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Airborne, Vehicle-Derived Fe-Bearing Nanoparticles in the Urban Environment: A Review

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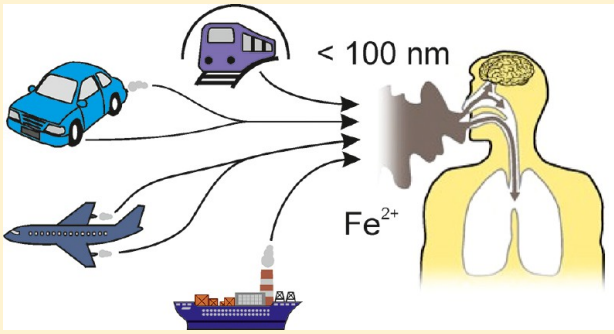
Supporting Information

ABSTRACT: Airborne particulate matter poses a serious threat to human health. Exposure to nanosized (<0.1 μm), vehicle-derived particulates may be hazardous due to their bioreactivity, their ability to penetrate every organ, including the brain, and their abundance in the urban atmosphere. Fe-bearing nanoparticles (<0.1 μm) in urban environments may be especially important because of their pathogenicity and possible association with neurodegenerative diseases, such as Alzheimer's and Parkinson's diseases. This review examines current knowledge regarding the sources of vehicle-derived Fe-bearing nanoparticles, their chemical and mineralogical compositions, grain size distribution and potential hazard to human health. We focus on data reported for the following sources of Fe-bearing nanoparticles: exhaust emissions (both diesel and gasoline), brake wear, tire and road surface wear, resuspension of roadside dust, underground, train and tram emissions, and aircraft and shipping emissions. We identify limitations and gaps in existing knowledge as well as future challenges and perspectives for studies of airborne Fe-bearing nanoparticles.



1. INTRODUCTION

Exposure to airborne particulate matter (PM) is known to pose a serious threat to human health, linked with damage not only to respiratory and cardiovascular health, but also to neurodevelopment and cognitive function. Metal-bearing, nanosized (<0.1 μm) particles may be especially hazardous to human health[1−7] because of their abundance in the urban atmosphere,[8−13] bioreactivity,[10,14−16] and their ability to reach all major organs of the human body by inhalation[10,17−21] and by circulation in the bloodstream.[22]

As evident from Figure 1, metal-bearing (e.g., Au-, Ba-, Cd-, Fe-, Mn-, Ti-bearing) particles may be translocated from the lungs to sites of vascular inflammation,[22] the central nervous system,[23−25] liver[22,26] and amniotic fluid.[27] Particles <200 nm can access the brain directly, bypassing the blood−brain barrier, via transport through the neuronal axons of the olfactory and/or trigeminal nerves.[10,24] Exposure to particulate air pollution may thus cause damage to neurodevelopment and cognitive functions.[10,28−33] Large population-scale epidemiological studies show that living close to heavy traffic is associated with a higher incidence of dementia.[33−36] Other epidemiological studies in the USA and Mexico indicate that young and old subjects living in areas with high airborne PM concentrations have cognitive deficits.[37−40] In a magnetic resonance imaging (MRI) analysis of elderly women (age from 71 to 89 years; the Women's Health Initiative Memory Study cohort), white matter loss was increased by 1% per 3 $\mu g/m^3$ of $PM_{2.5}$.[41] Post-mortem brain samples from clinically healthy subjects (and dogs) exposed to lifetime high pollution, while living in Mexico City[10,28,30] or Manchester, UK[10] display: (i) typical hallmarks of Alzheimer's disease (AD) pathogenesis (e.g., tau-P, β-amyloid deposition), (ii) inflammation, oxidative stress and DNA signaling damage;[30] and (iii) the abundant presence of metal-bearing NPs, including mixed Fe^{2+}/Fe^{3+} (magnetite) NPs, as determined by electron microscopy (EM) and magnetic analyses.[10] Mouse models for urban nanoparticulate air pollution show consistent induction of inflammatory responses in major brain regions[42] (see also Table SI 3 (Supporting Information)).

Due to their large surface area and potentially high reactivity with biomolecules and tissues, metal-bearing nanoparticles (NPs) may generate adverse health impacts related to oxidative stress, inflammation,[18,19,43−47] and generation of reactive oxygen species (ROS).[3,48−50] Exposure to bioreactive airborne NPs can occur at every life stage; in the womb,[27,51] as developing infants and children,[37,52−56] and through adulthood to terminal decline. Both the young and the elderly are particularly vulnerable to air pollution.[57−62] Prebirth exposures to both PM_{10} and $PM_{2.5}$ are associated with low birth weight (e.g., refs 62−65). Childhood exposures to traffic-derived PM (elemental carbon − EC, nitrogen dioxide − NO_2 and PM <

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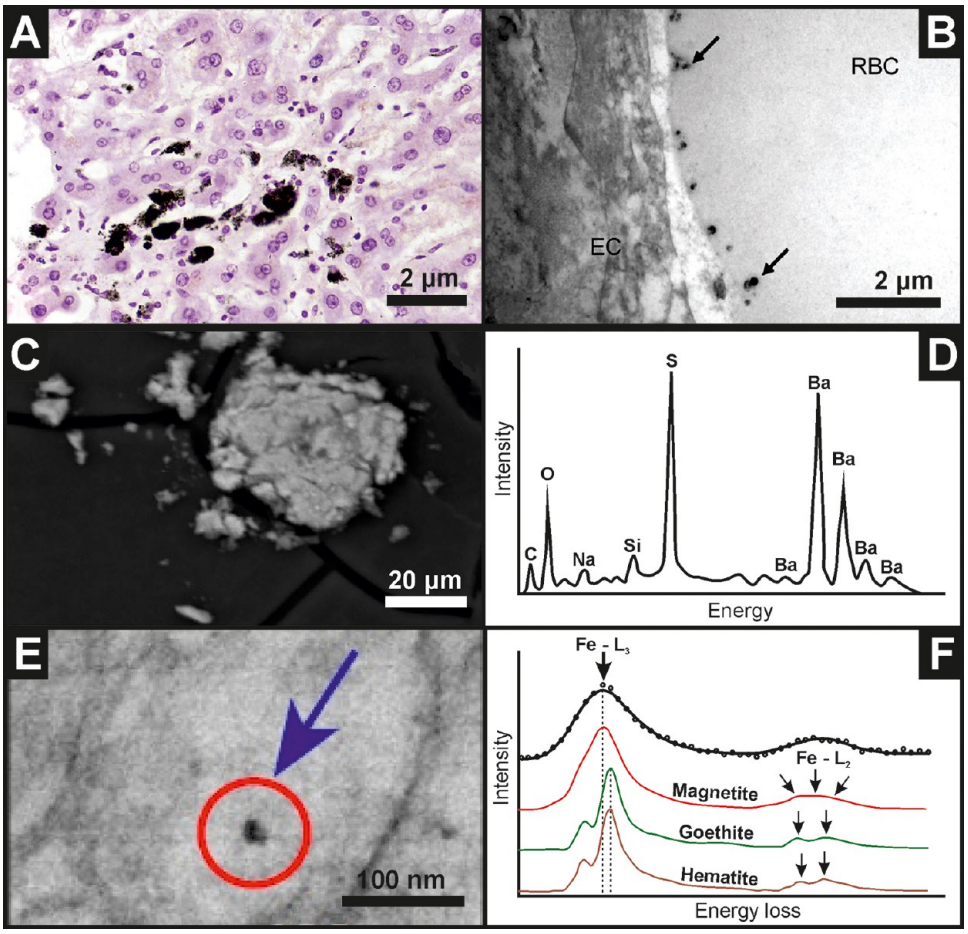

Figure 1. Metal-bearing particles found in the human body: (A) PM in the liver of a 32 year-old male (adapted from from Calderón-Garcidueñas et al.[30]); (B) PM spherules in a lung capillary, RBC = red blood cell; EC = endothelial cell (adapted from Calderón-Garcidueñas et al.[30]); (C) SEM image of Ba-bearing particles in the amniotic fluid with (D) the corresponding EDS spectrum (adapted from Barošová et al.;[27] (E) TEM image of magnetite NPs in frontal cortex brain tissue with (F) corresponding electron energy loss spectroscopy spectra (in black) for the rounded particle shown in E and for standard Fe oxide species (adapted from Maher et al.[10]).

700 nm) impair lung growth[66] and cognitive development.[55,56] These early life exposures to airborne PM can thus set in train a sequence of health problems which progressively develop through later life. Children spend more time outdoors, are more physically active, and they have an increased breathing rate and immature immune system, all of which result in a higher dosage in the respiratory tract of children compared to adults, especially in the case of NPs.[67] Compared to children living with clean air, those in polluted areas (e.g., Mexico City) display health problems, including systemic inflammation, and even Alzheimer's and Parkinson's hallmarks.[30,37,38,52]

Exposure specifically to Fe-bearing airborne NPs may be especially hazardous to human health. Fe is an essential biometal, as a component of diverse metalloproteins, from cytochromes to hemoglobin, and playing key roles in neuronal function, ranging from nerve impulse transduction to neurotransmitter synthesis and mitochondrial energy production. Fe is mobilized and stored via changes in valence, controlled by sophisticated regulatory homeostasis processes. Such processes are vital, since labile Fe^{2+}, i.e., in the unbound, redox-active state, can be toxic to living cells. In humans, nonhaem Fe is stored in a redox-inactive, oxidized (Fe^{3+}) form as 8 nm spherical cores of ferrihydrite ($5Fe_2O_3{\cdot}9H_2O$) within the 12 nm-diameter Fe-storage protein, ferritin. However, if redox-active, ferrous (Fe^{2+}) remains unbound and freely available, it can catalyze the formation of ROS, including the damaging hydroxyl radical, through the Fenton reaction.[68]

Problems with transportation and storage of Fe have been linked with a variety of diseases, including Alzheimer's, Parkinson's, Huntington's, multiple sclerosis, amyotrophic lateral sclerosis and Hallervorden−Spatz disease.[68−74] Some of the excess Fe found in the neurodegenerative brain occurs in the form of magnetite, a strongly magnetic, mixed Fe^{2+}/Fe^{3+} oxide (see Section 12 for more details).

Given the potential risks to human health posed by airborne Fe-bearing NPs, it is timely to examine information regarding their sources, abundance and composition, in order to identify major routes of exposure, and potential pathways to removing or reducing their particle number concentrations.

2. IRON-BEARING NANOPARTICLES IN THE ENVIRONMENT

Fe-bearing NPs occur both abundantly and widely in the urban atmosphere.[8−13] Fe, in oxide, hydroxide and metallic forms, is a major contributor not only to the aerosol mass in urban environments but also, because of its abundant occurrence within primary, emitted NPs, to the particle number concentrations in urban air.

Fe has been found in high concentrations (11,000−94,000 ppm) in the submicrometer fraction of roadside PM.[13] Fe-

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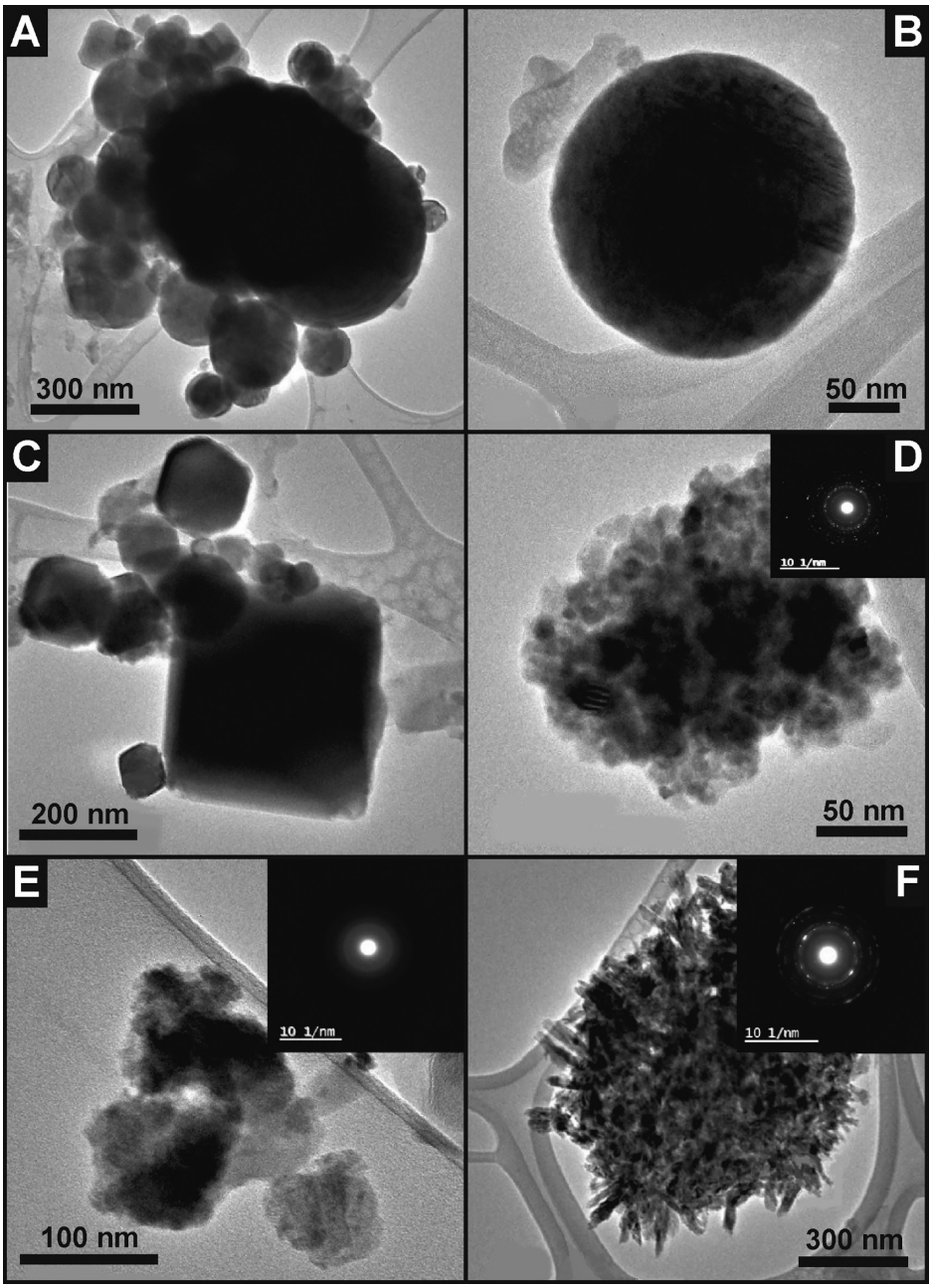

Figure 2. Fe-bearing NPs found in street dust: (A) aggregate of spherical magnetite particles, (B) Fe-rich fly ash, (C) magnetite particle with an octahedral shape, (D) hematite, (E) ferrihydrite, (F) goethite (adapted from Yang et al.[13]).

bearing NPs are frequently associated both with transition and heavy metals, including Cr, Ni, Cu, Zn, Pb, Mn.[10,13,75] Most of the Fe-bearing particles emitted from vehicles are ferromagnetic (*sensu lato*) Fe oxides, comprising mixtures of strongly magnetic magnetite (Fe_3O_4), its oxidized counterpart, maghemite (γ-Fe_2O_3) and hematite (α-Fe_2O_3), with some metallic Fe (α-Fe) also reported.[12,76−80] The magnetic ordering shown by these airborne Fe-oxide particles enables their measurement and quantification using a range of concentration- and particle size-dependent magnetic analyses (see Section 11).

Magnetic particles in indoor and outdoor airborne PM can originate from a range of domestic (e.g., wood/coal burning, cigarette smoking, printer emissions), workplace, transport and industry sources (see also SI 2, Supporting Information).[78,79,81−87] Such particles can occur over a great range of sizes, from several nm up to several hundred μm. Moreover, NPs <100 nm can agglomerate with each other and/or surround bigger particles. However, until recently, most magnetic studies have focused on PM fractions ≥1 μm.

Vehicles are a major source of Fe-bearing NPs in the environment.[12,13,80,86,88−96] Figure 2 shows Fe-bearing NPs in street dust, variously composed of magnetite, hematite, goethite and ferrihydrite.[13] Vehicle-derived, Fe-bearing NPs originate from exhaust emissions, both diesel[97,98] and gasoline,[86] brake wear,[96] tire wear,[99] resuspension of roadside dust,[8] underground, rail and tram systems,[94] aircraft[100] and shipping emissions.[91]

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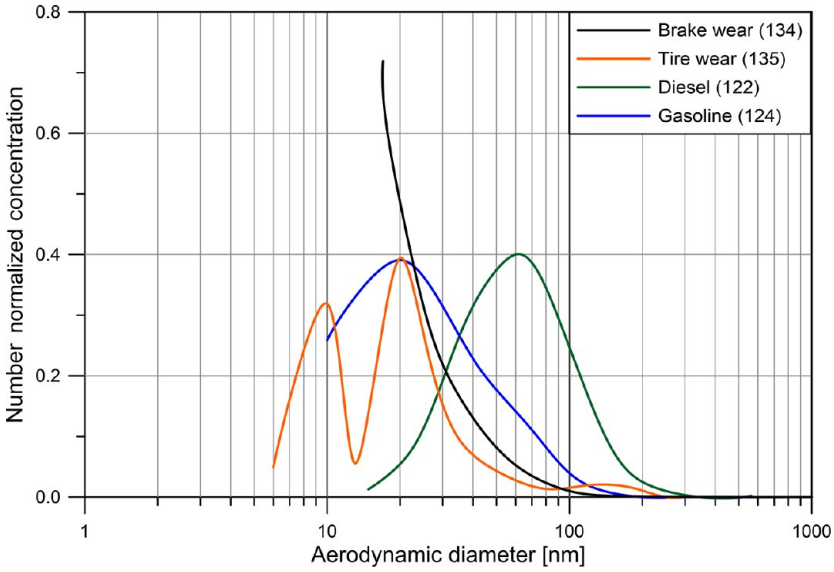

Figure 3. Examples of particle-size distributions for nonexhaust (brake and tire wear debris) and exhaust (diesel and gasoline) emissions. Brake wear was collected using a pin-on-disc machine (sliding speed of 13 m/s, contact pressure of 1.39 MPa, for 3 h).[134] Tire wear was collected using a tire test bench (inner diameter of 3.8 m, speed of 0, 30, 50 and 70 km/h).[135] Diesel emissions were collected using an AC electric dynamometer and the U.S. UDDS (urban dynamometer drive schedule) test cycle.[122] Gasoline emissions were collected using a test vehicle (acceleration from 30 to 90 km/h; hot start).[124]

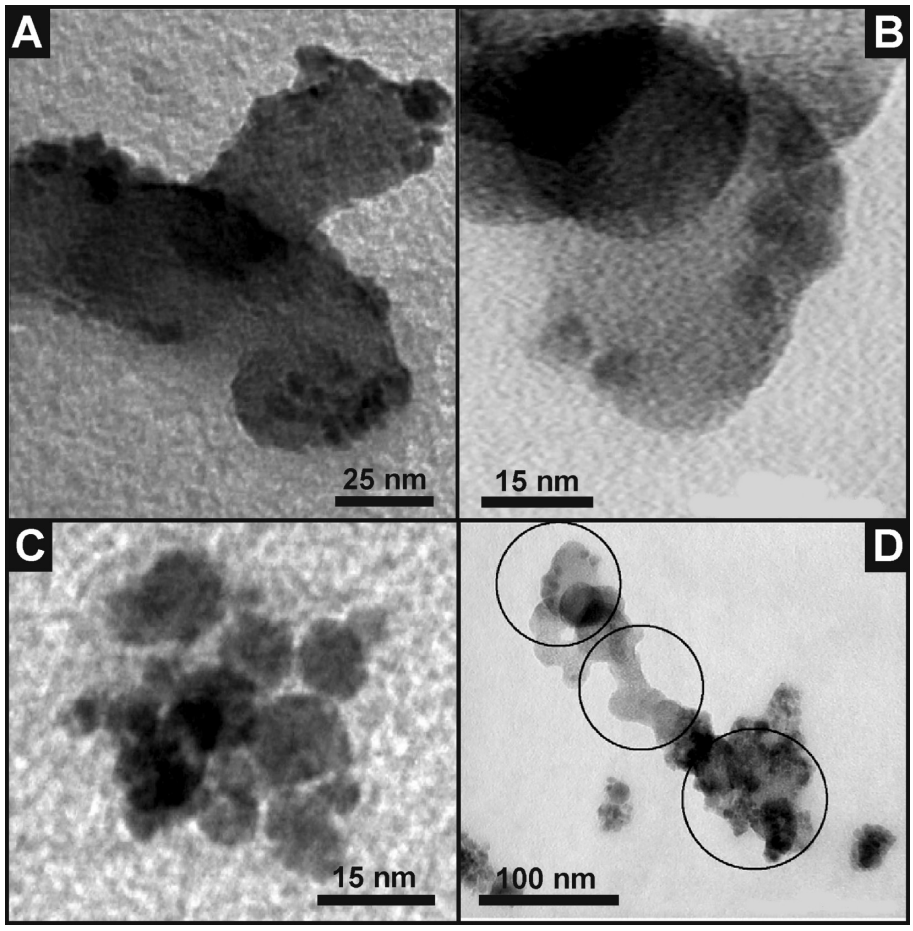

Figure 4. Examples of Fe-bearing NPs in diesel emissions: (A and B) carbon agglomerates decorated with primary Fe NPs, (C) nucleated primary particles of Fe and (D) combined carbon/iron agglomerates (adapted from Miller et al.[139]).

