THE INFLUENCE OF INDUSTRIAL IMMISSIONS ON THE MAGNETIC SUSCEPTIBILITY OF SOILS IN UPPER SILESIA

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Summary: *Samples of metallurgical dusts and fly ashes from coal power plants and iron works in Upper Silesia as well as soil profiles in the close vicinity of these plants and in Ojcow National Park (ca. 25 km east of the industrial area) have been studied magnetically and mineralogically. The metallurgical dusts and fly ashes are highly enriched in ferromagnetic minerals. The topsoils from profiles collected near the plants have very high values of magnetic susceptibility while susceptibility in the fermentation and humic subhorizons in soil profiles from Ojcow National Park is considerably increased. The magnetic properties of the metallurgical dusts and fly ashes such as frequency dependence of susceptibility, saturation remanence or coercivity are similar to those observed in the top horizons of the soils. They are mostly related to the occurrence of large (multidomain) grains of non-stoichiometric magnetite ranging from 1 to 20 μm. The similarity of the magnetic particles in the soils is taken as evidence of an anthropogenic origin. They are responsible for the high soil susceptibilities in Upper Silesia and in adjacent areas. Some of the magnetic particles carry substantial quantitities of trace elements such as Pb, Ni, Zn and Cu. Field and laboratory susceptibility measurements can therefore be used as a simple and costeffective method of detecting the presence of heavy metals in the soils of this area.*

Keywords: anthropogenic ferrimagnetics, metallurgical dust, fly ashes, trace elements

1. INTRODUCTION

Upper Silesia (Katowice voivodeship) is the most industrialised and most polluted region of Poland. Many heavy industry plants are concentrated in this small area covering ca. 6650 km^2. Environmental hazards are mostly linked to the old metallurgical plants, cokery plants and power plants which combust large amounts of hard coal and emit many types of gaseous and dusty pollutants. Dependent on character and size, the pollutants can be transported over variable distances as solid phase particles or in aerosols. After landing on the soil surface, the dust particles penetrate into the soil profile and accumulate mostly in the fermentation (O_f) and humic (O_h) subhorizons.

The industrial dust falls contain also a certain proportion of magnetic particles, which cause an increase of magnetic susceptibility in the topsoil. According to records at many monitoring stations in the voivodeship since 1981 the dust fall in the study area varied from 200 to 450 gm^{-2}year^{-1} (*Strzyszcz, 1994*) and the iron flux in dustfall varied from 10 to 200 gm^{-2}year^{-1} (Fig. 1). Earlier investigations of metallurgical dusts and fly ashes from power plants (*Strzyszcz et al., 1988; Strzyszcz, 1989a; Strzyszcz and Magiera, 1993*) demonstrated very high magnetic susceptibility

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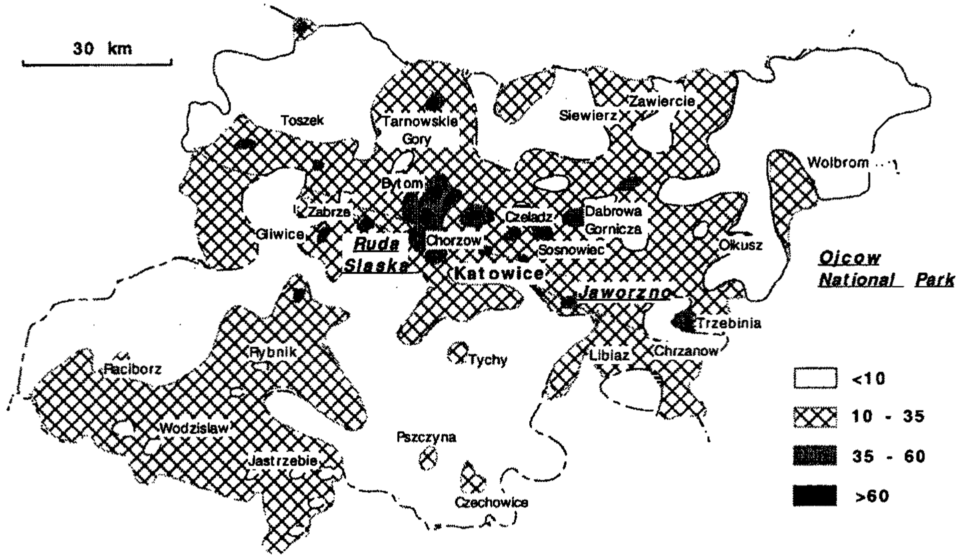

