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Lake sediment magnetism and atmospheric deposition

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Many recent lake sediment profiles contain atmospherically derived fly ash and various particles from industrial processes. All these include a magnetic fraction that can be studied by subjecting subsamples to controlled magnetic fields in the laboratory and measuring the isothermal remanences acquired. These provide a basis for partially characterizing and roughly quantifying the magnetic minerals preserved in the sediments. The results presented illustrate those obtained from some 70% of the 39 profiles taken from 32 sites mostly in upland Wales and the Scottish Highlands. They show widespread increases in magnetite and haematite deposition beginning from the mid-nineteenth century onwards and steepening in the last three to five decades.

Introduction

Fly ash from solid-fuel fired power stations, together with the particulates emitted by industrial processes such as iron and steel manufacture and non-ferrous metal smelting, contains high concentrations of magnetic minerals. Those in fly ash arise from the conversion of iron impurities in the feed coal through high temperature combustion. They include both ferrimagnetic ('magnetite') and imperfect anti-ferromagnetic ('haematite') components in concentrations and proportions that vary with coal source and combustion procedures. The upper layers of peat bogs (Oldfield *et al.* 1978) and of lake sediments contain a historical record of atmospherically deposited magnetic particulates that, in most cases, are indistinguishable under scanning electron microscopy (SEM) from power-station fly ash (Hunt 1988).

The magnetic properties of most lake sediments are largely controlled by the nature of the magnetic minerals washed in from the catchment. Only where the contributions from these and from any authigenic components are consistently sparse and relatively uniform will it be easy to distinguish an atmospherically deposited component. The magnetic properties of all the lake sediment profiles used in both the Surface Water Acidification Project (SWAP) and the U.K. Department of Environment research programmes were measured to assess the value of magnetic records as indicators of the deposition of air-borne pollutants from combustion and industrial sources. One of the main advantages of magnetic measurements is their non-destructive character. This allows the same subsamples to be used for dating by γ assay (also non-destructive) and subsequent trace-metal or other analyses. The main disadvantage arises from the failure of the technique to give precise quantitative information on unambiguously identified mineral phases.

Methods and interpretation

The most frequent approach to establishing magnetic concentrations in samples involves measurement of magnetic susceptibility. The majority of samples used in this study were too small or too poor in ferrimagnetic minerals to make routine susceptibility measurements reliable or informative. Magnetic remanence measurements alone were used.

Dried and powdered sediment samples were weighed into 10 ml polystyrene pots that had been acid washed, pre-measured and selected for minimum magnetic contamination. In all cases the samples were given an anhysteretic remanent magnetization (ARM) by using a peak AF field of 100 mT and a DC field of 0.04 mT. In some cases stepwise acquisition of isothermal remanent magnetization (IRM) was measured by using forward fields of 20 mT and 300 mT. 'Saturation' isothermal remanent magnetization (SIRM) was grown for all samples in a DC field of 1 T. A partial stepwise demagnetization of SIRM was done on all samples by using reverse fields of 20 mT, 40 mT, 100 mT and 300 mT; ARM acquisition was achieved by means of a suitably adapted Molspin AF demagnetizer and IRMs were generated by using Molspin pulse magnetizers. At each step, the remanence retained by the sample was measured by using a Minispin slow-speed spinner fluxgate magnetometer. Measurements of laboratory remanences such as ARM and IRM can be used both to characterize and, within the limits set by the between-sample variations in magnetic-grain size and mineralogy, to quantify magnetic mineral assemblages; SIRM measurements as defined and measured in this study integrate contributions from all the isothermal remanence bearing components of the magnetic mineral assemblages in a sample. These different components can, to some degree, be differentiated by using the approach outlined below.

In presenting and interpreting the results of the magnetic remanence studies it has been useful to distinguish three mutually independent measurements each one related to the concentration of one or more magnetic components.

1. That part of the SIRM that can be demagnetized in a reverse DC field of 20 mT ($\mathrm{SIRM} - \mathrm{IRM}_{-20\,\mathrm{mT}}$). For the samples measured in this study, this 'soft' remanence component provides the best available approximate indication of the relative importance of magnetite. Thompson's (1986) calculations suggest that this measurement will be roughly proportional to the concentration of magnetite across a wide range of grain sizes with diameters above 0.0625 μm.

2. That part of the SIRM that remains unreversed in a reverse DC field of 300 mT ($\mathrm{SIRM} + \mathrm{IRM}_{-300\,\mathrm{mT}}$). This 'hard' remanence component can be used as a rough guide to the relative importance of haematite in the sample as the remanence held by almost all forms of magnetite saturates in fields lower than this.