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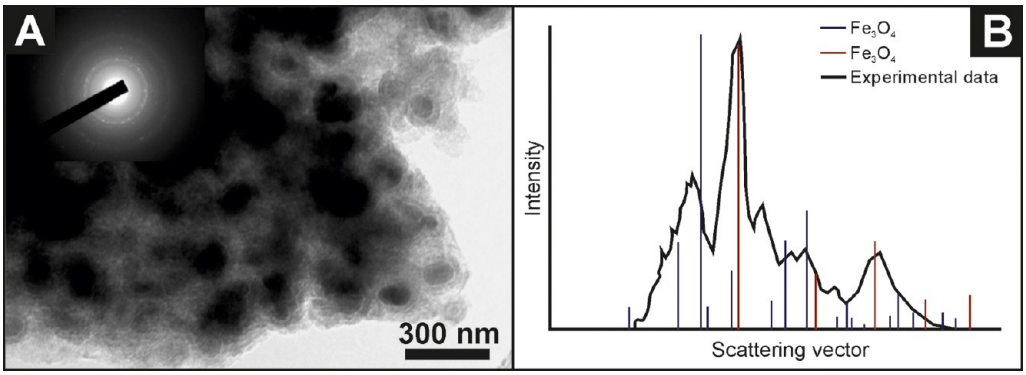

Figure 5. (A) TEM image of NP wear particles collected during a dynamometer test and (B) electron diffraction (EDX) intensity profile (adapted from Kukutschová et al.[154]).

3. EXHAUST EMISSIONS

Exhaust emissions constitute a substantial source of Fe-bearing NPs in urban environments[8,101−104]). Although about 90% of tailpipe emissions is carbonaceous material, exhaust emissions contain up to 10 wt % of trace metals,[105−109] *n*-alkanes and polycyclic aromatic hydrocarbons (PAHs) (e.g., refs 110−112). Diesel engines also generate soot nanospheres, of between 10 and 60 nm,[113,114] which are often associated with Mg, Fe, Cu, Ca and Zn.[104] Trace metals emitted by engines include Fe, Zn, Cr, Mo, Ti, Mg, Ni, Pb, Ca, Cu, Ba, Sb, Co, Cd, V, Pt and Pd (e.g., refs 114, 115−118).

Particle size distributions (PSD) can be measured in terms of mass and number. Number-normalized PSDs are more sensitive to the presence of nanoparticles (<100 nm), which contribute most to the particle number concentrations while contributing very little to the PM mass. Number-normalized PSDs of exhaust emissions are typically bimodal, with peaks at 20−40 nm and 50−90 nm[119,120] or unimodal, with a peak at 10−70 nm[121−124] (Figure 3). Number-normalized PSDs often depend on nucleation processes and atmospheric conditions (e.g., refs 125, 126). NP numbers are also heavily influenced by the proximity of traffic (or other emission sources), especially in the case of particles of diameter <30 nm;[103,127−131] a point of vital importance from the epidemiological point of view as the key exposure zone for traffic is reported to be within 500 m,[66,132] and more specifically, within 50 m[133] of major roadways.

Ntziachristos et al.[8] studied traffic-derived PM in Los Angeles, USA and reported that Fe was a predominant metal in the <0.18 μm fraction, accounting for 10−25% of $PM_{0.18}$ mass. Magnetite particles in car (diesel) exhaust emissions were first observed by Abdul-Razzaq and Gautam.[97] Later studies[98,136−138] found Fe oxide NPs in diesel emissions, and superparamagnetic (SP) NPs of magnetite and hematite[86] in gasoline emissions (cf. Figure 2A,B,D). Fe-bearing NPs often occur as rounded or even spherical particles, "nanospheres", of magnetite and/or hematite, depending on combustion/heating conditions (Figure 2A,B,D).[13,86,97] Miller et al.[139] identified 4 groups of NPs containing Fe: homogeneously nucleated primary NPs of Fe; carbon agglomerates decorated with primary Fe NPs; agglomerates of Fe primary particles; and combined Fe/carbon agglomerates (Figure 4). Fe-bearing particles emitted from tailpipes often comprise a mixture of all these particle types.

Fe-bearing NPs in exhaust emissions might originate from several sources. Magnetite was found in exhaust emissions when fuel additives (e.g., ferrocene) had been used (e.g., ref 140). Magnetic NPs may form due to Fe impurities in fuel which can convert to Fe oxides, such as magnetite and/or hematite, depending on combustion conditions.[13,78,86,97] Ferromagnetic NPs might also originate from aging of steel in the engine[141] and/or melting of engine fragments in the combustion chamber and subsequent crystallization during cooling.[137] In modern engines, however, various coatings (mainly Al, Cr, Mo, Ti, Zr alloy) reportedly reduce the wear rate by up to 94%.[142]

Studies on animals showed that exposure to exhaust emissions might be associated with developmental impairment in various ways, including changes in growth, sexual development, hormone levels, weight of the reproductive and accessory organs, expression of immune-related genes and susceptibility to allergies (e.g., refs 143−145). The exposure to diesel and biodiesel emissions might also lead to cardiovascular alterations as well as pulmonary and systemic inflammation in mice.[146] Gasoline-derived PM has also been associated with health problems, e.g., impairment of epithelial defense mechanisms.[147] Moreover, a study by Cheung et al.,[148] evaluating the oxidative potential of gasoline, diesel and biodiesel emissions, showed that soluble Fe is strongly associated (R = 0.99) with particulate ROS activity (see Table SI 3, Supporting Information for more details).

4. BRAKE WEAR EMISSIONS

Brake wear debris comprises primarily carbonaceous and metal-bearing components.[149] In contrast to tailpipe emissions, Fe often dominates, constituting >50 wt % of all brake wear emissions.[150−153] Besides Fe, brake dust emissions can also contain a wide range of elements, including Cu, C, Ba, Sb, Si, Al, Mo, S, Sn, Cd, Cr, Pb, Zr, Ti and Zn.[96,134,152,154−157]

Table SI 4 (Supporting Information) shows total particle number and mass of nonvolatile brake dust emissions during a pin-on-disc test of a commercial, low-metallic brake pad.[134] The NP fraction (<100 nm) constitutes >90% of total particle number. Similarly, Garg et al.[149] reported that most particles were <30 nm (cf. Figure 3).

Although Fe-rich particles have been widely reported in brake wear studies,[12,134,150,152,154,158] little attention has yet been paid to identification of their properties (phase/mineralogical composition, concentrations of particular phases, size distribution) or potential health impact. Figure 5 shows an agglomerate of NPs 20−100 nm in size, derived from brake wear collected during a dynamometer test of a widely available

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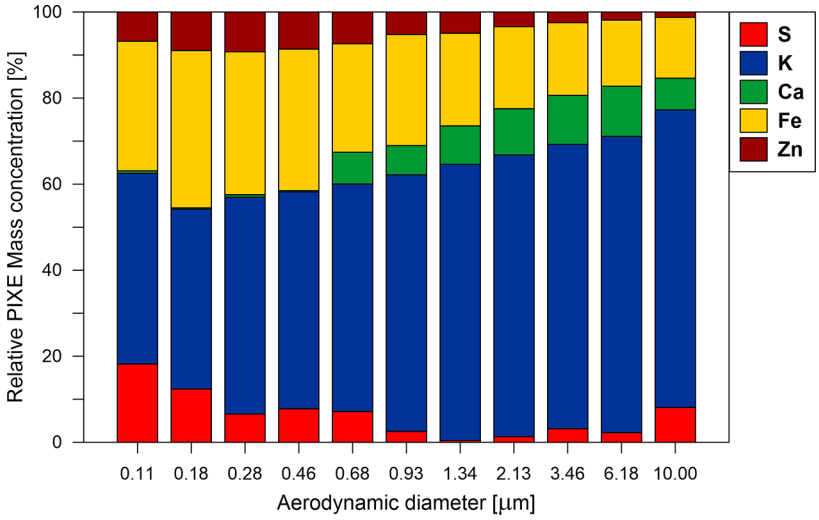

Figure 6. Relative mass concentration of Zn, Fe, Ca, K and S in tire wear, excluding dominating mineral elements Al and Si (adapted from Gustafsson et al.[99]).

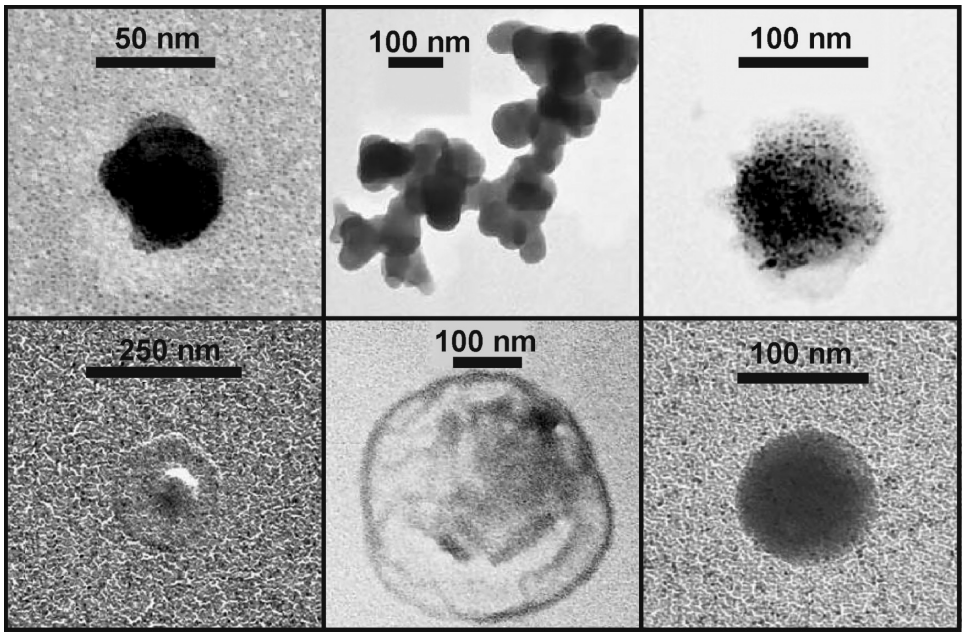

Figure 7. TEM images of tire wear NP generated by a road simulator (adapted from Dahl et al.[185]). No compositional data were provided by the authors.

brake pad. These brake-wear NPs contain mostly magnetite and elemental Fe.[154] Other authors have reported other Fe oxides, mainly hematite, wüstite (FeO) and maghemite in the NP fraction.[12,134,155]

Brake wear debris results from friction between a brake pad, of diverse chemical compositions, and the cast iron brake disc. Both members of this friction couple can be a source of Fe-bearing NPs. Moreover, nano- and submicrometer-sized Fe powder, as magnetite, is often added (in concentrations up to 50 wt %) to brake pads,[159] as a solid lubricant. At brake pad temperatures below 200 °C, abrasive processes dominate, and wear particles >1 μm are mostly generated. But at higher temperatures (>190 °C), the concentration of nanosized grains (<100 nm) increases due to evaporation, condensation and aggregation processes.[134,149,152,154] As a result, high numbers (10^{11} particles/stop/brake) of Fe-rich NPs are emitted.[96,134,149,152,154,160,161] The NP emission rate increases by 4−6 orders of magnitude at a critical temperature, usually 160 to 190 °C.[162−165]

In vitro and *in vivo* studies (see Table SI 3, Supporting Information) show that brake wear emissions can induce oxidative stress and chromosomal damage, invoke pro-inflammatory response and cause increased ROS production.[166−173] The effects were more severe for the emissions originating from nonasbestos organic brake pads, compared to low-metallic ones.[171,174]

Since NPs constitute the great majority of brake wear particle numbers, and Fe is the dominant component of those NPs, brake wear dust thus comprises a major source of airborne Fe-bearing NPs (see also Section 11).

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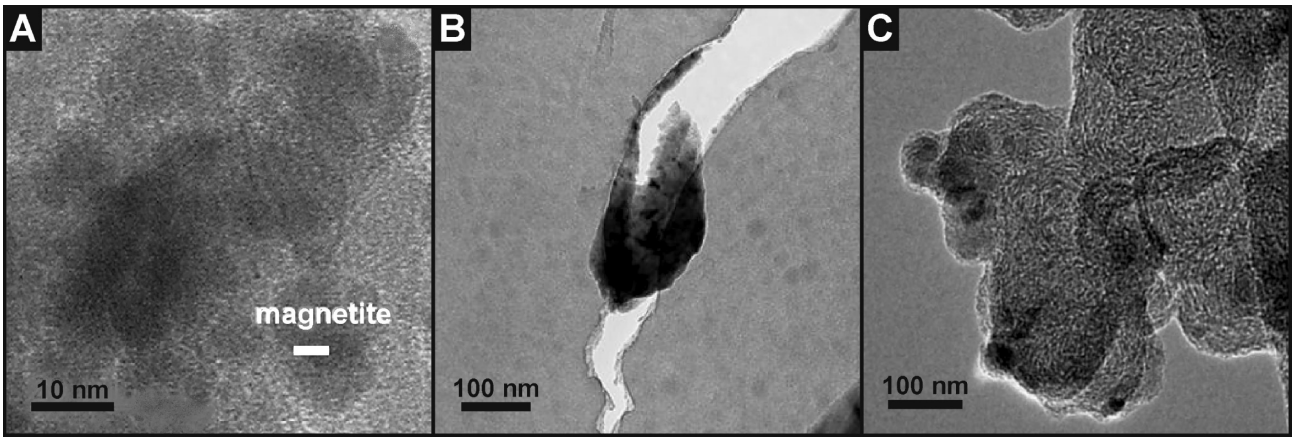

Figure 8. TEM images of (A) magnetite NPs found in subway PM (adapted from Moreno et al.[94]) and (B) aircraft-derived NPs containing mainly Fe, Cr and Ni (adapted from Mazaheri et al.[246]), (C) ship-derived soot particles with V-, Fe- and Ni-bearing speckles of 5−10 nm (adapted from Streibel et al.[247]).

5. TIRE AND ROAD SURFACE WEAR EMISSIONS

Other types of traffic-related NPs, from tire and road surface wear, contain lower concentrations of Fe-bearing NPs, but still display high genotoxicity and potential hazard to human health.[175−178]

Tire wear debris typically comprises 47% rubber, 21.5% carbon black and as much as 16.5% metals.[179] Dominant components of tire wear include *n*-alkanes, *n*-alkanoic acids, PAHs, benzothiazoles[110,120,151,157,178,180−184] and elements such as Al, Si, Zn, Fe, S, K, Ca, Ti and Mg (Figure 6).[99,157,176] Notably, studies showed that Fe occurs in all size fractions <10 μm of tire wear emissions, with concentrations up to 37 wt % in the fractions <460 nm (Figure 6).[99]

The PM (mass and number) emitted from tire/road surface wear depends on several factors, i.e., speed, slip angle, load and longitudinal force.[99,135,185] Tire/road surface wear particles >10 μm constitute 60−70%, PM_{10} 30% and $PM_{2.5}$ 4.5% of total tire emission mass.[186] However, tire wear accounts for a negligible portion (<5%) of road dust mass, contributing mainly to the NP (<100 nm) fraction[135,185] (cf. Figures 3 and 7), whereas road surface wear is responsible for emission of the larger grains.[99,186] Foitzik et al.[135] found that the PSD of tire NP emissions is bimodal (with peaks at 10 and 25 nm) or unimodal (peak at 10−15 nm) (cf. Figure 3). Other authors report number PSDs with slightly larger grains (main peak 25−45 nm).[99,185,187] Some authors, however, dispute NP generation during real-world conditions.[188]

The interaction between tires, road surface and traction sanding (sand introduced on the road surface to improve vehicle traction) results in emission of substantial particle numbers, especially in, for example, Nordic countries and Japan where studded tires are used regularly during winter.[189] NPs from tires might derive from the evaporation of semivolatile softening oils at temperatures >100 °C,[185] or inclusions of zinc oxides and sulfates.[99,176,185] Moreover, steel (and other metals) is often added (up to 25 wt %) as reinforcement material[190] so is likely to be an additional source of tire-derived Fe-bearing NPs.

Toxicological studies (see Table SI 3) suggest that tire/road surface wear poses a threat to human health via damage to epithelial cells and ROS formation.[175−178]

6. RESUSPENSION OF ROADSIDE DUST

Resuspension of road dust is one of the major sources (27−38 wt %) of airborne PM in urban environments (e.g., refs 109, 191−194). PM fractions >1 μm are predominant in resuspended dust,[75,109,186,195,196] with submicrometer particles constituting ∼10−15 wt %.[186]

High Fe concentrations (11,000−94,000 ppm) have been observed in the submicrometer fraction of roadside PM[13] in several different forms: magnetite, hematite, metallic Fe, ferrihydrite and goethite (Figure 2).[13,197−204]

Roadside dust can be a complex mixture of natural, soil-derived particles, and of traffic- (both exhaust and nonexhaust) and industry-derived emissions. Hence, exposure to resuspended dust may also be hazardous to human health.

