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**A MAGNETIC SUSCEPTIBILITY STUDY OF SOME
SWISS SOILS:
ENVIRONMENTAL ASPECTS**

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Editor's Preface

The present publication entitled "A magnetic susceptibility study of some Swiss soils: Environmental aspects " is report number. 37 of the "Contribution to the Geology of Switzerland - Geophysical Series", published by the Swiss Geophysical Commission.

It contains a very interesting application of the measurement of soil susceptibility for determining the extent of heavy metal pollution. The authors begin by giving an overview of the methodology used. In the second part of this publication they study in two test areas, (Basel and Biberist / Gerlafingen), the correlation between the magnetic susceptibility, determined at two different frequencies, and the lead content of the soils. The results are presented in the form of maps that show a clear correlation. Finally a study of the remanence properties of the iron oxides is presented.

This study clearly shows that the measurement of the magnetic susceptibility of the first 20 cm of the soil can be used as a first step in the determination of the pollution because it is simple, quick and inexpensive.

The Swiss Geophysical Commission is most grateful to the authors for having realized this very interesting work and demonstrating that simple geophysical methods can be useful for environmental purposes. The commission also hopes that this methodology will be extended in the future and routinely applied.

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Zurich, Mai 2002

In the name of the
Swiss Geophysical Commission
The President :

Prof. Emile Klingelé

1. Introduction

The magnetic study of sedimentary sequences from peat bogs, lakes and oceans allows one to trace the history of the deposition of magnetic particles from the atmosphere. For example, magnetic measurements on lake sediments in Great Britain, Scandinavia and North America have shown the presence of pollutants from the beginning of industrialization in these countries as well as the later increase in the atmospheric pollution.

Even before the suspended particles are deposited on the soil surface, they present a considerable health hazard and many studies have been carried out to understand their source and nature (Anonymous, 1979). Soils in urban areas can show high levels of pollutants, for example lead concentrations that even reach the percent level (Solomon et al. 1976) leading to the risk of lead intoxication, particularly in young children.

This pollution can come from heavy industry, thermal power stations, incineration plants, as well as vehicle exhaust gases. The fine magnetic particles carried by these hot gases are dispersed at varying distances on to the surrounding soil surface (Hunt et al., 1984, Beckwith et al., 1986) and they can be distinguished magnetically from the natural iron oxides present in the soil (Oldfield et al., 1985).

For example, recent studies carried out on a national scale in England (Hay et al., 1997), have shown that these anthropogenic particles in soils have a magnetic signature that can be readily recognized and also that there exists a correlation between the concentration of heavy metals and the magnetic fraction of the pollutants (Linton et al., 1980, Olsen and Skogerboe, 1975).

Increasing use of magnetic measurements on soils in Germany (Hoffmann et al., 1999) and the Czech Republic (Kapicka et al., 1999, Petrovsky et al., 2000) has led to the suggestion that they can be advantageously used as a proxy method for the evaluation of environmental pollution. Instead of onerous chemical analysis, magnetic methods, after a field calibration, could provide an inexpensive and rapid tool for delimiting the extent of atmospheric pollution in topsoils.

2. Methodology

This present study is part of the *Project Magsol* begun in 1998, whose aim is to use magnetic susceptibility measurements to examine the pollution of topsoils in Switzerland. All samples come from existing soil collections in the different cantons. These collections were assembled by each canton in response to the Federal directives concerning the quality of soils in Switzerland (OSOL, 1989), and the sampling procedure adopted follows strict guide-lines (OFEFP/FAC, 1997). The soils of this study are from the uppermost part of the soil profile; that is 0-20 cm.

In the laboratory a Bartington MS 2 susceptibility meter was used for the basic magnetic susceptibility measurements using a model B dual frequency sensor. The soil samples were held in standard 10ml cylindrical plastic bottles.

By using two different frequencies (0.46 kHz and 4.6 kHz) additional information is obtained on the granulometry of the very fine magnetic grains present. This concerns

those very fine grains of magnetite/titanomagnetite that are close to the superparamagnetic (SP)-stable single domain (SSD) transition, and which have a diameter of 20-30nm.

In the measurement of magnetic susceptibility a weak magnetic field (80 A/m) of the same order of magnitude as that of the earth is applied to the sample and all the mineral grains, in the complex mixture that constitutes a soil, contribute to a greater or smaller extent. It is the ferrimagnetic minerals, such as magnetite (Fe_3O_4), pyrrhotite (FeS_{1+x}) or greigite (Fe_3S_4) that contribute the most important signal followed by the more weakly magnetic haematite ($\alpha\text{-Fe}_2\text{O}_3$) and goethite ($\alpha\text{-FeOOH}$) but the paramagnetic ferromagnesian minerals such as biotite, olivine and pyroxenes can also contribute. The clay minerals have quite low susceptibilities whilst the diamagnetic minerals such as quartz and calcite have very low and negative susceptibilities and so can reduce the magnetic signal of the soil (Dearing, 1994).

A further more detailed magnetic analysis was also undertaken on certain samples to characterize the nature of the magnetic minerals present in these soils in order to understand their relationship to the source of pollution.

Two different settings are presented here, the first in an urban environment and the second in a clearly industrial one, that of iron smelting.

3. Basel

Many different sources of air pollution exist in built-up zones and originate mainly from various combustion processes; heavy industry, incineration plants, as well as road and rail traffic.

The urban conurbation of Basel was chosen as example because of the contrast of a dense city center that changes rapidly to the rural countryside as well as the availability of suitable soil samples. The area studied here (figure 1) consists of all of the city (BS) and the northern western part of the other half canton, that of Basel country (BL).

It is estimated that a total of 1000 tons of atmospheric dust per year (particles less than 10 micron, PM10) falls over the whole of the region of Basel, which amounts to an average of 2g per m^2 . Some 33% of this dust comes from the road traffic and another 30% from industry and incineration (Anonymous, 2001).

The magnetic susceptibility varies by more than one order of magnitude and generally shows increasing values with increasing lead content. A few of the samples of the BS survey come from family allotments (familiegarten), which are well known to be highly polluted due to varied “nutrients” added to the soil. Soils in urban areas can have a complex history and can be likened to a “witch’s brew” (C. Hohl, private communication). This could be a factor contributing to the scatter in the susceptibility versus lead plot, where the best fitting straight line (fig. 2) gives a correlation coefficient R^2 of only 0.59.

It should also be mentioned that one sample from BS has been eliminated as it is known to have been heavily contaminated by waste washed from medieval mining activity in the southern Black Forest.

A 2D representation of both the magnetic susceptibility (fig.3) and the lead concentration (fig. 4) clearly shows the close link between them. Higher values are clearly associated with the built-up areas. In the case of BL all but one of the sites whose lead content

