content in soil samples from 0-10 cm from two different areas



I figure 10, diffuse soil pollution caused by emissions from traffic and wastewater from the road surface is illustrated.

Figure 10: Content of Pb, Zn, Cu and BaP in soil samples (0 - 30 cm) as a function of distance from the road verge.



It is assessed that soil pollution is caused by deposition of emissions from petrol combustion motors and diesel motors, by road dust and water (contaminated with motor oils, tires, and gaseous emission) which is sprayed by the passage of traffic on to the verges. Road constructions materials especially roads that previous have been established with coal tar asphalt will also contribute to soil pollution.

As can be seen in figure 10, a large range of values are measured close to the road verge, but generally soil within 10 - 20 m of the road is contaminated with lead, copper, zinc and PAH (as illustrated by BaP) in the upper soil layer (0 – 30 cm´s). Figure 10 also indicates that elevated copper contents are only observed close to the road, presumably because spreading to the soil occurs via contaminated water spray from the wet roads and not by gaseous emission.

Generally some correlation was found between the content of lead, zinc and cadmium (R^2 >0.8) in samples taken from the 0 – 10 cm topsoil and within 10 m´s of the road verge, but there was no correlation between the content of metals and PAH (R^2 <0.2).

Discussion

Geostatistics allows an interpretation of the spatial variability across an area. It allows estimation of concentration levels at positions within the area, where no measurements have been made. It provides information on the range of influence for the correlated data and gives an estimate of the uncertainty of the prediction of concentration levels. In figure 11, the predicted concentration in soil samples at any given position in an area is illustrated. In figure 12-13, the probability that a soil sample at any given position in the area exceeds the soil guidance limit and is less than soil intervention limit is illustrated.

Figure 11: Prediction of lead content in a sample from a given position within the investigation area

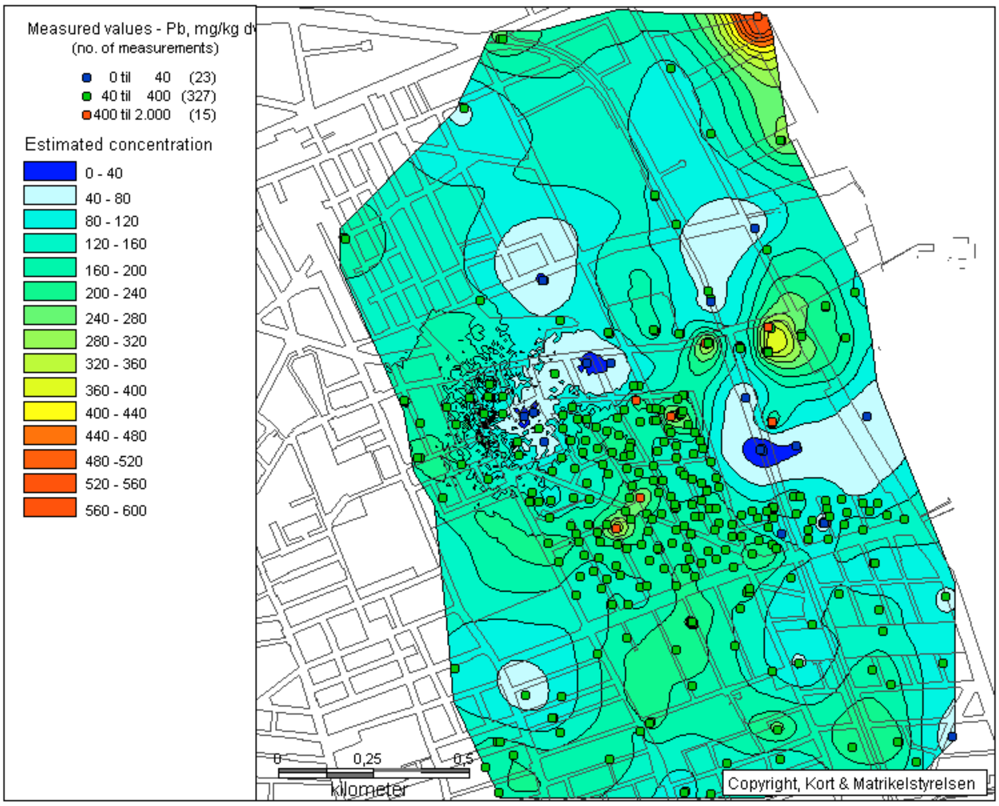


Figure 12: Probability that a sample from a given position within the area exceeds the soil guidance limit (40 mg Pb/kg dw).

Figure 13: Probability that a sample from a given position within the area is less than the soil intervention limit (400 mg Pb/kg dw).

Not all investigation areas proved to be amenable to a geostatistical analysis. The number of data position and the size of the area in comparison with the variation due to pollutant heterogeneity required a more detailed investigation to be performed.

Conclusions

- Urban soil pollution is often dependent on the age for land development
- Lead and PAH/BaP are the pollutants that most frequently and consistently exceed soil guidance and occasionally soil intervention limits
- Cadmium can exceed the soil guidance limit in some areas
- Mercury can exceed the soil intervention limits and occasionally the soil intervention limit in old parts of the city
- Elevated levels of copper and zinc are always seen in urban areas
- Only low levels or levels under detection limits are seen for PCB, phthalate, dioxins and pesticides
- Dioxin levels are slightly elevated in comparison with background levels in rural areas
- Lead, zinc and cadmium are often correlated
- PAH and lead are not correlated
- Pollution from traffic is usually restricted to a 10 - 20 m corridor
- Geostatistics is useful in the mapping of diffuse soil pollution

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