Environmental Science & Technology

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Critical Review

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Environ. Sci. Technol., **Just Accepted Manuscript** • Publication Date (Web): 25 May 2017

Downloaded from http://pubs.acs.org on May 31, 2017

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Biomagnetic monitoring of atmospheric pollution: a review of magnetic signatures from biological sensors

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KEYWORDS: Air pollution, PM, NO_x, PAHs, heavy metals, biomagnetic, monitoring, magnetism, SIRM, susceptibility, urban

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Abstract

Biomagnetic monitoring of atmospheric pollution is a growing application in the field of environmental magnetism. Particulate matter (PM) in atmospheric pollution contains readily-measurable concentrations of magnetic minerals. Biological surfaces, exposed to atmospheric pollution, accumulate magnetic particles over time, providing a record of location-specific, time-integrated air quality information. This review summarizes current knowledge of biological material ('sensors') used for biomagnetic monitoring purposes. Our work addresses: the range of magnetic properties reported for lichens, mosses, leaves, bark, trunk wood, insects, crustaceans, mammal and human tissues; their associations with atmospheric pollutant species (PM, NOx, trace elements, PAHs); the pros and cons of biomagnetic monitoring of atmospheric pollution; current challenges for large-scale implementation of biomagnetic monitoring; and future perspectives. A summary table is presented, with the aim of aiding researchers and policy makers in selecting the most suitable biological sensor for their intended biomagnetic monitoring purpose.

1. Introduction

Since 1950, the world population more than doubled, the number of cars increased tenfold and the proportion of people living in urban areas increased by a factor of four[1]. This growing urbanization has had detrimental consequences for urban air quality. The urban air quality database of the World Health Organisation (WHO, 2014), covering 1600 cities over 91 countries, reveals that only 12% of the urban population resides in cities that meet their air quality guidelines; about half of the urban population is exposed to levels >2.5 times those guidelines.

Urban atmospheric pollution levels vary both spatially and temporally[2–4]. The spatial variation is mainly linked to distance to contributing pollutant sources, differences in traffic intensity, and urban topology. Temporal variations reflect day-to-day (meteorological and urban background fluctuations), within-day (traffic dynamics) and microscale variability (single short-lived events)[5]. Air quality assessments are inherently challenging since high monitoring resolution needs, ideally, to be achieved in both space and time.

Current telemetric monitoring networks comprise accurate physicochemical monitoring instrumentation to trace atmospheric concentrations of, among others, particulate matter (PM), nitrogen oxides (NO_x), sulfur dioxide (SO_2) and ozone (O_3) at high temporal resolution. However, high investment and maintenance costs spatially limit this type of monitoring coverage in urban environments. Moreover, with regard to PM pollution, it is generally recognized that morphological and chemical aerosol properties are more relevant to human health than the total PM mass, yet so far the latter is the only parameter routinely monitored[6–9]. The morphological and chemical properties of PM are usually determined through time-consuming laboratory analysis, such as single-particle chemical or microscopic analysis, or bulk analysis of trace elements or isotope ratios[10]. Such studies indicate the need to monitor additional pollutant species, e.g., $PM_{2.5}$, PM_1, black carbon (BC), polycyclic aromatic hydrocarbons (PAHs), volatile organic compounds (VOCs), ultrafine particles (UFPs, <0.1 µm)[9,11–16].

In addition to telemetric monitoring networks, higher spatial resolution in air quality data is typically obtained using: (1) mobile and/or “low-cost” sensors[7,17–21]; (2) specific short-term monitoring campaigns[22,23]; and (3) air quality modelling[24–27]. However, these approaches have their limitations: (1) mobile-sensor platforms need repeated measurements to untwine spatial from temporal variability[5], (2) the representativeness of short-term campaigns is uncertain, and (3) air quality models require adequate validation data[25]. These limitations are particularly important for short-lived and/or highly-variable pollutant species, e.g., UFPs, BC and heavy metals, which are known to exert adverse health effects[11,15,16,28]. Current and future air quality monitoring strategies, therefore, face the dual need for greater spatial coverage and information on health-related pollutant species, at feasible levels of cost. One might, however, question the future feasibility of monitoring a growing number of pollutants at both high temporal and spatial resolution. Biomagnetic monitoring - evaluating magnetic properties of biological material - may potentially serve both purposes, acting as a widely-applicable, low-cost method for assessing health-relevant pollutant species.

Biomagnetic monitoring is a growing application in the field of environmental magnetism, i.e., the use of magnetic measurements to study environmental systems[29,30]. The ubiquitous presence of remanence-capable magnetic particles (including anthropogenic particles) in the air, soil, sediments, rocks and organisms provides the opportunity to identify and quantify the formation, sources, transport and deposition of these particles. Atmospheric pollution, in particular urban PM, often contains levels of magnetic minerals, e.g., iron oxides like magnetite, hematite and maghemite[30–32], that are easily measurable magnetically. For more information on the different properties of magnetic minerals, domain states and grain sizes, and their responses to induced magnetic fields, please refer to SI 1.

Exposed biological surfaces, e.g. lichens, mosses and leaves, accumulate atmospheric particles, providing a record of location-specific and time-integrated information of local air quality. Magnetic monitoring of these biological sensors can add valuable spatial data to existing air quality monitoring networks and has been successfully applied to evaluate local air quality model performances[33–36]. Trace metals, such as zinc (Zn), cadmium (Cd), lead (Pb) and chromium (Cr), are often directly associated with magnetic PM, e.g. due to their incorporation in the mineral structure during combustion processes[37,38]. Therefore, the magnetic signal may act not only as a PM proxy but be of direct, often health-related, interest in itself.

The aim of this work is to summarise the different biological sensors so far used in biomagnetic monitoring studies, their pros and cons, and reported associations with atmospheric pollutant species (PM, NOx, heavy metals and PAHs). Our review encompasses worldwide, active (introduced) and passive (extant) biomagnetic monitoring studies; including lichens, mosses, plant leaves, tree bark and trunk wood, insects, crustaceans, and mammal and human tissue. Current challenges and future perspectives regarding the application of biomagnetic monitoring in air quality assessments are discussed. Finally, an overview table is presented to assist researchers and policy makers in selecting suitable biological sensors for their envisaged biomagnetic monitoring purpose.

2. Sources of magnetic particles

Sources of magnetic minerals in the atmosphere include natural, crustal PM sources, including volcanic eruptions and wind erosion of soil and dust, and anthropogenic sources, including industrial and vehicular combustion, heating and abrasion processes[29]. Higher magnetic concentration values (SIRM, susceptibility) are typically measured with increasing proximity to PM sources, and with increasing source strength (e.g. traffic volume). Examples of such magnetic distance-decay abound, whether for PM emitted from volcanoes[39], industry[37,40,41], road dust[42–44] or traffic[31,38,45].

In urban environments, traffic-related PM results from both exhaust (fossil fuel combustion) and non-exhaust (brake heating and abrasion, and tyre and road abrasion) processes[46–49]. Ubiquitous and often abundant in urban PM, iron-rich particles (frequently spherical) exhibit strongly magnetic (ferrimagnetic) behaviour[43,44,50–52]. Magnetic and electron microscopic analyses of roadside dust identify contributions of anthropogenic PM both from fuel combustion processes[53], with higher magnetic emissions reported from petrol- rather than diesel-fuel vehicles[54], and from frictional heating and abrasion of brake pads[55]. Large magnetic contributions from railway traffic have been documented[56–58], as Mn-, Cu-, Cr- and Ba-containing ferruginous particles are emitted by wear of railway tracks, brakes, wheels and electric overhead lines[59–61]. The electrified tram/train fleets generate magnetic PM mainly through wear/abrasion rather than exhaust emissions[62].

Different types of industry (e.g. lignite/coal plants, cement production, coke production, Fe/Cu smelters, slag processing, steelworks) also emit distinctive magnetic PM[37,40,41,44,63,64], probably due to differences in fuel source, combustion temperature and/or redox conditions[63]. For example, higher magnetite contents are observed near power, cement and ore dressing plants, compared to steel or coal processing plants, probably reflecting different hematite concentrations between the sites. Traffic- and industry-derived magnetic PM have also shown to differ[44,63,65].