Fig. 1. Iron fall (Fe_2O_3) in Katowice District (Upper Silesia) in 1987, in $gm^{-2}a^{-1}$.

values: max. > $18000 \times 10^{-8} m^3kg^{-1}$ for metallurgical dusts and $2500 \times 10^{-8} m^3kg^{-1}$ for fly ashes. These values point at a considerable amount of ferrimagnetic iron oxides in the industrial dusts. The magnetic low field susceptibility in 1500 soil samples from Upper Silesia increases distinctly in the topsoils (mostly subhorizons O_f and O_h). This effect has been observed especially near the great industrial plants (*Strzyszcz et al., 1994*). There is also evidence of a close direct correlation between magnetic susceptibility of soils and concentration of some heavy metals.

The purpose of present work is to investigate the magnetic particles in the industrial dusts and ashes and in the topsoils of industrial areas, where these particles cause an artificial increase of magnetic susceptibility. The former field observations by *Strzyszcz (1988,1989a,1989b,1991)*, *Strzyszcz and Magiera (1993)* and *Strzyszcz et al. (1994)* will be supplemented by laboratory characterization of the ferromagnetic mineralogy.

2. MATERIALS AND METHODS

Two large industrial plants in Upper Silesia have been selected for this study: The "Pokoj" iron works in Ruda Slaska and the huge power plant Jaworzno III (underlined localities in Fig. 1). Metallurgical dusts from two blast-furnaces of the "Pokoj" iron works and ashes from the electro-filter of the Jaworzno III power plant have been sampled. Over 60 soil samples have been collected during an earlier study from the area near these plants between 1988 and 1992 (see references above). Two soil profiles collected in the prevailing wind direction 1·2 and 2·4 km NE from "Pokoj" iron works and samples from the top of a peat soil near the Jaworzno III power plant have been chosen for the present laboratory study. The third area selected for the present study is Ojcow National Park (ONP), which is situated 25 km east of the Upper Silesia industrial district in a region without large sources of pollution. According to *Grodzienska (1992)*, the heavy metal concentration in mosses from Ojcow National Park indicates the most strongly polluted national park in Poland. Soil pollution and magnetic susceptibility have been studied in this area in 20 soil profiles by *Strzyszcz and Magiera (1993)*. Two of them, which have the highest magnetic susceptibility values, have been selected for the present investigation. Soil samples from ONP and around the Jaworzno III power plant have been collected in forests, where the topsoil is not disturbed by agricultural activity and pollution including magnetic particles continued over a long time. The soils around the "Pokoj" iron works have been collected from deforested areas which were not cultivated and ploughed during the last 30 years.

During the first stage of this study the following magnetic parameters have been measured:

- χ - the specific magnetic bulk susceptibility measured using a low-frequency, low field susceptibility bridge KLY-2;
- χ_{fd} - the frequency dependence of magnetic low field susceptibility measured at two frequencies (difference between χ_{lf} at 0·47 kHz and χ_{hf} at 4·7 kHz, expressed as percentage of χ) using a Bartington susceptibility bridge;
- *SIRM* - the saturation isothermal remanent magnetization which was acquired at room temperature in fields up to 1 T measured on Digico fluxgate spinner magnetometer;
- *IRM* - isothermal remanent magnetisation measured as a function of the applied field in order to determine the IRM/SIRM ratios in variable fields;
- $(B_0)_{cr}$ - the coercivity of saturation remanent magnetization;
- $M_s(T)$ - the temperature dependence of saturation magnetization measured using a Curie balance whereby the samples were subjected to a heating and cooling cycle in air, in a steady magnetic field of 0·1 T;

Magnetic concentrates (using a hand magnet) were obtained from four samples for further mineralogical studies. This material has been subjected to the chemical and mineralogical analysis using a Cameca SX 50 electron microprobe.