7. UNDERGROUND, TRAIN AND TRAM EMISSIONS

Underground transport systems are used daily by approximately 4 million people in London,[205] 4.5 million in New York[206] and 7 million in Tokyo.[207] Although commuting typically constitutes only ∼10% of the day (e.g., refs 208, 209), subway commuters are exposed to high levels of pollutants,[210] especially Fe-bearing NPs.[94,211] Daily average $PM_{2.5}$ levels in subway environments reach 480 $\mu g/m^3$ and particle number concentrations (PNC) up to 31,000 particles/cm^3,[94,212−214] greatly (up to 30 times) exceeding nearby above-ground PM levels, e.g. in Helsinki,[212] Rome,[215] Seoul,[216] Buenos Aires,[217] Paris[218] or Mexico City.[219]

Fe is a dominant element in subway, train and tram emissions, in coarse (10−2.5 μm), fine (<2.5 μm) and NP (<100 nm) fractions,[94,214,217,219−229] constituting usually 65 wt % of PM_{10} and $PM_{2.5}$ (e.g., refs 213, 225, 228), sometimes reaching levels as high as 86 wt % of PM_{10},[221] 88 wt % of $PM_{2.5-1.0}$[211] and 95% of PNC.[211] Fe is usually accompanied by other metals, including Cu, Ba, Zn, Si, Al, Ca, Ni, Ti, Sb, V, Co, Cr, Mn.[94,214,225,230,231]

The PSD of subway emissions is dominated by NPs <100 nm.[220−222,232] Airborne PM at subway stations contains a mixture of magnetite, maghemite and hematite, with some additional evidence of the presence of metallic Fe, wüstite and Fe oxyhydroxides (goethite and/or lepidocrocite).[94,219,220,222,227,233−236] Fe-bearing NPs of 5−50 nm usually occur in the form of spherical, agglomerated grains of metallic Fe with oxidized rims of magnetite and/or hematite

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(Figure 8A).[94,220,222,227,235] Some studies identify hematite as the dominant Fe phase in subway airborne NPs;[220,235] others find magnetite in the highest concentrations.[227,233] It is evident that the composition of particulate air pollution is variable with location, and hence, that impacts on human health may also differ by location. The Fe oxidation level is important in terms of health hazard (see Section 12); thus more data are needed to quantify the magnetite/hematite contributions to total subway PM and if necessary, to identify and introduce mitigation measures limiting subway PM emissions.[237,238]

Subway-derived Fe-bearing NPs may form from braking,[94,211,230,231] wheel/rail track interactions[232,235,239] or sparking between a third-rail and collectors.[220] The presence of coexisting metallic Fe (often with oxidized rim), magnetite, maghemite and hematite suggests that particular Fe oxides reflect increasing stages of oxidation.[94]

Fe-rich subway particles have been found to be toxic (see Table SI 3, Supporting Information), causing mitochondrial depolarization, inflammatory responses and generation of ROS,[229,233,240−245] especially in the case of fractions <2.5 μm and <0.18 μm.[244] Karlsson et al.[233] found that subway particles are 8 times more genotoxic than roadside PM collected nearby a very busy street. The high genotoxicity of subway-derived PM probably results from synergistic effects of Fe-, Cu- and Zn-bearing, subway-derived pollution, possibly in the fraction <0.1 μm.[241,244]

Trams and trains generate similar Fe-bearing NPs (e.g., refs 76, 223, 224, 248), which may be particularly hazardous at subway stations and/or tunnels due to confined spaces, poor ventilation and high congestion. Hence, commuters using underground systems are exposed to high levels of metal-rich, potentially hazardous airborne particulates.[210]

8. AIRCRAFT EMISSIONS

Masiol and Harrison[249] have reviewed aircraft emissions and other airport-related contributions to ambient air pollution. Airports, and their associated road traffic, are a source of Fe-bearing NPs.

Nanoparticulate aircraft emissions are dominated by soot particles, accompanied by smaller amounts (4−6 wt %) of Fe[100,246,250−253] and other metals, including Cr, Ni, Ca and K.[246,250]

Number-normalized PSDs of aircraft emissions are dominated by NPs (<100 nm), with a main peak at 10−20 nm.[246,254−259]

From TEM and EDX, aircraft-derived Fe-bearing NPs occur in the form of magnetite and/or maghemite (Figure 8B).[100] The presence of Fe in soot agglomerates might arise from chemical/mechanical corrosion of the liquid fuel atomizer.[100] Fe-bearing NPs in aircraft emissions might also originate from impurities in the kerosene fuel.[251]

Aircraft emissions are reported to have deleterious health impacts both globally and locally.[260,261] Aircraft-generated NPs, at Santa Monica Airport, Southern California, USA, occurred at concentrations 11, 10, 5 and 2.5 times the background value for sites at distances from the airport of 80, 100, 380 and 660 m, respectively.[256] Enhanced, aircraft-derived levels of NPs (up to 20× background concentrations) persisted up to 900 m from a takeoff area,[255−257] thus likely exerting an adverse influence on adjacent neighborhoods (see Table SI 3, Supporting Information). Furthermore, there have recently been several lawsuits related to "aerotoxic syndrome", linked to occupational exposure to polluted cabin air and associated with severe headaches, mental confusion, sight problems, insomnia, digestive and respiratory problems, and even death.[262] The impacts of aircraft emissions on human health are poorly understood, but some evidence indicates that occupational exposure of airport personnel and aircraft crew has detrimental effects.[249,262−266]

9. SHIPPING EMISSIONS

Shipping emissions, derived from combustion of so-called "bunker fuel", affect air quality both locally[267−271] and globally.[272−274]

Most studies to date have focused on NO_x, SO_2, CO and CO_2 as they constitute the majority of ship-derived emissions.[267,273,275−279] Fe-bearing particles occur in the $PM_{2.5}$ fraction in concentrations up to 10 $\mu g/m^3$,[91,247,270,280−286] usually accompanied by other metals, including V, Ni, Zn, Ca, Na, P.[91,282−284,286] Using cluster analysis of elemental composition and electron microscopy observations, Popovicheva et al.[284] distinguished 5 groups of particles in shipping emissions. The Fe-rich group, constituting 3−4% of all PM mass, comprised 50% Fe, with smaller portions of Si, S and Ca.

Shipping-derived Fe-bearing particles occur usually in the fraction <100 nm, in the form of 5−10 nm inclusions in soot particles (Figure 8C).[247,285] The NPs have been observed in the forms of Fe_2O_3, Fe_2S, Ni_2Fe, Ni_3Fe, $Ni_3Fe_2O_3$ and $NiFe_2O_4$.[91,284]

Those inclusions are formed as a result of the decomposition or oxidation of Fe-bearing minerals in heavy fuel oil, and/or corrosion of the engine.[284]

Exposure to shipping-derived particles (see Table SI 3, Supporting Information) results in acute cytotoxic effects in cell cultures *in vitro*[286,287] and oxidative stress.[288] Shipping emissions are related to 60,000 cardiopulmonary and lung cancer deaths annually.[289] In busy ports, the health impacts of shipping-derived Fe-bearing NPs are additive to the NP loadings from associated road/freight traffic.

10. SOURCE APPORTIONMENT OF ROADSIDE-DERIVED PM

Estimation of the contributions made by specific sources to total airborne PM or PNC is often difficult, and variable between different studies (Figure 9).[191,194,290−295] For a street canyon in Zurich, and using elemental data and positive matrix factorization (PMF) to distinguish between sources, Bukowiecki et al.[191] found that brake wear, exhaust emissions and

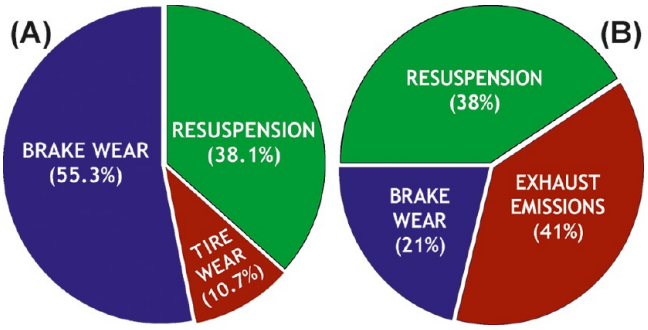

Figure 9. Source apportionment of (A) nonexhaust PM emissions (adapted from Harrison et al.[292]) and (B) total traffic-derived PM (adapted from Bukowiecki et al.[191]).

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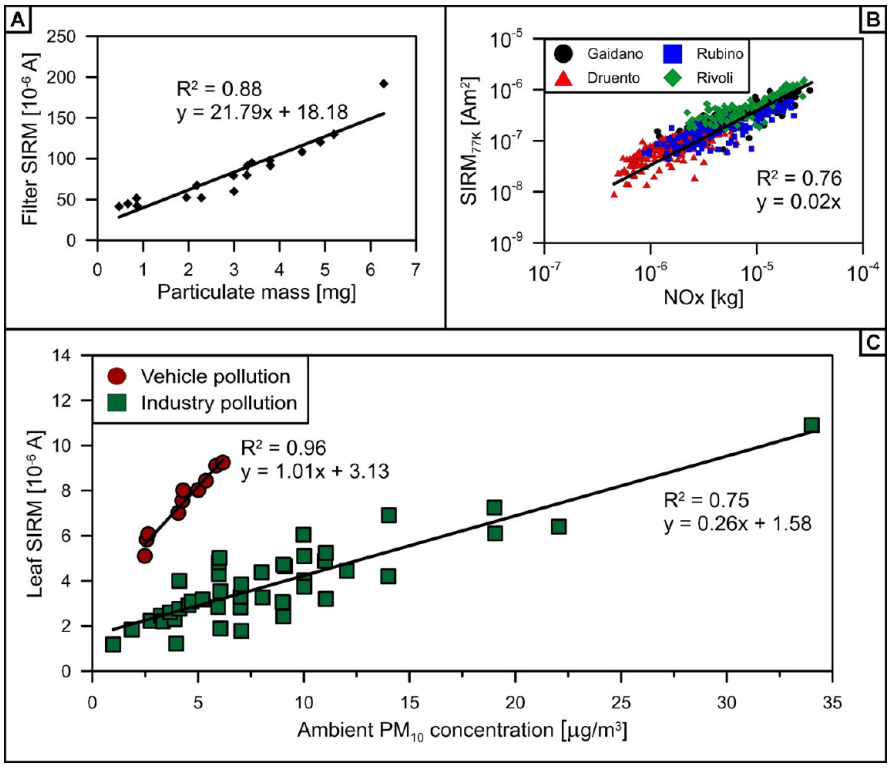

Figure 10. Correlation between: (A) air filter SIRM and roadside PM_{10} mass in Lancaster, UK (adapted from Mitchell and Maher[79]); (B) air filter SIRM at 77K ($SIRM_{77K}$) and concentration of total nitrogen oxides (NO_x) at the roadside, in Turin, Italy (adapted from Saragnese et al.[315]); (C) leaf SIRM and ambient PM_{10} concentrations, derived from vehicle pollution and industrial stack point source pollution (adapted from Mitchell et al.[322] and Hansard et al.[80]).

resuspension contributed 21%, 41% and 38% of total traffic-derived PM_{10} mass, respectively (Figure 9B). Lawrence et al.[194] studied PM in a tunnel in Hatfield, UK, relying on principal component analysis (PCA) and multiple linear regression analysis, and reported that resuspension was responsible for 27% of PM_{10} mass, diesel emissions for 21%, gasoline emissions for 12%, brake wear for 11%, road surface wear for 11%, with 18% of PM_{10} mass not attributed to any source. Harrison et al.[292] studied roadside PM at a busy London street, assessing the contribution of nonexhaust emission sources using specific elemental tracers, i.e., Ba for brake emissions and Zn for tire emissions. They found that the mass of brake wear (0.9−11.5 μm) was higher than that of resuspended dust (55.3 ± 7.0% versus 38.1 ± 9.7% of nonexhaust emissions). In this study, 10.7 ± 2.3% of nonexhaust PM (0.9−11.5 μm) was attributed to tire wear (Figure 9A).

Some source apportionment studies have considered particle number and volume of PM_{10} (e.g., refs 291, 293). The authors found that exhaust emissions accounted for 22.4% of total volume and 65.4% of total number, brake dust 13.7% of total volume and 1.7% of total number, and resuspension 4.4% of total volume and 4.8% of total number. Tire wear emissions were not identified in this study.[291]

Discrepancies between these estimates probably reflect the differing methodologies, instruments and characteristics of the sampling sites. There is presently no unambiguous tracer of resuspended dust, which makes estimation of the resuspension contribution challenging.[109] Pant and Harrison[109] provide a detailed description of methodologies for the assessment of source apportionment of urban PM.

Around the world, there are numerous emission standards limiting exhaust emissions, e.g., European Emission Standard (EURO) in European Union (EU), Tier motor vehicle emission and fuel standards in USA, WLTC-based (Worldwide Harmonized Light Vehicles Test Cycle) regulation in Japan, or Bharat Stage regulation in India.[296] In response to increasing regulation, the automotive industry has sought, with varying measures and outcomes, to reduce exhaust emissions. Non-tailpipe emissions are currently unregulated; yet constitute a similar proportion of total mass traffic-derived PM as exhaust emissions (Figure 9B).[191,297,298] Resuspension may become a more substantial source of roadside PM, as electrification (both electric engines and regenerative braking systems) of the vehicle fleet will decrease both exhaust and brake wear emissions.[299] Further research is needed to quantify the resuspension contribution to roadside PM.

11. MAGNETIC MEASUREMENTS OF VEHICLE-DERIVED, IRON-BEARING NANOPARTICLES

The majority of the Fe-bearing particles emitted by vehicles occur as ferromagnetic (*sensu lato*), i.e., magnetically ordered, minerals, especially as magnetite (Fe_3O_4), maghemite (γ-Fe_2O_3), hematite (α-Fe_2O_3) and metallic α-Fe.[12,76−80,87,92,300] Thus, magnetic analyses, based on concentration- and magnetic grain size-dependent parameters, enable characterization and quantification of vehicle-derived PM (e.g., refs 76, 79, 300−302).

Magnetic methods have enabled mapping of heavy metal contaminations in soils (e.g., refs 303−305), as well as PM concentrations in the urban atmosphere (e.g., refs 80, 92, 248,

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306−308). Hofman et al.[309] review data from biomagnetic monitoring studies of air pollution (see also Table SI 6, Supporting Information).

Significant correlation has frequently been found between PM_{10}, NO_x, heavy metal content and concentration-dependent magnetic properties (e.g., magnetic susceptibility, saturation isothermal remanent magnetization) of PM (Figure 10).[78−80,248,300−302,310] The magnetite content of roadside PM_{10} appears greater than PM_{10} from a power generation emissions stack (Figure 10c).[80] An early study identified strong correlation ($n = 55$; $R^2 = 0.79$) between magnetic susceptibility of airborne PM and mutagenicity.[311] Moreover, other composition- and magnetic grain size-dependent magnetic parameters (e.g., hysteresis properties, first-order reversal curves) have often been helpful in the distinction between different sources of vehicle-derived pollutants (e.g., refs 312−314). For example, Sagnotti et al.[312] and Sagnotti and Winkler[314] showed that exhaust emissions have distinctive magnetic hysteresis properties compared to brake wear emissions.

Magnetic NPs <30 nm in size display a distinctive magnetic behavior; they are "superparamagnetic" (SP), unable to hold a magnetic remanence at room temperature, due to thermal agitation of their magnetic moments. Several magnetic studies have reported on the ultrafine, SP fraction of airborne roadside pollution.[76,300,312−317] In Munich, Germany, SP ferrites contributed 22% to the total saturation isothermal remanent magnetization (SIRM) of roadside PM and the SP fraction was dominated by 10−16 nm grains of maghemite and 5−8 nm grains of α-Fe.[76] Higher SP contributions to the SIRM of roadside PM, even up to 88%, have since been reported,[312,315] possibly reflecting different measurement temperatures. Muxworthy et al.[76] measured SIRM at 77 K, Sagnotti et al.[312] at 4 K and Saragnese et al.[315] at 15 K. Due to thermal agitation, grains of magnetite <16 nm, maghemite <22 nm and hematite <11 nm are unable to retain magnetic remanence at 77 K[318−321] and therefore are magnetically "invisible" at this temperature. It should be noted that the SP fraction might occur not only as discrete magnetic grains <30 nm in size but also as oxidized rims around larger particles.[319]

Magnetic PM, containing redox-active Fe, might not only be harmful to human health on its own; it is also usually associated with other toxic components, including Ba, Br, Cr, Cu, Mn, Mo, Ni, Pb, Zn[10,13,75,302,323] and PAHs.[310,324]

Critically, although Fe-bearing fine (<2.5 μm) and nanosized (<0.1 μm) particles are present in almost all types of traffic-derived emissions (see previous sections), the magnetic signatures of specific particle sources appear to be distinctive (Figure 11; see also Table SI 5, Supporting Information). For example, the ferrimagnetic content of brake wear PM greatly exceeds that of many other PM sources (Figure 11). Furthermore, the SP fraction of brake wear emissions contributes 35% of SIRM, compared to 20% SP contribution in gasoline tailpipe emissions.[313,314] Component analysis of magnetic coercivity distributions of roadside PM_{10} in Switzerland enabled identification of two magnetic components/PM sources: the first (C1), resuspended dust (a mixture of natural and anthropogenic street dust); the second (C2), traffic exhaust emissions (Figure 12).[302]

Conventional, elemental analysis of Fe, which is present in high concentrations at urban roadsides (see previous sections) cannot differentiate between its different vehicular sources. In contrast, magnetic methods of PM analysis can distinguish

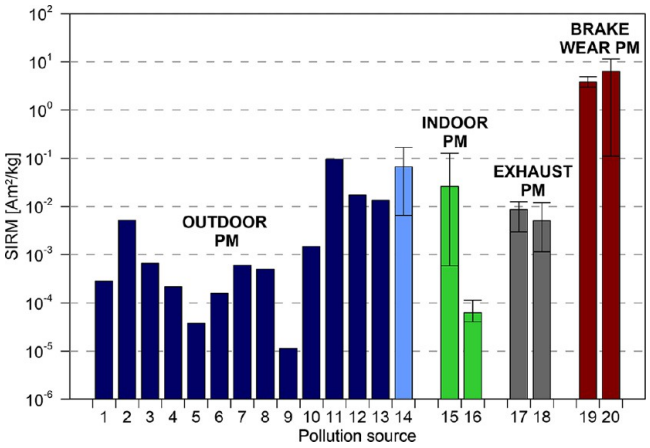

Figure 11. Saturation magnetic remanence (SIRM) for indoor and outdoor PM (adapted from Halsall et al.[77] and Hansard et al.[92]), exhaust and brake wear emissions:[325] 1, wood fired boiler; 2, coal fired boiler; 3, gas/oil boiler; 4, oil fired boiler; 5, gas kettle incinerator; 6, coal fired power station; 7, gas/oil power station; 8, graphite plant; 9, smelting plant; 10, coke; 11, sinter; 12, BOS slag; 13, iron ore pellets; 14, roadside filters from Halsall et al.;[77] 15, indoor filters from Halsall et al.;[77] 16, printer dust from Gonet et al.;[325] 17, diesel emissions from Gonet et al.;[325] 18, gasoline emissions from Gonet et al.;[325] 19, airborne brake wear emissions, from Gonet et al.;[326] and 20, nonairborne brake emissions, from Gonet et al.[326]

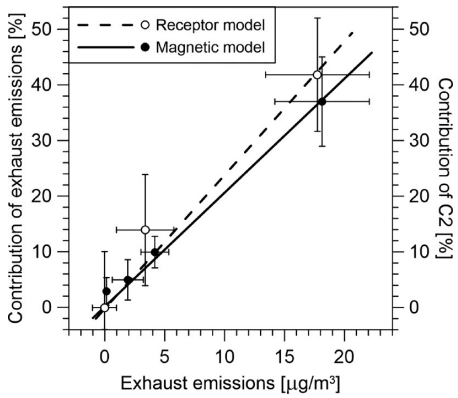

Figure 12. Absolute and relative contributions of exhaust emissions to PM_{10} at several sites in Switzerland. Open circles and dashed best-fit line refer to the receptor model (chemically estimated PM_{10} mass contribution of exhaust emissions). Solid dots and the solid best-fit line refer to the magnetic model, where component C2 (identified using magnetic component analysis) corresponded to exhaust emissions (adapted from Spassov et al.[302]).

between different Fe minerals and oxidation states (Fe^{2+} vs Fe^{3+}), information on particular potential value regarding Fe-bearing NP toxicity and human health impacts (see Section 12). Moreover, given distinctive magnetic fingerprints from different PM sources, combining elemental and magnetic analyses may enable more accurate and precise source apportionment (cf. Figure 12).

12. SPECIFIC TOXICITY OF IRON-BEARING NANOPARTICLES

Exposure to airborne Fe-bearing NPs may be particularly hazardous to human health due to their high bioreactivity and potential toxicity, combined with their abundance in the urban environment. Numerous studies have shown that Fe plays an

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important role in the generation and pathogenicity of ROS.[3,327−335] Dusseldorp et al.[328] found that airborne Fe was associated with exacerbation of some respiratory symptoms. Fe-rich particles have been shown to generate ROS and provoke an inflammatory response.[331] These findings are supported by the results obtained by Pelclova et al.[334] who found elevated oxidative stress markers in exhaled breath condensate of workers exposed to NPs during Fe oxide pigment production (see also Table SI 3, Supporting Information).