Basel region: Topsoil sites

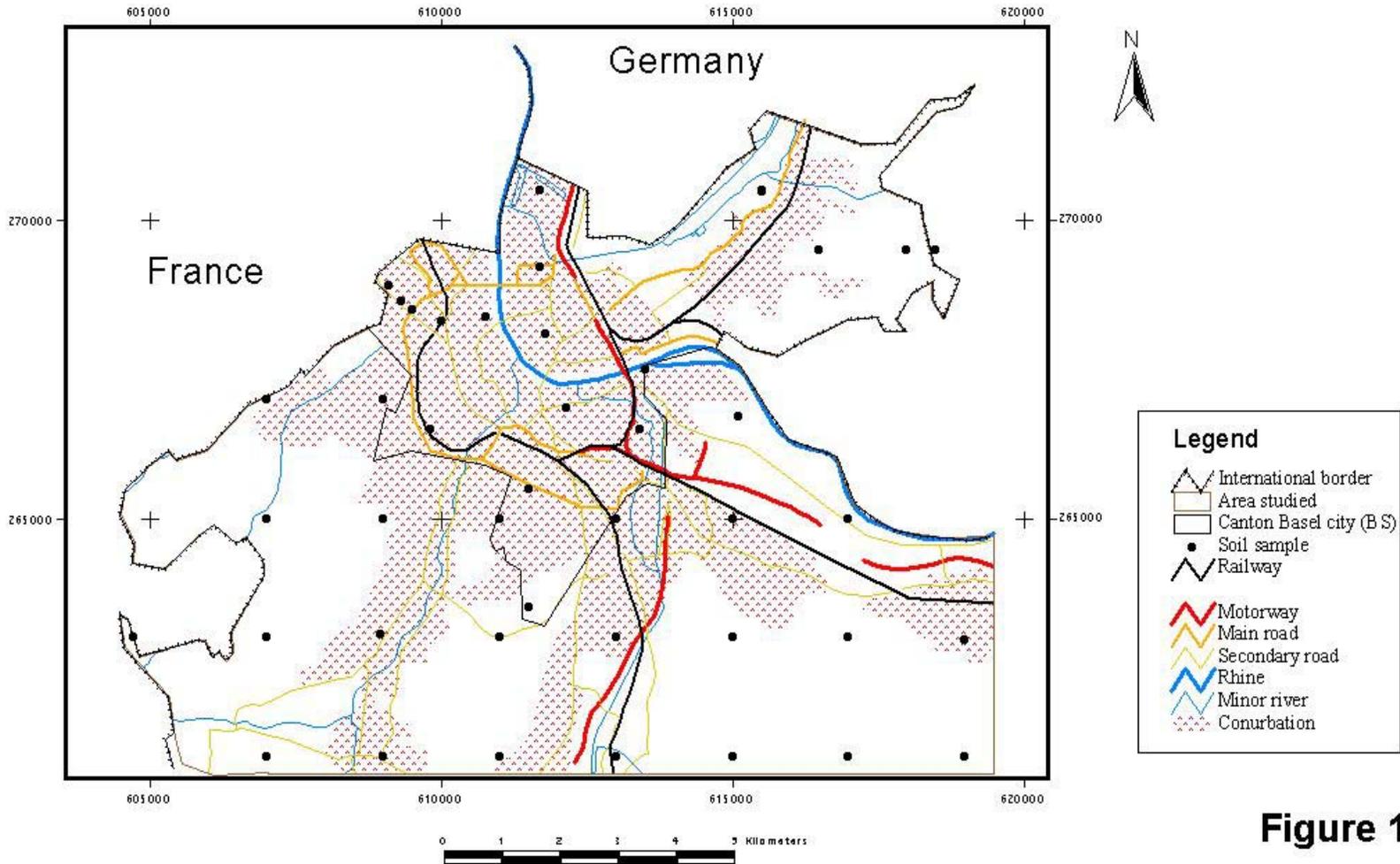


Figure 1

exceeds the federal guide value (50 ppm) are to be found in built-up areas (Bono, 1993).

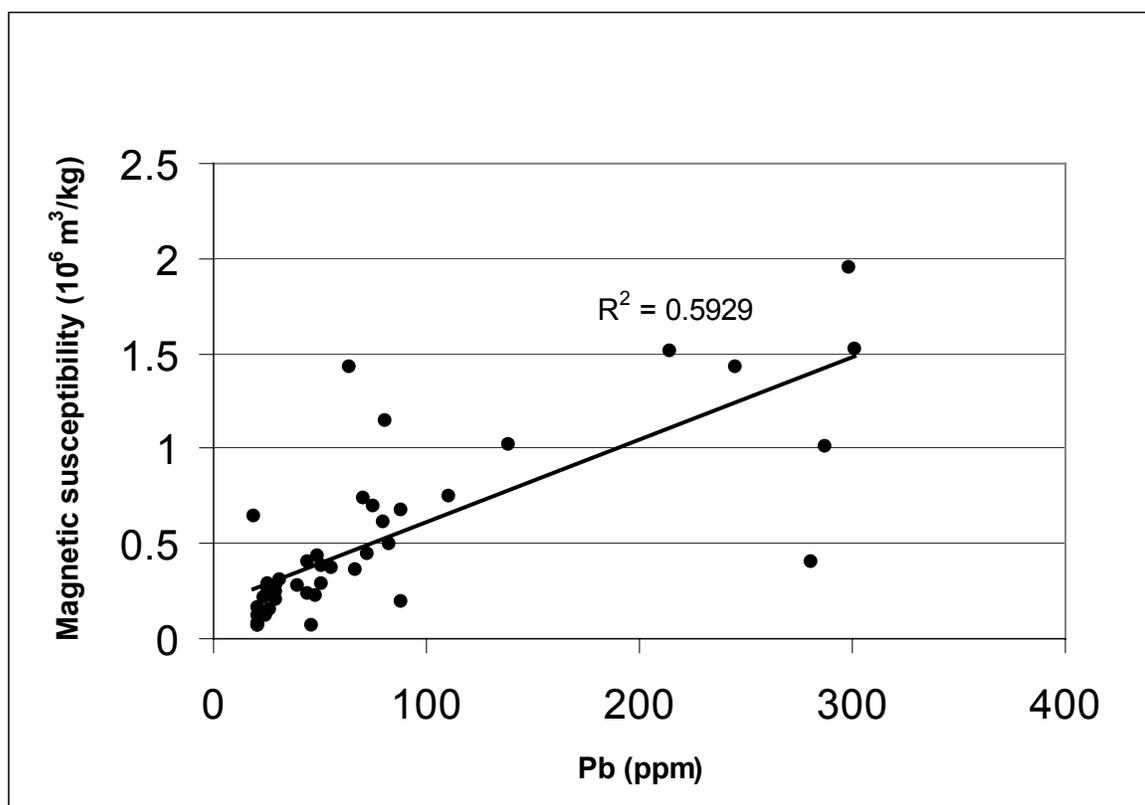


Figure 2. Basel region: topsoil magnetic susceptibility versus lead content.

A geostatistical study of the same BL soil collection but with additional sampling (Genolet, 1995) shows a similar map of the lead distribution to that of the present study. A correlation between the concentrations of lead, zinc (fig 5) and cadmium suggests an anthropogenic origin of these heavy metals, coming from industry or road traffic. The source of the lead is most probably the previous use of leaded petrol, whilst the zinc and cadmium come from lorry and car tyres. Genolet, *loc cit* concludes that the sampling density needs to be increased even further to determine the reasons for the observed spatial distribution, requiring an important investment in time and money.

Further evidence for the link between vehicular traffic and pollution in an urban environment comes from a study by Knab et al.(1998) in SW Germany which showed a high degree of linear correlation between magnetic susceptibility and the Pb, Cu, Cd, and Zn contents of soils from a roadside with heavy traffic. Analysis of urban dust from a moderately busy road intersection (7000 vehicles/day) in Urbana, Illinois (Hopke et al. 1980) showed that the lead (1000ppm total) was preferentially concentrated in the magnetic fraction of the dust. This suggests an intimate link between the magnetic iron oxides and the lead and the reason for this association needs further investigation (Georgeaud et al.,1997).