3. RESULTS AND DISCUSSION

3.1 Metallurgical dusts

The metallurgical dust samples from the "Pokoj" iron works have high magnetic susceptibilities ranging from 5900 to $6400 \times 10^{-8} m^3 kg^{-1}$. These samples reach or are closely to magnetic saturation in a field of 300 mT. Figure 2a presents the backfield demagnetization for the metallurgical dust sample HP2 (blast-furnace dust). The coercivity $(B_0)_{cr}$ amounts to 25 mT and the S-ratio which is defined as the ratio of isothermal remanence at a backfield of 0·1 T versus SIRM (Stober and Thompson, 1979), equals −0·72. Coercivity values between 20 and 40 mT are characteristic of relatively coarse magnetite grains (Scoullos et al., 1979). The *S*-value suggests that magnetite is the predominant magnetic mineral in the sample and the presence of high coercivity antiferromagnetic minerals is marginal.

Low χ_{fd} values between 1·07 and 1·44% are also characteristic. The small frequency dependence is characteristic for single- to multidomain grains of magnetite - mostly grains over 1 μm (*Kittel, 1949; Banerjee et al., 1981*). Very small grains below the single-domain/superparamagnetic threshold would show greater changes of magnetic susceptibility at variable field frequency.

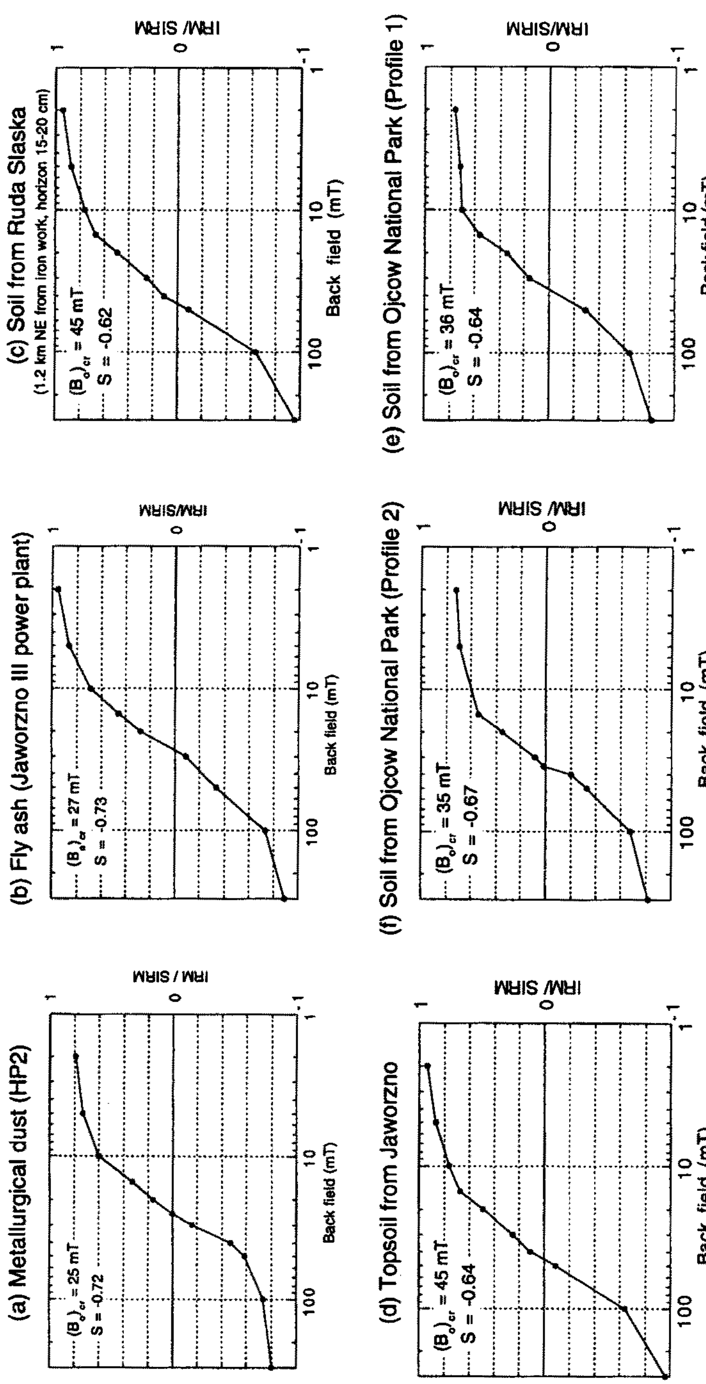

Fig. 2. Back-field demagnetization of (a) metallurgical dust, (b) fly ash and (c-f) polluted sooils.

Thermomagnetic analyses show that the Curie points of metallurgical dusts range between 600 and 610°C (Fig. 3a). These values are between the Curie temperature for pure magnetite (585°C) and that of pure haematite (675°C). The thermomagnetic curves are non-reversible. After heating and cooling to 650°C, the room temperature magnetization value represents only about 80% of the initial value. Some proportions of the ferromagnetic iron minerals must have been transformed into minerals of lower spontaneous magnetization during heating. This behaviour is typical for maghemite and is connected with gradual transformation of maghemite to hematite in the temperature range 300 – 500°C The often observed negligible magnetization loss during heating at temperatures below 400°C combined with a distinct kink of the magnetization curve suggests the presence of at least two ferrimagnetic minerals in the metallurgical dusts which according to the above magnetic results must be characterized by low coercivity and high *S* values.