These two calculated remanence properties provide the main basis for reconstructing the record of atmospheric deposition at sites where it is possible to distinguish either or both components from the catchment input. Simpler calculations by using remanences grown during stepwise acquisition of IRM can be used for the same purpose, but reverse-field measurements have been used in this study, largely because they can be precisely remeasured and confirmed much more easily. Moreover, the forward field ratios are available only for the cores measured towards the end of the programme.

3. The ARM values. For a given concentration of magnetite, ARM values reach a maximum both in absolute terms and also relative to IRM values, in true stable single domain grains with diameters around 0.02–0.04 μm (Maher 1988). The magnetic properties of most fly-ash samples and of most igneous rocks are dominated by coarser grains whereas secondary ferrimagnetic oxides formed in soils often include higher concentrations of these fine grains. A catchment-derived input of magnetic minerals may therefore have a wide range of ARM and SIRM/ARM values depending on the sources and types of material represented. Both catchment and atmospheric sources will contribute to ARM values; moreover, SIRM/ARM quotients will be

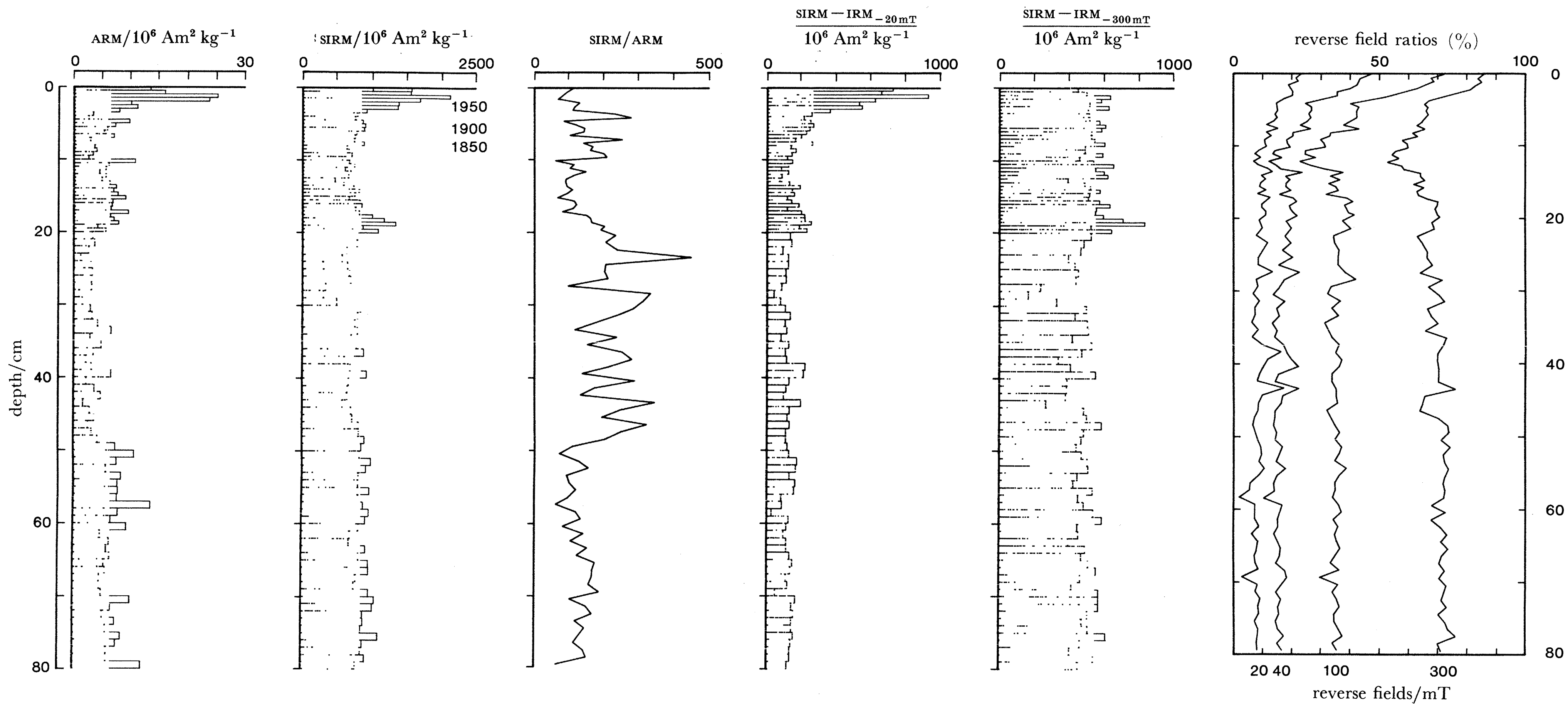

FIGURE 1. Magnetic remanence measurements done on subsamples from Lochan Uaine, Core UAI2. The sequence of measurements is described and explained in the text. In the reverse-field ratio plot the upper scale signifies percentage reverse saturation. Thus 50% represents the stage at which demagnetization of the original SIRM has reduced net remanent magnetization to zero; 100% represents the point at which the original SIRM is fully reversed. The figures below the graph are the reverse fields in millitesla (mT) used at each stage in demagnetization. The dates shown are based on a ^{210}Pb chronology derived from gamma measurements (Appleby *et al.*, this symposium).

