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Recent Applications of Mineral Magnetic Methods in Sediment Pollution Studies: a Review

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Abstract
This paper reviews recent progress in applying mineral magnetic methods in sediment pollution studies. Such applications include its use as a dating marker, as a proxy for heavy metal concentrations and to trace metal pollutant dispersal. The mineral magnetic method has been found to be a promising tool in a wide range of sediment metal pollution studies. However, its use as a proxy of heavy metal concentrations is not always straightforward. This reflects the potentially mixed origins of magnetic minerals in sediments which may have an anthropogenic, natural or mixed source. Furthermore, anthropogenic magnetic particles may not have a common source with heavy metals. The possible linkage between magnetic minerals and heavy metals is discussed. The role of sorting, sorption/desorption and post-depositional diagenesis on the magnetic mineral-heavy metal linkage is highlighted as still requiring careful consideration. It is suggested that detailed characterisation of magnetic mineralogy using combined magnetic, geochemical and mineralogical methods is critical to the optimization of sediment pollution studies using a mineral magnetic approach.

Keywords Mineral magnetic method · Heavy metals · Sediment pollution · Dating marker · Proxies · Tracer studies

Introduction

Since the 1970s, mineral magnetic methods have been applied to a wide range of sediment pollution studies (e.g. [1, 2]). The key concept is that anthropogenic magnetic particles released into the environment generally lead to the magnetic enhancement of the sediments in which they accumulate. Since anthropogenic magnetic particles are generally associated with heavy metals, the magnetic properties (e.g. magnetic susceptibility) of such deposits have subsequently been used as proxies for heavy metal concentrations. The linkage between magnetic properties and heavy metal pollution has been examined in various environmental materials, including peat, atmospheric dust, tree bark and leaves, soils and sediments. With such a range of applications and the use of this technique over several decades, a number of reviews of environmental magnetism has been published, e.g. Thompson and Oldfield [3], Verosub and Roberts [4], Petrovsky and Ellwood [5], Evans and Heller [6], Maher [7], Liu et al. [8] and Hofman et al. [9], in which the principles of applying the mineral magnetic method and development of applications are summarised.

In this paper, we examine recent progress (i.e. the last 5 years) in the use of mineral magnetic methods in sediment pollution studies. The aim is to highlight the challenges and prospects for environmental magnetism in such studies and inform future research directions.

This article is part of the Topical Collection on *Sediment Pollution*

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Recent Progress

Environmental magnetism has a wide application in heavy metal pollution studies in lacustrine, riverine, estuarine and coastal environments (Table 1). In this review, we will discuss developments in such studies under three main aspects.

Magnetic Properties as an Anthropocene Dating Marker

The term of ‘Anthropocene’ has been coined to describe recent geological history strongly impacted by human activities [25]. Although the starting point for the Anthropocene is still debated [26–28], some suggest that the Anthropocene reflects the impact of the Industrial Revolution over the last 200 years [29]. In order to find a global dating marker, for practical purposes, the 1950s has been suggested as the onset of the Anthropocene since potential markers such as artificial radionuclides (e.g. ^{137}Cs, ^{241}Am, $^{239+240}Pu$) were emitted into the global environment for the first time in this decade [29]. Spherical carbonaceous particles (SCP), a product of coal combustion, have also been proposed as a potential stratigraphic signal [30]. Since anthropogenic magnetic particles (e.g. Fe_3O_4) are also produced during coal combustion, magnetic measurements can also offer a potential dating method. Furthermore, the distinct morphology (commonly a spherical appearance under scanning electron microscopy) can confirm their anthropogenic origin. In fact, peats, river and lake sediments receiving magnetic fly ash that source from solid fuel combustion and metal smelting have long been used to reconstruct atmospheric pollution histories (e.g. [1, 31]). In the Upper Sangamon River Basin, central Illinois, USA, magnetic fly ash has been used to identify post-settlement alluvium deposits [13]. Peaks in spherical magnetic particle content were found to correspond to maxima in magnetic susceptibility (χ) and Pb concentration that occurred in the mid-twentieth century. A coincident decline in magnetic fly ash sphere content and magnetic susceptibility was observed in the upper part of sediment profile in the Upper Sangamon River Basin, which could be attributed to decreased atmospheric pollution following control measures and the diluting effects of enhanced river bank erosion since the 1980s [13]. Chudaničová et al. [12] studied the magnetic profile of floodplain deposits in the Czech Republic and UK to explore the application of environmental magnetism as dating tools. In combination with ^{137}Cs dating, they found that some sites showed magnetic susceptibility enhancement in the profile of the overbank deposits and a single peak in χ values was comparable with the timeframe of regional industrial activity. These magnetic profiles had the greatest dating potential in aggrading river environments, but the approach was not successful at all sites.