The composition of the mostly spherical magnetic grains in the metallurgical dusts has been determined by microprobe analysis. Variable results of the stoichiometric quantitative chemical analysis of the percentage composition are obtained for different particles. The total analysis closes between 89·90 to 94·39% (Table 1) very near to the theoretical values of pure magnetite at 93·06% and of pure maghemite at 90·41%. Either of the two phases may constitute the magnetic grains. Some particles, however, are made up of a mixture of both phases.

3.2 Fly ashes

The magnetic susceptibility of the fly ashes from Jaworzno III power station range around $2500 \times 10^{-8} m^3 kg^{-1}$. This value is reduced at higher frequency only little by 1·6 – – 1·7% (χ_{fd}). $(B_0)_{cr}$ and *S* (Fig. 2b) are similar to the corresponding values measured for metallurgical dusts. They suggest that similar magnetic particles occur in the fly ashes.

The thermomagnetic analysis for fly ash from Jaworzno III suggests that magnetite is the predominant ferromagnetic mineral in the sample (Fig. 3c). The Curie point is slightly below 600°C, and the magnetization curve is almost reversible upon cooling.

Granulometric analysis of the magnetic fraction from fly ash from Jaworzno III shows that 75% of ferromagnetic grains are contained in the range 2 – 50 μm whereas the fraction < 2 μm amounts only to 6% (Table 2).

3.3 Soils around "Pokoj" iron works

Sandy clays and sands constitute the substrate in the area of "Pokoj" iron works. Their specific magnetic susceptibilities vary between 15 and $30 \times 10^{-8} m^3 kg^{-1}$. These values are inadequate to account for the magnetic susceptibility values observed in the uppermost 20 cm of the soil profiles (Fig. 4a,b).

In profile 1, the susceptibility values are high across the uppermost 20 cm of the soil with the maximum between 15 – 20 cm. In profile 2, considerable susceptibility increase is observed only in the topmost 5 cm of the soil. The high bulk susceptibility χ is accompanied by very low susceptibility frequency-dependence (χ_{fd}), which varies between 1·10 and 1·78% in profile 1. In profile 2, χ_{fd} seems to be inversely proportional to the χ. χ_{fd} has a maximum (4·04%) in the horizon where the bulk susceptibility is minimal.