The presence in the human brain specifically of magnetite, a strongly magnetic, mixed Fe^{2+}/Fe^{3+} oxide, is important because it has been causally linked with neurodegenerative diseases. Magnetite in brain tissue has been observed to be directly associated with AD plaques (e.g., refs 336−338) and correlated (albeit with rather small sample size) with the incidence of AD.[339,340] Neuropathological changes associated with AD include the formation of senile plaques, containing amyloid-β fibrils. When associated with redox-active transition metal ions, such as the Fe^{2+} ions contained within magnetite, amyloid-β can generate damaging ROS, directly contributing to oxidative brain damage, a key early feature of AD (e.g., refs 74, 341, 342). *In vitro* experimental data additionally show that magnetite acts synergistically to enhance the toxicity of amyloid-β.[341,343] Moreover, higher concentrations of magnetite have been also detected in brain tumor tissue, in meningioma[344,345] and in diseased hippocampal tissues,[346] compared to healthy brain tissue.

In vitro studies on A549 human epithelial cells have shown that exposure to magnetite (Fe_3O_4) leads to acute cytotoxic effects and generation of ROS.[347] In the case of the NP fraction (<0.1 μm), the health response (DNA damage) was more severe than with larger particles (0.2−10 μm).[348] Assessment of the "theranostic" use of SP iron oxide nanoparticles (SPIONs) has also shown that exposure to these particles can lead to toxic effects, including DNA damage, ROS generation, chromosome condensation, formation of apoptotic bodies, inflammation, impaired mitochondrial function and membrane leakage of lactate dehydrogenase (e.g., refs 349−352; see also Table SI 3, Supporting Information).

Fe^{2+} is toxic as it may cause the generation of ROS via the Fenton reaction:[68]

$$H_2O_2 + Fe^{2+} \rightarrow HO\cdot + OH^- + Fe^{3+} \quad (1)$$

The hydroxyl radical (HO·) is especially dangerous in brain tissue as it might cause oxidative stress, which is a hallmark of neurodegenerative diseases like AD (e.g., refs 74, 341, 342). Until recently, the magnetite particles found in the human brain were assumed to be entirely of *in situ*, biogenic origin.[353,354] It has been suggested that the normal processes for safe storage (within ferritin cores) and transport of the brain Fe become dysfunctional as a result of neurodegeneration (e.g., ref 74). However, Maher et al.[10] identified, for the first time, the abundant presence in the human brain of externally derived, magnetite pollution NPs. These magnetite NPs displayed a range of particles sizes (from 10 to 150 nm), were frequently rounded or even spherical, and often associated with other "exotic" transition metal NPs, containing Pt, Ni, Co and Cu. Some have fused surface crystallites that would be very difficult to reconcile with *in situ*, low temperature growth or dissolution formation processes.

Thus, airborne magnetic pollution NPs may pose a threat to human health due to their bioreactivity, their ability to penetrate every organ, including the brain, their link with neurodegenerative diseases and their abundance in the urban atmosphere.

13. PERSPECTIVES AND OUTLOOK

Many studies have identified adverse health effects associated with exposure to particulate air pollution (e.g., refs 28, 355). Traffic-derived emissions are a major source of urban PM, constituting up to 80% of airborne concentrations of PM in the urban environment.[109] Hence, improved understanding of the biological interactions induced in the human body by traffic-derived PM is of critical importance for identifying their specific pathways of impact, and relative priority in terms of their abatement and/or mitigation.

Metal-bearing NPs may be especially hazardous to human health. Studies on mouse models and human epithelial cells have demonstrated the detrimental impact of NPs (e.g., refs 20, 356), with Fe-bearing NPs of particular potential hazard (e.g., refs 3, 330). Combustion- and friction heating-derived, Fe-bearing NPs found in human brain[10] are especially worrisome, due to the high redox activity of magnetite and its potential association with neurodegenerative diseases, such as Alzheimer's or Parkinson's diseases.[10,68,338] Given the abundant occurrence of Fe-bearing NPs in the environment (indoor and outdoor), workplace, and in biomedical applications, and the inconsistency of the data reporting their effects on biological systems, more systematic studies on nanosized, Fe-bearing particles are needed in order to define their toxic potential.

Traffic-related air pollution has been shown to be associated with respiratory, cardiovascular and neurological problems (e.g., refs 28, 357). However, little is known about long-term health effects of ambient NPs. Recently, studies have examined regression models of health outcomes and the spatial distribution of NPs (e.g., refs 358−363). These models can be useful in the assessment of chronic health effects of NPs using population-based cohorts. Ostro et al.[364] observed statistically significant associations between ischemic heart disease (IHD) mortality and both $PM_{2.5}$ and $PM_{0.1}$ fractions. Moreover, some specific components were observed to be independently associated with IHD mortality, including gasoline/diesel emissions and metals (Fe and Cu).[364] The results of Weichenthal et al.[361] suggest that exposure to ambient NPs is directly associated with the incidence of prostate cancer. Bai et al.[365] found that chronic exposure to NPs may increase the incidence of hypertension and diabetes. Land use regression models can potentially be used, along with actual NP concentration measurements, as a basis for an assessment of human chronic exposures to NPs. Interestingly, Weichenthal et al.[362] did not observe evidence of positive associations between modeled long-term NP exposure and the incidence of respiratory disease.

It is worth emphasizing that humans are variably exposed not to a single, individual traffic-related source of particulates, but to a complex mixture of them. Some of the deleterious effects of particular particles/compounds can be accelerated by the presence of others. For example, adverse effects (for instance, lipid peroxidation in brain tissue) of Fe-bearing particles (for instance, from brake wear) are promoted by the presence of Al (for instance, from tire wear or natural, soil dust).[366] Although increasing numbers of epidemiological

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studies have identified associations between exposure to traffic-derived air pollution and detrimental health impacts, the specific mechanisms of these associations remain imperfectly understood.

Furthermore, PM composition is variable not only from city to city but also within cities, such that different populations are exposed to different pollution mixes. Hence, the impacts of exposure to PM on human health likely also differ by location.

Exhaust emissions have been legally regulated for over 25 years (e.g., in EU since 1992, in USA since 1994, in India since 1991).[296] Initially, only the mass of CO (carbon monoxide), HC (hydrocarbons; in USA), NOx (nitrogen oxides; in EU and USA), PM (in EU and USA) and NMHC (nonmethane hydrocarbons; in USA) was limited. Subsequently, more parameters have been covered by these regulations, e.g., mass of VOC (volatile organic compounds) and PN (particle number).[296] It seems highly probable that legal regulation concerning brake wear emissions will be introduced in the near future, especially given their increasing importance as exhaust emissions decline with electrification of vehicle fleets. Given the growing evidence that Fe-bearing NPs may constitute a particular hazard to human health, regulation to limit these Fe-rich, strongly magnetic emissions may also become important and urgent, on an international scale.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.9b01505.

Search strategy; nonvehicle sources of Fe-bearing nanoparticles in the urban environment; summary table of selected toxicological studies of vehicle-derived PM; total particle number and mass of brake dust emissions during a dynamometer test; selected magnetic parameters of exhaust and brake wear emissions; inventory table of reported magnetic studies on pumped-air filters and tree leaves (PDF)

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Notes

The authors declare no competing financial interest.

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■ REFERENCES

(1) Ferin, J.; Oberdörster, G.; Penney, D. P. Pulmonary retention of ultrafine and fine particles in rats. *Am. J. Respir. Cell Mol. Biol.* **1992**, *6*, 535−542.

(2) Seaton, A.; MacNee, W.; Donaldson, K.; Godden, D. Particulate air pollution and acute health effects. *Lancet* **1995**, *345*, 176−178.

(3) Gilmour, P. S.; Brown, D. M.; Lindsay, T. G.; Beswick, P. H.; MacNee, W.; Donaldson, K. Adverse health effects of PM_{10} particles: involvement of iron in generation of hydroxyl radical. *Occup. Environ. Med.* **1996**, *53*, 817−822.

(4) Gilmour, P. S.; Brown, D. M.; Beswick, P. H.; Benton, E.; MacNee, W.; Donaldson, K. Surface free radical activity of PM_{10} and ultrafine titanium dioxide: a unifying factor in their toxicity? *Ann. Occup. Hyg.* **1997**, *41* (1), 32−38.

(5) Stone, V.; Shaw, J.; Brown, D. M.; MacNee, W.; Faux, S. P.; Donaldson, K. The role of oxidative stress in the prolonged inhibitory effect of ultrafine carbon black on epithelial cell function. *Toxicol. In Vitro* **1998**, *12*, 649−659.

(6) Donaldson, K.; Stone, V.; Seaton, A.; MacNee, W. The pulmonary toxicology of ultrafine particles. *J. Aerosol Med.* **2002**, *15* (2), 213−220.

(7) Brook, R. D.; Rajagopalan, S.; Pope, A. C., III; Brook, J. R.; Bhatnagar, A.; Diez-Roux, A. V.; Holguin, F.; Hong, Y.; Luepker, R. V.; Mittleman, M. A.; Peters, A.; Siscovick, D.; Smith, S. C., Jr; Whitsel, L.; Kaufman, J. D. Particulate matter air pollution and cardiovascular disease. An update to the scientific statement from the American Heart Association. *Circulation* **2010**, *121*, 2331−2378.

(8) Ntziachristos, L.; Ning, Z.; Geller, M. D.; Sheesley, R. J.; Schauer, J. J.; Sioutas, C. Fine, ultrafine and nanoparticle trace element compositions near a major freeway with a high heavy-duty diesel fraction. *Atmos. Environ.* **2007**, *41*, 5684−5696.

(9) Hays, M. D.; Cho, S. H.; Baldauf, R.; Schauer, J. J.; Shafer, M. Particle size distributions of metal and non-metal elements in an urban near-highway environment. *Atmos. Environ.* **2011**, *45*, 925−934.

(10) Maher, B. A.; Ahmed, I. A. M.; Karloukovski, V.; MacLaren, D. A.; Foulds, P. G.; Allsop, D.; Mann, D. M. A.; Torres-Jardón, R.; Calderón-Garcidueñas, L. Magnetite pollution nanoparticles in the human brain. *Proc. Natl. Acad. Sci. U. S. A.* **2016**, *113* (39), 10797−10801.

(11) Sanderson, P.; Delgado-Saborit, J. A.; Harrison, R. M. A review of chemical and physical characterisation of atmospheric metallic nanoparticles. *Atmos. Environ.* **2014**, *94*, 353−365.

(12) Sanderson, P.; Su, S. S.; Chang, I. T. H.; Delgado Saborit, J. M.; Kepaptsoglou, D. M.; Weber, R. J. M.; Harrison, R. M. Characterisation of iron-rich atmospheric submicrometre particles in the roadside environment. *Atmos. Environ.* **2016**, *140*, 167−175.

(13) Yang, Y.; Vance, M.; Tou, F.; Tiwari, A.; Liu, M.; Hochella, M. F., Jr. Nanoparticles in road dust from impervious urban surfaces: distribution, identification, and environmental implications. *Environ. Sci.: Nano* **2016**, *3*, 534−544.

(14) Ferin, J.; Oberdörster, G.; Soderholm, S. C.; Gelein, R. Pulmonary tissue access of ultrafine particles. *J. Aerosol Med.* **1991**, *4* (1), 57−68.

(15) Donaldson, K.; Stone, V.; Clouter, A.; Renwick, L.; MacNee, W. Ultrafine particles. *Occup. Environ. Med.* **2001**, *58*, 211−216.

(16) Nel, A.; Xia, T.; Mädler, L.; Li, N. Toxic potential of materials at the nanolevel. *Science* **2006**, *311* (5761), 622−627.

(17) Oberdörster, G.; Ferin, J.; Gelein, R.; Soderholm, S. C.; Finkelstein, J. Role oft he alveolar macrophage in lung injury: studies of ultrafine particles. *Environ. Health Perspect.* **1992**, *97*, 193−199.

(18) Oberdörster, G.; Ferin, J.; Lehnert, B. E. Correlation between particle size, *in vivo* particle persistence, and lung injury. *Environ. Health Perspect.* **1994**, *102*, 173−179.

(19) Oberdörster, G.; Celein, R. M.; Ferin, J.; Weiss, B. Association of particulate air pollution and acute mortality: Involvement of ultrafine particles? *Inhal. Toxicol.* **1995**, *7*, 111−124.

(20) Oberdörster, G.; Oberdörster, E.; Oberdörster, J. Nanotoxicology: An emerging discipline evolving from studies of ultrafine particles. *Environ. Health Perspect.* **2005**, *113* (7), 823−839.

(21) Elder, A.; Gelein, R.; Silva, V.; Feikert, T.; Opanashuk, L.; Carter, J.; Potter, R.; Maynard, A.; Ito, Y.; Finkelstein, J.; Oberdörster, G. Translocation of inhaled ultrafine manganese oxide particles to the central nervous system. *Environ. Health Perspect.* **2006**, *114* (8), 1172−1178.

(22) Miller, M. R.; Raftis, J. B.; Langrish, J. P.; McLean, S. G.; Samutrtai, P.; Connell, S. P.; Wilson, S.; Vesey, A. T.; Fokkens, P. H. B.; Boere, A. J. F.; Krystek, P.; Campbell, C. J.; Hadoke, P. W. F.; Donaldson, K.; Cassee, F. R.; Newby, D. E.; Duffin, R.; Mills, N. L.

Inhaled nanoparticles accumulate at sites of vascular disease. *ACS Nano* **2017**, *11*, 4542−4552.
(23) Tjälve, H.; Henriksson, J.; Tallkvist, J.; Larsson, B. S.; Lindquist, N. G. Uptake of manganese and cadmium from the nasal mucosa into the central nervous system via olfactory pathways in rats. *Pharmacol. Toxicol.* **1996**, *79*, 347−356.
(24) Oberdörster, G.; Sharp, Z.; Atudorei, V.; Elder, A.; Gelein, R.; Kreyling, W.; Cox, C. Translocation of inhaled ultrafine particles to the brain. *Inhalation Toxicol.* **2004**, *16*, 437−445.
(25) Geiser, M.; Kreyling, W. G. Deposition and biokinetics of inhaled nanoparticles. *Part. Fibre Toxicol.* **2010**, *7* (2), 2.
(26) Oberdörster, G.; Sharp, Z.; Atudorei, V.; Elder, A.; Gelein, R.; Lunts, A.; Kreyling, W.; Cox, C. Extrapulmonary translocation of ultrafine carbon particles following whole-body inhalation exposure of rats. *J. Toxicol. Environ. Health, Part A* **2002**, *65*, 1531−1543.
(27) Barošová, H.; Dvořáčková, J.; Motyka, O.; Kutláková, K. M.; Peikertová, P.; Rak, J.; Bielniková, H.; Kukutschová, J. Metal-based particles in human amniotic fluids of foetuses with normal karyotype and congenital malformation - a pilot study. *Environ. Sci. Pollut. Res.* **2015**, *22* (10), 7582−7589.
(28) Calderón-Garcidueñas, L.; Azzarelli, B.; Acuna, H.; Garcia, R.; Gambling, T. M.; Osnaya, N.; Monroy, S.; Del Rosario Tizapantzi, M.; Carson, J. L.; Villarreal-Calderón, A.; Rewcastle, B. Air pollution and brain damage. *Toxicol. Pathol.* **2002**, *30* (3), 373−389.
(29) Calderón-Garcidueñas, L.; Reed, W.; Maronpot, R. R.; Henríquez-Roldán, C.; Delgado-Chavez, R.; Calderón-Garcidueñas, A.; Dragustinovis, I.; Franco-Lira, M.; Aragón-Flores, M.; Solt, A. C.; Altenburg, M.; Torres-Jardón, R.; Swenberg, J. A. Brain inflammation and Alzheimer's-like pathology in individuals exposed to severe air pollution. *Toxicol. Pathol.* **2004**, *32*, 650−658.
(30) Calderón-Garcidueñas, L.; Solt, A. C.; Henríquez-Roldán, C.; Torres-Jardón, R.; Nuse, B.; Herritt, L.; Villarreal-Calderón, R.; Osnaya, N.; Stone, I.; García, R.; Brooks, D. M.; González-Maciel, A.; Reynoso-Robles, R.; Delgado-Chávez, R.; Reed, W. Long-term air pollution exposure is associated with neuroinflammation, an altered innate immune response, disruption of the blood-brain barrier, ultrafine particulate deposition, and accumulation of amyloid β-42 and α-synuclein in children and young adults. *Toxicol. Pathol.* **2008**, *36*, 289−310.
(31) Block, M. L.; Calderón-Garcidueñas, L. Air pollution: Mechanisms of neuroinflammation & CNS disease. *Trends Neurosci.* **2009**, *32* (9), 506−516.
(32) Guxens, M.; Sunyer, J. A review of epidemiological studies on neuropsychological effects of air pollution. *Swiss Med. Wkly* **2012**, *141*, No. w13322.
(33) Jung, C.-R.; Lin, Y.-T.; Hwang, B.-F. Ozone, particulate matter, and newly diagnosed Alzheimer's disease: A population-based cohort study in Taiwan. *J. Alzheimer's Dis.* **2015**, *44*, 573−584.
(34) Wu, Y.-C.; Lin, Y.-C.; Yu, H.-L.; Chen, J.-H.; Chen, T.-F.; Sun, Y.; Wen, L.-L.; Yip, P.-K.; Chu, Y.-M.; Chen, Y.-C. Association between air pollutants and dementia risk in the elderly. *Alzheimers Dement. (Amst)* **2015**, *1*, 220−228.
(35) Oudin, A.; Forsberg, B.; Nordin Adolfsson, A.; Lind, N.; Modig, L.; Nordin, M.; Nordin, S.; Adolfsson, R.; Nilsson, L.-G. Traffic-related air pollution and dementia incidence in Northern Sweden: A longitudinal study. *Environ. Health Perspect.* **2016**, *124*, 306−312.
(36) Chen, H.; Kwong, J. C.; Copes, R.; Tu, K.; Villeneuve, P. J.; van Donkelaar, A.; Hystad, P.; Martin, R. V.; Murray, B. J.; Jessiman, B.; Wilton, A. S.; Kopp, A.; Burnett, R. T. Living near major roads and the incidence of dementia, Parkinson's disease, and multiple sclerosis: a population-based study. *Lancet* **2017**, *389* (10070), 718−726.
(37) Calderón-Garcidueñas, L.; Villarreal-Calderón, R.; Valencia-Salazar, G.; Henríquez-Roldán, C.; Gutiérrez-Castrellón, P.; Torres-Jardón, R.; Osnaya-Brizuela, N.; Romero, L.; Torres-Jardón, R.; Solt, A.; Reed, W. Systematic inflammation, endothelial dysfunction, and activation in clinically healthy children exposed to air pollutants. *Inhal. Toxicol.* **2008**, *20*, 499−506.
(38) Calderón-Garcidueñas, L.; Kavanaugh, M.; Block, M.; D'Angiulli, A.; Delgado-Chávez, R.; Torres-Jardón, R.; González-Maciel, A.; Reynoso-Robles, R.; Osnaya, N.; Villarreall-Calderón, R.; Guo, R.; Hua, Z.; Zhu, H.; Perry, G.; Diaz, P. Neuroinflammation, hyperphosphorylated tau, diffuse amyloid plaques, and down-regulation of the cellular prion protein in air pollution exposed children and young adults. *J. Alzheimers Dis.* **2012**, *28*, 93−107.
(39) Suglia, S. F.; Gryparis, A.; Wright, R. O.; Schwartz, J.; Wright, R. J. Association of black carbon with cognition among children in a prospective birth cohort study. *Am. J. Epidemiol.* **2007**, *167*, 280−286.
(40) Ailshire, J. A.; Crimmins, E. M. Fine particulate matter air pollution and cognitive finction among older US adults. *Am. J. Epidemiol.* **2014**, *180* (4), 359−366.
(41) Chen, J.-C.; Wang, X.; Wellenius, G. A.; Serre, M. L.; Driscoll, I.; Casanova, R.; McArdle, J. J.; Manson, J. E.; Chui, H. C.; Espeland, M. A. Ambient air pollution and neurotoxicity on brain structure: evidence from Women's Health Initiative Memory Study. *Ann. Neurol.* **2015**, *78*, 466−476.
(42) Cheng, H.; Saffari, A.; Sioutas, C.; Forman, H. J.; Morgan, T. E.; Finch, C. E. Nanoscale particulate matter from urban traffic rapidly induces oxidative stress and inflammation in olfactory epithelium with concomitant effects on brain. *Environ. Health Perspect.* **2016**, *124* (10), 1537−1546.
(43) Morrow, P. E. Possible mechanisms to explain dust overloading of the lungs. *Fundam. Appl. Toxicol.* **1988**, *10*, 369−384.
(44) Ferin, J.; Oberdörster, G.; Soderholm, S. C.; Gelein, R. Pulmonary tissue access of ultrafine particles. *J. Aerosol Med.* **1991**, *4* (1), 57−68.
(45) Donaldson, K.; Li, X. Y.; MacNee, W. Ultrafine (nanometre) particle mediated lung injury. *J. Aerosol Sci.* **1998**, *29* (5/6), 553−560.
(46) Brown, D. M.; Stone, V.; Findlay, P.; MacNee, W.; Donaldson, K. Increased inflammation and intracellular calcium caused by ultrafine carbon black is independent of transition metals or other soluble components. *Occup. Environ. Med.* **2000**, *57*, 685−691.
(47) Kreyling, W. G.; Semmler, M.; Möller, W. Dosimetry and toxicology of ultrafine particles. *J. Aerosol Med.* **2004**, *17* (2), 140−152.
(48) Li, X. Y.; Gilmour, P. S.; Donaldson, K.; MacNee, W. Free radical activity and pro-inflammatory effects of particulate air pollution (PM_{10}) in vivo and in vitro. *Thorax* **1996**, *51*, 1216−1222.
(49) Shuster-Meiseles, T.; Shafer, M. M.; Heo, J.; Pardo, M.; Antkiewicz, D. S.; Schauer, J. J.; Rudich, A.; Rudich, Y. ROS-generating/ARE-activating capacity of metals in roadway particulate matter deposited in urban environment. *Environ. Res.* **2016**, *146*, 252−262.
(50) Zhang, Y.; Demokritou, P.; Ryan, D. K.; Bello, D. Comprehensive assessment of short-lived ROS and H_2O_2 in laser emissions: Assessing the relative contribution of metal oxides and organic constituents. *Environ. Sci. Technol.* **2019**, *53*, 7574−7583.
(51) Pinkerton, K. E.; Joad, J. P. Influence of air pollution on respiratory health during perinatal development. *Clin. Exp. Pharmacol. Physiol.* **2006**, *33*, 269−272.
(52) Calderón-Garcidueñas, L.; Serrano-Sierra, A.; Torres-Jardón, R.; Zhu, H.; Yuan, Y.; Smith, D.; Delgado-Chávez, R.; Cross, J. V.; Medina-Cortina, H.; Kavanaugh, M.; Guilarte, T. R. The impact of environmental metals in young urbanities' brains. *Exp. Toxicol. Pathol.* **2013**, *65* (5), 503−511.
(53) Calderón-Garcidueñas, L.; Reynoso-Robles, R.; Vargas-Martínez, J.; Gómez-Maqueo-Chew, A.; Pérez-Guillé, B.; Mukherjee, P. S.; Torres-Jardón, R.; Perry, G.; Gónzalez-Maciel, A. Prefrontal white matter pathology in air pollution exposed Mexico City young urbanities and their potential impact on neurovascular unit dysfunction and the development of Alzheimer's disease. *Environ. Res.* **2016**, *146*, 404−417.
(54) Mohai, P.; Kweon, B.-S.; Lee, S.; Ard, K. Air pollution around schools is linked to poorer student health and academic performance. *Health Aff* **2011**, *30* (5), 852−862.
(55) Sunyer, J.; Esnaola, M.; Alvarez-Pedrerol, M.; Forns, J.; Rivas, I.; López-Vicente, M.; Suades-González, E.; Foraster, M.; Garcia-