Basel region: Topsoil Magnetic susceptibility

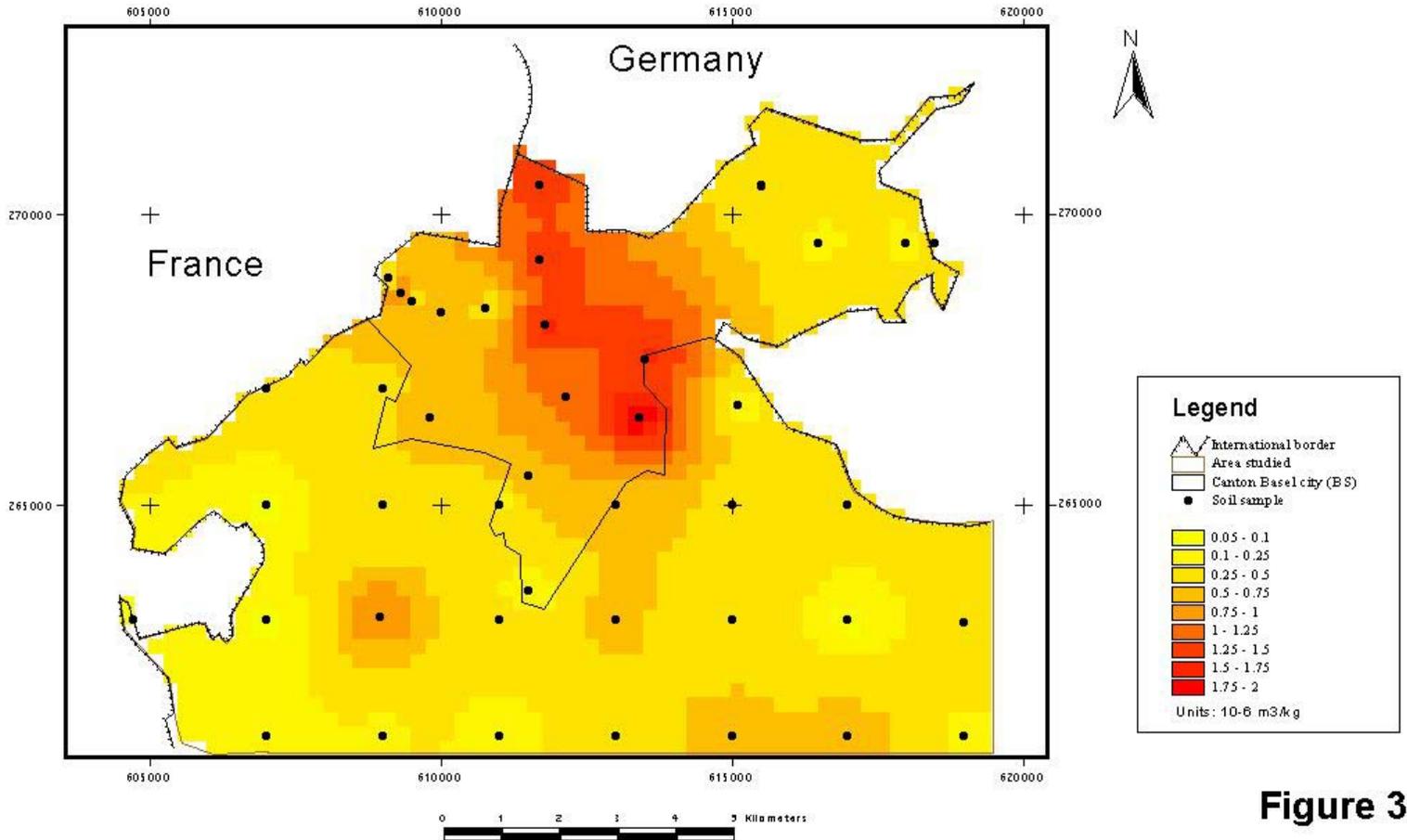


Figure 3

Basel region: Topsoil Lead content

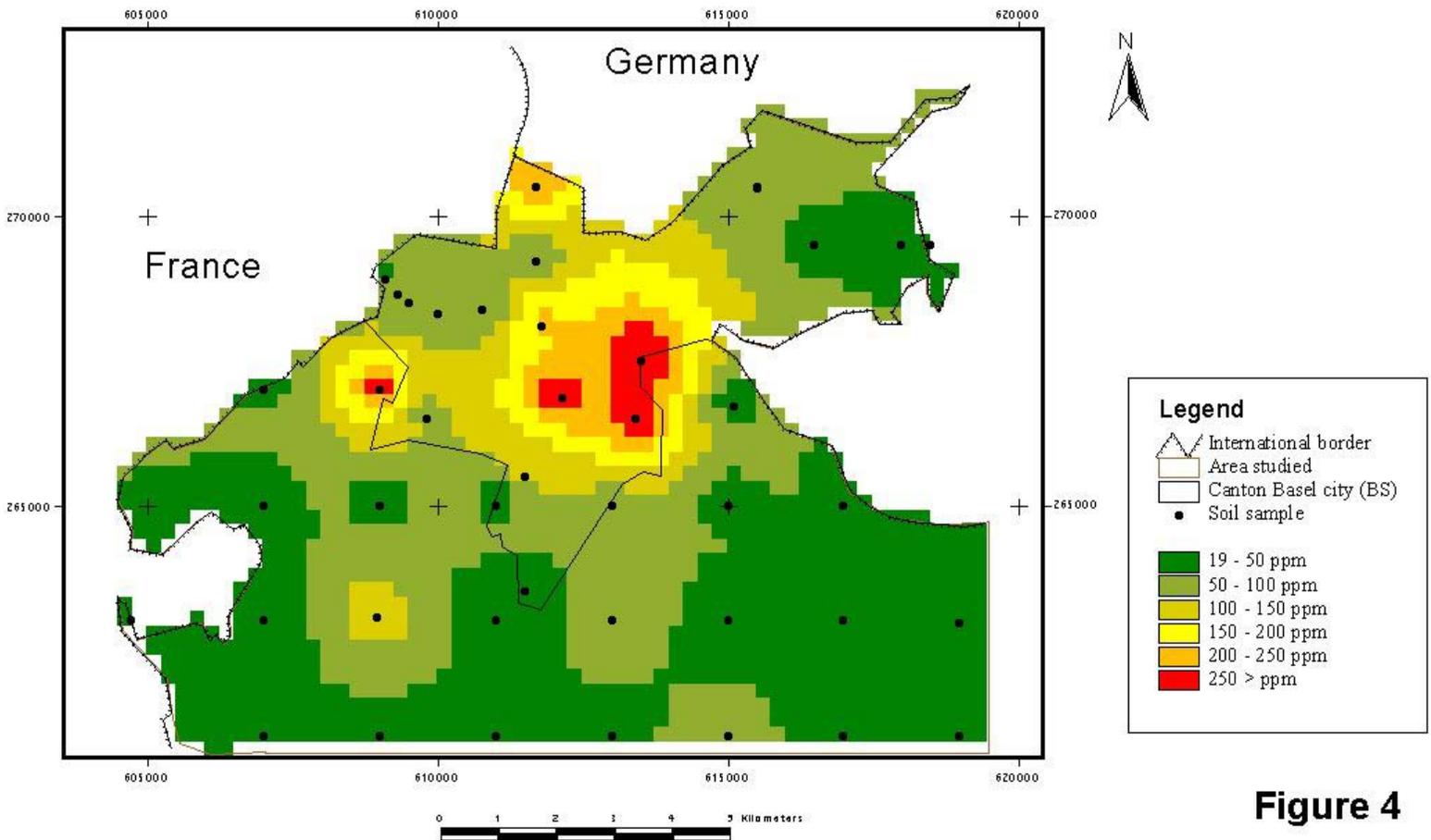


Figure 4

Basel region: Topsoil Zinc content

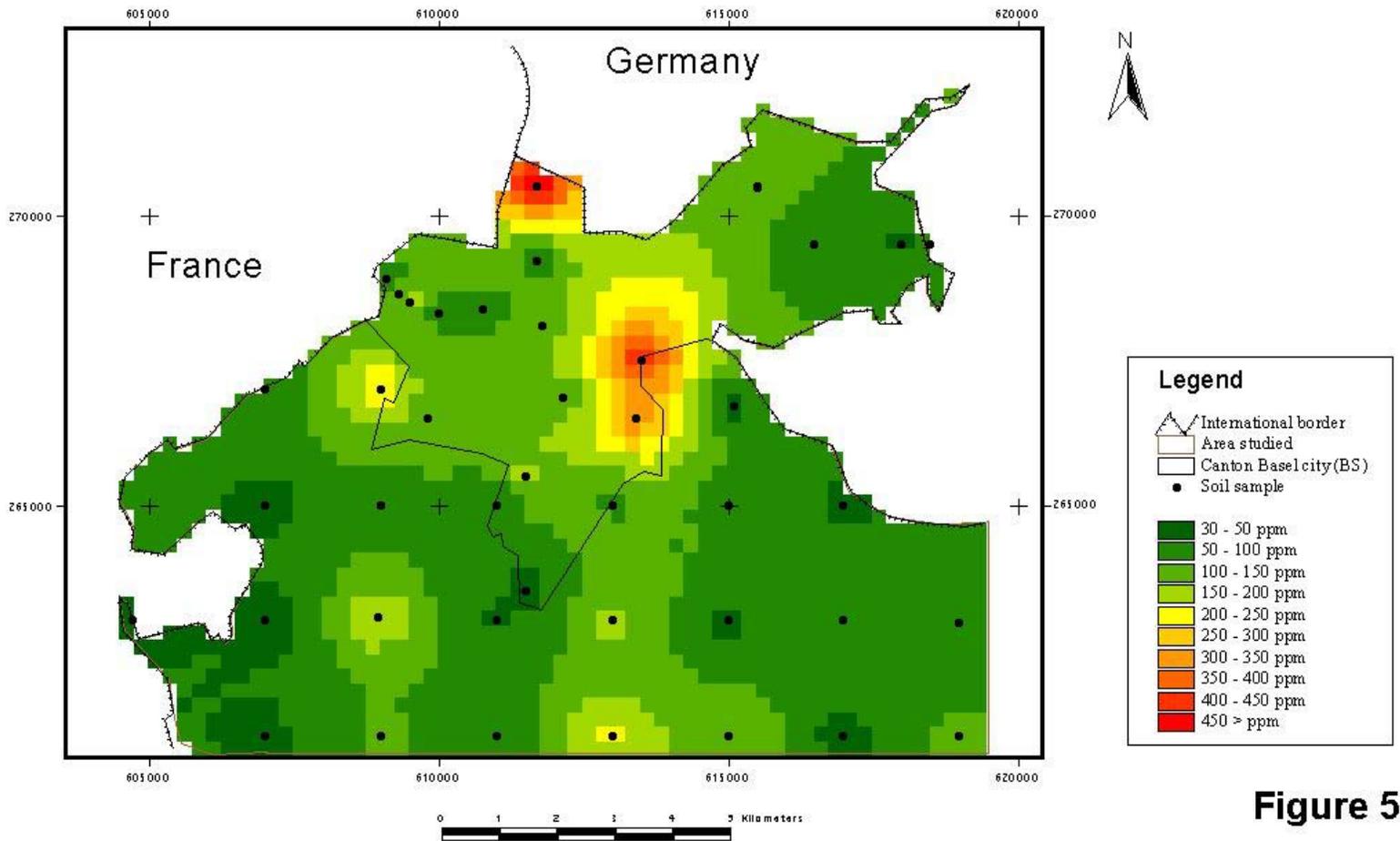


Figure 5

Basel region: Magnetic susceptibility criteria classification

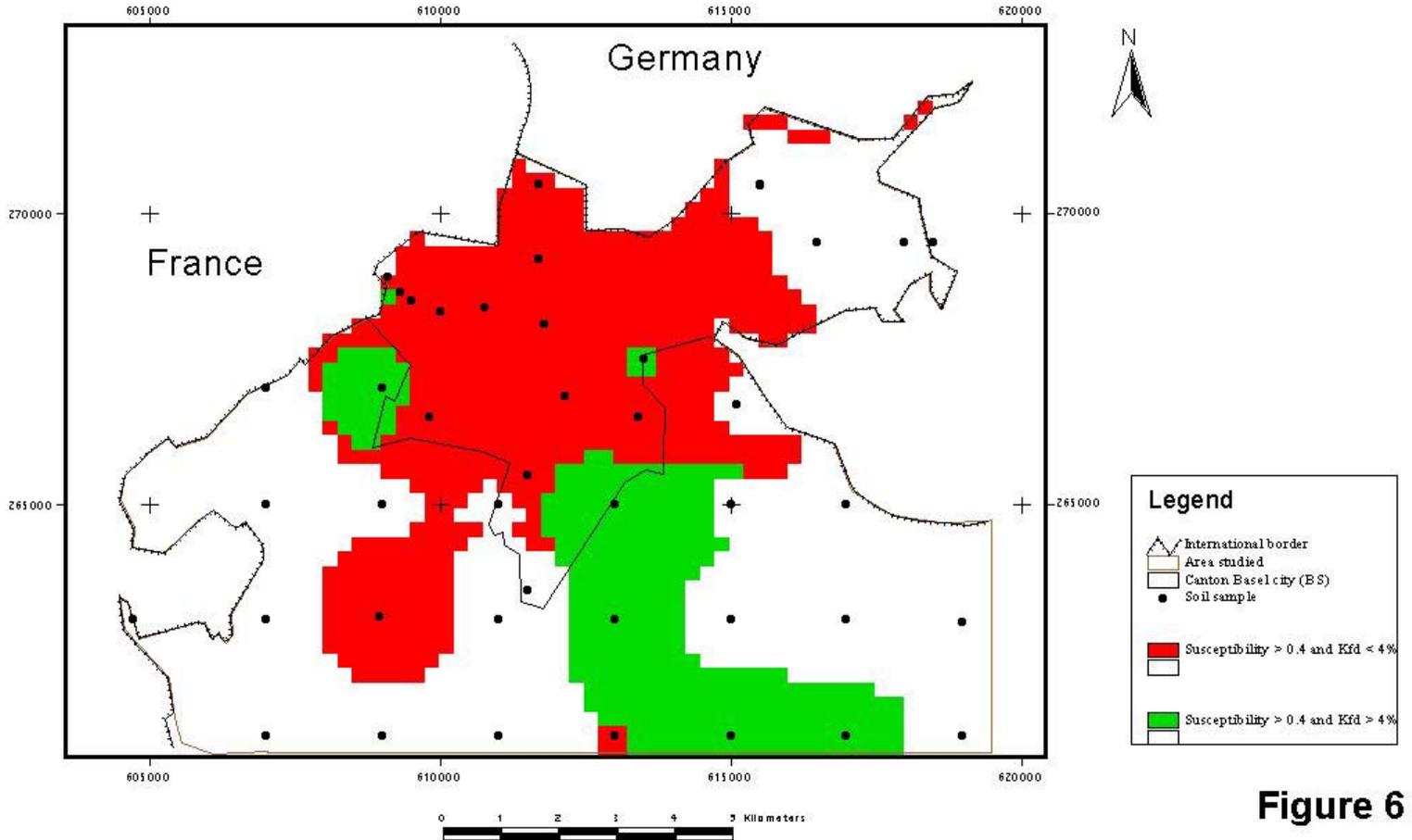


Figure 6

It is interesting to use the other parameter obtained from the susceptibility measurements, that is the frequency dependent parameter k_{fd} . When expressed as a percentage, k_{fd} is given by:

$$k_{fd}\% = \frac{(k_{lf} - k_{hf})}{k_{lf}} \times 100$$

Where k_{lf} and k_{hf} are the initial magnetic susceptibilities measured at 0.46 and 4.6 kHz respectively.

Large grains of magnetite, which have sizes much greater than the superparamagnetic (SP) limit, do not exhibit a variation of susceptibility with frequency and so have a small k_{fd} , usually less than 2%. However, the very fine grains present naturally in soils have sizes that span this critical limit and their k_{fd} can reach 12-14%. As the particles produced