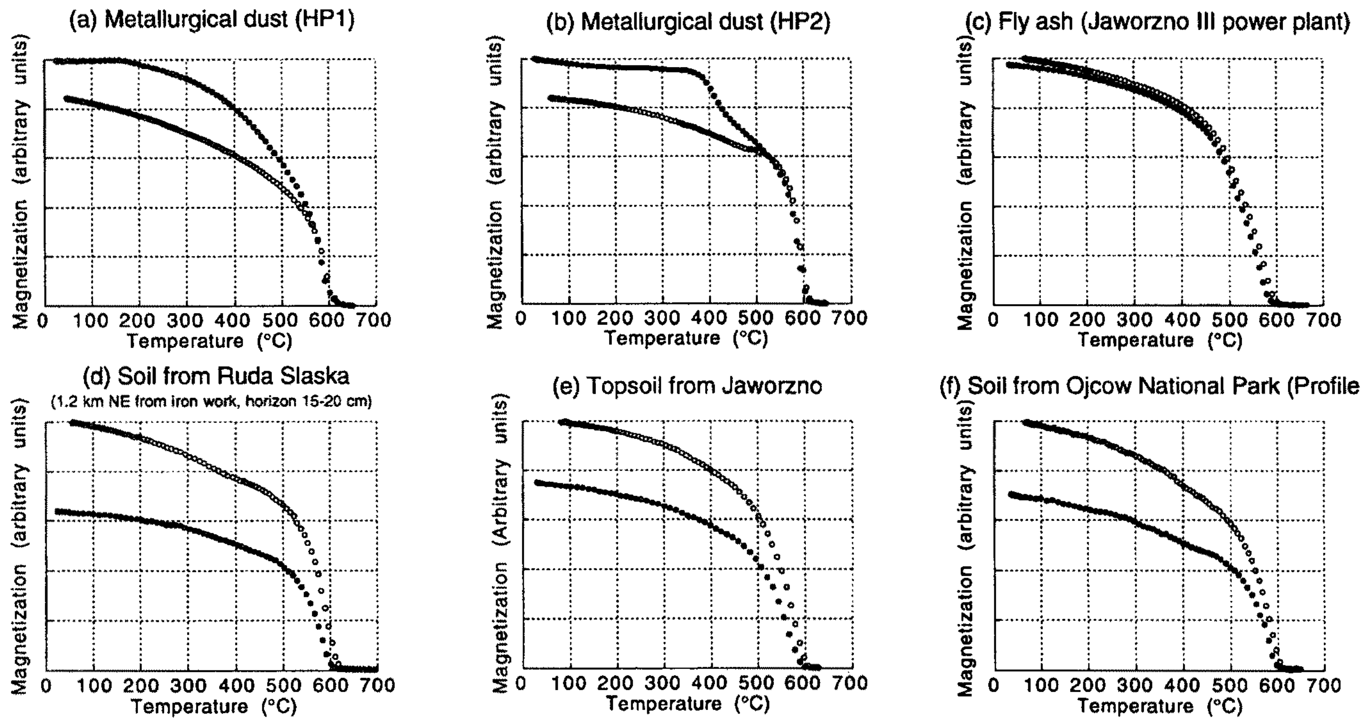

Fig. 3. Thermomagnetic curves of (a,b) metallurgical dust, (c) fly ash, (d-f) polluted soils. Closed symbols represent heating, the open ones cooling. Applied field: 0·1 T, treatment in air, heating and cooling rate 12°C/min.

Table 1. Stochiometric analysis of five different magnetic particles (in %) from metallurgical dust.