In terms of natural PM sources, aeolian dust plumes can contribute to high ambient PM concentrations, such as occur in areas of China, downwind of desert and loess crustal sources, where the PM toxicity is estimated to be much less (0.22 % increase in premature mortality with every 10 μg m^{-3} $PM_{2.5}$), compared with cities in Europe dominated by anthropogenic PM (6% increase)[66]. Biomagnetic monitoring of sweet chestnut leaves (*Castanea sativa*) has been used to map volcanic ash deposition from Mt. Etna, Sicily (Italy). The ash contains coarse-grained (~ 5 to 15 μm) magnetite-like particles contributing > 90% of the leaf SIRM[39].

3. Health effects of magnetic particles

Nano- and micrometer-sized magnetic PM may itself comprise a source of toxicological hazard to human health. Additionally, magnetic PM can be used as a proxy for atmospheric pollution if co-associations with other pollutant species are displayed.

3.1 Inherent toxicological properties

Magnetic iron oxide particles can exert adverse health effects, by inducing oxidative stress pathways, free radical formation and DNA damage[67–69]. Free radical formation results from the Fenton reaction, where iron(II) is stoichiometrically oxidized by H_2O_2 to iron(III), producing a hydroxyl radical (OH·)[70]. In vitro experiments examining the oxidative stress pathway of size-fractionated (0.2-10; 0.2-3; 0.5-1 μm; 20-60 nm) magnetite on human lung cells indicated acute cytotoxicity (within 24 hours), due to endocytosis, followed by reactive oxygen species (ROS) formation for all size fractions[71]. Smaller grains (<100 nm) were more cytotoxic than larger grains (~5 μm)[72].

Links have been reported between increased brain concentrations of magnetic iron compounds and brain tumors[73,74], and neurodegenerative diseases like Alzheimer's, Parkinson's and Huntington's[75–79], the latter possibly through the damaging action of magnetite-amyloid-ß complexes on neuronal circuits[80].

3.2 Biomagnetism as a proxy metric for atmospheric pollution

Notwithstanding the possible direct health impacts of airborne magnetic iron oxides, most studies have so far focused on measuring the concentration of magnetic particles (through SIRM and χ), as a proxy metric for more conventionally-monitored pollutant species, e.g. PM, NO_x, heavy metals and PAHs, co-emitted with, and/or adsorbed onto, the magnetic particles. Biomagnetic techniques, measuring the passive accumulation of airborne magnetic PM on biological surfaces, enable sensitive, rapid, and relatively cheap environmental monitoring, providing a valuable addition to conventional monitoring networks[81].

3.2.1 Particulate matter (PM)

The link between magnetic properties and PM has been investigated both directly (on filter-collected PM) and by using biological accumulation surfaces (e.g. leaves).

3.2.1.1 Filter-collected PM

The magnetisable fraction of PM_{10} often comprises a mixture of low-coercivity, magnetite-like, ferrimagnetic particles with a wide spectrum of grain sizes, related to a variety of natural and anthropogenic sources[82]. Several studies have reported the magnetic properties of atmospheric PM, collected on high-volume, pumped-air filters (SI 2). Magnetic and chemical analyses of automated urban pumped-air PM_{10}, $PM_{2.5}$ and PM_1 filters could distinguish between vehicular and crustal (local and North African wind-blown dust) particle sources[50,82]. As magnetic particles occur mainly in the fine ($PM_{2.5}$) and ultrafine ($PM_{0.1}$) particle size range, magnetic properties provide information on the most health-relevant particle size fractions[83,84]. In absence of natural inputs (e.g. sea salt, aeolian dust), strong associations are reported between the PM_{10} concentrations of pumped air samples and their susceptibility ($R^2 > 0.88$) and SIRM ($R^2 = 0.90$, n = 54, p = 0.01)[36,81,82]. For air samples from Munich, the magnetic PM concentration in PM_{10}, collected on pumped-air filters, was between 0.3 and 0.6% by mass, mainly consisting of magnetite in the size range 0.2-5 µm[85,86].

Only a few studies exist on self-designed PM collectors, based on passive particle deposition (fallout). Such artificial collectors are comparable to biological exposure surfaces as particles are collected passively and non-selectively in terms of particle size. For example, circular fallout collectors covered with plastic sheets were exposed for about 3-4 weeks in Munich (Germany) and subsequently washed with isoproponal and analysed by Mössbauer spectroscopy and magnetic techniques, yielded primarily maghemite and metallic iron particles with mean magnetic grain sizes in the range 0.1–0.7 µm[56]. Another study using small filter bags with natural wool sorbents, collected mainly 2-25 µm-sized particles and yielded consistent magnetic susceptibility and coercivity results, when compared to co-located leaf samples[87].

3.2.1.2 Leaf-deposited PM

Biological materials, such as plant leaves, accumulate airborne PM passively (but efficiently), often displaying associations between their magnetic PM and the ambient airborne PM concentrations. Depending on location (and especially climatic conditions), this accumulation process is cumulative.

A couple of studies in the U.K. reported short-term associations between magnetic properties and daily or even instantaneous PM measurements have been reported. After an initial build-up period of ~ 6 days, strong correlations ($R^2 = 0.8$–0.9, n = 10, p = 0.01) were obtained between the daily-averaged atmospheric PM_{10} concentration (collected by a high-volume sampler at 1133 l min^{-1}) and daily repeated measurements of leaf SIRM of birch (*Betula pendula*) and lime (*Tilia platyphyllos*) trees[81]. Another study around at 37 locations around a coal-fired power station[40], reported a correlation (R^2 = 0.71, n = 37, p = 0.01) between leaf SIRM values and co-located handheld PM_{10} measurements (TSI SidePak AM_{510}).

Conversely, in mainland Europe, many studies suggest that leaf magnetic concentration properties reflect a time-integrated pollution exposure. A study on monthly-sampled *Nerium oleander* leaves[88] obtained no correlation between the leaf susceptibility and daily PM_{10} concentrations. Another study[84] found magnetic concentration increased with *Pinus nigra* needle exposure time (up to 55 months) and reflected exposure to environmental pollutant load at 6 locations with different emission backgrounds. For deciduous leaves, with a shorter lifespan of only several months, increases in magnetic PM content with time have been observed[45,89]. Associations have also been documented between two-weekly[90] or monthly[91] leaf SIRM and cumulative atmospheric $PM_{2.5}$ and PM_{10} concentrations throughout an entire in-leaf season. Moreover, significant correlations were also obtained between the gravimetric leaf-deposited dust load (mg m^{-2}) and the resulting SIRM (A m^2 kg^{-1}), within the 0.2 – 3, 3 – 10 and >10 µm particle size fractions[92].

3.2.2 Relationship with NO_x

As magnetic particles in urban environments are frequently associated with vehicular emissions [38,42,43,50,93], associations have been evaluated as well between magnetic concentration parameters and traffic-related gaseous pollutants (mainly NO_x: NO + NO_2). The latter namely exhibits greater spatial variation than PM[94].

In Madrid (Spain), associations were observed between *Platanus x hispanica* leaf magnetic content (SIRM and χ) and cumulative daily NO_x concentrations[45]; the relationship was weaker for PM_{10} concentrations. Similarly, stronger association between SIRM of ivy leaves and modelled atmospheric NO_2 concentrations was observed, compared to modelled PM_{10} concentrations, in a city-scale biomonitoring and modelling study in Antwerp, Belgium[58]. A significant correlation ($n = 29$, $r = 0.92$, $p < 0.001$) was found between SIRM of *Carpinus betulus* leaves at 6 monitoring locations along a vehicular traffic-gradient, and modelled NO_2 concentrations in Antwerp, Belgium[95]. In Bulgaria, a linear association (n=10) between the average magnetic susceptibility from multiple street dust samples collected in 10 different cities and the average annual atmospheric NO_2 concentrations, derived from telemetric air monitoring stations[52]. Stronger correlations with NO_x rather than PM concentrations are likely in locations where PM is not only traffic-related but has contributions from secondary aerosols, sea spray and crustal matter[45].