DOI: 10.1021/acs.est.9b01505
Environ. Sci. Technol. 2019, 53, 9970−9991

Esteban, R.; Basagaña, X.; Viana, M.; Cirach, M.; Moreno, T.; Alastuey, A.; Sebastian-Galles, N.; Nieuwenhuijsen, M.; Querol, X. Association between traffic-related air pollution in schools and cognitive development in primary school children: A prospective cohort study. *PLoS Med.* **2015**, *12* (3), No. e1001792.

(56) Sunyer, J.; Suades-González, E.; García-Esteban, R.; Rivas, I.; Pujol, J.; Alvarez-Pedrerol, M.; Forns, J.; Querol, X.; Basagaña, X. Traffic-related air pollution and attention in primary school children. *Epidemiology* **2017**, *28* (2), 181−189.

(57) Bateson, T. F.; Schwartz, J. Children's response to air pollution. *J. Toxicol. Environ. Health A* **2007**, *71*, 238−243.

(58) Sunyer, J. The neurological effects of air pollution in children. *Eur. Respir. J.* **2008**, *32*, 535−537.

(59) Burtscher, H.; Schüepp, K. The occurrence of ultrafine particles in the specific environment of children. *Paediatr. Respir. Rev.* **2012**, *13*, 89−94.

(60) Pieters, N.; Koppen, G.; van Poppel, M.; de Prins, S.; Cox, B.; Dons, E.; Nelen, V.; Int Panis, L.; Plusquin, M.; Schoeters, G.; Nawrot, T. S. Blood pressure and same-day exposure to air pollution at school: Associations with nano-sized to coarse PM in children. *Environ. Health Perspect.* **2015**, *123* (7), 737−742.

(61) Cacciottolo, M.; Wang, X.; Driscoll, I.; Woodward, N.; Saffari, A.; Reyes, J.; Serre, M. L.; Vizuete, W.; Sioutas, C.; Morgan, T. E.; Gatz, M.; Chui, H. C.; Shumaker, S. A.; Resnick, S. M.; Espeland, M. A.; Finch, C. E.; Chen, J. C. Particulate air pollutants, APOE alleles and their contributions to cognitive impairment in older women and to amyloidogenesis in experimental models. *Transl. Psychiatry* **2017**, *7*, No. e1022.

(62) Li, X.; Huang, S.; Jiao, A.; Yang, X.; Yun, J.; Wang, Y.; Xue, X.; Chu, Y.; Liu, F.; Liu, Y.; Ren, M.; Chen, X.; Li, N.; Lu, Y.; Mao, Z.; Tian, L.; Xiang, H. Association between ambient fine particulate matter and preterm birth or term low birth weight: An updated systematic review and meta-analysis. *Environ. Pollut.* **2017**, *227*, 596−605.

(63) Wang, X.; Ding, H.; Ryan, L.; Xu, X. Association between air pollution and low birth weight: A community-based study. *Environ. Health Perspect.* **1997**, *105* (5), 514−520.

(64) Lee, B. E.; Ha, E. H.; Park, H. S.; Kim, Y. J.; Hong, Y. C.; Kim, H.; Lee, J. T. Exposure to air pollution during different gestational phases contributes to risks of low birth weight. *Hum. Reprod.* **2003**, *18* (3), 638−643.

(65) Bell, M. L.; Ebisu, K.; Belanger, K. Ambient air pollution and low birth weight in Connecticut and Massachusetts. *Environ. Health Perspect.* **2007**, *115* (7), 1118−1124.

(66) Gauderman, W. J.; Vora, H.; McConnell, R.; Berhane, K.; Gilliland, F.; Thomas, D.; Lurmann, F.; Avol, E.; Kunzli, N.; Jerrett, M.; Peters, J. Effect of exposure to traffic on lung development from 10 to 18 years of age: a cohort study. *Lancet* **2007**, *369*, 571−577.

(67) Ginsberg, G. L.; Perkovich Foos, B.; Firestone, M. P. Review and analysis of inhalation dosimetry methods for application to children's risk assessment. *J. Toxicol. Environ. Health, Part A* **2005**, *68*, 573−615.

(68) Smith, M. A.; Harris, P. L. R.; Sayre, L. M.; Perry, G. Iron accumulation in Alzheimer disease is a source of redox-generated free radicals. *Proc. Natl. Acad. Sci. U. S. A.* **1997**, *94*, 9866−9868.

(69) Swaiman, K. F. Hallervorden-Spatz syndrome and brain iron metabolism. *Arch. Neurol.* **1991**, *48*, 1285−1293.

(70) Beard, J. L.; Connor, J. R.; Jones, B. C. Iron in the brain. *Nutr. Rev.* **1993**, *51* (6), 157−170.

(71) Rouault, T. A. Systemic iron metabolism: A review and implications for brain iron metabolism. *Pediat. Neurol.* **2001**, *25* (2), 130−137.

(72) Dobson, J. Magnetic iron compounds in neurological disorders. *Ann. N. Y. Acad. Sci.* **2004**, *1012*, 183−192.

(73) Zecca, L.; Youdim, M. B. H.; Riederer, P.; Connor, J. R.; Crichton, R. R. Iron, brain ageing and neurodegenerative disorders. *Nat. Rev. Neurosci.* **2004**, *5*, 863−873.

(74) Castellani, R. J.; Moreira, P. I.; Liu, G.; Dobson, J.; Perry, G.; Smith, M. A.; Zhu, X. Iron: The redox-active center of oxidative stress in Alzheimer disease. *Neurochem. Res.* **2007**, *32*, 1640−1645.

(75) Chen, J.; Zheng, H.; Wang, W.; Liu, H.; Lu, L.; Bao, L.; Ren, L. Resuspension method for road surface dust collection and aerodynamic size distribution characterization. *China Particuol.* **2006**, *4* (6), 300−303.

(76) Muxworthy, A. R.; Schmidbauer, E.; Petersen, N. Magnetic properties and Mössbauer spectra of urban atmospheric particulate matter: a case study from Munich, Germany. *Geophys. J. Int.* **2002**, *150*, 558−570.

(77) Halsall, C. J.; Maher, B. A.; Karloukovski, V. V.; Shah, P.; Watkins, S. J. A novel approach to investigating indoor/outdoor pollution links: Combined magnetic and PAH measurements. *Atmos. Environ.* **2008**, *42*, 8902−8909.

(78) Maher, B. A.; Moore, C.; Matzka, J. Spatial variation in vehicle-derived metal pollution identified by magnetic and elemental analysis of roadside tree leaves. *Atmos. Environ.* **2008**, *42*, 364−373.

(79) Mitchell, R.; Maher, B. A. Evaluation and application of biomagnetic monitoring of traffic-derived particulate pollution. *Atmos. Environ.* **2009**, *43*, 2095−2103.

(80) Hansard, R.; Maher, B. A.; Kinnersley, R. Biomagnetic monitoring of industry-derived particulate pollution. *Environ. Pollut.* **2011**, *159*, 1673−1681.

(81) Buseck, P. R.; Adachi, K. Nanoparticles in the atmosphere. *Elements* **2008**, *4*, 389−394.

(82) Maher, B. A. Rain and dust: Magnetic records of climate and pollution. *Elements* **2009**, *5*, 229−234.

(83) Morawska, L.; He, C.; Johnson, G.; Jayaratne, R.; Salthammer, T.; Wang, H.; Uhde, E.; Bostrom, T.; Modini, R.; Ayoko, G.; McGarry, P.; Wensing, M. An investigation into the characteristics and formation mechanisms of particles originating from the operation of laser printers. *Environ. Sci. Technol.* **2009**, *43*, 1015−1022.

(84) Zhang, H.; Wang, S.; Hao, J.; Wan, L.; Jiang, J.; Zhang, M.; Mestl, H. E. S.; Alnes, L. W. H.; Aunan, K.; Mellouki, A. W. Chemical and size characterization of particles emitted from the burning of coal and wood in rural households in Guizhou, China. *Atmos. Environ.* **2012**, *51*, 94−99.

(85) Kumar, P.; Pirjola, L.; Ketzel, M.; Harrison, R. M. Nanoparticle emissions from 11 non-vehicle exhaust sources - A review. *Atmos. Environ.* **2013**, *67*, 252−277.

(86) Zajzon, N.; Márton, E.; Sipos, P.; Kristály, F.; Németh, T.; Kis-Kovács, V.; Weiszburg, T. G. Integrated mineralogical and magnetic study of magnetic airborne particles from potential pollution sources in industrial-urban environment. *Carpath. J. Earth Env.* **2013**, *8* (1), 179−186.

(87) Jeleńska, M.; Górka-Kostrubiec, B.; Werner, T.; Kądziałko-Hofmokl, M.; Szczepaniak-Wnuk, I.; Gonet, T.; Szwarczewski, P. Evaluation of indoor/outdoor urban air pollution by magnetic, chemical and microscopic studies. *Atmos. Pollut. Res.* **2017**, *8*, 754−766.

(88) Shi, J. P.; Khan, A. A.; Harrison, R. M. Measurements of ultrafine particle concentration and size distribution in the atmosphere. *Sci. Total Environ.* **1999**, *235*, 51−64.

(89) Cass, G. R.; Hughes, L. A.; Bhave, P.; Kleeman, M. J.; Allen, J. O.; Salmon, L. G. The chemical composition of atmospheric ultrafine particles. *Philos. Trans. R. Soc., A* **2000**, *358*, 2581−2592.

(90) Harrison, R. M.; Shi, J. P.; Xi, S.; Khan, A.; Mark, D.; Kinnersley, R.; Yin, J. Measurement of number, mass and size distribution of particles in the atmosphere. *Philos. Trans. R. Soc., A* **2000**, *358*, 2567−2580.

(91) Popovicheva, O.; Kireeva, E.; Shonija, N.; Zubareva, N.; Persiantseva, N.; Tishkova, V.; Demirdjian, B.; Moldanová, J.; Mogilnikov, V. Ship particulate pollutants: Characterization in terms of environmental implication. *J. Environ. Monit.* **2009**, *11*, 2077−2086.

(92) Hansard, R.; Maher, B. A.; Kinnersley, R. P. Rapid magnetic biomonitoring and differentiation of atmospheric particulate pollutants at the roadside and around two major industrial sites in the U.K. *Environ. Sci. Technol.* **2012**, *46*, 4403−4410.

DOI: 10.1021/acs.est.9b01505
Environ. Sci. Technol. 2019, 53, 9970−9991

(93) Mazaheri, M.; Johnson, G. R.; Morawska, L. An inventory of particle and gaseous emissions from large aircraft thrust engine operations at an airport. *Atmos. Environ.* **2011**, *45*, 3500−3507.
(94) Moreno, T.; Martins, V.; Querol, X.; Jones, T.; BéruBé, K.; Minguillón, M. C.; Amato, F.; Capdevila, M.; de Miguel, E.; Centelles, S.; Gibbons, W. A new look at inhalable metalliferous airborne particles on rail subway platforms. *Sci. Total Environ.* **2015**, *505*, 367−375.
(95) Peikertova, P.; Filip, P. Influence of the automotive brake wear debris on the environment - A review of recent research. *SAE Int. J. Mater. Manf.* **2016**, *9* (1), 133−146.
(96) Kukutschová, J.; Filip, P. Review of brake wear emissions: A review of brake emission measurement studies: Identification of gaps and future needs. In *Non-exhaust emissions*; Amato, F., Ed.; Academic Press, 2018; pp 123−146.
(97) Abdul-Razzaq, W.; Gautam, M. Discovery of magnetite in the exhausted material from a diesel engine. *Appl. Phys. Lett.* **2001**, *78* (14), 2018−2019.
(98) Lee, D.; Miller, A.; Kittelson, D.; Zachariah, M. R. Characterization of metal-bearing diesel nanoparticles using single-particle mass spectrometry. *J. Aerosol Sci.* **2006**, *37*, 88−110.
(99) Gustafsson, M.; Blomqvist, G.; Gudmundsson, A.; Dahl, A.; Swietlicki, E.; Bohgard, M.; Lindbom, J.; Ljungman, A. Properties and toxicological effects of particles from the interaction between tires, road pavement and winter traction material. *Sci. Total Environ.* **2008**, *393*, 226−240.
(100) Demirdjian, B.; Ferry, D.; Suzanne, J.; Popovicheva, O. B.; Persiantseva, N. M.; Shonija, N. K. Heterogeneities in the microstructure and composition of aircraft engine combustor soot: Impact on the water uptake. *J. Atmos. Chem.* **2006**, *56*, 83−103.
(101) Harris, S. J.; Maricq, M. M. Signature size distributions for diesel and gasoline engine exhaust particulate matter. *J. Aerosol Sci.* **2001**, *32*, 749−764.
(102) Kam, W.; Liacos, J. W.; Schauer, J. J.; Delfino, R. J.; Sioutas, C. Size-segregated composition of particulate matter (PM) in major roadways and surface streets. *Atmos. Environ.* **2012**, *55*, 90−97.
(103) Keuken, M. P.; Henzing, J. S.; Zandveld, P.; van den Elshout, S.; Karl, M. Dispersion of particle numbers and elemental carbon from road traffic, a harbour and an airstrip in the Netherlands. *Atmos. Environ.* **2012**, *54*, 320−327.
(104) Lombaert, K.; Morel, S.; Le Moyne, L.; Adam, P.; Tardieu de Maleissye, J.; Amouroux, J. Nondestructive analysis of metallic elements in diesel soot collected on filter: Benefits of laser induced breakdown spectroscopy. *Plasma Chem. Plasma Process.* **2004**, *24* (1), 41−56.
(105) Brook, J. R.; Graham, L.; Charland, J. P.; Cheng, Y.; Fan, X.; Lu, G.; Li, S. M.; Lillyman, C.; MacDonald, P.; Caravaggio, G.; MacPhee, J. A. Investigation of the motor vehicle exhaust contribution to primary fine particle organic carbon in urban air. *Atmos. Environ.* **2007**, *41*, 119−135.
(106) Robert, M. A.; VanBergen, S.; Kleeman, M. J.; Jakober, C. A. Size and composition distributions of particulate matter emissions: Part 1 - Light-duty gasoline vehicles. *J. Air Waste Manage. Assoc.* **2007**, *57*, 1414−1428.
(107) Fulper, C. R.; Kishan, S.; Baldauf, R. W.; Sabisch, M.; Warila, J.; Fujita, E. M.; Scarbro, C.; Crews, W. S.; Snow, R.; Gabele, P.; Santos, R.; Tierney, E.; Cantrell, B. Methods of characterizing the distribution of exhaust emissions from light-duty, gasoline-powered motor vehicles in the U.S. fleet. *J. Air Waste Manage. Assoc.* **2010**, *60*, 1376−1387.
(108) Peltier, R. E.; Cromar, K. R.; Ma, Y.; Fan, Z.-H.; Lippmann, M. Spatial and seasonal distribution of aerosol chemical components in New York City: (2) Road dust and other tracers of traffic-generated air pollution. *J. Exposure Sci. Environ. Epidemiol.* **2011**, *21*, 484−494.
(109) Pant, P.; Harrison, R. M. Estimation of the contribution of road traffic emissions to particulate matter concentrations from field measurements: A review. *Atmos. Environ.* **2013**, *77*, 78−97.
(110) Rogge, W. F.; Hildemann, L. M.; Mazurek, M. A.; Cass, G. R.; Simoneit, B. R. T. Sources of fine organic aerosol. 3. Road dust, tire debris, and organometallic brake lining dust: Roads as sources and sinks. *Environ. Sci. Technol.* **1993**, *27*, 1892−1904.
(111) Lough, G. C.; Schauer, J. J.; Park, J.-S.; Shafer, M. M.; Deminter, J. T.; Weinstein, J. P. Emissions of metals associated with motor vehicle roadways. *Environ. Sci. Technol.* **2005**, *39*, 826−836.
(112) Alam, M. S.; Zeraati-Rezaei, S.; Stark, C. P.; Liang, Z.; Xu, H.; Harrison, R. M. The characterisation of diesel exhaust particles - composition, size distribution and partitioning. *Faraday Discuss.* **2016**, *189*, 69−84.
(113) Su, D. S.; Müller, J.-O.; Jentoft, R. E.; Rothe, D.; Jacob, E.; Schlögl, R. Fullerene-like soot from Euro IV diesel engine: consequences for catalytic automotive pollution control. *Top. Catal.* **2004**, *30/31*, 241−245.
(114) Liati, A.; Spiteri, A.; Dimopoulos Eggenschwiler, P.; Vogel-Schäuble, N. Microscopic investigation of soot and ash particulate matter derived from biofuel and diesel: implications for the reactivity of soot. *J. Nanopart. Res.* **2012**, *14* (11), 1224.
(115) Maricq, M. M. Chemical characterization of particulate emissions from diesel engines: A review. *Aerosol Sci.* **2007**, *38*, 1079−1118.
(116) Wang, Y.-F.; Huang, K.-L.; Li, C.-T.; Mi, H.-H.; Luo, J.-H.; Tsai, P.-J. Emissions of fuel metals content from a diesel vehicle engine. *Atmos. Environ.* **2003**, *37*, 4637−4643.
(117) Sharma, M.; Agarwal, A. K.; Bharathi, K. V. L. Characterization of exhaust particulates from diesel engine. *Atmos. Environ.* **2005**, *39*, 3023−3028.
(118) Prichard, H. M.; Fisher, P. C. Identification of platinum and palladium particles emitted from vehicles and dispersed into the surface environment. *Environ. Sci. Technol.* **2012**, *46*, 3149−3154.
(119) Kumar, P.; Fennell, P.; Langley, D.; Britter, R. Pseudo-simultaneous measurements for the vertical variation of coarse, fine and ultrafine particles in an urban street canyon. *Atmos. Environ.* **2008**, *42*, 4304−4319.
(120) Ondráček, J.; Schwarz, J.; Ždímal, V.; Andělová, L.; Vodička, P.; Bízek, V.; Tsai, C.-J.; Chen, S.-C.; Smolík, J. Contribution of the road traffic to air pollution in the Prague city (busy speedway and suburban crossroads). *Atmos. Environ.* **2011**, *45*, 5090−5100.
(121) Bagley, S. T.; Baumgard, K. J.; Gratz, L. D.; Johnson, J. H.; Leddy, D. G. Characterization of fuel and aftertreatment device effects on diesel emissions. *Res. Rep. Health Eff. Inst.* **1996**, *76*, 1−75.
(122) Harris, S. J.; Maricq, M. M. Signature size distributions for diesel and gasoline engine exhaust particulate matter. *J. Aerosol Sci.* **2001**, *32*, 749−764.
(123) El Haddad, I.; Marchand, N.; Dron, J.; Temime-Roussel, B.; Quivet, E.; Wortham, H.; Jaffrezo, J. L.; Baduel, C.; Voisin, D.; Besombes, J. L.; Gille, G. Comprehensive primary particulate organic characterization of vehicular exhaust emissions in France. *Atmos. Environ.* **2009**, *43*, 6190−6198.
(124) Karjalainen, P.; Pirjola, L.; Heikkilä, J.; Lähde, T.; Tzamkiozis, T.; Ntziachristos, L.; Keskinen, J.; Rönkkö, T. Exhaust particles of modern gasoline vehicles: A laboratory and on-road study. *Atmos. Environ.* **2014**, *97*, 262−270.
(125) Imhof, D.; Weingartner, E.; Prévôt, A. S. H.; Ordóñez, C.; Kurtenbach, R.; Wiesen, P.; Rodler, J.; Sturm, P.; McCrae, I.; Ekström, M.; Baltensperger, U. Aerosol and NO_x emission factors and submicron particle number size distributions in two road tunnels with different traffic regimes. *Atmos. Chem. Phys.* **2006**, *6*, 2215−2230.
(126) Reche, C.; Querol, X.; Alastuey, A.; Viana, M.; Pey, J.; Moreno, T.; Rodriguez, S.; González, Y.; Fernández-Camacho, R.; Sánchez de la Campa, A. M.; de la Rosa, J.; Dall'Osto, M.; Prévôt, A. S. H.; Hueglin, C.; Harrison, R. M.; Quincey, P. New considerations for PM, black carbón and particle number concentration for air quality monitoring across different European cities. *Atmos. Chem. Phys.* **2011**, *11*, 6207−6227.
(127) Zhu, Y.; Hinds, W. C.; Kim, S.; Sioutas, C. Concentration and size distribution of ultrafine particles near a major highway. *J. Air & Waste Manage. Assoc.* **2002**, *52*, 1032−1042.
(128) Puustinen, A.; Hämeri, K.; Pekkanen, J.; Kulmala, M.; de Hartog, J.; Meliefste, K.; ten Brink, H.; Kos, G.; Katsouyanni, K.;