by combustion processes (vehicles and industrial plants) are many microns in size and they are much larger than the SP limit, k_{fd} could be used to detect the presence of such pollution. This technique has been used in England and it has been shown that the soils in heavily populated industrial areas have higher susceptibilities with a low k_{fd} indicating the dominance of coarse minerals probably originating from industrial emissions (Hay et al. 1997).

Using this condition the map of magnetic susceptibility has been redrawn (fig 6) showing only where the susceptibility is greater than $0.4 \cdot 10^{-6} \text{ m}^3/\text{kg}$ and is divided into two zones, namely where $k_{fd} < 2\%$ (red) and where $k_{fd} > 2\%$ (green). The former corresponds to those areas where a dominant magnetic signal comes from atmospheric pollution. This is seen to cover the central conurbation of Basel and to extend south-westwards towards the town of Oberwil. In the built up area along the Birs valley towards Reinach the higher values of k_{fd} suggest that the soils contain more of the very fine magnetic grains produced by pedogenesis, a natural process, and that the contribution from atmospheric pollution is less important here.

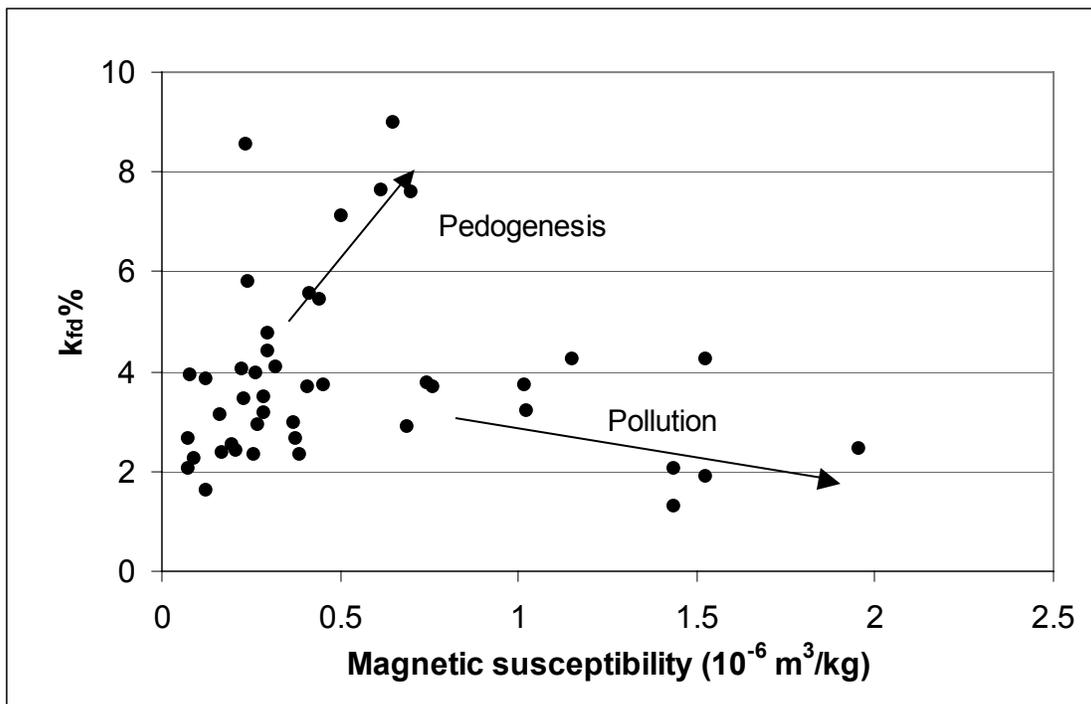


Figure 7. Basel region: Topsoil frequency dependent susceptibility versus magnetic susceptibility.