3.2.3 Particle-bound trace elements and PAHs

Numerous studies have reported associations between different magnetic parameters and particle-bound trace elements[42,93,96–101]. Trace elements, e.g. heavy metals, can be incorporated into the crystalline structure of magnetic particles during formation (e.g. combustion), and/or by subsequent surface adsorption[97,100,102]. Magnetic properties and magnetic-metal correlations may be valuable in PM source attribution. As, Cu, Mn, Ni, Pb, and Zn are linked to combustion particulates[99], while traffic-related heavy metals include emissions from the abrasion of tyres (Zn, Cd and Cu), brake pads and linings (Sb, Cu, Zn, Fe, Ba and Cr), corrosion (Fe, Cd, Zn, Cu, V and Ni), lubricating oils (V, Cd, Cu, Zn and Mo) or fuel additives (V, Cd, Zn and Pb)[46,103,104]. Although Fe and Mn are common in the natural

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environment, their co-occurrence with Ni, Cu, Zn, Cr, Cd, and Pb is typically associated with road traffic [49].

Relations between trace elements and magnetic parameters have been evaluated statistically by means of fuzzy models[105,106], fuzzy clustering[107,108] and principal component analysis[109]. Associations between magnetic parameters and elemental Fe, As, Cu, Mn, Ni, Pb and Zn content or the Tomlinson pollution load index (PLI) confirm that much urban heavy metal contamination is linked to combustion-derived particulate emissions[52,65,102]. High magnetic susceptibility was found to correlate with mutagenicity of atmospheric PM collected on air-pumped filters[110]. Co-association between traffic-derived Pb and resulting leaf SIRMs were found[51], despite the introduction of unleaded petrol (since 1986 in the UK). Possible non-fuel sources of Pb include lead plating of fuel tanks and lead in vulcanized fuel hoses, piston coatings, valve seats and spark plugs[51]. A recent study[111], combining SEM/EDX with leaf magnetic concentrations from different land use classes, obtained significant correlations between leaf SIRM and Fe, Zn, Pb, Mn and Cd content of deposited particles. This is in line with observed correlations between leaf susceptibility and Fe, Zn, Pb and Cu [112]; and between Cu and Fe and leaf SIRM and susceptibility[88]. Significant correlations were reported between the magnetic susceptibility of leaf and topsoil samples and Fe, Cr, Ni, Pb, Cu levels in Linfen, China[113–115]. Conversly, another study[89] related leaf susceptibility and IRM to Al and Cu in the leaf-wash solution, suggesting that in arid regions with high lithogenic PM contribution, the relationships between metal concentrations and magnetic susceptibility could be obscured.

Association was found between the PAH content of lichens and poplar leaves in Bulgaria and their SIRM[116]. Likewise, in Cologne (Germany)[117], covariance between pine needle SIRM

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and pyrene content was observed, the latter a proxy for urban PAH load. This covariance broke down for railway-proximal locations where PM originated mostly from wear and not combustion. Similarly, consistency was reported between modelled pollutant distribution (ADMS-Road model), instrumental PM_{10} monitoring and biomonitoring of 11 metals and 14 PAHs from tree (*Quercus ilex*) leaves and moss bag samples in a street canyon in Naples, Italy[35]. Washing of *Quercus ilex* leaves[118] indicates that most particle-bound trace elements (Cr, Cu, Fe, Pb, V and Zn) are deposited on the leaf surface (and therefore removed by washing), while PAHs seem to migrate more easily into epicuticular waxes.

4. Application as biological sensors

Magnetic characterization of atmospheric pollution by a few pioneering studies[31,98,119–121] was followed by magnetic studies of pumped-air filters[50,55,82,85,86,122] and subsequently a host of environmental substrates. The latter include soils; river and marine sediments; indoor and outdoor settled dust; roadside snow[123] and biological material (SI 2) including mosses and lichens; plant leaves; tree bark and trunk wood; insects; crustaceans; mammal (of which human) tissues.

The inventory table (SI 2) provides an overview of different reported biological sensors. The magnetic properties, influencing processes, identified associations with atmospheric pollutants, and applied monitoring protocols are described below for each biological sensor.

4.1 Mosses and lichens

Mosses and lichens have been used as environmental biomonitors for over 40 years; they are efficient accumulators and sensitive to multiple atmospheric pollutants[124]. They lack a rooting system, so nutrients are sourced from the atmosphere through wet and dry deposition, similar to atmospheric pollution pathways. They have a high capacity to retain metals due to the absence of a cuticle. Strong associations are usually reported between elemental levels in moss or lichen samples and bulk atmospheric deposition samples[125].

4.1.1 Trace elements, PAHs, PCBs, dioxins, furans and PBDEs

Since the 1970s, mosses and lichens have been used to monitor levels of, amongst others, metals or metalloids (Pb, Zn, Cu, Cd, Fe, Ni), NO_x and persistent organic pollutants (POPs), such as PAHs, polychlorinated biphenyls (PCBs), dioxins and furans (PCDD/Fs) and polybrominated diphenyl ethers (PBDEs)[124,126–131]. As mosses and lichens are not ubiquitous in urban environments and their identification and age difficult to determine, transplant techniques are often applied to monitor urban atmospheric pollution levels. Most frequently, pioneered by Goodman and Roberts[129], exposure bags containing lichens or mosses are hung in the urban environment to evaluate ambient pollutant levels (Figure 1).

Figure 1. Sphagnum *girgensohnii* moss bag exposure in different urban microenvironments (from [128]).

Spatial variation in moss and lichen elemental content ranges in scale from within single street canyons[34,132,133] to different land use classes[127,134]. Bulk chemical analysis (e.g. by ICP-MS) dominates but particle-based characterization (e.g. by SEM/EDX) has also been reported. For *Hypnum cupressiforme* moss bags, exposed in different roadside, industrial and green area sites in Trieste, Italy, the majority of entrapped particles (up to 98.2%) were <10 μm, dominated by Al, Ca, Fe and Si- containing particles[134]. Similarly, enrichments of Al, Cr, Fe, Na, Ni and Pb, and magnetic content were obtained in moss bag samples after snowmelt with increased road dust resuspension, and near heavily-trafficked sites in Turku, Finland[135]. Coarser particles (0.1 - 5 μm) are often observed in roadside- or industry-exposed moss samples (Figure 2), compared to less-polluted samples (particles <0.1 μm)[134,136].