	1	2	3	4	5
SiO_2	0·00	0·02	1·93	0·19	0·24
TiO_2	0·00	0·05	0·04	0·06	0·12
Al_2O_3	0·10	0·44	0·52	0·11	0·87
FeO	89·76	93·73	88·34	90·05	88·79
MnO	0·00	0·02	0·01	0·08	0·03
MgO	0·00	0·05	0·00	0·00	0·01
CaO	0·01	0·06	0·53	0·03	0·03
Na_2O	0·02	0·02	0·00	0·00	0·04
K_2O	0·01	0·00	0·00	0·02	0·00
Total	89·90	94·39	91·36	90·54	90·13

The values of the S parameter and coercivity of remanence $(B_0)_{cr}$ (Fig. 2c) are higher than in the metallurgical dust, but they are still in a range characteristic of relatively coarse grained magnetite (*Scoullos et al., 1979*). The value of $(B_0)_{cr}$ (45 mT) in profile 1 and the position of the horizon with maximum susceptobility in the soil profile (15 – 20 cm) could suggest that possibly also finer ferrimagnetics of pedogenic origin exist. The very low χ_{fd}, however, shows that these magnetic minerals are not in the range of superparamgnetic grains. Therefore a pedogenic origin is excluded. In profile 2, χ_{fd} (2·69%) is a little bit higher, but is still too low to give definite evidence for the occurrence of fine-grained pedogenic magnetic minerals.

Thermomagnetic analysis (Fig. 3d) shows that the Curie point is just above 600°C like in the metallurgical dust. This suggests that the predominant magnetic mineral is non-stoichiometric magnetite. The thermomagnetic curve is irreversible and after cooling to the room temperature, the magnetisation increases by 40% in relation to the initial value. This increase has probably resulted from the conversion of paramagnetic material at high temperature to ferrimagnetic minerals. Scanning electron microscope observations have shown the presence of spherical magnetic particles which have diameters between 1 and 20 μm.

3.4 Soils around Jaworzno III power station

Peat soils which developed on sands, predominate around the Jaworzno III power plant. They are very good collectors of industrial pollution. The organic horizon in soils from Jaworzno shows very high susceptibilities between $1300 - 1500 \times 10^{-8} m^3 kg^{-1}$. The

Table 2. Granulometric composition of the magnetic fraction of fly ash from Jaworzno III power plant.

Analysis	Percent of grainsize fraction (in μm)							
	1000–500	500–250	250–100	100–50	50–20	20–6	6–2	<2
1	0	1	14	5	20	31	23	6
2	0	1	14	5	19	32	23	6
3	0	1	14	5	20	31	23	6

susceptibility decreases abruptly below the organic horizon to a value $250 - 280 \times \times 10^{-8} m^3 kg^{-1}$. After removal of organic matter and concentration of the magnetic particles using a hand magnet, soil samples from the organic horizon show almost identical values of magnetic parameters like the soil samples collected near the iron works (Figs. 2d, 3d). The coercivity $(B_0)_{cr}$ is 45 mT and the S parameter equals $-0{\cdot}64$. It suggests that again a high concentration of anthropogenic magnetic particles is responsible for the abnormally high magnetic susceptibility. Besides of the activity of the power plant and the specific soil type, the geographic position of Jaworzno in the eastern part of Upper Silesian Industrial District plays an additional role for the higher concentration of ferrimagnetics in this area. Since westerly winds predominate in this area, this part of Upper Silesia is especially exposed to an influx of pollution from the central part of Upper Silesia, where many big emitters are located.

3.5 Soils from Ojcow National Park

Soil profiles have been taken in Ojcow National Park, which is situated 25 km from the east border of the Upper Silesian Industrial District. They show a significant susceptibility increase in the uppermost few centimeters of the fermentation (O_f) and humic (O_h) subhorizons (Fig. 4c,d). The susceptibility in these subhorizons is several times higher than in the other profile parts which developed on extremely weakly magnetic Jurassic limestones. The susceptibility of these bedrocks is about $7 \times 10^{-8} m^3 kg^{-1}$. The χ_{fd} value in subhorizons with maximum susceptibility is very low ($1{\cdot}5 - 2\%$). The maximum χ_{fd} occurs below 10 cm, mostly in the B-horizon, where the susceptibility is low and probably due to the existence of natural, fine-grained antiferromagnetic and paramagnetic grains.