A plot of the k_{fd} parameter against initial magnetic susceptibility (fig. 7) shows two tendencies. Firstly increasingly larger values of susceptibility associated with lower $k_{fd}\%$ ($< 4\%$) typical of coarser magnetic grains possibly from atmospheric pollution. Secondly a less important increase in susceptibility accompanied by increase in $k_{fd}\%$. This latter

signature is typical in soil collections and the increase in susceptibility is due to an increasing amount of very fine magnetic grains, the so-called secondary ferromagnetic minerals (SFM) produced by chemical processes in the soil (Dearing et al 1996).

4. Biberist /Gerlafingen (SO)

This second study concerns soils from around the site of a large iron foundry in the canton of Solothurn. An environmental study has shown the presence of considerable amounts of heavy metals in the soils around the foundry that are clearly associated with the industrial activity (Borer, 1997). Highest lead concentrations are to be found near to the foundry and at a site 1 km to the southwest, which is thought to be a waste dump of slag from the metallurgical processes (fig. 8). This contaminated site was chosen as a good example for the application of environmental magnetic methods.

Susceptibility measurements were carried out on the same samples as were used in the original site investigation (fig. 9).

Biberist/Gerlafingen, Soil Pb content (ppm)

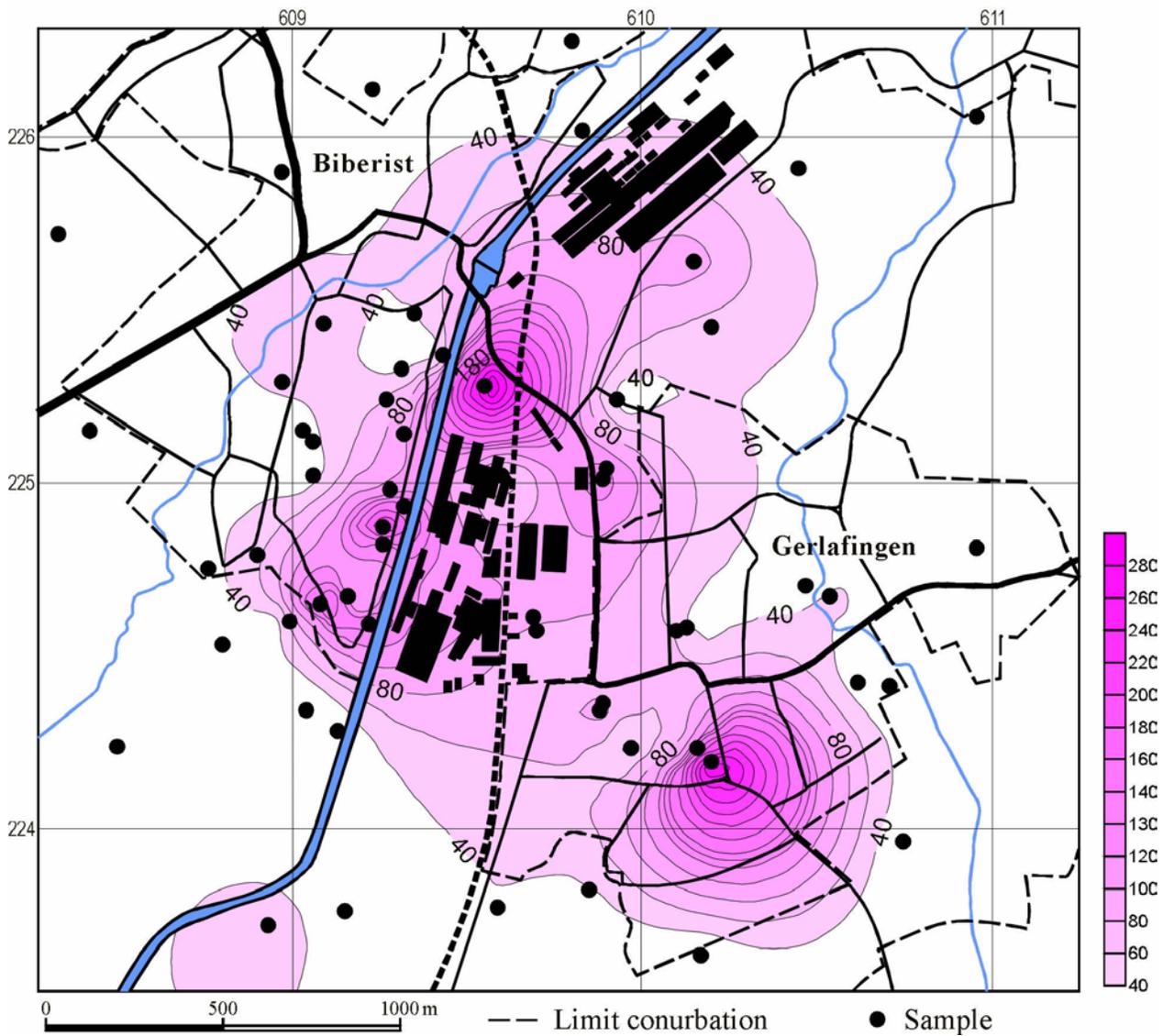


Figure 8 Biberist/Gerlafingen: Topsoil lead content.

Biberist/Gerlafingen, Topsoil magnetic susceptibility ($10^{-6} \text{ m}^3/\text{kg}$)

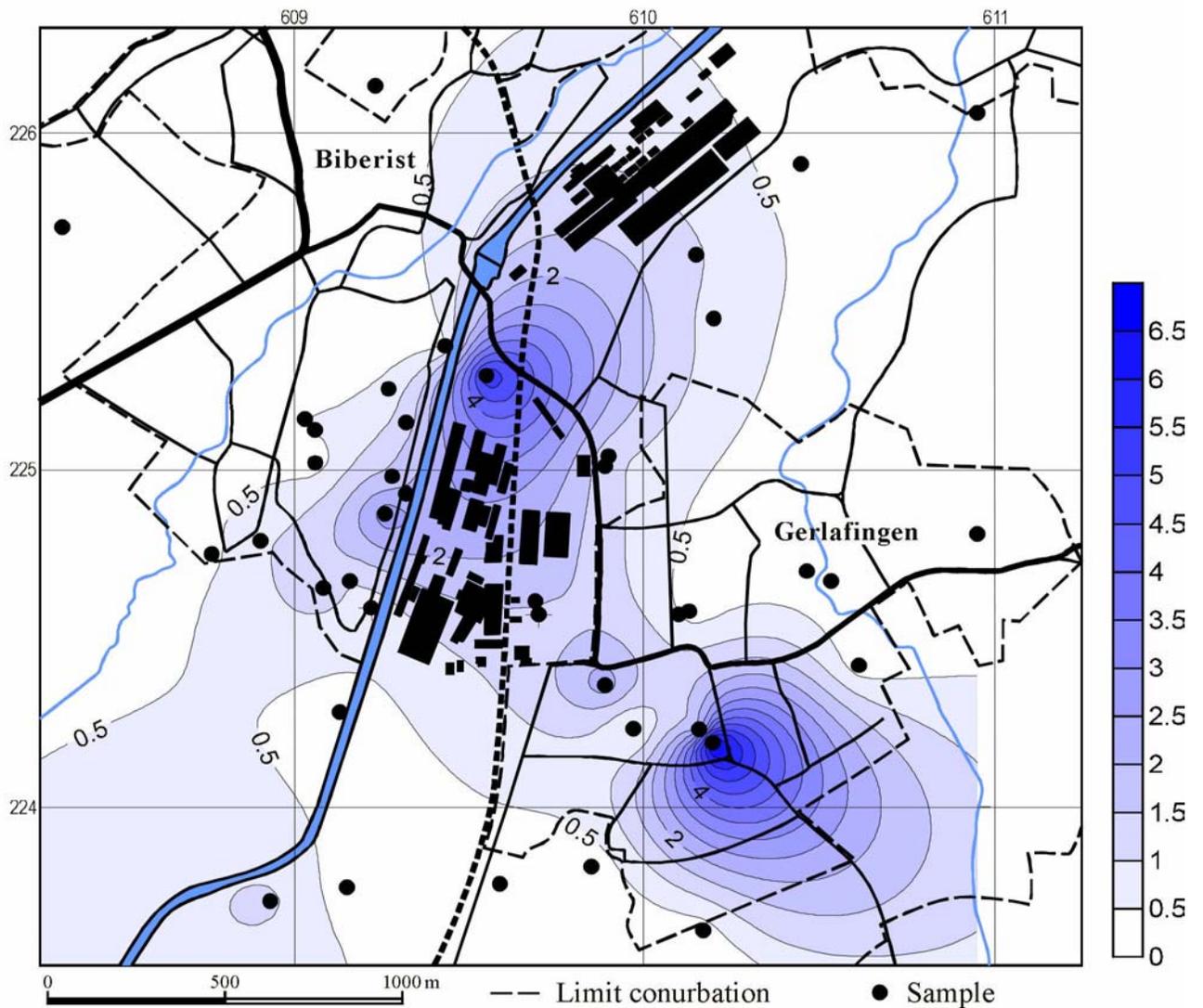


Figure 9. Biberist/Gerlafingen: Magnetic susceptibility of topsoil.