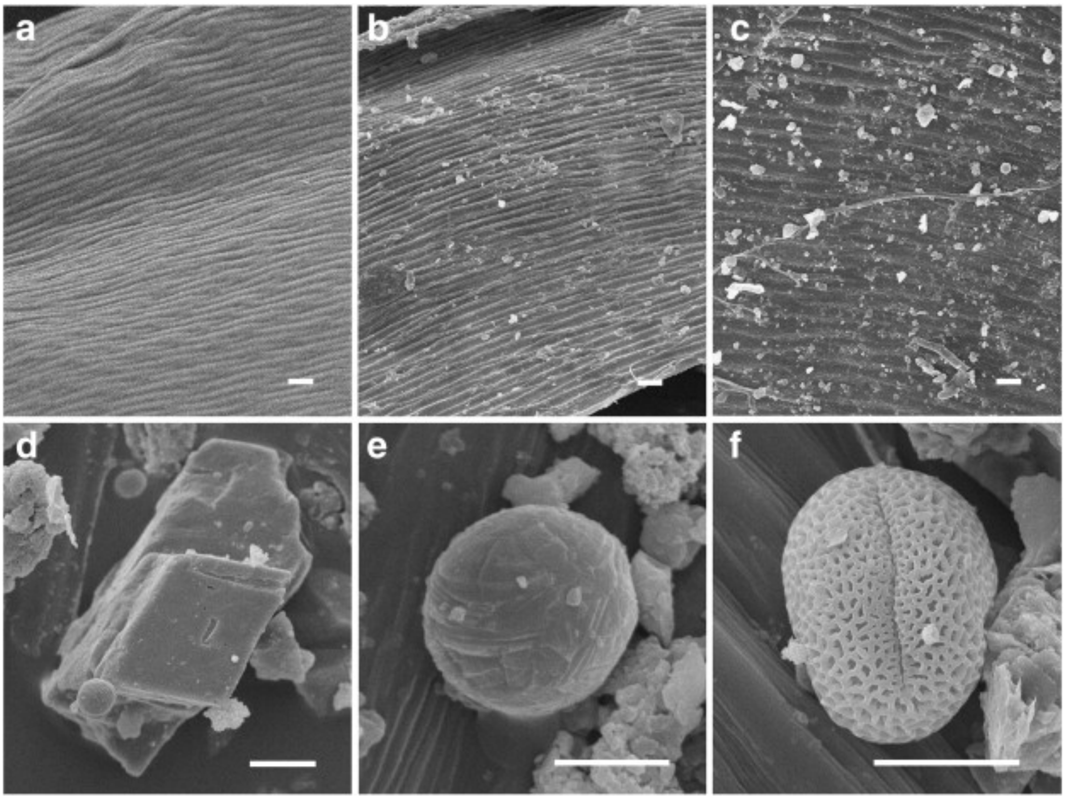

Figure 2. SEM pictures of moss leaflets before (a) and after exposure (b, c) in the green (b) and roadside (c) site with enlargement of particulate matter (d, e) and a pollen grain (f). Scale bar = 10 µm for a–d, and f and 3 µm for e (From [134]).

4.1.2 Magnetic signatures of mosses and lichens

Magnetic properties have been reported recently of terrestrial mosses and lichens[116,136,137] and moss bags[41,127,135,138–141]. Because of their high accumulation capacity and high surface:volume ratio, mosses and lichens are suitable for magnetic evaluation of environmental pollution[116]. Reported moss and lichen SIRMs range from 0.1 to 855 x 10^{-3} A m^2 kg^{-1}, while magnetic susceptibility ranges from -1.5 to 1161 x 10^{-8} m^3 kg^{-1} (SI 2).

Like tree leaves, moss and lichen magnetic properties appear species-dependent[139]. They show seasonal variations, due to changes in emissions and meteorology[124,142], and spatial variations, influenced by land use and pollutant sources' strength and proximity.

Magnetic measurements on moss samples collected along a 120 km transect through Oslo, Norway, showed higher magnetic susceptibility and IRM near the city, up to a distance of 20 km from the city center[137]. SEM analyses revealed differences in morphology, grain size (Figure 2) and chemical composition between urban and rural moss-collected dust[137,143]. Magnetic and chemical composition differences between both native and transplanted lichen samples and neighboring soil and rock samples[141], indicating an alternative source of lichen-accumulated magnetic particles, identified as the nearby cement production industry. They confirmed the cumulative nature of the magnetic PM content as the native lichen samples exhibited higher concentration-dependent magnetic properties, compared to transplanted lichens which experienced a shorter exposure period[141].

Regarding spatial variability of moss and lichen magnetism, distinct enrichment factors have been found near metallurgic factories and road traffic, with evidence of source-distance and source strength (e.g. traffic intensity) effects[41,135,136]. Associations were reported between magnetic properties of mosses and their heavy metal[138] and PAH content[116]. Magnetic content decreased with distance from the contributing anthropogenic sources (Cu-Ni smelter and road traffic) in Finland. Directional wind effects on the Cu-Ni smelter plume were observed in the moss susceptibility values and heavy metal levels[138].

4.1.3 Selection criteria and protocol

Selection of biomonitoring species appears governed by its presence/abundance in the considered study region[124], or by its availability from reference backgrounds or commercial sources. The most frequently used moss bag species belong to the *Sphagnum* genus (SI 2). Mosses and lichens display similar spatiotemporal variation in element accumulation and magnetic properties[138,142]. Mosses tend to have a higher accumulation capacity, but are more sensitive to environmental stressors (e.g. drought) than lichens[124,140]. Lichens appear more sensitive to gaseous pollutants (specifically SO_2)[144,145] and potentially lose more surface-deposited particles due to rain or wind resuspension[146].

Reviewing112 scientific studies, a standardized protocol has been presented for the preparation, exposure and post-exposure treatment of moss bags in environmental biomonitoring studies[124]. The use of a *Sphagnum palustre* clone for trace element analysis is recommended for its low and constant background element composition, and homogenous morphological characteristics[147].

4.2 Plant leaves

4.2.1 Studies and reported magnetic properties

Due to its large specific surface area (leaf area density; LAD), urban vegetation is an efficient collector of PM, and thus valued as an additional ecosystem service in terms of phytoremediation[148–155]. Plant leaves (mostly from trees) have been used in a variety of biomagnetic monitoring studies (SI 2). Needle-deposited fly ash, from power plants, has shown to result in enhanced magnetic susceptibility of the needle samples[156]. When compared with artificial PM collectors in an industrial area in Linfen, China, co-located tree leaves showed similar magnetic properties[87].

Published leaf SIRM results range widely from 0.002 to 27.50 x 10^{-3} A m^2 kg^{-1} (mass-normalised) or 4.17 x 10^{-10} to 777 x 10^{-6} A (area-normalised), whereas mass specific susceptibility ranges from -0.9 to 846 x 10^{-8} m^3 kg^{-1} (SI 2, 46 studies). Although these ranges are large (depending on the applied plant species, sampling location and exposure time), leaf surface particle accumulation capacity appears lower than moss and lichen tissues. This might be explained by the absence of a cuticle in mosses and lichens, since particle deposition processes (dry and wet deposition, impaction and interception) and accumulation periods are similar or at least comparable.

4.2.2 Influencing factors

The particle accumulation efficiency of the leafy biomass varies between plant species, influenced by their phenology (deciduous vs evergreen), leaf area density (LAD) and leaf characteristics, e.g. wax layer properties, micro-surface roughness and presence of trichomes (Figure 3), i.e. hair-like features on the leaf surface[148,149,157–159].

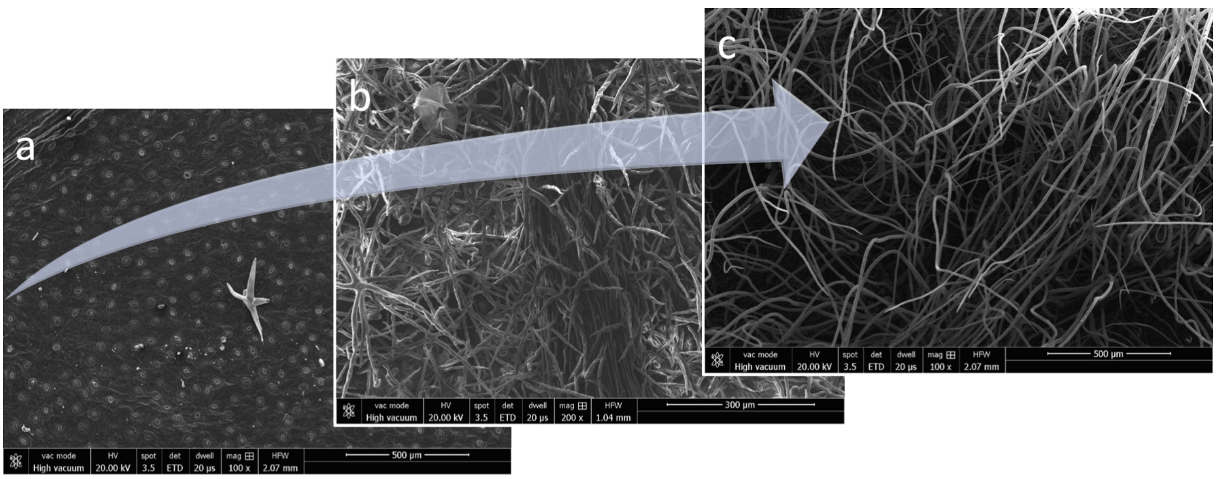

Figure 3. SEM pictures illustrating the hairiness (trichome) gradient observed between abaxial leaf surfaces of *Hedera hibernica* (a), *Buddleja davidii* (b) and *Stachis byzantina* (c).