Variations in the concentration-related magnetic parameters in sediment cores can therefore record a history of air pollution related to coal combustion and industrial activities. However, it has also been found that the onset of air pollution is site dependent, with developed countries showing an earlier start time for elevated air pollution levels than developing countries [31]. Whether anthropogenic magnetic particles can be used as a global stratigraphic marker should therefore be considered with caution. The possible post-depositional dissolution of magnetic particles under reducing environment also needs to be considered [31, 32].

Table 1 Examples of the application of environmental magnetism to pollution studies

Research purpose	Environment	Region	References
Pollution history/dating	Lake	China	[10]
		Romania	[11]
	Floodplain	Czechia	[12]
		USA	[13]
	Coast	China	[14]
		India	[15]; [16]
Heavy metal proxies	River	Bulgaria	[17]
		Spain	[18]
		UK	[19]
	Coast	China	[20]; [21]
		India	[22]
Pollution transport	River	Croatia	[23]
		France	[24]

Magnetic Properties as Proxies of Heavy Metal Concentrations

Due to the mineral magnetic method’s attributes of being simple, rapid and non-destructive, magnetic properties have been widely used as proxies of heavy metal concentrations in surface and sediment cores. Commonly, magnetic mineral concentration-related parameters, such as magnetic susceptibility and saturation isothermal remanent magnetisation (SIRM), are used for this purpose. They are correlated either with heavy metal concentrations or combined indices such as the Tomlinson Pollution Load Index (PLI) [10, 20, 22]. In the lakes in the Romanian Carpathians, Akinyemi et al. [11] found that the magnetic properties of sediment cores can indicate recent atmospheric particulate deposition, with SIRM values showing similar patterns to Pb and Zn concentrations. However, relationships between SIRM and Co, Cr, Cu and Ni were poor, suggesting that Pb and Zn have a source different to that of Co, Cr, Cu and Ni. A study by Ma et al. [10] of reservoir sediment in northern China, downwind of a steel plant in Linfen City, showed that concentration-related magnetic parameters χ, SIRM and anhysteretic remanent magnetisation (ARM) had significant correlations with heavy metal (Al, Ni, V, Pb, Cr, Fe and Cu) concentrations and the PLI.

In addition to concentration-related magnetic parameters, grain size-sensitive parameters have also been found to correlate with heavy metals in certain environments. In a study of surface sediments from the Anllóns River, Spain, Costanzo-Alvarez et al. [18] found that κ_{FD}% (frequency dependent susceptibility) best reflected the changes in the PLI values. Jordanova et al. [17] studied soil profiles from the alluvial plain of the Ogosta River, Bulgaria, which is impacted by Fe-Pb mining activities. They found good relationships between As and magnetic susceptibility, which was interpreted as reflecting the absorption of As by iron oxides. In an earlier study, Zhang et al. [33] revealed that susceptibility of ARM (χ_{ARM}) displayed significant correlations with Cu, Zn and Pb in intertidal sediments in the Yangtze Estuary, China. Similarly, the ratio of χ_{ARM}/SIRM and χ_{ARM}/χ, and, to a lesser degree, frequency dependent susceptibility (χ_{fd}), displayed significant correlations with heavy metals as well as the fine sediment fraction (< 16 μm). In India, similar studies of tidal mud flats [15], estuarine [34] and continental shelf sediments [35] also confirm such an association. This relationship can be explained by the roles of sediment particle size and iron oxides in controlling metal concentrations. These studies indicate that heavy metals are linked to magnetic particles either through a common source or by the sorption of heavy metals on fine-grained iron oxides in the sediments. The former relationship is most pronounced in areas with a strong direct pollution source, while the latter tends to be found in environments where heavy metals show a strong affinity for clay minerals and iron oxides.