strongly influenced by the haematite component of the sample. Because of the variety and complexity of magnetic phases in the present samples, the ARM measurements therefore contribute to an understanding of the range of variation in the magnetic mineralogy of the samples without identifying any specific magnetic component.

In the figures presented here, these three properties are plotted on a mass specific basis as $10^{-6}\,Am^2\,kg^{-1}$. The results as compiled from the full range of measurements done are illustrated by the diagram for Lochan Uaine (figure 1).

Results

In all, 39 cores from 32 sites have been measured. In terms of their magnetic record, the cores fall into the following categories.

1. Those in which the catchment input of magnetic minerals is too high and variable to permit the isolation of an atmospheric component (Flower *et al.* 1987).

2. Those where an atmospherically derived component in the magnetic record can be inferred only circumstantially through a comparison between the magnetic and the trace metal or carbonaceous particle deposition histories.

3. Those where, despite significant catchment input of magnetic minerals, one component (either magnetite or haematite) of the atmospheric deposition record can be distinguished on the basis of its magnetic properties. The records from Lochan Uaine in the Cairngorms (figure 1) and from Lilla Öresjön (Renberg *et al.*, this symposium) illustrate this.

4. Those where the catchment input of magnetic minerals is negligible compared with that from atmospheric deposition. At such sites, both the magnetite and haematite components of the atmospheric deposition record can be identified confidently (figure 2).

Discussion and conclusions

Previously published results from Loch Tanna (Arran), Lochnagar (Cairngorms), Scoat Tarn (Lake District) and Llyn Dulyn (N. Wales) illustrate the consistency of the magnetic record from widely scattered sites (Battarbee *et al.* 1988). In each lake, concentrations and fluxes of magnetic oxides increase up to the present day from late 19th and early 20th century levels. At the two Scottish sites the magnetic accumulation rate accelerates steeply after 1960, whereas at the more southerly sites, the acceleration in magnetic accumulation begins earlier, between 1915 and 1930, but still steepens around 1960. At Lochan Uaine (figure 1), one of the SWAP sites, the 'soft' remanence component ($\text{SIRM}-\text{IRM}_{-20\,\text{mT}}$), representing changing magnetite input, increases steeply in the top 3.5 cm, i.e. from *ca.* 1940 onwards. In the sediments from Dubh Loch in the Cairngorms (figure 2), the SIRM values and those for both the magnetically 'soft' magnetite component indicated by $\text{SIRM}-\text{IRM}_{-20\,\text{mT}}$ and the 'hard' haematite component indicated by $\text{SIRM}+\text{IRM}_{-300\,\text{mT}}$ begin to increase gently around 1920–1930, then more steeply from *ca.* 1960 onwards. The record closely parallels that previously published from Lochnagar (Battarbee *et al.* 1988).

Figure 2 also plots results from undated cores from Loch na Larach, Scotland (National Grid ref. NC 217583 and Llyn Irddyn (National Grid ref. SH 629222) and Llyn Glas, Wales (National Grid ref. SH 600546). In all cases there is a steep increase in SIRM close to the sediment surface following uniformly low values below. Comparison between the traces from