Comparing particle loadings on leaves of 22 trees and 25 shrub species[148], *Pinus mugo, Pinus sylvestris, Taxus media, Taxus baccata, Stephanandra incisa* and *Betula pendula* were identified as most efficient accumulators of PM_{10}, $PM_{2.5}$ and PM_1, while *Acer platanoides, Prunus avium* and *Tilia cordata* were less efficient collectors. Another comparative study of 11 deciduous tree species, using leaf SIRM as a proxy for particle capture[81], identified *Betula pendula* as the most efficient particle accumulator. Greater particle accumulation was observed for leaves with hairy and ridged surfaces, and aphid 'honeydew' contributing to leaf stickiness[81]. Compared to deciduous species, longer accumulation histories can be obtained from evergreen species, like pine needles or ivy leaves[84]. Although particle accumulation, and therefore magnetic properties, are species-specific, inter-calibration of leaf SIRM results between different co-located species has been successfully applied in urban environments[81,91]. Particles typically appear concentrated within hollows and along ridges in the leaf surface, nerves and stomata, probably due to fluid flow past the leaf[36,89]. Particles <10 μm in size, deposited on the leaf surface, can become encapsulated inside the leaf's epicuticular wax layer, preventing any wind or rain resuspension[84,149,151,160]. This encapsulated fraction was found to account for 33-38% of the leaf SIRM of London plane (*Platanus x acerifolia*)[90,161]. These magnetic results agree with gravimetric PM measurements[153], indicating 36-45% mass contribution of in-wax PM to the total deposited leaf PM, based on a three-year study on seven tree and six shrub species. Ultrasonic washing off of surface-deposited particles resulted in leaf susceptibility/SIRM decreases of 50-89% for *Pinus pumila* needle samples[162], 65-80% for Betula pendula (Matzka & Maher, 1999) and 30-50% for *Quercus ilex* leaf samples[163]. Wax layer thickness varies both in time and space, depending on species and abiotic stress factors like temperature, humidity, wind stress and gaseous air pollution[164]. Waxes are subject to ongoing degradation, potentially removing wax-incorporated particles, but are also periodically renewed by the plant. Nevertheless, no effect of the temperature-induced seasonal decline of surface wax concentration was found on the magnetic properties of *Pinus nigra* needles[62]. Continuous increases in SIRM, ARM and magnetic susceptibility were obtained for *Pinus nigra* needles over 4 years, while the wax amount reached an equilibrium after 26 months of exposure[84].

Leaf magnetic concentration is influenced by the exposure time[45,84,90], source distance[31,38,51,57,63,64,163], source strength (e.g. traffic volume)[57] and leaf sampling height[81,165].

Particle accumulation with leaf/needle exposure time is observed for both surface-deposited and wax-encapsulated particles; biomagnetic monitoring can thus act as a proxy for the time-integrated particulate pollution exposure. A 2- to 4-fold increase in SIRM, ARM and magnetic susceptibility of *Pinus nigra* needles was observed during 55 months at 6 sampling sites with varying ambient atmospheric pollution in Cologne, Germany[84]. Similarly, a 263 % higher leaf SIRM for unwashed *Platanus x acerifolia* leaves collected in September versus May, and a 380 % leaf SIRM increase for washed samples during the same sampling period in Antwerp, Belgium[90]. These findings are in line with another study, which obtained a 288% and 393% increase in leaf SIRM between May and September for (unwashed) *Carpinus betulus* and *Tilia platyphyllos*, respectively[91]. This seasonal accumulation favours leaf collection towards the end of the in-leaf season, as it will optimize magnetic differentiation between contrasting sites[91]. Nevertheless, controversy remains about the influence of removal processes of leaf-deposited particles, due to wind, rain or leaf wax degradation. According to the latter, leaf sampling should be conducted before leaf senescence sets in. Some studies found a considerable wash-off effect due to precipitation events, resulting in leaf SIRM decreases in the order of 5 to 64%[31,45,51,81,89,162], while others observed a negligible or nonexistent effect of rain on the leaf SIRM or susceptibility[38,62,90,91,163]. The magnitude of these removal processes is likely determined by weather conditions, and both leaf surface properties (e.g. micro-surface roughness, presence of trichomes, ridges and hydrophobicity) and PM properties (e.g. particle size distribution). Meteorological factors which influence the leaf-deposited dust load, and thus the resulting magnetic properties, include number and intensity of rainfall events, wind velocity and direction[89,91,96,165].

Although the particle trapping efficiency of several species has been investigated in several experiments[148–151,153,157], further work is needed to clarify which leaf anatomical-morphological (e.g. size, trichomes, surface roughness) and physiological (e.g. wax characteristics, wax encapsulation and regeneration) characteristics, and which PM properties, drive the accumulation and/or entrapment processes, and how this is influenced by meteorological conditions (e.g. rain, wind, drought) and seasonal dynamics (e.g. leaf senescence).

4.2.3 Applications

As tree leaves are common across many urban areas, and provide a good interface for particle deposition, biomagnetic leaf monitoring is well-suited for spatial explorative studies of atmospheric pollution. The magnetic variability observed between different sampling sites appears larger than that observed within sampling sites[84], individual tree crowns[165] and within a single leaf[166]. Single leaf-measurements can be, therefore, considered to be representative for their specific location.

Leaf magnetic parameters exhibit high spatial variation throughout cities[45,57,58], urban street canyons[165] and even individual tree crowns[51,165]. In urban environments, lowest magnetic concentrations are commonly reported in green areas; highest values near congested roads, industrial sites or railway traffic[31,38,51,57,58,101]. City-scale maps of leaf magnetic concentration have been obtained for e.g. Antwerp (Belgium)[58], Cologne (Germany)[62], Ghent (Belgium)[57], Kathmandu (Nepal)[96], Madrid (Spain), Rome (Italy)[45], Vigo (Spain)[101], Linfen (China)[114], and Isfahan (Iran)[112]. At the street scale, for two adjacent birch (*Betula pendula*) trees at a dual carriageway, a study[51] observed consistently higher leaf SIRMs results next to the uphill lanes, while the tree near the downhill lanes exhibited lower SIRM results, indicating the traffic exhaust-based origin of magnetic particles in this location. Temporal variation can be studied by combining soil magnetic measurements (recording longer-term PM accumulation history) with leaf samples (reflecting current PM levels), enabling the retrieval of pollution histories[113].

4.2.4 Biogenic vs anthropogenic sources

Without deposited PM, leaves exhibit a diamagnetic signal (i.e. low, negative magnetic susceptibility). Biological magnetite can be found associated with ferritin (also present in animals), an intracellular iron storage protein occurring in plants as plastids (e.g. chloroplasts in leaves, amyloplasts in tubers and seeds)[167,168]. Such magnetite typically occurs as micrometer-sized agglomerates of nanocrystalline grains[169,170]. To separate biogenic from anthropogenic contributions, various authors have calculated elemental or magnetic enrichment factors (EFs) for leaf samples[51,89,171,172].

4.3 Trunk wood and bark

In contrast to leafy material, woody biomass encompasses plant tissues exposed to atmospheric pollution year-round and for multiple years, although the exact duration of exposure is difficult to assess for some species. Using moist tissue wipes, branch and trunk bark was found to exhibit higher magnetisation (respectively, 50 and 200 times) compared to leaf samples of the same trees[119].

4.3.1 Influencing factors

Chemical and SEM/EDX studies have identified the superficial deposition of atmospheric particles and internal accumulation of heavy metals in bark, in association with land use class, traffic intensity, source type, direction and distance for *Fraxinus pennsylvanica, Fraxinus excelsior*, *Cupressus sempervirens*, *Pinus sylvestris, Populus nigra and Quercus ilex*[173–176].