Considering that the particle size of sediments can play an important role in influencing heavy metal concentrations, it is necessary to distinguish particle size effects from those related to the influence of pollution. This is normally carried out by normalising heavy metal concentrations to conservative elements such as Al. Since magnetic parameters like χ_{ARM} are a good proxy of particle size as well as iron oxides [36], it has been proposed that χ_{ARM} can be used to normalise particle size effects [33]. However, as mentioned above, post-depositional dissolution may also detract from the use of magnetic parameters as a particle size normaliser.

Tracing Pollutant Transport Using Mineral Magnetic Methods

During a 6-month survey, Nizou et al. [24] used magnetic properties as fingerprints to trace the dispersal of dredged sediments in the Bay of the Seine, northern France. They found that dredged navigation channel sediments had a relatively high magnetic susceptibility so that the dumping sites showed contrasting magnetic properties before and after dumping. A study by Frančišković-Bilinski et al. [23] showed that magnetic parameter χ declined with increasing distance from a coal slag pollution source in river bed sediments near Duga Resa, Croatia, while χ_{fd} % showed the opposite trend. They interpreted the magnetic signals as reflecting a decrease in magnetic pollutant particles downstream, while the proportion of soil-derived fine-grained magnetic particle increased. They found that χ showed a strong correlation with Co, Fe, Sr, Al, U and B, while other metals Cd, Ni, Cr and Mo increased downstream. In an earlier study, Franke et al. [37] examined suspended matter in the Seine River system, France, and found an increase in magnetic concentration from upstream to downstream regions. They also found that magnetic mineralogy changed from high-coercivity minerals toward magnetite-like minerals from upstream to downstream, and interpreted this trend as an indication of increasing input of pollution toward the mouth of the river. Such spatial sampling can provide insights into pollutant transport processes and deposition patterns from source to sink, which are critical to assessment of the pollution and the surrounding environment. In another study of the salt marshes in the southern Bay of Biscay, Spain, magnetic susceptibility was found to be capable of indicating subtle anthropogenic disruptions in the sedimentary record, although it did not correlate with heavy metal concentrations in the sediments [38].

Challenges and Perspectives in Applying Environmental Magnetism in Sediment Pollution Studies

The breadth of the examples illustrated in this review indicates that the mineral magnetic method remains a promising tool in sediment pollution studies. However, its application as proxy of heavy metal concentrations needs careful consideration for several reasons (Fig. 1). First, magnetic minerals in sediments are derived from lithogenic, pedogenic and anthropogenic sources. Therefore, it is necessary to disentangle these signals for the best application of the technique. Normally, bulk magnetic parameters cannot discriminate these sources. In addition, variations in sediment particle size composition can influence magnetic properties [36, 39]. When sediments show greater temporal/spatial variations in particle size, magnetic measurements on a single particle size fraction can help to reduce such a particle size effect [40, 41]. Alternatively, Chudaničová and Hutchinson [42] developed a novel approach, based on data mining methods, to identify a characteristic magnetic signature in overbank sediments exhibiting anthropogenically induced magnetic enhancement, distinguishing them from background values and providing a means of rapid pollution determination.