DOI: 10.1021/acs.est.9b01505
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Karakatsani, A.; Kotronarou, A.; Kavouras, I.; Meddings, C.; Thomas, S.; Harrison, R.; Ayres, J. G.; van der Zee, S.; Hoek, G. Spatial variation of particle number and mass over four European cities. *Atmos. Environ.* **2007**, *41*, 6622−6636.

(129) Cyrys, J.; Pitz, M.; Heinrich, J.; Wichmann, H.-E.; Peters, A. Spatial and temporal variation of particle number concentration in Augsburg, Germany. *Sci. Total Environ.* **2008**, *401*, 168−175.

(130) Rose, D.; Wehner, B.; Ketzel, M.; Engler, C.; Voigtländer, J.; Tuch, T.; Wiedensohler, A. Atmospheric number size distributions of soot particles and estimation of emission factors. *Atmos. Chem. Phys.* **2006**, *6*, 1021−1031.

(131) Beddows, D. C. S.; Dall'Osto, M.; Harrison, R. M. Cluster analysis of rural, urban and curbside atmospheric particle size data. *Environ. Sci. Technol.* **2009**, *43*, 4694−4700.

(132) HEI Panel on the health effects of traffic-related air pollution. *Traffic-related air pollution: A critical review of the literature on emissions, exposure, and health effects*; HEI Special Report, 17; Health Effects Institute: Boston, MA, 2010.

(133) Ranft, U.; Schikowski, T.; Sugiri, D.; Krutmann, J.; Krämer, U. Long-term exposure to traffic-related particulate matter impairs cognitive function in the elderly. *Environ. Res.* **2009**, *109*, 1004−1011.

(134) Verma, P. C.; Alemani, M.; Gialanella, S.; Lutterotti, L.; Olofsson, U.; Straffelini, G. Wear debris from brake system materials: A multi-analytical characterization approach. *Tribol. Int.* **2016**, *94*, 249−259.

(135) Foitzik, M.-J.; Unrau, H.-J.; Gauterin, F.; Dörnhöfer, J.; Koch, T. Investigation of ultra fine particulate matter emission of rubber tires. *Wear* **2018**, *394−395*, 87−95.

(136) Liati, A.; Schreiber, D.; Dimopoulos Eggenschwiler, P.; Dasilva, Y. A. R. Metal particle emissions in the exhaust stream of diesel engines: An electron microscopy study. *Environ. Sci. Technol.* **2013**, *47*, 14495−14501.

(137) Liati, A.; Pandurangi, S. S.; Boulouchos, K.; Schreiber, D.; Arroyo Rojas Dasilva, Y. Metal nanoparticles in diesel exhaust derived by in-cylinder melting of detached engine fragments. *Atmos. Environ.* **2015**, *101*, 34−40.

(138) Liati, A.; Schreiber, D.; Arroyo Rojas Dasilva, Y.; Dimopoulos Eggenschwiler, P. Ultrafine particle emissions from modern gasoline and diesel vehicles: An electron microscopic perspective. *Environ. Pollut.* **2018**, *239*, 661−669.

(139) Miller, A.; Ahlstrand, G.; Kittelson, D.; Zachariah, M. The fate of metal (Fe) during diesel combustion: Morphology, chemistry, and formation pathways of nanoparticles. *Combust. Flame* **2007**, *149*, 129−143.

(140) Braun, A.; Huggins, F. E.; Kelly, K. E.; Mun, B. S.; Ehrlich, S. N.; Huffman, G. P. Impact of ferrocene on the structure of diesel exhaust soot as probed with wide-angle X-ray scattering and C(1s) NEXAFS spectroscopy. *Carbon* **2006**, *44*, 2904−2911.

(141) Filippelli, G. M.; Laidlaw, M. A. S.; Latimer, J. C.; Raftis, R. Urban lead poisoning and medical geology: Un unfinished story. *GSA Today* **2005**, *15* (1), 4−11.

(142) Mehran, Q. M.; Fazal, M. A.; Bushroa, A. R.; Rubaiee, S. A critical review on physical vapour deposition coatings applied on different engine components. *Crit. Rev. Solid State Mater. Sci.* **2018**, *43* (2), 158−175.

(143) Sugamata, M.; Ihara, T.; Takano, H.; Oshio, S.; Takeda, K. Maternal diesel exhaust exposure damages newborn murine brains. *J. Health Sci.* **2006**, *52* (1), 82−84.

(144) Ema, M.; Naya, M.; Horimoto, M.; Kato, H. Developmental toxicity of diésel exhaust: A review of studies in experimental animals. *Reprod. Toxicol.* **2013**, *42*, 1−17.

(145) Li, C.; Li, X.; Suzuki, A. K.; Zhang, Y.; Fujitani, Y.; Nagaoka, K.; Watanabe, G.; Taya, K. Effects of exposure to nanoparticle-rich diesel exhaust on pregnancy in rats. *J. Reprod. Dev.* **2013**, *59* (2), 145−150.

(146) Brito, J. M.; Belotti, L.; Toledo, A. C.; Antonangelo, L.; Silva, F. S.; Alvim, D. S.; Andre, P. A.; Saldiva, P. H. N.; Rivero, D. H. R. F. Acute cardiovascular and inflammatory toxicity induced by inhalation of diesel and biodiesel exhaust particles. *Toxicol. Sci.* **2010**, *116* (1), 67−78.

(147) Künzi, L.; Krapf, M.; Daher, N.; Dommen, J.; Jeannet, N.; Schneider, S.; Platt, S.; Slowik, J. G.; Baumlin, N.; Salathe, M.; Prévôt, A. S. H.; Kalberer, M.; Strähl, C.; Dümbgen, L.; Sioutas, C.; Baltensperger, U.; Geiser, M. Toxicity of aged gasoline exhaust particles to normal and diseased airway epithelia. *Sci. Rep.* **2015**, *5* (1), 11801.

(148) Cheung, K. L.; Ntziachristos, L.; Tzamkiozis, T.; Schauer, J. J.; Samaras, Z.; Moore, K. F.; Sioutas, C. Emissions of particulate trace elements, metals and organic species from gasoline, diesel, and biodiesel passenger vehicles and their relation to oxidative stress. *Aerosol Sci. Technol.* **2010**, *44* (7), 500−513.

(149) Garg, B. D.; Cadle, S. H.; Mulawa, P. A.; Groblicki, P. J.; Laroo, C.; Parr, G. A. Brake wear particulate matter emissions. *Environ. Sci. Technol.* **2000**, *34* (21), 4463−4469.

(150) Sanders, P. G.; Xu, N.; Dalka, T. M.; Maricq, M. M. Airborne brake wear debris: Size distributions, composition, and a comparison of dynamometer and vehicle tests. *Environ. Sci. Technol.* **2003**, *37*, 4060−4069.

(151) Adachi, K.; Tainosho, Y. Characterization of heavy metal particles embedded in tire dust. *Environ. Int.* **2004**, *30*, 1009−1017.

(152) Kukutschová, J.; Moravec, P.; Tomášek, V.; Matějka, V.; Smolík, J.; Schwarz, J.; Seidlerová, J.; Šafářová, K.; Filip, P. On airborne nano/micro-sized wear particles released from low-metallic automotive brakes. *Environ. Pollut.* **2011**, *159*, 998−1006.

(153) Österle, W.; Deutsch, C.; Gradt, T.; Orts-Gil, G.; Schneider, T.; Dmitriev, A. I. Tribological screening tests for the selection of raw materials for automotive brake pad formulations. *Tribol. Int.* **2014**, *73*, 148−155.

(154) Kukutschová, J.; Roubíček, V.; Mašláň, M.; Jančík, D.; Slovák, V.; Malachová, K.; Pavlíčková, Z.; Filip, P. Wear performance and wear debris of semimetallic automotive brake materials. *Wear* **2010**, *268*, 86−93.

(155) Peikertová, P.; Kukutschová, J.; Vávra, I.; Matějka, V.; Životský, O.; Vaculík, M.; Lee, P. W.; Filip, P. Water suspended nanosized particles released from nonairborne brake wear debris. *Wear* **2013**, *306*, 89−96.

(156) Mosleh, M.; Blau, P. J.; Dumitrescu, D. Characteristics and morphology of wear particles from laboratory testing of disk brake materials. *Wear* **2004**, *256*, 1128−1134.

(157) Thorpe, A.; Harrison, R. M. Sources and properties of non-exhaust particulate matter from road traffic: A review. *Sci. Total Environ.* **2008**, *400*, 270−282.

(158) Hagino, H.; Oyama, M.; Sasaki, S. Laboratory testing of airborne brake wear particle emissions using a dynamometer system under urban city driving cycles. *Atmos. Environ.* **2016**, *131*, 269−278.

(159) Jang, H. Brake friction materials. In *Encyclopedia of Tribology*; Wang, Q. J., Chung, Y.-W., Eds.; Springer: New York, Heidelberg, Dordrecht, London, 2013; pp 263−273.

(160) Nosko, O.; Olofsson, U. Effective density of airborne wear particles from car brake materials. *J. Aerosol Sci.* **2017**, *107*, 94−106.

(161) Perricone, G.; Alemani, M.; Metinöz, I.; Matějka, V.; Wahlström, J.; Olofsson, U. Towards the ranking of airborne particle emissions from car brakes - a system approach. *Proc. Inst. Mech. Eng., Part D* **2017**, *231* (6), 781−797.

(162) Nosko, O.; Alemani, M.; Olofsson, U. Temperature effect on emission of airborne wear particles from car brakes. *Europe's Braking Conference and Exhibition*; EB2015-TEF-014; 2015.

(163) Nosko, O.; Vanhanen, J.; Olofsson, U. Emission of 1.3−10 nm airborne particles from brake materials. *Aerosol Sci. Technol.* **2017**, *51* (1), 91−96.

(164) Alemani, M.; Nosko, O.; Metinoz, I.; Olofsson, U. A study of emission of airborne wear particles from car brake friction pairs. *SAE Int. J. Mater. Manf.* **2016**, *9* (1), 147−157.

(165) Perricone, G.; Matějka, V.; Alemani, M.; Valota, G.; Bonfanti, A.; Ciotti, A.; Olofsson, U.; Söderberg, A.; Wahlström, J.; Nosko, O.; Straffelini, G.; Gialanella, S.; Metinoz, I. A concept for reducing PM_{10} emissions for car brakes by 50%. *Wear* **2018**, *369−397*, 135−145.

DOI: 10.1021/acs.est.9b01505
Environ. Sci. Technol. 2019, 53, 9970−9991

(166) Gasser, M.; Riediker, M.; Mueller, L.; Perrenoud, A.; Blank, F.; Gehr, P.; Rothen-Rutishauser, B. Toxic effects of brake wear particles on epithelial lung cells *in vitro*. *Pert. Fibre Toxicol.* **2009**, *6* (30), 30.
(167) Kukutschová, J.; Roubíček, V.; Malachová, K.; Pavlíčková, Z.; Holuša, R.; Kubačková, J.; Mička, V.; MacCrimmon, D.; Filip, P. Wear mechanism in automotive brake materials, wear debris and its potential environmental impact. *Wear* **2009**, *267*, 807−817.
(168) Zhao, J.; Lewinski, N.; Riediker, M. Physico-chemical characterization and oxidative reactivity evaluation of aged brake wear particles. *Aerosol Sci. Technol.* **2015**, *49*, 65−74.
(169) Kazimirova, A.; Peikertova, P.; Barancokova, M.; Staruchova, M.; Tulinska, J.; Vaculik, M.; Vavra, I.; Kukutschova, J.; Filip, P.; Dusinska, M. Automotive airborne brake wear debris nanoparticles and cytokinesis-block micronucleus assay in peripheral blood lymphocytes: A pilot study. *Environ. Res.* **2016**, *148*, 443−449.
(170) Malachova, K.; Kukutschova, J.; Rybkova, Z.; Sezimova, H.; Placha, D.; Cabanova, K.; Filip, P. Toxicity and mutagenicity of low-metallic automotive brake pad materials. *Ecotoxicol. Environ. Saf.* **2016**, *131*, 37−44.
(171) Barosova, H.; Chortarea, S.; Peikertova, P.; Clift, M. J. D.; Petri-Fink, A.; Kukutschova, J.; Rothen-Rutishauser, B. Biological response of an in vitro human 3D lung cell model exposed to brake wear debris varies based on brake pad formulation. *Arch. Toxicol.* **2018**, *92*, 2339−2351.
(172) Puisney, C.; Oikonomou, E. K.; Nowak, S.; Chevillot, A.; Casale, S.; Baeza-Squiban, A.; Berret, J.-F. Brake wear (nano)particle characterization and toxicity on wirway epithelial cells *in vitro*. *Environ. Sci.: Nano* **2018**, *5*, 1036−1044.
(173) Rajhelová, H.; Peikertová, P.; Čabanová, K.; Kuzníková, L.; Čech Barabaszová, K.; Mamulová Kutláková, K.; Vaculík, M.; Kukutschová, J. Determination of oxidative potential caused by brake wear debris in non-cellular systems. *J. Nanosci. Nanotechnol.* **2019**, *19*, 2869−2875.
(174) Gerlofs-Nijland, M. E.; Bokkers, B. G. H.; Sachse, H.; Reijnders, J. J. E.; Gustafsson, M.; Boere, A. J. F.; Fokkens, P. F. H.; Leseman, D. L. A. C.; Augsburg, K.; Cassee, F. R. Inhalation toxicity profiles of particulate matter: a comparison between brake wear with other sources of emission. *Inhal. Toxicol.* **2019**, *31*, 89−98.
(175) Gualtieri, M.; Andrioletti, M.; Mantecca, P.; Vismara, C.; Camatini, M. Impact of tire debris on *in vitro* and *in vivo* systems. *Part. Fibre Toxicol.* **2005**, *2* (1), 1.
(176) Lindbom, J.; Gustafsson, M.; Blomqvist, G.; Dahl, A.; Gudmundsson, A.; Swietlicki, E.; Ljungman, A. G. Exposure to wear particles generated from studded tires and pavement induces inflammatory cytokine release from human macrophages. *Chem. Res. Toxicol.* **2006**, *19*, 521−530.
(177) Lindbom, J.; Gustafsson, M.; Blomqvist, G.; Dahl, A.; Gudmundsson, A.; Swietlicki, E.; Ljungman, A. G. Wear particles generated from studded tires and pavement induces inflammatory reactions in mouse macrophage cells. *Chem. Res. Toxicol.* **2007**, *20*, 937−946.
(178) Wik, A.; Dave, G. Occurrence and effects of tire wear particles in the environment - A critical review and an initial risk assessment. *Environ. Pollut.* **2009**, *157*, 1−11.
(179) Milani, M.; Pucillo, F. P.; Ballerini, M.; Camatini, M.; Gualtieri, M.; Martino, S. First evidence of tire debris characterization at the nanoscale by focused ion beam. *Mater. Charact.* **2004**, *52*, 283−288.
(180) Reddy, C. M.; Quinn, J. G. Environmental chemistry of benzothiazoles derived from rubber. *Environ. Sci. Technol.* **1997**, *31*, 2847−2853.
(181) Camatini, M.; Crosta, G. F.; Dolukhanyan, T.; Sung, C.; Giuliani, G.; Corbetta, G. M.; Cencetti, S.; Regazzoni, C. Micro-characterization and identification of tire debris in heterogeneous laboratory and environmental specimens. *Mater. Charact.* **2001**, *46*, 271−283.
(182) Kumata, H.; Yamada, J.; Masuda, K.; Takada, H.; Sato, Y.; Sakurai, T.; Fujiwara, K. Benzothiazolamines as tire-derived molecular markers: Sorptive behavior in Street runoff and application to source apportioning. *Environ. Sci. Technol.* **2002**, *36*, 702−708.
(183) Allen, J. O.; Alexandrova, O.; Kaloush, K. E. *Tire wear emissions for asphalt rubber and Portland cement concrete pavement surfaces. Final report*; Arizona Dept. of Transportation: Phoenix, AZ, 2006.
(184) Hjortenkrans, D. S. T.; Bergbäck, B. G.; Häggerud, A. V. Metal emissions from brake linings and tires: Case studies of Stockholm, Sweden 1995/1998 and 2005. *Environ. Sci. Technol.* **2007**, *41*, 5224−5230.
(185) Dahl, A.; Gharibi, A.; Swietlicki, E.; Gudmundsson, A.; Bohgard, M.; Ljungman, A.; Blomqvist, G.; Gustafsson, M. Traffic-generated emissions of ultrafine particles from pavement-tire interface. *Atmos. Environ.* **2006**, *40*, 1314−1323.
(186) Kupiainen, K. J.; Tervahattu, H.; Räisänen, M.; Mäkelä, T.; Aurela, M.; Hillamo, R. Size and composition of airborne particles from pavement wear, tires, and traction sanding. *Environ. Sci. Technol.* **2005**, *39*, 699−706.
(187) Dall'Osto, M.; Beddows, D. C. S.; Gietl, J. K.; Olatunbosun, O. A.; Yang, X.; Harrison, R. M. Characteristics of tyre dust in polluted air: Studies by single particle mass spectrometry (ATOFMS). *Atmos. Environ.* **2014**, *94*, 224−230.
(188) Mathissen, M.; Scheer, V.; Vogt, R.; Benter, T. Investigation on the potential generation of ultrafine particles from the tire-road interface. *Atmos. Environ.* **2011**, *45*, 6172−6179.
(189) Hosiokangas, J.; Ruuskanen, J.; Pekkanen, J. Effects of soil dust episodes and mixed fuel sources on source apportionment of PM10 particles in Kuopio, Finland. *Atmos. Environ.* **1999**, *33*, 3821−3829.
(190) Evans, A.; Evans, R. The composition of a tyre: Typical components; *Report of The Waste & Resources Action Programme*; WRAP: Banbury, UK, 2006.
(191) Bukowiecki, N.; Gehrig, R.; Lienemann, P.; Hill, M.; Figi, R.; Buchmann, B.; Furger, M.; Richard, A.; Mohr, C.; Weimer, S.; Prévôt, A.; Baltensperger, U. *PM10 emission factors of abrasion particles from road traffic*; TRIMIS: Schweiz, Eidgenoss, 2009; p 1268.
(192) Martuzevicius, D.; Kliucininkas, L.; Prasauskas, T.; Krugly, E.; Kauneliene, V.; Strandberg, B. Resuspension of particulate matter and PAHs from street dust. *Atmos. Environ.* **2011**, *45*, 310−317.
(193) Chen, J.; Wang, W.; Liu, H.; Ren, L. Determination of road dust loadings and chemical characteristics using resuspension. *Environ. Monit. Assess.* **2012**, *184*, 1693−1709.
(194) Lawrence, S.; Sokhi, R.; Ravindra, K.; Mao, H.; Prain, H. D.; Bull, I. D. Source apportionment of traffic emissions of particulate matter using tunnel measurements. *Atmos. Environ.* **2013**, *77*, 548−557.
(195) Tanner, P. A.; Ma, H.-L.; Yu, P. K. N. Fingerprinting metals in urban street dust of Beijing, Shanghai, and Hong Kong. *Environ. Sci. Technol.* **2008**, *42*, 7111−7117.
(196) Duong, T. T. T.; Lee, B.-K. Determining contamination level of heavy metals in road dust from busy traffic areas with different characteristics. *J. Environ. Manage.* **2011**, *92*, 554−562.
(197) Xie, S.; Dearing, J. A.; Bloemendal, J.; Boyle, J. F. Association between the organic matter content and magnetic properties in street dust, Liverpool, UK. *Sci. Total Environ.* **1999**, *241*, 205−214.
(198) Xie, S.; Dearing, J. A.; Boyle, J. F.; Bloemendal, J.; Morse, A. P. Association between magnetic properties and element concentrations of Liverpool street dust and its implications. *J. Appl. Phys.* **2001**, *48*, 83−92.
(199) Goddu, S. R.; Appel, E.; Jordanova, D.; Wehland, F. Magnetic properties of road dust from Visakhapatnam (India) - relationship to industrial pollution and road traffic. *Phys. Chem. Earth* **2004**, *29*, 985−995.
(200) Yang, T.; Liu, Q.; Li, H.; Zeng, Q.; Chan, L. Anthropogenic magnetic particles and heavy metals in the road dust: Magnetic identification and its implications. *Atmos. Environ.* **2010**, *44*, 1175−1185.
(201) Bućko, M. S.; Magiera, T.; Johanson, B.; Petrovský, E.; Pesonen, L. J. Identification of magnetic particulates in road dust