Decreasing magnetite concentrations in *Acer rubrum* tree and co-located topsoil samples (upper 1 cm) were observed with increasing distance from a major highway between Washington and Baltimore (US)[177]. Apparently, atmospheric particles are not only intercepted and collected by tree bark, but enter the xylem during the growing season to become lignified into the tree ring[178]. Because little or no lateral redistribution of magnetic particles has been observed between adjacent tree rings, magnetic properties of tree ring cores could act as annual recordings of atmospheric pollution. Indeed, the authors[178] found a good correlation ($n = 19$, $r = 0.91$, $p = 0.01$) between the temporal variation of SIRM in *Salix matsudana* tree ring cores and annual iron production of an iron-smelting plant in Xinglong (China). Although root-absorption might be an alternative pathway for magnetic particle uptake, the reported iron oxides are found to be insoluble in soil-solutions[176]. Moreover, the SIRM directionality of tree ring cores towards atmospheric particle sources confirms that magnetic particles enter the tree trunk through encapsulation of bark-accumulated particles[178]. The adhesiveness of trunk bark may be influenced by moisture[177], in turn influenced by ambient airflows (e.g. traffic turbulence).

4.3.2 Bark vs trunk wood

Bark tissue displays magnetic values many times higher than wood tissue. Up to 28-fold higher SIRM results were obtained when comparing *Platanus x acerifolia* bark (188-2048 x10^{-6} A m^2 kg^{-1}, n=9) to its trunk wood (45-128 x 10^{-6} A m^2 kg^{-1}, n=9) at three sites with differing pollution levels in Antwerp, Belgium[179]. For the same species, another study[180] demonstrated that SIRM of entire branch internodes was mainly confined to the bark tissue (by 78-93%). The branch internode SIRM of *Platanus x acerifolia*, normalised by the branch area, ranged from 18 to 650 x 10^{-6} A and increased with each year of exposure, even after 5 years. A study[181] however states that superficial particle loading on bark cannot represent a full several-year-accumulation of atmospheric contaminants and suggests that meteorological conditions such as rain play an important role.

Both weight-normalised SIRM (0.43 to 298 x 10^{-5} A m^2 kg^{-1}) and susceptibility (-3.5 to -2.5) are ~2 orders of magnitude lower for bark than the results obtained from leaf, moss and lichen samples. Nevertheless, when normalising for the projected surface area[180], a similar range (18-650 x 10^{-6} A) and 2 x higher results were obtained compared to neighbouring and simultaneously exposed leaf samples. Although absolute values can differ, similar spatial variation in SIRM is observed between tree bark and trunk samples and co-located soil[177] and leaf[180] samples. Moreover, correlations were obtained for trace element concentrations between bark tissue and lichens[182,183].

4.4 Insects

Since 1962, bees (*Hymenoptera, Apoidea*) have been increasingly employed for monitoring of e.g. heavy metals in territorial and urban surveys, pesticides in rural areas and radionuclides[184–187]. However, biogenic magnetite has been reported in the abdomen of bees[188], as well as the thorax of butterflies[189,190], abdomen and thorax of termites[191] and cockroaches[192]. A study[190] tested five migratory (moths and butterflies) and four non-migratory (crickets) insect species and found evidence for biogenic magnetism in only one migrant, the monarch butterfly (*Danaus plexippus*). Biogenic magnetic particles are thought to be used for navigation purposes, or so-called magnetoreception – the ability to perceive the Earth's magnetic field[190,193].

Although an atmospheric pathway for exogenous magnetic minerals (through plant and pollen) is suggested[194] and remanent magnetisation is measurable in insects, no evidence yet exists that insect magnetism can be applied as a proxy for atmospheric pollution. Another research gap concerns potential uptake of atmospheric particles through insect food intake or inhalation (through spiracles in cuticle and underlying tracheal system).

Reported SIRMs of insect tissues (Appendix 2) range from 0.09 – 13.98 A m^2 (volume-normalised) or 46 – 320 x 10^{-6} A m^2 kg^{-1} (mass-normalised). These values are much lower than plant accumulation surfaces; unsurprising as the particle uptake pathway (through plant and pollen) is indirect and less efficient.

4.5 Crustaceans: Isopods

Isopods are considered good bioindicators of metal contamination in the terrestrial environment due to their widespread occurrence in Europe (both in rural and urban areas), their size, conspicuousness, easy collection and high tolerance to heavy metals[195–198]. Analysis of bioavailable metals (Cd, Cr, Cu, Fe, Pb and Zn) from different isopod species (*Oniscus asellus* and *Porcellio scaber*), collected at urban and rural locations in Renfrewshire, UK, showed varying concentrations of natural and anthropogenic metal concentrations, in the order Cu > Cd > Pb > Cr > Zn > Fe for *Oniscus asellus* and Cu > Zn > Cd > Cr > Fe for *Porcellio scaber*[197]. Seasonal fluctuations in isopod metal bioaccumulation are observed[195], ascribed to temperature fluctuations. An isopod study[198] quantified Cd, Cr, Cu and Ni levels in cultivated *Porcellio scaber* and *Porcellio dilatatus* and suggested moulting as a way of detoxification for Cr and Ni (but not for Cd and Cu). Detoxification by excretion of accumulated Cd and Pb has been reported as well[199]. Use of isopod samples as biomonitors for atmospheric pollution requires understanding of these detoxification pathways, which will weaken any association between sample content and atmospheric pollution.

Two exploratory studies (Appendix 2) on biomagnetic monitoring of isopods report mass-normalised SIRMs ranging from 19 x 10^{-6} to 28 390 x 10^{-6} A m^2 kg^{-1} [200,201]; higher than the reported bee SIRM results. A study[200] collecting 5315 isopods, belonging to *Porcellio scaber* (1804), *Oniscus asellus* (1758), *Trachelipus rathkki* (1833) and *Philoscia muscorum* (1763) species, at 33 locations situated at varying wind directions and distances from a metallurgical plant in Antwerp, Belgium, observed a decrease in mass-normalized isopod SIRM with increasing distance from the plant and significant directional effects. Another study[201] collected two isopod species (*Porcellio scaber* and *Oniscus asellus*) and soil samples at 17 locations along an urbanization gradient in Antwerp, Belgium. Combining biomagnetic with elemental analysis (ICP-MS), the authors found a higher accumulation capacity of *Oniscus asellus*, significant variation between the sampled locations (depending on traffic volume, green areas and railway traffic) and significant associations between SIRM and Al, Ti, V, Mn, Fe, Ni, Ga, As, Sb, Bi and U[201]. Both studies report significantly higher SIRM results for *Oniscus asellus* (higher accumulation capacity) compared to co-located *Porcellio scaber*.

The magnetic content of isopods is thus species-specific, exhibits spatial variation along urbanisation gradients and shows associations with trace elemental content. Nevertheless, as with insects, questions remain regarding both detoxification and potential uptake pathways of atmospheric particles through food intake or inhalation.

4.6 Mammal tissues

An exploratory study using mammal tissues[202] reported SIRMs (at 77 K) for lung tissue obtained from four deceased mammals (three cats and a dog) near Munich, Germany. SIRMs ranged from 2 - 44 x 10^{-6} A m^2 kg^{-1}, attributed to <100 nm, magnetite-like minerals at ~100 ppb concentrations. A difference was observed between the rural (~2.9 × 10^{-6} A m^2 kg^{-1}) and urban (~4.4 and 4.9 × 10^{-6} A m^2 kg^{-1}) SIRMs in cats, but possibly reflecting a shorter exposure period for the younger rural cat.

Although based on only four individuals, these results demonstrate that biomagnetic monitoring can obtain information about PM in mammal lung tissue. As with the insects and isopods, atmospheric pollution dose might be obscured through non-stationarity of the animal, detoxification (lung clearance) or other metabolic pathways.

4.7 Human tissues

Biogenic magnetite has been reported inside human brain tissues[68,203,204] and the heart, liver and spleen[205]. Identification of magnetite was achieved through histological preparations, transmission electron microscopy, magnetic resonance and SQUID magnetometry[73].

4.7.1 Range of reported magnetic results and applications

SIRM and susceptibility values from human tissues (Appendix 2) range from 1.1 to 170 x 10^{-6} A m^2 kg^{-1} (mostly obtained at 77 K) and 0.2 to 5.2 x 10^{-8} m^3 kg^{-1}, respectively. Low temperature remanence is frequently measured in order to capture the SP magnetic component.