A good correlation is seen between the lead content and susceptibility with a correlation coefficient of 0.908 (fig. 10).

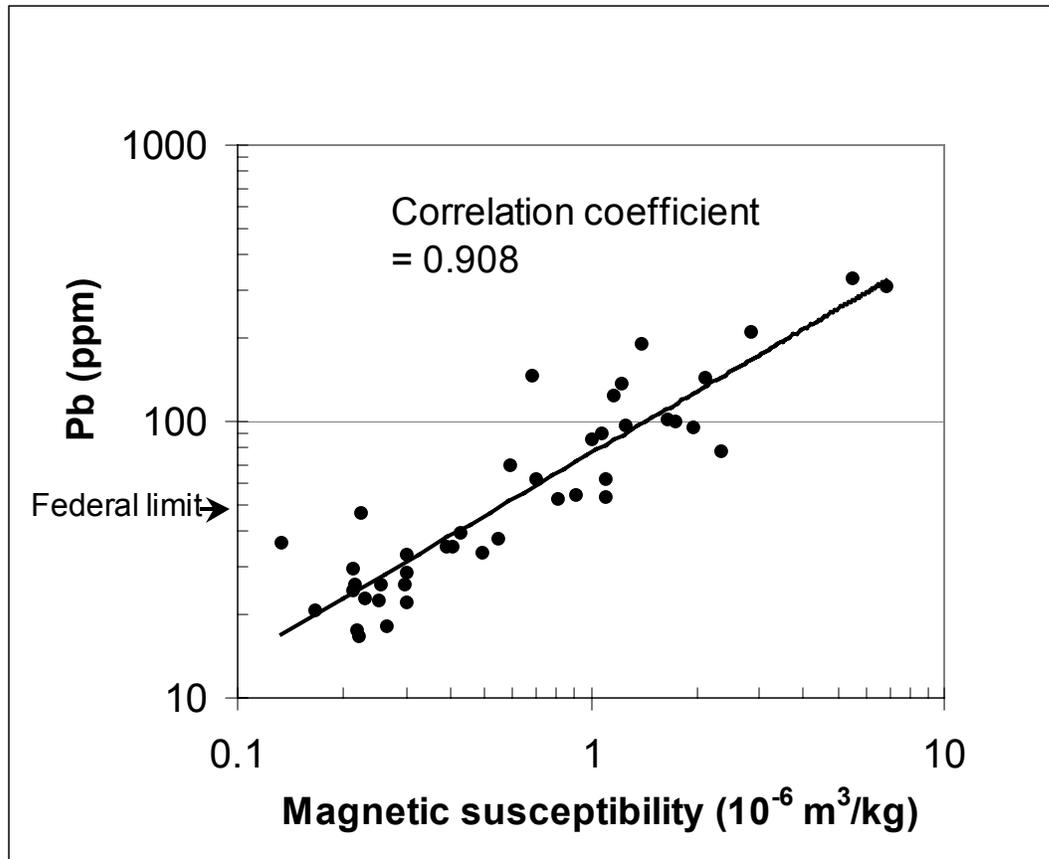


Figure 10. Biberist/Gerlafingen: Correlation of topsoil lead to magnetic susceptibility

Field measurements were also made on selected sites from the original soil survey using the Bartington D probe pressed on to the ground surface. In the form of a coil of mean diameter 18.5 cm this probe operates at a higher frequency (0.958 kHz) and effectively “sees” down to a depth of around 10 cm into the ground. The field measurements showed a reasonable agreement with the laboratory measurements using the B sensor on samples from the same sites. Due to the different conditions concerning the effective sample measured and the susceptibility measurement probes a good agreement is not to be expected (Lecoanet et al.1999).

The susceptibilities rise to high values, practically $10^{-5} \text{ m}^3/\text{kg}$, which is the highest yet observed in the *Magsol* project, which covers soils from practically the whole of Switzerland.

These elevated values are accompanied by low $k_{fd}\%$ (fig. 11) indicating the absence of very fine SP particles and the dominance of much larger magnetic grains associated with industrial atmospheric pollution.

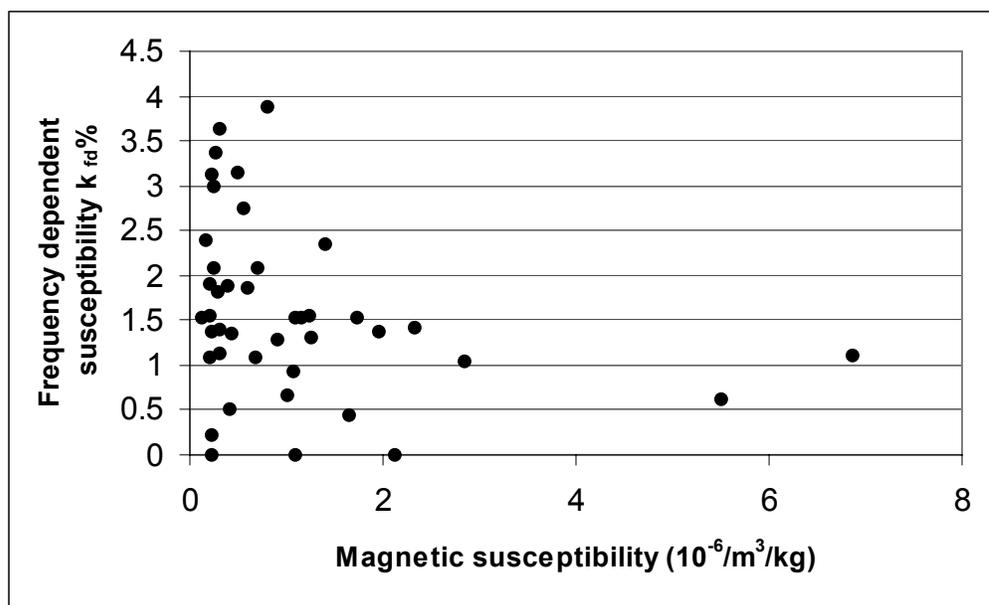


Figure 11. Biberist/Gerlafingen: Topsoil frequency dependent susceptibility versus magnetic susceptibility.

Even higher values of susceptibility have been observed in heavily polluted soils in Poland where the source is coal-burning power plants (Heller et al.1998).

5. Rock magnetic study

Complementing the magnetic susceptibility study important information can be gleaned from an investigation of the remanence properties of the iron oxides in the soils. In effect the grain size variation of susceptibility for the major magnetic carrier, magnetite, is not very pronounced whilst the parameters associated with the remanence change by almost an order of magnitude (Heider et al. 1992). This is an important aid in attempting magnetic granulometry.

Before any treatment a wad of polyethylene foam was pressed into the open plastic container on top of the soil and then the lid was clipped tightly closed to prevent movement of the soil grains during the magnetic analyses.

5.1 Anhyseretic magnetization

First an anhyseretic magnetization (ARM) was applied to the soil sample using an alternating field demagnetiser constructed in Geneva and fitted with a permanent magnet assembly around the sample. This latter device applies a constant direct field of 0.2 mT parallel to the alternating field that is applied during the ARM acquisition. The maximum alternating field applied was 100mT and this was decreased at a constant rate over a period of 50 seconds. The exact conditions for the production of an ARM are important (Liu, 1995) and for this reason they are given here.

After measurement of the ARM and its susceptibility a demagnetisation in an AF field of 40mT was carried out followed by a complete AF demagnetization. Surprisingly it was

sometimes necessary to physically stir the sample to destroy its remanence, as the maximum alternating field (0.18 T) available was not sufficient.