Second, even when magnetic mineral particles are primarily derived from anthropogenic sources, the linkage with heavy metals is still poorly understood [23]. Some heavy metals may have a common source with magnetic particles, while others may not. In certain environments, the linkage is

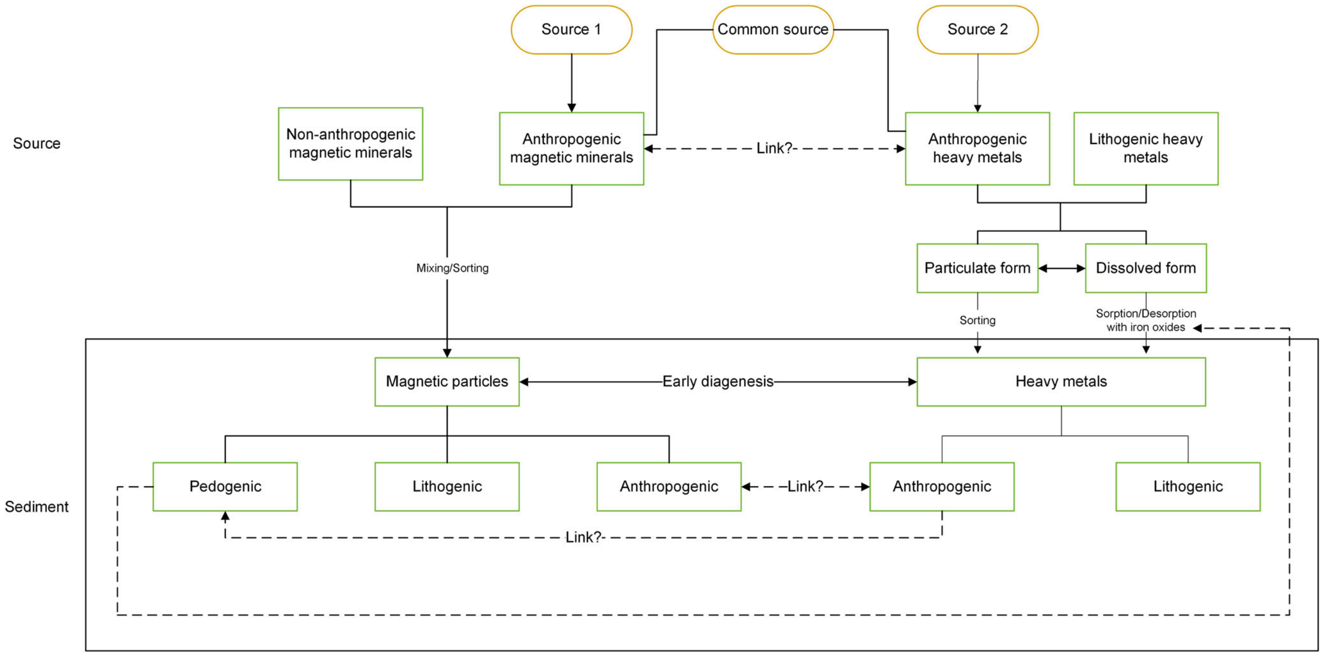

Fig. 1 Possible magnetic mineral-heavy metal linkages in sediments: a source-to-sink view of the influencing factors. The orange rounded rectangles represent the notional sources of anthropogenic magnetic particles and heavy metals; yellow boxes indicate the components of the source-to-sink pathway of magnetic particles and heavy metals and the blue boxes highlight key influencing processes

due to the absorption of heavy metals by iron oxides. This is reflected in the diverse relationships between heavy metals and magnetic parameters. As a result, detailed characterisation of magnetic mineralogy is needed to optimise pollution studies. Magnetic measurements, in combination with XRD and SEM, have commonly been applied to pollution studies. In a study of dust collected from underground train stations in Shanghai, China, thermomagnetic and XRD analysis confirmed the presence of an iron phase in the dust caused by abrasion of the rails [43]. More advanced techniques, such as synchrotron-based techniques, have also been used for microscale studies. For example, Yu and Lu [44] applied micro-X-ray fluorescence (μ-XRF) and micro-X-ray absorption near-edge structure spectroscopy (μ-XANES) to study the distribution of heavy metals on anthropogenic magnetic particles in soils contaminated by the steel industry in China. They found that the magnetic particles included iron, ferroalloy and magnetite by applying the μ-XANES technique. The μ-XRF mapping of the magnetic particles indicated that Co, Pb, Cu, Cr and Ni were mainly derived from ferroalloy particles, while As had an additional association with the iron phase. Such a high resolution study at a micrometre scale is helpful in identifying the mineralogy of magnetic particles as well as their source and linkage with heavy metals. It should also be noted that such a microscale study should ensure that its sampling is adequately representative to allow for the heterogeneity of magnetic particles [45].