In terms of pollution exposure, most research has focused on exogenous pneumotoxic constituents, particularly trace metals[206] and magnetic particles, inhaled in lung tissues. The ferromagnetic remanence of in vivo and *post mortem* lung tissues can be measured externally by magnetometers, as an indicator of the inhaled dust load. Such magnetopneumography (MPG) identifies influences of exposure to welding, asbestos and coal mining, steel industry and smoking habits on the lung magnetic remanence[207–212]. Lung magnetite concentrations between 10 and 800 µg g^{-1} have been reported in 20 ashed post-mortem lung samples from asbestos miners[208], substantially higher than the concentrations reported for heart, spleen and liver tissues[205]. In vivo particle migration and lung clearance were also investigated[212]. An investigation on lung clearance[211] compared lung clearance in smokers and nonsmokers, through magnetite dust inhalation experiments. After 11 months, smokers still retained 50% of the inhaled magnetite, while non-smokers retained 10%.

5.7.2 Associated health effects

Recently, IRM and susceptibility measurements on different human *post mortem* brain, liver, spleen, pancreas, heart and lung tissues[213] showed highest susceptibility values, while lowest values were obtained for the pancreas. These results are in line with a previous study[205], reporting highest magnetite concentrations (SIRM, at 77K) for human heart tissue samples (13-343 ng g^{-1}; 5-16 x 10^{-6} A m^2 kg^{-1}), compared to spleen (14-308 ng g^{-1}; 0.6-14 x 10^{-6} A m^2 kg^{-1}) or liver (34-158 ng g^{-1}; 1.5-7.3 x 10^{-6} A m^2 kg^{-1}) samples. Higher SIRM and susceptibility results are typically obtained for lungs of smokers or certain professions (e.g. car painters), confirming the presence of exogeneous magnetic particles. While susceptibility can be influenced by the amount of blood and water (para-/diamagnetic behaviour), magnetic remanence (IRM) will only quantify magnetite- or hematite-like minerals.

Besides their presence in human lung tissues, exogenous magnetite nanoparticles have recently been identified in human brain tissues[78]. Magnetite can have potentially large impacts on the brain due to its unique combination of redox activity, surface charge and strongly magnetic behaviour. Previous work has shown a correlation between the amount of brain magnetite (up to ~ 7 μg g^{-1}) and the incidence of Alzheimer's disease (AD), albeit for small sample sizes[76,79]. Magnetite nanoparticles, ascribed to biogenic formation, have been found directly associated with AD plaques[214]. However, new evidence identifies the presence of magnetite nanoparticles in the human brain consistent with an external, not internal, source. Magnetometry, high-resolution transmission electron microscopy (HRTEM), electron energy loss spectroscopy (EELS) and energy dispersive x ray analysis (EDX) were used to examine the mineralogy, morphology, and composition of magnetic nanoparticles in and from the frontal cortex of 37 human brain samples, from subjects who lived in Mexico City and in Manchester, U.K. These analyses identified the abundant presence (up to ~10 μg g^{-1}) of magnetite nanoparticles that are consistent with high-temperature formation, suggesting therefore an external, not internal, source. This brain magnetite, often found with other transition metal nanoparticles, display a range of sizes (~ 10 – 150 nm), and rounded morphologies, some with fused surface textures, likely reflecting condensation from an initially heated, iron-bearing source material. Such high-temperature magnetite 'nanospheres' are ubiquitous and abundant in airborne PM. Because of their combination of ultrafine size, specific brain toxicity, and ubiquity within airborne PM, pollution-derived magnetite nanoparticles might be a possible AD risk factor. In addition to occupational settings (including, for example, exposure to printer toner powders), higher concentrations of magnetite pollution nanoparticles may arise in the indoor environment from open fires or poorly-sealed stoves used for cooking and/or heating, and in the outdoor environment from vehicle (especially diesel) and/or industrial PM sources. Epidemiological studies have identified associations between exposure to vehicle-derived PM and cognitive decline[215], and between residence in proximity to major roads and the incidence of dementia[216]. The latter study, based on a large population-cohort in Ontario, Canada, estimates that between 7 and 11% of dementia cases in patients who live < 50 m from heavily-trafficked roads were attributable to traffic exposure. Further work is needed in order to examine if there are causal links between vehicle-derived magnetite nanoparticles and the widespread incidence of later-age neurological damage

5. Challenges and future perspectives

Although, since 1973, a variety of environmental magnetic studies has been reported, the application of biomagnetic monitoring for atmospheric pollution assessment has only been explored during recent decades. This review, based on 83 biomagnetic studies and 230+ references, demonstrates the potential of this approach for fast qualitative or semi-quantitative atmospheric pollution monitoring. Table 1 presents a summary table on currently available biological sensors, encompassing uptake pathways, influencing factors, advantages, limitations, applications and major challenges, to assist researchers and policy makers in selecting the most suitable biological material for their specific monitoring application. As various and complex influencing factors need to be considered when setting up biomagnetic monitoring campaigns, more elaboration is provided within the following paragraphs.

5.1 Experimental design

So far, most biomagnetic research has focused on plant leaves (46 of 84 studies). As these biological accumulation surfaces are stationary and often cumulative, they are used in spatiotemporal campaigns in environments with large atmospheric pollution gradients (e.g. urban areas; near industrial sites). Depending on the envisaged monitoring period, deciduous leaves (in-leaf season), evergreen needles (year-round) or bark (year-round or multiple years) can be sampled. Leaves and bark are frequently available across urban environments (allowing both active and passive biomonitoring), in contrast to mosses/ lichens which require active installation.

Besides the stationary sensors, mobile biological sensors can be distinguished as well; small-radius (insects and crustaceans) and large-radius (mammals, including humans) sensors. Small-radius sensors can still be applied for spatial monitoring of pollution gradients, investigating possible relations with pollination or evaluate the persistence of contaminants within ecosystems or food chain. Nevertheless, limited data are currently available (only on isopods and bees) and questions remain about metabolic pathways of atmospheric pollution (e.g. food intake, inhalation, internal transport and detoxification through excretion or moulting). Compared to stationary biological sensors, small-radius sensors show much lower magnetic concentrations, with less resulting magnetic sensitivity to pollution gradients. Nevertheless, reported associations between isopod biomagnetic properties and urbanization gradients or trace elemental content, make it an interesting area for future research.

Finally, large-radius sensors generally exhibit lowest magnetic concentrations (and therefore, lowest sensitivity) as atmospheric pollutants need to be inhaled and transported through the body. On the one hand, this allows for personalized air pollution monitoring, quantifying the exhibited pollution exposure, having important considerations for human health studies. This is similar to traditional atmospheric pollution monitoring which is not restricted to fixed-site monitoring, but evolves into portable or mobile instrumentation as well [21,e.g. 217–222], enabling quantificatuon of personal air pollution exposure. On the other hand, internal body transport, detoxification pathways (e.g. lung clearance) and metabolism (between and within individuals and individual organs) will need additional consideration when interpreting the magnetic results. Size selection of atmospheric particles will, for example, occur during inhalation (<10 μm), deposition in the alveoli (<2.5 μm) and uptake in the bloodstream (<0.1 μm), while leaf-deposited magnetic particle sizes are reported up to 50 μm (SI 2)). Tracking of research subjects will be required to obtain information on their pollution exposure routes, while ethical issues might hinder some types of experimental design.

Table 1. Summary of considerations (e.g. sensitivity, influencing factors, limitations) on the use of current available biological sensors for biomagnetic monitoring of atmospheric pollution. The sensitivity of the considered sensors was judged quantitatively, based on the reported SIRM and susceptibility ranges. See text for additional elaboration.