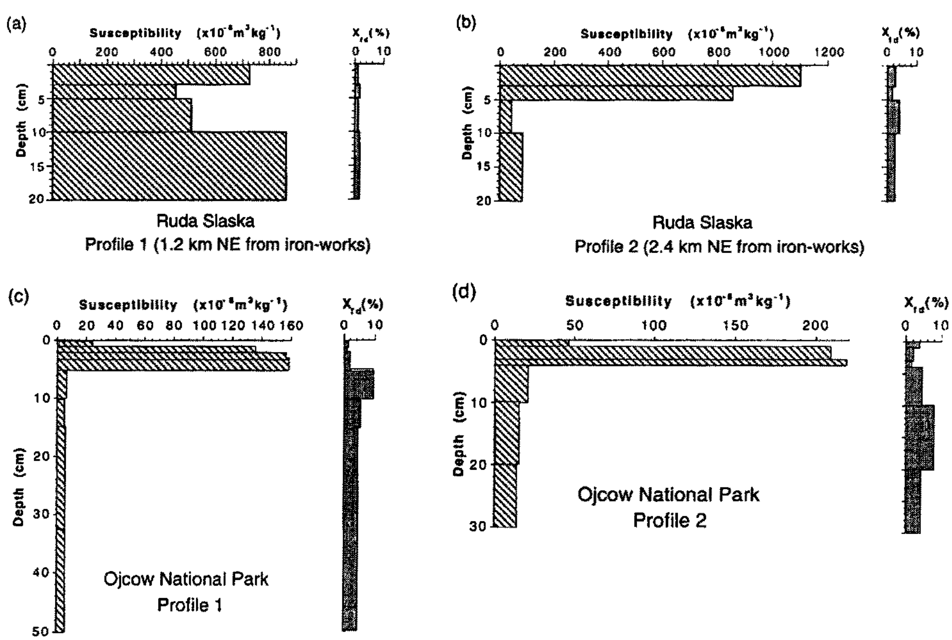

Fig. 4. Bulk and frequency dependent magnetic susceptibility in soil profiles near an ironwork (a,b) and in the Ojcow National Park (c,d).

The coercivity of remanence (ca. 35 mT) in the layers of maximum susceptibility proves that magnetite multidomain grains are predominant like in the soils around the "Pokoj" iron works and Jaworzno III power plant. The *S*-values at −0·63 and −0·70 (Fig. 2e,f) witness to the preponderance of ferrimagnetic over antiferromagnetic iron oxides. The thermomagnetic curve is also similar to those presented before. The same ferrimagnetics of anthropogenic origin are responsible for the susceptibility increase of the topsoils from Ojcow National Park.

4. MAGNETIC PARTICLES AND HEAVY METALS

Many authors have emphasized the connection between the increase of magnetic susceptibility and the concentration of certain heavy metals in various environments. *Beckwith et al. (1986)* show a linear relationship between the concentration of ferrimagnetics and the Cu, Fe, Pb and Zn content in sediments collected in an urban area as well as near an highway. A similar relationship between *SIRM* and susceptibility on the one hand and Pb, Cu, Zn, and Cd in atmospheric particulates on the other has been observed by *Hunt et al. (1984)*. The relation between the presence of magnetic minerals and the concentration of heavy metals in the marine environment has been confirmed by *Scoullos et al. (1979)*. A linear relationship between some magnetic parameters and the Pb and Cu concentration in modern lacustrine deposits has been pointed out by *Oldfield et al. (1984)*. *Strzyszcz and Magiera (1993)* have observed a positive correlation between susceptibility and the concentration of metals such as Zn, Pb and Cd in the topsoils of Ojcow National Park. *Hullet et al. (1980)* observed that the first row transition elements - V, Cr, Mn, Co, Ni, Zn and Cu - present in fly ashes from American power plants are mostly connected with magnetic mineral phases, and occur usually in the form of substituted spinels $Fe_{3-x}M_xO_4$.