DOI: 10.1021/acs.est.9b01505
Environ. Sci. Technol. 2019, 53, 9970−9991

accumulated on roadside snow using magnetic, geochemical and micro-morphological analyses. *Environ. Pollut.* **2011**, *159*, 1266−1276.
(202) Bućko, M. S.; Mattila, O.-P.; Chrobak, A.; Ziółkowski, G.; Johanson, B.; Čuda, J.; Filip, J.; Zbořil, R.; Pesonen, L. J.; Leppäranta, M. Distribution of magnetic particulates in a roadside snowpack based on magnetic, microstructural and mineralogical studies. *Geophys. J. Int.* **2013**, *195*, 159−175.
(203) Wawer, M.; Magiera, T.; Ojha, G.; Appel, E.; Bućko, M. S.; Kusza, G. Characteristics of current roadside pollution using test-monitoring plots. *Sci. Total Environ.* **2015**, *505*, 795−804.
(204) Dytłow, S.; Górka-Kostrubiec, B. Effective and universal tool for evaluating heavy metals - passive dust samplers. *Environ. Pollut.* **2019**, *247*, 188−194.
(205) Transport for London. https://tfl.gov.uk/modes/tube (accessed February 5, 2019).
(206) The New York City Subway System. https://www.ny.com/transportation/subways/ (accessed February 5, 2019).
(207) Tokyo's Metro Advertising Media. https://www.metro-ad.co.jp/en/ (accessed February 5, 2019).
(208) Klepeis, N. E.; Nelson, W. C.; Ott, W. R.; Robinson, J. P.; Tsang, A. M.; Switzer, P.; Behar, J. V.; Hern, S. C.; Engelmann, W. H. The National Human Activity Pattern Survey (NHAPS): a resource for assessing exposure to environmental pollutants. *J. Exposure Sci. Environ. Epidemiol.* **2001**, *11*, 231−252.
(209) Saksena, S.; Quang, T. N.; Nguyen, T.; Dang, P. N.; Flachsbart, P. Commuters' exposure to particulate matter and carbon monoxide in Hanoi, Vietnam. *Transp. Res. D* **2008**, *13*, 206−211.
(210) Knibbs, L. D.; Cole-Hunter, T.; Morawska, L. A review of commuter exposure to ultrafine particles and its health effects. *Atmos. Environ.* **2011**, *45*, 2611−2622.
(211) Moreno, T.; Querol, X.; Martins, V.; Minguillón, M. C.; Reche, C.; Ku, L. H.; Eun, H. R.; Ahn, K. H.; Capdevila, M.; de Miguel, E. Formation and alteration of airborne particles in the subway environment. *Environ. Sci.: Processes Impacts* **2017**, *19*, 59−64.
(212) Aarnio, P.; Yli-Tuomi, T.; Kousa, A.; Mäkelä, T.; Hirsikko, A.; Hämeri, K.; Räisänen, M.; Hillamo, R.; Koskentalo, T.; Jantunen, M. The concentrations and composition of and exposure to fine particles ($PM_{2.5}$) in the Helsinki subway system. *Atmos. Environ.* **2005**, *39*, 5059−5066.
(213) Seaton, A.; Cherrie, J.; Dennekamp, M.; Donaldson, K.; Hurley, J. F.; Tran, C. L. The London Underground: dust and hazards to health. *Occup. Environ. Med.* **2005**, *62*, 355−362.
(214) Abbasi, S.; Jansson, A.; Sellgren, U.; Olofsson, U. Particle emissions from rail traffic: A literature review. *Crit. Rev. Environ. Sci. Technol.* **2013**, *43*, 2511−2544.
(215) Ripanucci, G.; Grana, M.; Vicentini, L.; Magrini, A.; Bergamaschi, A. Dust in the underground railway tunnels of an Italian Town. *J. Occup. Environ. Hyg.* **2006**, *3* (1), 16−25.
(216) Kim, K. Y.; Kim, Y. S.; Roh, Y. M.; Lee, C. M.; Kim, C. N. Spatial distribution of particulate matter (PM_{10} and $PM_{2.5}$) in Seoul Metropolitan Subway stations. *J. Hazard. Mater.* **2008**, *154*, 440−443.
(217) Murruni, L. G.; Solanes, V.; Debray, M.; Kreiner, A. J.; Davidson, J.; Davidson, M.; Vázquez, M.; Ozafrán, M. Concentrations and elemental composition of particulate matter in the Buenos Aires underground system. *Atmos. Environ.* **2009**, *43*, 4577−4583.
(218) Raut, J.-C.; Chazette, P.; Fortain, A. Link between aerosol optical, microphysical and chemical measurements in an underground railway station in Paris. *Atmos. Environ.* **2009**, *43*, 860−868.
(219) Mugica-Álvarez, V.; Figueroa-Lara, J.; Romero-Romo, M.; Sepúlveda-Sánchez, J.; López-Moreno, T. Concentrations and properties of airborne particles in the Mexico City subway system. *Atmos. Environ.* **2012**, *49*, 284−293.
(220) Salma, I.; Pósfai, M.; Kovács, K.; Kuzmann, E.; Homonnay, Z.; Posta, J. Properties and sources of individual particles and some chemical species in the aerosol of a metropolitan underground railway station. *Atmos. Environ.* **2009**, *43*, 3460−3466.
(221) Gustafsson, M.; Blomqvist, G.; Swietlicki, E.; Dahl, A.; Gudmundsson, A. Inhalable railroad particles at ground level and subterranean stations - Physical and chemical properties and relation to train traffic. *Transp. Res. D* **2012**, *17*, 277−285.
(222) Midander, K.; Elihn, K.; Wallén, A.; Belova, L.; Karlsson, A.-K. B.; Wallinder, I. O. Characterisation of nano- and micron-sized airborne and collected subway particles, a multi-analytical approach. *Sci. Total Environ.* **2012**, *427−428*, 390−400.
(223) Bukowiecki, N.; Gehrig, R.; Hill, M.; Lienemann, P.; Zwicky, C. N.; Buchmann, B.; Weingartner, E.; Baltensperger, U. Iron, manganese and copper emitted by cargo and passenger trains in Zürich (Switzerland): Size-segregated mass concentrations in ambient air. *Atmos. Environ.* **2007**, *41*, 878−889.
(224) Gehrig, R.; Hill, M.; Lienemann, P.; Zwicky, C. N.; Bukowiecki, N.; Weingartner, E.; Baltensperger, U.; Buchmann, B. Contribution of railway traffic to local PM10 concentrations in Switzerland. *Atmos. Environ.* **2007**, *41*, 923−933.
(225) Abbasi, S.; Olander, L.; Larsson, C.; Olofsson, U.; Jansson, A.; Sellgren, U. A field test study of airborne wear particles from a running regional train. *Proc. IMechE* **2012**, *226* (F), 95−109.
(226) Kam, W.; Ning, Z.; Shafer, M. M.; Schauer, J. J.; Sioutas, C. Chemical characterization and redox potential of coarse and fine particulate matter (PM) in underground and ground-level rail systems of the Los Angeles Metro. *Environ. Sci. Technol.* **2011**, *45*, 6769−6776.
(227) Zhang, W.; Jiang, H.; Dong, C.; Yan, Q.; Yu, L.; Yu, Y. Magnetic and geochemical characterization of iron pollution in subway dusts in Shanghai, China. *Geochem. Geophys. Geosyst.* **2011**, *12*, Q06Z25.
(228) Jung, H.-J.; Kim, B.; Malek, M. A.; Koo, Y. S.; Jung, J. H.; Son, Y.-S.; Kim, J.-C.; Kim, H.; Ro, C.-U. Chemical speciation of size-segregated floor dusts and airborne magnetic particles collected at underground subway stations in Seoul, Korea. *J. Hazard. Mater.* **2012**, *213-214*, 331−340.
(229) Loxham, M.; Cooper, M. J.; Gerlofs-Nijland, M. E.; Cassee, F. R.; Davies, D. E.; Palmer, M. R.; Teagle, D. A. H. Physicochemical characterization of airborne particulate matter at a mainline underground railway station. *Environ. Sci. Technol.* **2013**, *47*, 3614−3622.
(230) Namgung, H.-G.; Kim, J.-B.; Woo, S.-H.; Park, S.; Kim, M.; Kim, M.-S.; Bae, G.-N.; Park, D.; Kwon, S.-B. Generation of nanoparticles from friction between railway brake disks and pads. *Environ. Sci. Technol.* **2016**, *50*, 3453−3461.
(231) Namgung, H.-G.; Kim, J. B.; Kim, M.-S.; Kim, M.; Park, S.; Woo, S.-H.; Bae, G.-N.; Park, D.; Kwon, S.-B. Size distribution analysis of airborne wear particles released by subway brake system. *Wear* **2017**, *372−373*, 169−176.
(232) Lee, Y.; Choi, K.; Jung, W.; Versoza, M. E.; Barabad, M. L. M.; Kim, T.; Park, D. Generation characteristics of nanoparticles emitted from subways in operation. *Aerosol Air Qual. Res.* **2018**, *18*, 2230−2239.
(233) Karlsson, H. L.; Nilsson, L.; Möller, L. Subway particles are more genotoxic than street particles and induce oxidative stress in cultured human lung cells. *Chem. Res. Toxicol.* **2005**, *18*, 19−23.
(234) Kang, S.; Hwang, H.; Park, Y.; Kim, H.; Ro, C.-U. Chemical compositions of subway particles in Seoul, Korea determined by a quantitative single particle analysis. *Environ. Sci. Technol.* **2008**, *42*, 9051−9057.
(235) Querol, X.; Moreno, T.; Karanasiou, A.; Reche, C.; Alastuey, A.; Viana, M.; Font, O.; Gil, J.; de Miguel, E.; Capdevila, M. Variability of levels and composition of PM_{10} and $PM_{2.5}$ in the Barcelona metro system. *Atmos. Chem. Phys.* **2012**, *12*, 5055−5076.
(236) Eom, H.-J.; Jung, H.-J.; Sobanska, S.; Chung, S.-G.; Son, Y.-S.; Kim, J.-C.; Sunwoo, Y.; Ro, C.-U. Iron speciation of airborne subway particles by the combined use of Energy Dispersive Electron Probe X-ray microanalysis and Raman microspectrometry. *Anal. Chem.* **2013**, *85*, 10424−10431.
(237) Tokarek, S.; Bernis, A. An example of particle concentration reduction in Parisian subway stations by electrostatic precipitation. *Environ. Technol.* **2006**, *27*, 1279−1287.
(238) Moreno, T.; de Miguel, E. Improving air quality in subway systems: An overview. *Environ. Pollut.* **2018**, *239*, 829−831.

DOI: 10.1021/acs.est.9b01505
Environ. Sci. Technol. 2019, 53, 9970−9991

(239) Lee, H.; Namgung, H.-G.; Kwon, S.-B. Effect of train velocity on the amount of airborne wear particles generated from wheel-rail contacts. *Wear* **2018**, *414−415*, 296−302.
(240) Karlsson, H. L.; Ljungman, A. G.; Lindbom, J.; Möller, L. Comparison of genotoxic and inflammatory effects of particles generated by wood combustion, a road simulator and collected from street and subway. *Toxicol. Lett.* **2006**, *165*, 203−211.
(241) Karlsson, H. L.; Holgersson, Å.; Möller, L. Mechanisms related to the genotoxicity of particles in the subway and from other sources. *Chem. Res. Toxicol.* **2008**, *21*, 726−731.
(242) Nieuwenhuijsen, M. J.; Gómez-Perales, J. E.; Colvile, R. N. Levels of particulate air pollution, its elemental composition, determinants and health effects in metro systems. *Atmos. Environ.* **2007**, *41*, 7995−8006.
(243) Janssen, N. A. H.; Yang, A.; Strak, M.; Steenhof, M.; Hellack, B.; Gerlofs-Nijland, M. E.; Kuhlbusch, T.; Kelly, F.; Harrison, R.; Brunekreef, B.; Hoek, G.; Cassee, F. Oxidative potential of particulate matter collected at sites with different source characteristics. *Sci. Total Environ.* **2014**, *472*, 572−581.
(244) Loxham, M.; Morgan-Walsh, R. J.; Cooper, M. J.; Blume, C.; Swindle, E. J.; Dennison, P. W.; Howarth, P. H.; Cassee, F. R.; Teagle, D. A. H.; Palmer, M. R.; Davies, D. E. The effects on bronchial epithelial mucociliary cultures of coarse, fine, and ultrafine particulate matter from an underground railway station. *Toxicol. Sci.* **2015**, *145* (1), 98−107.
(245) Moreno, T.; Kelly, F. J.; Dunster, C.; Oliete, A.; Martins, V.; Reche, C.; Minguillón, M. C.; Amato, F.; Capdevila, M.; de Miguel, E.; Querol, X. Oxidative potential of subway $PM_{2.5}$. *Atmos. Environ.* **2017**, *148*, 230−238.
(246) Mazaheri, M.; Bostrom, T. E.; Johnson, G. R.; Morawska, L. Composition and morphology of particle emissions from in-use aircraft during takeoff and landing. *Environ. Sci. Technol.* **2013**, *47*, 5235−5242.
(247) Streibel, T.; Schnelle-Kreis, J.; Czech, H.; Harndorf, H.; Jakobi, G.; Jokiniemi, J.; et al. Aerosol emissions of a ship diesel engine operated with diesel fuel of heavy fuel oil. *Environ. Sci. Pollut. Res.* **2017**, *24*, 10976−10991.
(248) Kardel, F.; Wuyts, K.; Maher, B. A.; Samson, R. Intra-urban spatial variation of magnetic particles: Monitoring via leaf saturation isothermal remanent magnetisation (SIRM). *Atmos. Environ.* **2012**, *55*, 111−120.
(249) Masiol, M.; Harrison, R. M. Aircraft engine exhaust emissions and other airport-related contributions to ambient air pollution: A review. *Atmos. Environ.* **2014**, *95*, 409−455.
(250) Lelièvre, C. P.; Magnani, G.; Ramaroson, R.; Gouriou, F.; Talbaut, M.; Fréjafon, E.; Schuermann, G.; Schaefer, K.; Cornier, I.; Emeis, S.; Vannier, F.; Paux, E.; Copalle, A. Air quality and engine emission at Paris CDG airport during AIRPUR field campaigns. In Air Pollution; Longhurst, J. W. S.; Brebbia, C. A., Eds.; *WIT Transactions on Ecology and the Environment*, 2006.
(251) Popovicheva, O. B.; Persiantseva, N. M.; Lukhovitskaya, E. F.; Shonija, N. K.; Zubareva, N. A.; Demirdjian, B.; Ferry, D.; Suzanne, J. Aircraft engine soot as contrail nuclei. *Geophys. Res. Lett.* **2004**, *31*, DOI: 10.1029/2003GL018888.
(252) Amato, F.; Moreno, T.; Pandolfi, M.; Querol, X.; Alastuey, A.; Delgado, A.; Pedrero, M.; Cots, N. Concentrations, sources and geochemistry of airborne particulate matter at a major European airport. *J. Environ. Monit.* **2010**, *12*, 854−862.
(253) Fine, P.; Polidori, A.; Teffera, S. General aviation airport air monitoring study; *Final Report for the U.S. Environmental Protection Agency, Region IX*; U.S. EPA: San Francisco, 2010.
(254) Petzold, A.; Ström, J.; Schröder, F. P.; Kärcher, B. Carbonaceous aerosol in jet engine exhaust: emission characteristics and implications for heterogeneous chemical reactions. *Atmos. Environ.* **1999**, *33*, 2689−2698.
(255) Westerdahl, D.; Fruin, S. A.; Fine, P. L.; Sioutas, C. The Los Angeles International Airport as a source of ultrafine particles and other pollutants to nearby communities. *Atmos. Environ.* **2008**, *42*, 3143−3155.
(256) Hu, S.; Fruin, S.; Kozawa, K.; Mara, S.; Winer, A. M.; Paulson, S. E. Aircraft emission impacts in a neighborhood adjacent to a general aviation airport in Southern California. *Environ. Sci. Technol.* **2009**, *43*, 8039−8045.
(257) Biswas, S.; Fine, P. M.; Geller, M. D.; Hering, S. V.; Sioutas, C. Performance evaluation of a recently developed water-based condensation particle counter. *Aerosol Sci. Technol.* **2005**, *39*, 419−427.
(258) Masiol, M.; Vu, T. V.; Beddows, D. C. S.; Harrison, R. M. Source apportionment of wide range particle size spectra and black carbon collected at the airport of Venice (Italy). *Atmos. Environ.* **2016**, *139*, 56−74.
(259) Masiol, M.; Harrison, R. M.; Vu, T. V.; Beddows, D. C. S. Sources of sub-micrometre particles near a major international airport. *Atmos. Chem. Phys.* **2017**, *17*, 12379−12403.
(260) Unal, A.; Hu, Y.; Chang, M. E.; Odman, M. T.; Russell, A. G. Airport related emissions and impacts on air quality: Application to the Atlanta International Airport. *Atmos. Environ.* **2005**, *39*, 5787−5798.
(261) Barrett, S. R. H.; Britter, R. E.; Waitz, I. A. Global mortality attributable to aircraft cruise emissions. *Environ. Sci. Technol.* **2010**, *44*, 7736−7742.
(262) Ramsden, J. J. An estimate of the chronic exposure to neurotoxins of a neurologically injured pilot. *J. Biol. Phys. Chem.* **2016**, *16* (2), 95−102.
(263) Cavallo, D.; Ursini, C. L.; Carelli, G.; Iavicoli, I.; Ciervo, A.; Perniconi, B.; Rondinone, B.; Gismondi, M.; Iavicoli, S. Occupational exposure in airport personnel: Characterization and evaluation of genotoxic and oxidative effects. *Toxicology* **2006**, *223*, 26−35.
(264) Schindler, B. K.; Weiss, T.; Schütze, A.; Koslitz, S.; Broding, H. C.; Bünger, J.; Brüning, T. Occupational exposure of air crews to tricresyl phosphate isomers and organophosphate flame retardants after fume events. *Arch. Toxicol.* **2013**, *87*, 645−648.
(265) Harrison, R. M.; Masiol, M.; Vardoulakis, S. Civil aviation, air pollution and human health. *Environ. Res. Lett.* **2015**, *10*, 041001.
(266) He, R.-W.; Shirmohammadi, F.; Gerlofs-Nijland, M. E.; Sioutas, C.; Cassee, F. R. Pro-inflammatory responses to $PM_{0.25}$ from airport and urban traffic emissions. *Sci. Total Environ.* **2018**, *640−641*, 997−1003.
(267) Saxe, H.; Larsen, T. Air pollution from ships in three Danish ports. *Atmos. Environ.* **2004**, *38*, 4057−4067.
(268) Healy, R. M.; Hellebust, S.; Kourtchev, I.; Allanic, A.; O'Connor, I. P.; Bell, J. M.; Healy, D. A.; Sodeau, J. R.; Wenger, J. C. Source apportionment of $PM_{2.5}$ in Cork Harbour, Ireland using a combination of single particle mass spectrometry and quantitative semi-continuous measurements. *Atmos. Chem. Phys.* **2010**, *10*, 9593−9613.
(269) Pandolfi, M.; Gonzalez-Castanedo, Y.; Alastuey, A.; de la Rosa, J. D.; Mantilla, E.; de la Campa, A. S.; Querol, X.; Pey, J.; Amato, F.; Moreno, T. Source apportionment of PM_{10} and $PM_{2.5}$ at multiple sites in the strait of Gibraltar by PMF: impact of shipping emissions. *Environ. Sci. Pollut. Res.* **2011**, *18*, 260−269.
(270) Zhang, F.; Chen, Y.; Tian, C.; Wang, X.; Huang, G.; Fang, Y.; Zong, Z. Identification and quantification of shipping emissions in Bohai Rim, China. *Sci. Total Environ.* **2014**, *497−498*, 570−577.
(271) Zhang, Y.; Yang, X.; Brown, R.; Yang, L.; Morawska, L.; Ristovski, Z.; Fu, Q.; Huang, C. Shipping emissions and their impacts on air quality in China. *Sci. Total Environ.* **2017**, *581−582*, 186−198.
(272) Corbett, J. J.; Koehler, H. W. Updated emissions from ocean shipping. *J. Geophys. Res.* **2003**, *108* (D20), 4650.
(273) Endresen, Ø; Sørgård, E.; Sundet, J. K.; Dalsøren, S. B.; Isaksen, I. S. A.; Berglen, T. F.; Gravir, G. Emission from international sea transportation and environmental impact. *J. Geophys. Res.* **2003**, *108* (D17), 4560.
(274) Eyring, V.; Isaksen, I. S. A.; Berntsen, T.; Collins, W. J.; Corbett, J. J.; Endresen, O.; Grainger, R. G.; Moldanova, J.; Schlager, H.; Stevenson, D. S. Transport impacts on atmosphere and climate: Shipping. *Atmos. Environ.* **2010**, *44*, 4735−4771.