Sensor / Considerations	Mosses and lichens	Plant leaves	Bark and wood	Insects	Crustaceans	Mammal tissue	Human tissues
Monitoring technique	Mostly active	Passive/active	Mostly passive	Mostly passive	Mostly passive	Mostly passive	Mostly passive
Uptake pathway	Deposition, impaction, interception	Deposition, impaction, interception Root uptake negligible?	Deposition, impaction, interception Root uptake negligible?	Food intake? Inhalation?	Food intake? Inhalation?	Inhalation Internal transport	Inhalation Internal transport
Sensitivity	++++	+++	+++	++	++	+	+
Accumulation period	Period of exposure	Period of exposure (min: 6 days, max: in-leaf season)	Period of exposure	Lifetime	Lifetime	Lifetime	Lifetime
Influencing factors	Exposure time Environmental conditions Species characteristics Moss bags/transplants	Exposure time Environmental conditions Plant species Leaf-surface properties Sampling height Leaf morphology Cuticular wax encapuslation	Exposure time Environmental conditions Tree characteristics Bark characteristics	Exposure time Way of feeding Metabolism	Exposure time Way of feeding Metabolism	Exposure time Life/work habits Metabolism Tissue selection	Exposure time Life/work habits Metabolism Tissue selection
Advantages	Stationary Absence of cuticle No rooting system High surface to volume ratio	Stationary High availability High surface to volume ratio Standardized protocol	Stationary High availability Root-adsorption negligible Surface accumulation	High availability	High availability	Personal monitoring Link with exposure	High availability Personal monitoring Link with exposure

	Standardized protocol Surface accumulation	Surface accumulation	Multiannual accumulation				
Limitations	Not omnipresent in urban areas Resuspension?	Wash off? Resuspension?	Wash off? Resuspension?	Mobility	Mobility Detoxification pathways?	Mobility Ethics Tissue selection	Mobility Ethics Tissue selection
Application	Spatiotemporal campaigns	Spatiotemporal campaigns	Spatiotemporal studies Long-term studies (multiannual)	Spatial campaigns Relation with pollination?	Spatial campaigns	Personal monitoring Exposure	Human health Personal monitoring Exposure
Challenges	Transplant techniques	Spatial distribution Active: maintenance, vandalism	Spatial distribution	Metabolism	Metabolism	Ethics Mobility Metabolism Activities	Ethics Mobility Metabolism Activities

5.2 Sampling strategy

Sampling strategies must always consider how atmospheric pollutants accumulate in biological sensors. All biomagnetic results covered here have shown species-specific accumulation capacities, reflecting PM collection through differing sets of morphological and/or physiological properties. Monitoring campaigns should thus use a single monitoring species or seek inter-calibration between multiple monitoring species. Based on this review, we can recommend efficient accumulator species as biological sensors, e.g. *Sphagnum palustre* when aiming for moss biomagnetic monitoring or e.g. *Betula pendula* or evergreen species (e.g. *Hedera sp*.) for leaf biomagnetic monitoring[81,148]. However, the species selection will depend on the envisaged research objective; e.g. winter campaigns will require evergreen species; short-term campaigns (e.g. 1 month) demand for high accumulators (e.g. hairy leaf species) in order to obtain quantifiable magnetic signals; and spatial monitoring campaigns will require a widespread occurrence (e.g. *Platanus acerifolia*).

Biological sensors can record exposure periods from ~ 6 days (leaves) to an in-leaf season (leaves) or multiple years (bark) and up to individual lifetimes (mammal and human tissues). By combining leaf, bark, wood and soil samples, a pollution history can be retrieved (current vs historical). For surface-accumulating sensors (e.g. mosses, lichens, leaves and bark), samples can be obtained from existing species (passive biomonitoring) or actively-introduced monitor species (active biomonitoring). Active biomonitoring guarantees similar exposure periods, provides for spatially-ordered sampling and allows for better standardization of the applied biomonitoring materials (similar background conditions before pollution exposure), ultimately leading to more reliable data. Active biomonitoring can further reduce biological variations by working with clonal material.

For magnetically weak samples (e.g. leaves, human/insect tissues), where magnetic susceptibility is below the detection limit of existing instrumentation, concentration-dependent magnetic information can be obtained from SIRM, at room or low temperature. At low temperatures (often 77 K), magnetic particles small enough to be superparamagnetic at room temperature block in, and contribute to higher induced magnetization values.

5.3 Associations with atmospheric pollutant species

A challenge in biomagnetic monitoring arises from the determination of the association between concentration-dependent magnetic properties (χ, SIRM, ARM) and ambient PM or gaseous pollutant concentrations. Reported associations may not be generalized but are often specific for each considered environment or contributing sources. This can be observed when looking at the differences in associated elements from the table in SI 2. Due to a spatiotemporal variation and source-specific physicochemical composition of atmospheric dusts, and the fact that magnetic particles only make up part of the dust emissions, the magnetic response will vary accordingly. This implies that spatial maps of magnetic concentration parameters are only reliable in environments with similar (or at least comparable) source contributions. Within such "single source" environments (e.g. highway transects, street canyon studies), quantification of magnetic concentration parameters will be sufficient to obtain an idea about the bulk particle/elemental deposition. When considering larger monitoring scales (e.g. urban/regional mapping), inclusion of multiple sources with heterogeneous chemical and magnetic particle characteristics will complicate the associations with atmospheric pollutants, which increases the need for an extended magnetic characterisation (e.g. using different magnetic parameters, ratios or coercivity spectra to obtain information on the magnetic mineralogy, domain state and grain size).

Combining analytical techniques (e.g. SEM/EDX, EELS, ICP-MS, X-ray diffraction, Mössbauer spectroscopy) with magnetic parameters can provide valuable supplementary information on PM composition and contributing sources[111,171,223]. Magnetic differentiation between industrial and traffic PM sources, based on the magnetite:hematite ratio, has already proven feasible[40,63]. Interesting work was also performed by magnetically and chemically analyzing filter-collected PM_{10} at different monitoring sites in Switzerland[224], calculating two magnetic components from the magnetic coercivity distributions using skewed generalized Gaussian (SGG) functions developed earlier[225]. Based on these magnetic components, together with elemental information, anthropogenic and natural PM_{10} contributions could be identified. The magnetic contribution of the anthropogenic component was shown to be proportional to the chemically-estimated PM_{10} mass contribution of traffic exhaust emissions, while the other component was attributed to a mix of natural dust and resuspended anthropogenic street dust. Moreover, the anthropogenic magnetic components were significantly associated with traffic-related elements; Ba, Cu, Mo, Br and elemental carbon[224].

We encourage further development of magnetic fingerprints from different atmospheric pollution sources. Such source-specific magnetic information will be essential for the holistic interpretation of biomagnetic results, it will increase the magnetic power for source attribution in mixed-source environments and for measuring impacts of PM mitigation policies.

6. Outlook

Biomagnetic monitoring provides substantial worldwide potential to address the growing need for cost-effective methodologies to capture high spatial resolution variation and compositional changes of atmospheric pollution across urban environments. It comprises a rapid, cost-effective and non-destructive tool, providing qualitative or semi-quantitative information on magnetic concentration, mineralogy, domain state and grain size of airborne PM. In most cases, biomagnetic monitoring should not be regarded as a stand-alone methodology, but might serves as a valuable addition to existing monitoring networks, analytical techniques or modelling frameworks. So far magnetic techniques have been applied to: spatial mapping of atmospheric pollution; validation of air quality models; tracing of historical vs current pollutant levels (e.g. soil vs leaf samples); mapping of emission plumes from point sources; and personal (exposure) monitoring. Magnetic properties often display strong linkages with PM, NO_x, PAHs and heavy metals, and can thus act as an effective proxy. Source-related chemical and magnetic heterogeneity can be regarded as the major challenge of biomagnetic monitoring and should be targeted in further research. Additional direct significance may be attributed to magnetic PM if exogenous magnetite nanoparticles, present in human brain tissue, are causally linked with neurodegenerative diseases.

Acknowledgements

The corresponding author (JH) acknowledges the Research Foundation Flanders (FWO) for his postdoctoral fellowship (12I4816N). AC receives a FWO doctoral fellowship grant (SB, 1S15122716N).

Supporting Information

A theoretical background on environmental magnetism and an inventory table of reported magnetic studies on pumped-air filters and biological sensors is available free of charge on the ACS Publications website.

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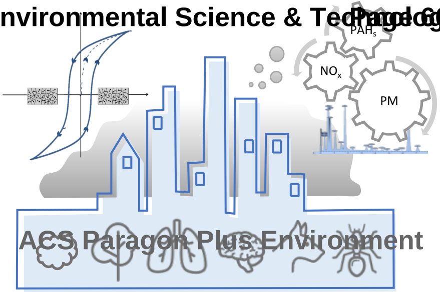
PAHs
NOx
PM