Third, the possible post-depositional diagenesis of magnetic minerals must be considered. It has been well established that the formation/dissolution of magnetic minerals can lead to mineralogical as well as magnetic grain size changes [46, 47]. As a result, heavy metals can be either absorbed onto magnetic particles or released, thus altering the original relationship between sediment magnetic properties and heavy metal levels [48].

Fourth, hydrodynamic sorting effects along the source-to-sink pathway can alter the linkage between magnetic minerals and heavy metals. A number of studies have shown that sorting can lead to the selective transport of magnetic minerals [49]. In certain energetic environments, coarse-grained magnetic minerals are enriched due to the winnowing of fine-grained particles [49–51]. On the other hand, the selective removal of coarse-grained magnetic minerals in suspended sediment can lead to fine-grained magnetic particles being transported greater distances [14, 50–53]. Such a sorting effect may not only influence the grain size of sediments, but also magnetic mineral assemblages and heavy metal concentrations. In a study of pollution in a reservoir in the Czech Republic, Sebastian et al. [16] found a significant spatial variation in sediment grain size, geochemistry and magnetic susceptibility, reflecting changing hydrodynamic conditions alongside sediment provenance. It is anticipated that ultrafine-grained magnetic minerals, such as nanoparticles, will be enriched in hydrodynamically weak environments. Due to their large specific surface area and affinity for heavy metals, their role in heavy metal transport and accumulation should be given proper attention.

Lastly, the relative importance of anti-ferromagnetic minerals (e.g., hematite and goethite) should be emphasised. These minerals are normally more abundant than magnetite in pristine sediments and they are also effective carriers of heavy metals [54]. Magnetic measurements, together with other techniques such as diffuse reflective spectroscopy (DRS), can characterise these magnetically weak minerals [14, 50, 51, 55, 56]. The proper combination of these techniques can provide a better and rapid insight into the source, transformation and storage of heavy metals in the environment.

Conclusions

Environmental magnetism has gained increasing use in sediment pollution studies since its early development as a tool in palaeoscience. The rapid and cost-effective nature of mineral magnetic measurements suggests that the technique can be used to analyse a relatively large number of samples over a study area, which enhances the investigation resolution on both temporal and spatial scales. It has been increasingly found that local-scale issues and a finer level of resolution are important in environmental pollution studies. This method is powerful in detecting pollution caused by industrial activities such as coal-combustion and steel smelting. However, its use as proxy of heavy metal concentrations is not always straightforward. This is partly due to the multiple pedogenic, lithogenic and anthropogenic origins of magnetic minerals. On the other hand, heavy metals discharged into the sediments may come from sources other than those associated with the emission of anthropogenic magnetic particles. Furthermore, hydrodynamic sorting and post-depositional diagenesis may affect both magnetic mineral assemblages and heavy metal behaviour. To make optimum use of environmental magnetism in pollution studies, a detailed characterisation of the sediment's magnetic mineralogy is critical and can provide information regarding sediment provenance, transport and diagenesis. In addition to magnetically strong minerals such as iron and magnetite, the more abundant, but magnetically weaker minerals such as hematite and goethite, merit further attention considering their close link with heavy metals in the environment. Consequently, an interdisciplinary approach including magnetic, geochemical and mineralogical analyses is recommended to provide more insight into mineral magnetic-heavy metal linkages and to advance sediment pollution studies and the mineral magnetic approach.

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Compliance with Ethical Standards

Conflict of Interest On behalf of all authors, the corresponding author states that there is no conflict of interest.

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