5.2 Coercivity

The soil sample was then progressively magnetized in a direct magnetic field to induce a saturation remanence (SIRM), first using a coil (0-60 milliTesla) and then a laboratory electromagnet (Bruker B-E 10 C8) to reach a maximum magnetic field of 1.3 Tesla. Finally a reverse direct magnetic field was used to reduce the remanence to zero enabling the coercivity of the carriers of remanence (H_{cr}) to be estimated.

The two samples from the Basel region: BA 16 and BL 30 gave an H_{cr} of 24 and 20 mT respectively. The SIRM/ χ ratios for these soils are 14.6 and 8.2 kA/m respectively. Referring to the biplots of Peters (1995) based on published data for sized magnetite, these values correspond to grains of around 0.1 micron in size.

Samples from soils around the steel works at Biberist/Gerlafingen gave an H_{cr} of 24 (BA 11) and 29 mT (BA 5) with SIRM/ χ values of 15.7 and 10.6 kA/m respectively. This suggests that the magnetite here is slightly magnetically harder, that is smaller in grain size than in the Basel soils.

Hysteresis loops on the Biberist samples using a PAR vibrating sample magnetometer give further information on the magnetic constituents present. The coercive forces of samples BA 11 and BA 5 are 9.4 and 7.7 mT, in agreement with the above rock magnetic measurements and place the dominant magnetic component in the so-called pseudo-single domain field for magnetite.

5.3 Decomposition of isothermal remanence

From the progressive acquisition of magnetization experiment it is possible to separate the different magnetic components present in the soil. Although the paramagnetic minerals in the soil, which may contribute to the susceptibility, are not concerned here this analysis can still give useful information on the presence of weakly magnetic but magnetically hard minerals, such as hematite and goethite. The programme IRM-CLG (Kruiver et al. 2001) was used to decompose the isothermal remanence acquisition curves. The results show that the major remanence carrier corresponds to a magnetite-like phase with 1-3 % of a second phase, which judging from its high coercivity must be goethite.

5.4 High temperature magnetic susceptibility variation

After completion of the high field experiments a small part of the 10ml sample of soil was used for high temperature susceptibility measurements to help identify the magnetic oxides present. The measurements were made with a KLY-2 susceptibility bridge equipped with a furnace.

In general all samples showed a Curie point of magnetite with a substantial increase in susceptibility after heating with the colour of some of the samples turning black. This is probably due to a reduction, by the organic content of the soil, of iron minerals to magnetite. For this reason the cooling curve is not very informative.

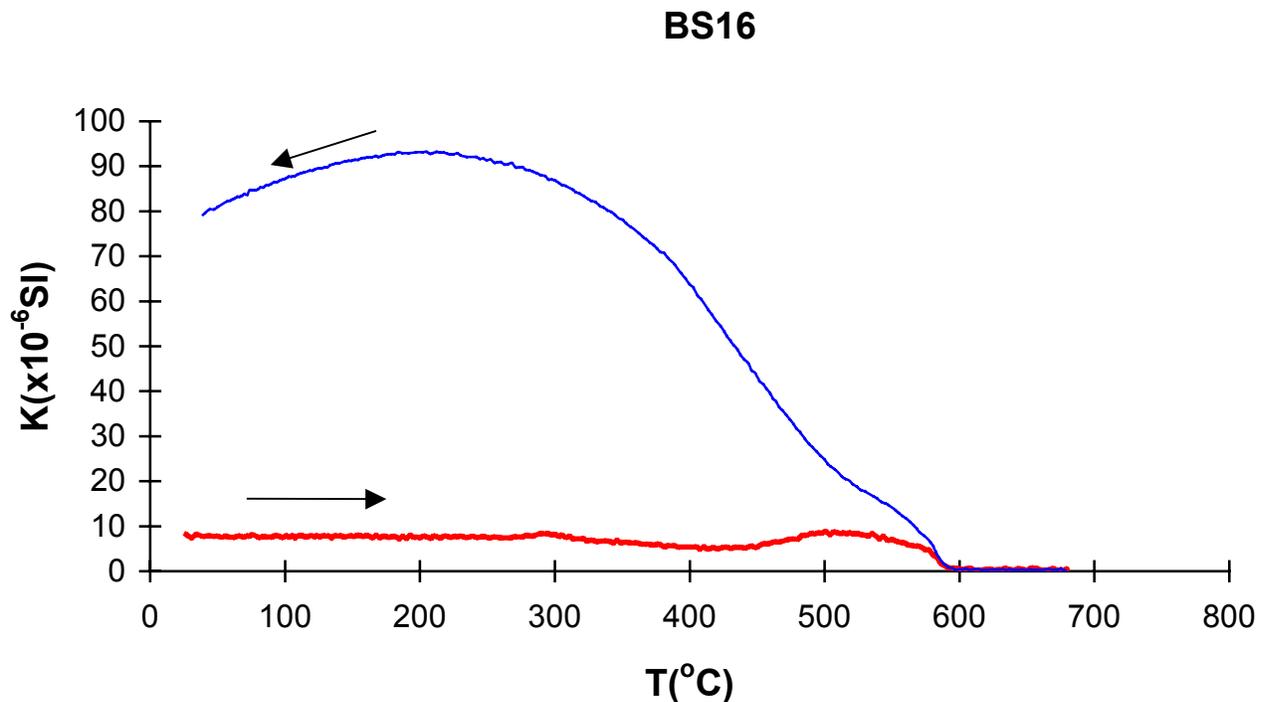


Figure 12. High temperature magnetic susceptibility variation of sample BS 16 from Basel city.

A small broad peak at around 280°C resembles that of the dehydration of poorly crystallised lepidocrocite (γ -FeOOH) to the highly magnetic maghemite (γ -Fe₂O₃) followed by its subsequent inversion to hematite (α -Fe₂O₃). Figure 12 shows the high temperature variation of a sample from Basel city (BS16). Lepidocrocite is a less frequent form of FeOOH than the ubiquitous goethite α -FeOOH and its formation in soils is usually attributed to waterlogged conditions, with fine crystallites being produced by rapid oxidation of Fe²⁺ (Schwertmann et al. 1979).

However, the high temperature measurements indicate a dominant presence of magnetite and the paramagnetic lepidocrocite can only play a minor role in the observed susceptibility of the soils at room temperature.

High temperature susceptibility measurements on topsoil from the edge of a busy road (24,000 vehicles daily) in SW Germany (Hoffmann et al. 1999) give a quite similar curve to that of BS 16, although the authors do not identify the peak observed between 250 and 300°C. As the source of pollution at this roadside site is thought to come solely from the traffic the similarity in the susceptibility-temperature curves suggests that the major part of the pollution in Basel city comes also comes from the road traffic.

The broad Hopkinson peak before the Curie point of magnetite, particularly visible in the sample from Biberist (BA11d) is typical of single domain (SD) particles (Dunlop, 1974) that is of size 50-90 nm (Fig. 13).

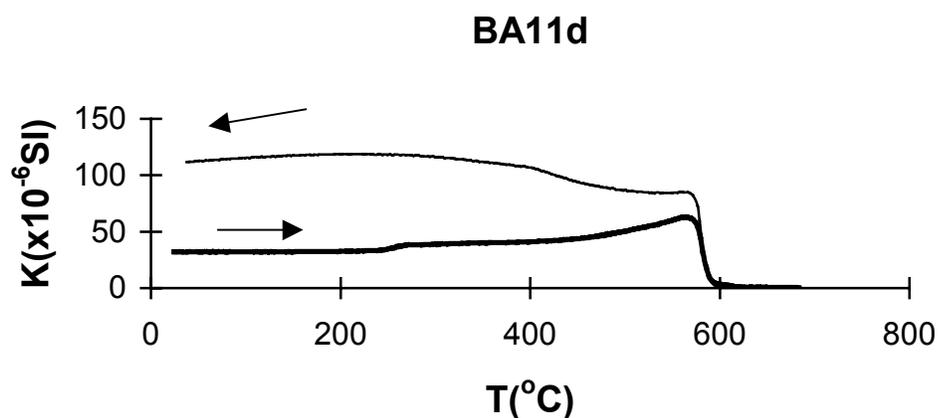


Figure 13. High temperature magnetic susceptibility variation of sample BA 11d from Biberist.

6 Scanning electron microscope study

Magnetic extracts from the soils were obtained by dispersing some of the soil sample in water and using a ferrite permanent magnet covered with plastic film. This extract was studied using a JEOL scanning electron microscope fitted with an energy dispersive analysis system (Oxford Inca 300).