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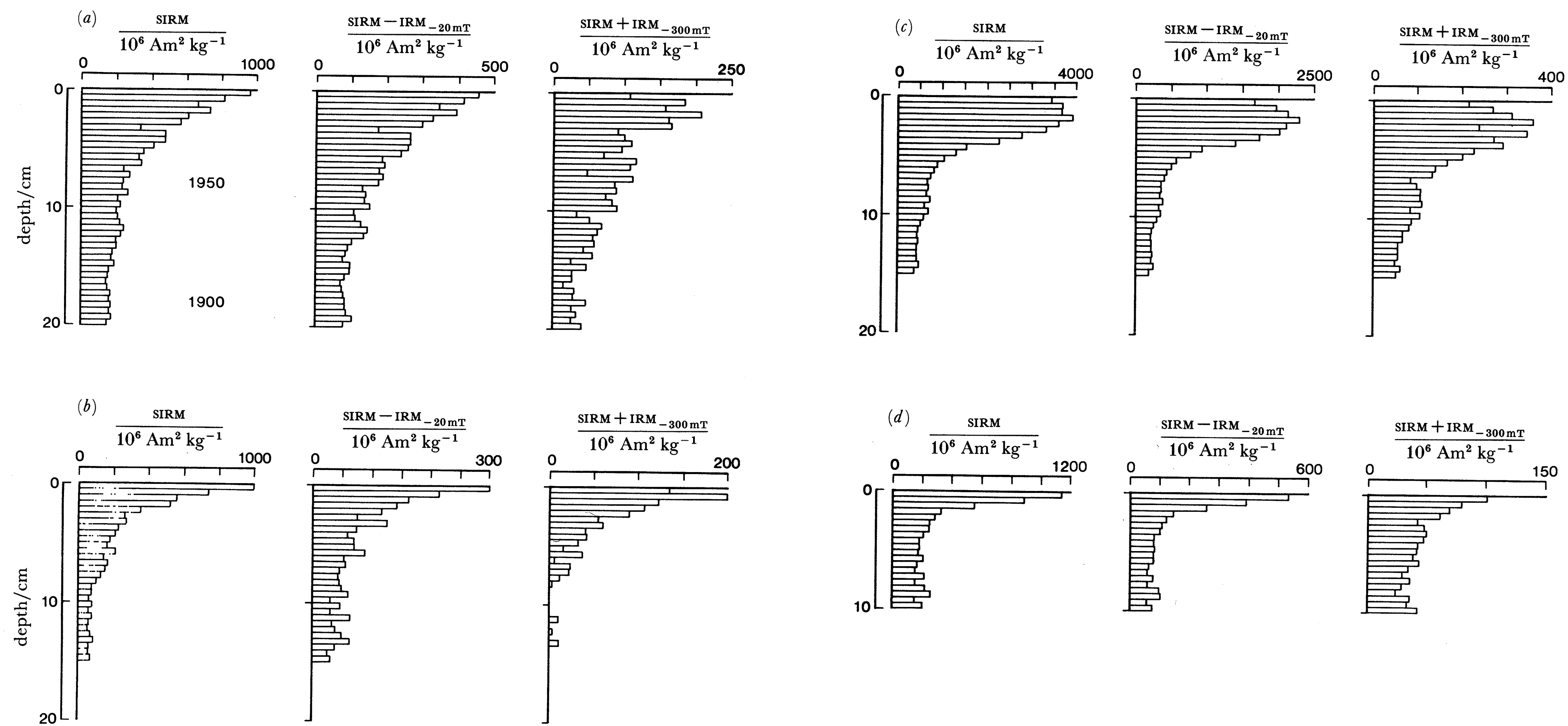

FIGURE 2. Profiles of SIRM, SIRM − $IRM_{-20\ mT}$ and SIRM + $IRM_{-300\ mT}$ for cores from (*a*) Dubh Loch (Grid ref. NO 238825) and (*b*) Loch na Larach (Grid ref. NC 217583) in the Scottish Highlands and from (*c*) Llyn Irddyn (Grid ref. SSH 629222) and (*d*) Llyn Glas (Grid ref. SH 600546), in upland Wales. The dates on the Dubh Loch graph are based on a ^{210}Pb chronology derived from γ measurements (Appleby *et al.*, this symposium). The other three cores, which form part of the U.K. Department of Environment programme, have not yet been dated.

Loch na Larach and Llyn Glas illustrates well the effect of catchment lithology on the record at different sites. At Loch na Larach, catchment input before measurable atmospheric deposition is dominated by 'soft' magnetite, with little or no input of unambiguously defined haematite. The 'hard' magnetic component then rises from values at or close to zero to near-surface peak values approaching 200×10^{-6} Am^2 kg^{-1}. At Llyn Glas, the catchment input is dominated by hard remanence minerals and in consequence the 'background' hard IRM values average 30–40 % of the surface peaks. In this respect, the magnetic record from Lochan Uaine resembles that from Llyn Glas in having a very strong catchment input of haematite upon which the record of atmospheric deposition is superimposed. In contrast, the record from Lilla Öresjön (Renberg *et al.*, this symposium) has a negligible haematite but relatively strong magnetite input from the catchment and is thus more comparable with Loch na Larach.

Although the value of the magnetic record as an indicator of atmospherically derived industrial particulate deposition in recent lake sediments is strongly dependent on the lithology of, and surface processes operating within, the lake catchment, in some 70 % of the cores studied it indicates an increase in deposition from the early–mid 19th century onwards followed by a steep rise in the last 3–5 decades.

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