Therefore some particles from a magnetic concentrate of fly ash from Jaworzno III power plant were analyzed microchemically. Eleven particles which according to stoichiometric chemical analysis and magnetic investigations are made of non-stoichiometric magnetite or/and magnetite - maghemite transition minerals, have been selected for trace element microprobe analysis. The sizes of these particles are 10 – – 40 μm. Seven most common heavy metals (Cr, Mn, Ni, Cu, Zn, Cd, Pb) have been

Table 3. Heavy metals in magnetioc particles of fly ash from Jaworzno III power plant (in ppm).

	Cr	Mn	Ni	Cu	Zn	Cd	Pb
1	< 100	< 100	< 100	< 100	< 100	< 100	340
2	< 100	< 100	< 100	< 100	330	< 100	260
3	< 100	< 100	110	250	< 100	< 100	< 100
4	< 100	< 100	< 100	< 100	< 100	< 100	130
5	< 100	< 100	< 100	< 100	< 100	< 100	320
6	< 100	< 100	100	< 100	350	< 100	220
7	< 100	< 100	< 100	< 100	< 100	< 100	< 100
8	< 100	< 100	640	170	120	< 100	730
9	< 100	< 100	< 100	< 100	< 100	< 100	320
10	< 100	< 100	100	< 100	< 100	< 100	500
11	< 100	< 100	880	< 100	< 100	< 100	< 100

determined (Table 3). It was found that lead, nickel, zinc and copper occur in substantial quantities in some of the 11 particles. Concentrations of > 100 ppm of lead were measured in eight particles, those of nickel in five, those of zinc in three, and copper in two. The other investigated metals were below resolution of the microprobe used. In magnetic particles over 300 μm in diameter, manganese occurs in quantities even up to 2·3%. In the metallurgical dusts, the magnetic particles contain up to 0·08% of manganese (Table 2).

5. CONCLUSIONS

1. Accumulation of anthropogenic ferrimagnetics due to immission of industrial dusts over long time periods causes considerable increase of magnetic susceptibility in soils, especially in the fermentation (O_f) and humic (O_h) subhorizons of soil litter. The susceptibility of soils in industrialized regions can be taken as a measure of the industrial immissions over a large area and a wide range of emissions. The soils of the Ojcow National Park, although placed already some 25 km east from the main sources of emission in Upper Silesia, indicate rather extensive pollution.
2. Mineralogical and chemical studies of magnetic particles, which are responsible for the considerable increase of susceptibility in top subhorizons of the soils in Upper Silesia show clearly that these are mostly relatively large (multidomain) magnetite grains ranging from 1 to 20 μm in size. The magnetite generally is non-stoichiometric, being an intermediate phase between Fe_3O_4 and γFe_2O_3. Magnetic particles with the same magnetic characteristics and chemical composition are present in metallurgical dusts which are emitted from iron works, in fly ashes originating from hard-coal combustion in power plants and also in the soils of the industrial areas. The similarity of magnetic particles is taken as evidence for an anthropogenic origin of particles which are responsible for the high magnetic susceptibility of soils in Upper Silesia and in adjacent areas.
3. The study of additional magnetic parameters such as χ_{fd}, *SIRM*, *IRM* and $(B_0)_{cr}$ and thermomagnetic analysis is able to characterize the type of the magnetic particles, their domain status, and their anthropogenic or natural origin.
4. Considerable amounts of heavy metals which can be a potential hazard for the environment, are bound to the magnetic particles. The fast and relatively cheap measurements of the magnetic low field susceptibility can be used as a preliminary method of studying pollution effects in soils before starting time-consuming and expensive chemical methods. The anthropogenic ferrimagnetics can be treated as a tracer for the presence of heavy metals.

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