Amongst the many different shapes and sizes of grains seen, the most striking were the ones with an almost perfect spherical shape. The production of magnetic spherules by combustion processes is well documented (Petrovsky et al. 2000), particularly those emitted by coal-fired power stations (fly-ash). However, municipal waste incineration also produces particles that are thought to be responsible for urban atmospheric lead pollution, for example in New York City (Chillrud et al. 1999). The spherical shapes are produced by melting at the high temperatures reached in the combustion process.

Sample BL 30 (fig. 14) shows spheres 5-30 microns in size and the major elements detected are Fe and O, with Al and Mg as minor elements.

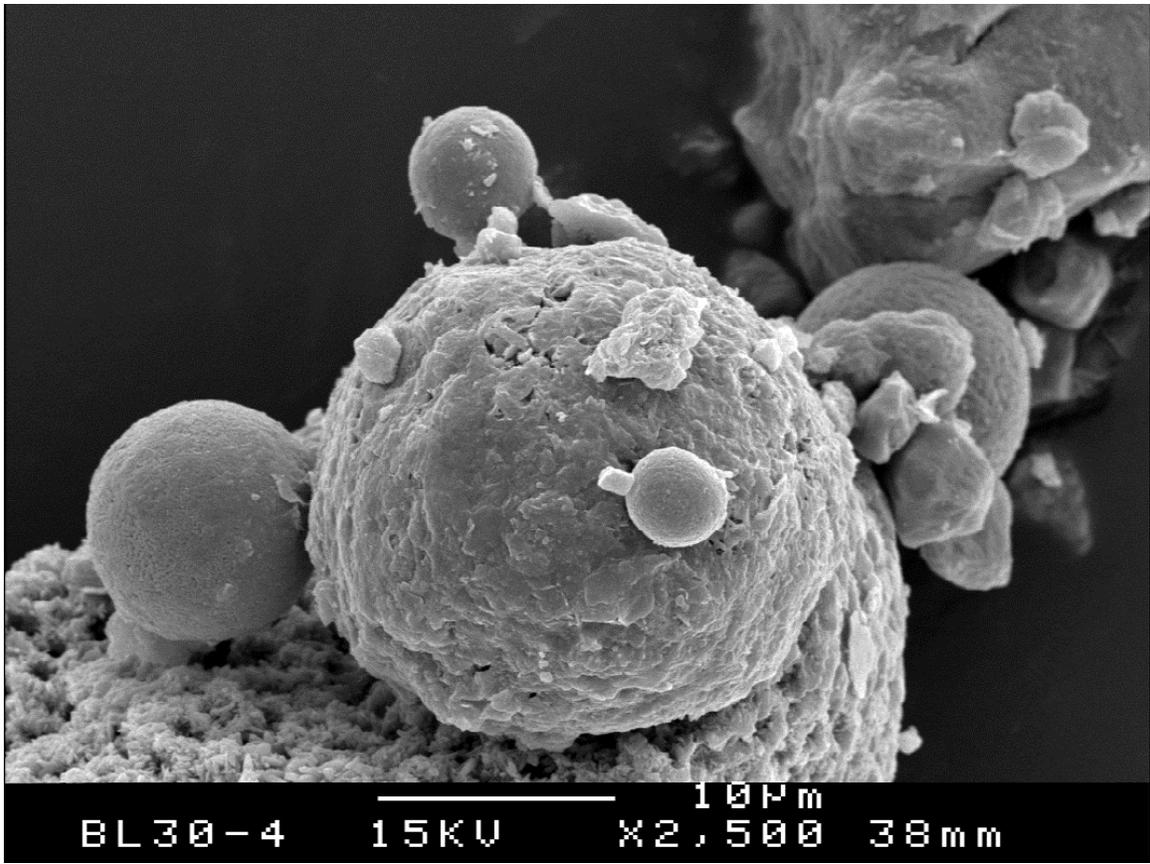
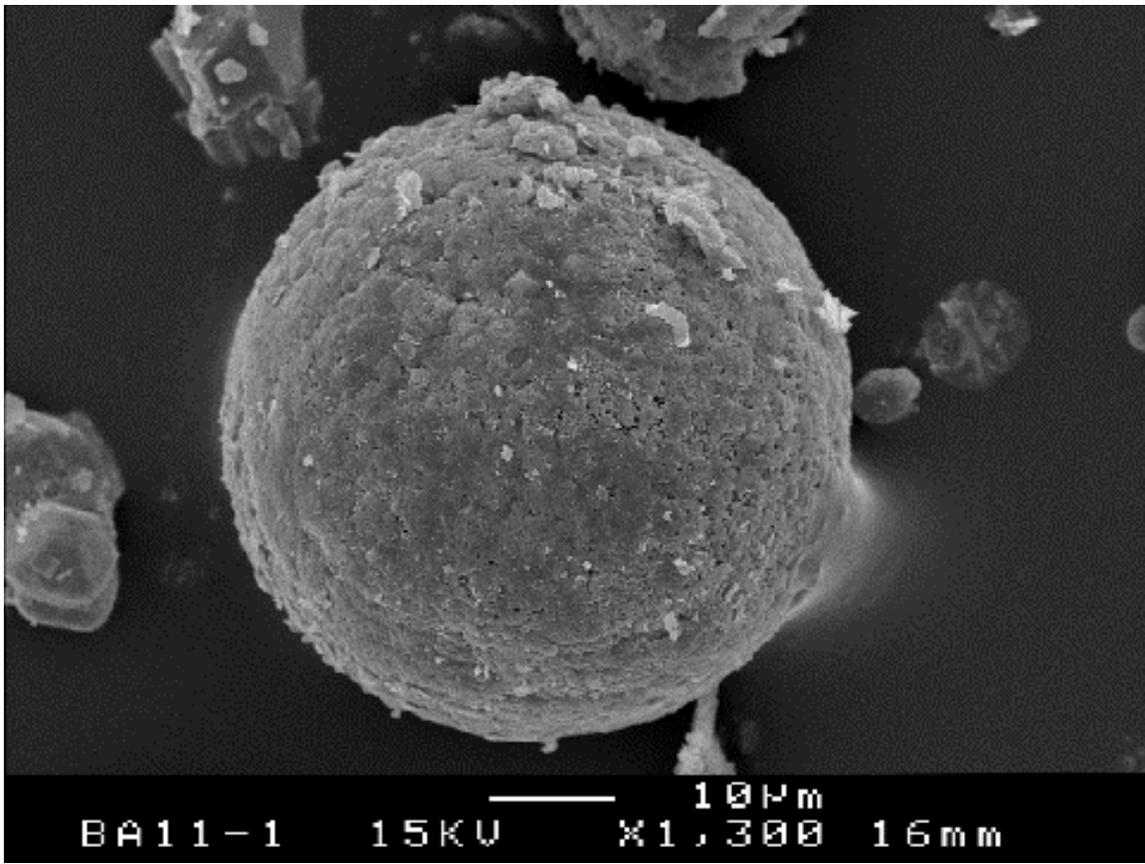


Figure 14. Scanning electron microscope image of magnetic extract from sample BL 30 Basel country

The sample from Biberist/Gerlafingen BA-11 reveals the presence of large spherical particles (fig. 15) 20-180 microns in diameter. EDAX analysis of the spherule in fig. 15 indicated Fe and O as major elements with minor Al and Mg.



The surface of the spherule has a glassy etched appearance but there surprisingly there appears to be no Si in the chemical composition.

Their chemical composition corresponding to an iron oxide distinguishes these spherules from those found in fly ash where Si is a major element (Fisher et al. 1976).

7. Conclusion

Magnetic measurements carried out on topsoils from the region of Basel urban setting and also near an iron smelting factory in the canton of Soleure show a correlation between the magnetic susceptibility and the heavy metal content of the soils (particularly that of lead). Most probably due to the multi-source nature of the pollutants in the region of Basel, the correlation is poorer than for the simpler example of the industrial site, where a strong correlation is observed.

However, the distribution map of magnetic susceptibility for the region of Basel corresponds closely to the map of the soil lead content. This is also seen for the area around the factory at Biberist/Gerlafingen where the high values of susceptibility and the limited topographic extent of the pollution make it a good example of the magnetic “proxy” method for delimiting the polluted area.

The rock magnetic study reveals the presence of sub-micron magnetite with a minor amount of goethite and possibly a trace of lepidocrocite. The magnetic spherules seen in the magnetic extract appear to be solely iron oxide, unlike the common fly ash spherules

which are silicates.

This study confirms utility of magnetic susceptibility mapping as a rapid and cost effective method for determining the extent of heavy metal pollution of soils, in agreement with the previous results from Central and Eastern Europe

8. Acknowledgements

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