DOI: 10.1021/acs.est.9b01505
Environ. Sci. Technol. 2019, 53, 9970−9991

(275) Isakson, J.; Persson, T. A.; Selin Lindgren, E. Identification and assessment of ship emissions and their effects in the harbour of Göteborg, Sweden. *Atmos. Environ.* **2001**, *35*, 3659−3666.
(276) Chen, G.; Huey, L. G.; Trainer, M.; Nicks, D.; Corbett, J.; Ryerson, T. An investigation of the chemistry of ship emission plumes during ITCT 2002. *J. Geophys. Res.* **2005**, *110* (D10), D10S90.
(277) González, Y.; Rodríguez, S.; Guerra García, J. C.; Trujillo, J. L.; García, R. Ultrafine particles pollution in urban coastal air due to ship emissions. *Atmos. Environ.* **2011**, *45*, 4907−4914.
(278) González, Y.; Rodríguez, S. A comparative study on the ultrafine particle episodes induced by vehicle exhaust: A crude oil refinery and ship emissions. *Atmos. Res.* **2013**, *120−121*, 43−54.
(279) Juwono, A. M.; Johnson, G. R.; Mazaheri, M.; Morawska, L.; Roux, F.; Kitchen, B. Investigation of the airborne submicrometer particles emitted by dredging vessels using a plume capture method. *Atmos. Environ.* **2013**, *73*, 112−123.
(280) Agrawal, H.; Malloy, Q. G. J.; Welch, W. A.; Miller, J. W.; Cocker, D. R., III. In-use gaseous and particulate matter emissions from a modern ocean going container vessel. *Atmos. Environ.* **2008**, *42*, 5504−5510.
(281) Healy, R. M.; O'Connor, I. P.; Hellebust, S.; Allanic, A.; Sodeau, J. R.; Wenger, J. C. Characterisation of single particles from in-port ship emissions. *Atmos. Environ.* **2009**, *43*, 6408−6414.
(282) Moldanová, J.; Fridell, E.; Popovicheva, O.; Demirdjian, B.; Tishkova, V.; Faccinetto, A.; Focsa, C. Characterisation of particulate matter and gaseous emissions from a large ship diesel engine. *Atmos. Environ.* **2009**, *43*, 2632−2641.
(283) Moldanová, J.; Fridell, E.; Winnes, H.; Holmin-Fridell, S.; Boman, J.; Jedynska, A.; Tishkova, V.; Demirdjian, B.; Joulie, S.; Bladt, H.; Ivleva, N. P.; Niessner, R. Physical and chemical characterisation of PM emissions from two ships operating in European Emission Control Areas. *Atmos. Meas. Tech.* **2013**, *6*, 3577−3596.
(284) Popovicheva, O.; Kireeva, E.; Persiantseva, N.; Timofeev, M.; Bladt, H.; Ivleva, N. P.; Niessner, R.; Moldanová, J. Microscopic characterization of individual particles from multicomponent ship exhaust. *J. Environ. Monit.* **2012**, *14*, 3101−3110.
(285) Corbin, J. C.; Mensah, A. A.; Pieber, S. M.; Orasche, J.; Michalke, B.; Zanatta, M.; et al. Trace metals in soot and $PM_{2.5}$ from heavy-fuel-oil combustion in a marine engine. *Environ. Sci. Technol.* **2018**, *52*, 6714−6722.
(286) Oeder, S.; Kanashova, T.; Sippula, O.; Sapcariu, S. C.; Streibel, T.; Arteaga-Salas, J. M.; Passig, J.; Dilger, M.; Paur, H.-C.; Schlager, C.; et al. Particulate matter from both heavy fuel oil and diesel fiel shipping emissions show strong biological effects on human lung cells at realistic and comparable *in vitro* exposure conditions. *PLoS One* **2015**, *10* (6), e0126536.
(287) Sapcariu, S. C.; Kanashova, T.; Dilger, M.; Diabaté, S.; Oeder, S.; Passig, J.; Radischat, C.; Buters, J.; Sippula, O.; Streibel, T.; et al. Metabolic profiling as well as stable isotope assisted metabolic and proteomic analysis of RAW 264.7 macrophages exposed to ship engine aerosol emissions: Different effects of heavy fuel oil and refined diesel fuel. *PLoS One* **2016**, *11* (6), e0157964.
(288) Mousavi, A.; Sowlat, M. H.; Hasheminassab, S.; Polidori, A.; Shafer, M. M.; Schauer, J. J.; Sioutas, C. Impact of emissions from the Ports of Los Angeles and Long Beach on the oxidative potential of ambient $PM_{0.25}$ measured across the Los Angeles County. *Sci. Total Environ.* **2019**, *651*, 638−647.
(289) Corbett, J. J.; Winebrake, J. J.; Green, E. H.; Kasibhatla, P.; Eyring, V.; Lauer, A. Mortality from ship emissions: A global assessment. *Environ. Sci. Technol.* **2007**, *41*, 8512−8518.
(290) Ketzel, M.; Omstedt, G.; Johansson, C.; Düring, I.; Pohjola, M.; Oettl, D.; Gidhagen, L.; Wåhlin, P.; Lohmeyer, A.; Haakana, M.; Berkowicz, R. Estimation and validation of $PM_{2.5}/PM_{10}$ exhaust and non-exhaust emission factors for practical street pollution modelling. *Atmos. Environ.* **2007**, *41*, 9370−9385.
(291) Harrison, R. M.; Beddows, D. C. S.; Dall'Osto, M. PMF analysis of wide-range particle size spectra collected on a major highway. *Environ. Sci. Technol.* **2011**, *45*, 5522−5528.
(292) Harrison, R. M.; Jones, A. M.; Gietl, J.; Yin, J.; Green, D. C. Estimation of the contributions of brake dust, tire wear, and resuspension to nonexhaust traffic particles derived from atmospheric measurements. *Environ. Sci. Technol.* **2012**, *46*, 6523−6529.
(293) Vu, T. V.; Delgado-Saborit, M.; Harrison, R. M. Review: Particle number size distributions from seven major sources and implications for source apportionment studies. *Atmos. Environ.* **2015**, *122*, 114−132.
(294) Squizzato, S.; Masiol, M.; Agostini, C.; Visin, F.; Formenton, G.; Harrison, R. M.; Rampazzo, G. Factors, origin and sources affecting PM_1 concentrations and composition at an urban background site. *Atmos. Res.* **2016**, *180*, 262−273.
(295) Crilley, L. R.; Lucarelli, F.; Bloss, W. J.; Harrison, R. M.; Beddows, D. C.; Calzolai, G.; Nava, S.; Valli, G.; Bernardoni, V.; Vecchi, R. Source apportionment of fine and coarse particles at a roadside and urban background site in London during the 2012 summer ClearfLo campaign. *Environ. Pollut.* **2017**, *220*, 766−778.
(296) DieselNet. Engine & emission technology online. https://www.dieselnet.com/ (accessed June 10, 2019).
(297) Harrison, R. M.; Yin, J.; Mark, D.; Stedman, J.; Appleby, R. S.; Booker, J.; Moorcroft, S. Studies of the coarse particle (2.5−10 õm) component un UK urban atmospheres. *Atmos. Environ.* **2001**, *35*, 3667−3679.
(298) Querol, X.; Alastuey, A.; Ruiz, C. R.; Artiñano, B.; Hansson, H. C.; Harrison, R. M.; Buringh, E.; ten Brink, H. M.; Lutz, M.; Bruckmann, P.; Straehl, P.; Schneider, J. Speciation and origin of PM10 and PM2.5 in selected European cities. *Atmos. Environ.* **2004**, *38*, 6547−6555.
(299) Timmers, V. R. J. H.; Achten, P. A. J. Non-exhaust emissions from electric vehicles. *Atmos. Environ.* **2016**, *134*, 10−17.
(300) Muxworthy, A. R.; Matzka, J.; Davila, A. F.; Petersen, N. Magnetic signature of daily sampled urban atmospheric particles. *Atmos. Environ.* **2003**, *37*, 4163−4169.
(301) Matzka, J.; Maher, B. A. Magnetic biomonitoring of roadside tree leaves: identification of spatial and temporal variations in vehicle-derived particulates. *Atmos. Environ.* **1999**, *33*, 4565−4569.
(302) Spassov, S.; Egli, R.; Heller, F.; Nourgaliev, D. K.; Hannam, J. Magnetic identification of urban pollution sources in atmospheric particulate matter. *Geophys. J. Int.* **2004**, *159*, 555−564.
(303) Hoffmann, V.; Knab, M.; Appel, E. Magnetic susceptibility mapping of roadside pollution. *J. Geochem. Explor.* **1999**, *66*, 313−326.
(304) Hanesch, M.; Scholger, R. Mapping of heavy metal loadings in soils by means of magnetic susceptibility measurements. *Environ. Geol.* **2002**, *42*, 857−870.
(305) Magiera, T.; Strzyszcz, Z.; Kapicka, A.; Petrovsky, E. Discrimination of lithogenic and anthropogenic influences on topsoil magnetic susceptibility in Central Europe. *Geoderma* **2006**, *130*, 299−311.
(306) Hanesch, M.; Scholger, R.; Rey, D. Mapping dust distribution around an industrial site by measuring magnetic parameters of tree leaves. *Atmos. Environ.* **2003**, *37*, 5125−5133.
(307) Moreno, E.; Sagnotti, L.; Dinarés-Turell, J.; Winkler, A.; Cascella, A. Biomonitoring of traffic air pollution in Rome using magnetic properties of tree leaves. *Atmos. Environ.* **2003**, *37*, 2967−2977.
(308) Davila, A. F.; Rey, D.; Mohamed, K.; Rubio, B.; Guerra, A. P. Mapping the sources of urban dust in a coastal environment by measuring magnetic parameters of *Platanus hispanica* leaves. *Environ. Sci. Technol.* **2006**, *40*, 3922−3928.
(309) Hofman, J.; Maher, B. A.; Muxworthy, A. R.; Wuyts, K.; Castanheiro, A.; Samson, R. Biomagnetic monitoring of atmospheric pollution: A review of magnetic signatures from biological sensors. *Environ. Sci. Technol.* **2017**, *51*, 6648−6664.
(310) Jordanova, D.; Jordanova, N.; Lanos, P.; Petrov, P.; Tsacheva, T. Magnetism of outdoor and indoor settled dust and its utilization as a tool for revealing the effect of elevated particulate air pollution on cardiovascular mortality. *Geochem. Geophys. Geosyst.* **2012**, *13*, Q08Z49.

DOI: 10.1021/acs.est.9b01505
Environ. Sci. Technol. 2019, 53, 9970−9991

(311) Morris, W. A.; Versteeg, J. K.; Bryant, D. W.; Legzdins, A. E.; McCarry, B. E.; Marvin, C. H. Preliminary comparisons between mutagenicity and magnetic susceptibility of respirable airborne particulate. *Atmos. Environ.* **1995**, *29* (23), 3441−3450.
(312) Sagnotti, L.; Macri, P.; Egli, R.; Mondino, M. Magnetic properties of atmospheric particulate matter from automatic air sampler stations in Latium (Italy): Toward a definition of magnetic fingerprints for natural and anthropogenic PM_{10} sources. *J. Geophys. Res.* **2006**, *111*, B12S22.
(313) Sagnotti, L.; Taddeucci, J.; Winkler, A.; Cavallo, A. Compositional, morphological, and hysteresis characterization of magnetic airborne particulate matter in Rome, Italy. *Geochem. Geophys. Geosyst* **2009**, *10*, Q08Z06.
(314) Sagnotti, L.; Winkler, A. On the magnetic characterization and quantification of the superparamagnetic fraction of traffic-related urban airborne PM in Rome, Italy. *Atmos. Environ.* **2012**, *59*, 131−140.
(315) Saragnese, F.; Lanci, L.; Lanza, R. Nanometric-sized atmospheric particulate studied by magnetic analyses. *Atmos. Environ.* **2011**, *45*, 450−459.
(316) Szönyi, M.; Sagnotti, L.; Hirt, A. M. On leaf magnetic homogeneity in particulate matter biomonitoring studies. *Gephys. Res. Lett.* **2007**, *34*, L06306.
(317) Szönyi, M.; Sagnotti, L.; Hirt, A. M. A refined biomonitoring study of airborne particulate matter pollution in Rome, with magnetic measurements on *Quercus Ilex* tree leaves. *Geophys. J. Int.* **2008**, *173*, 127−141.
(318) Dunlop, D. J. Superparamagnetic and single-domain threshold sizes in magnetite. *J. Gephys. Res.* **1973**, *78* (11), 1780−1793.
(319) Özdemir, Ö.; Dunlop, D. J.; Moskowitz, B. M. The effect of oxidation on the Verwey transition in magnetite. *Gephys. Res. Lett.* **1993**, *20* (16), 1671−1674.
(320) Bødker, F.; Mørup, S. Size dependence of the properties of hematite nanoparticles. *Europhys. Lett.* **2000**, *52* (2), 217−223.
(321) Pankhurst, Q.; Hautot, D.; Khan, N.; Dobson, J. Increased levels of magnetic iron compounds in Alzheimer's disease. *J. Alzheimer's Dis.* **2008**, *13*, 49−52.
(322) Mitchell, R.; Maher, B. A.; Kinnersley, R. Rates of particulate pollution deposition onto leaf surfaces: Temporal and inter-species magnetic analyses. *Environ. Pollut.* **2010**, *158*, 1472−1478.
(323) Kim, W.; Doh, S.-J.; Park, Y.-H.; Yun, S.-T. Two-year magnetic monitoring in conjunction with geochemical and electron microscopic data of roadside dust in Seoul, Korea. *Atmos. Environ.* **2007**, *41*, 7627−7641.
(324) Lehndorff, E.; Schwark, L. Biominitoring of air quality in the Cologne Conurbation using pine needles as a passive sampler - Part II: polycyclic aromatic hydrocarbons (PAH). *Atmos. Environ.* **2004**, *38*, 3793−3808.
(325) Gonet, T.; Maher, B. A.; et al. Source apportionment of magnetite in the roadside particulate matter [in prep.].
(326) Gonet, T.; Maher, B. A.; Kukutschová, J. Quantitative evaluation of magnetic mineralogy of airborne brake wear emissions [in prep.].
(327) Ghio, A. J.; Kennedy, T. P.; Whorton, A. R.; Crumbliss, A. L.; Hatch, G. E.; Hoidal, J. R. Role of surface complexed iron in oxidant generation and lung inflammation induced by silicates. *Am. J. Physiol.* **1992**, *263*, 511−518.
(328) Dusseldorp, A.; Kruize, H.; Brunekreef, B.; Hofschreuder, P.; de Meer, G.; van Oudvorst, A. B. Associations of PM10 and airborne iron with respiratory health of adults living near a steel factory. *Am. J. Respir. Crit. Care Med.* **1995**, *152*, 1932−1939.
(329) Donaldson, K.; Brown, D. M.; Mitchell, C.; Dineva, M.; Beswick, P. H.; Gilmour, P.; MacNee, W. Free radical activity of PM_{10}: Iron-mediated generation of hydroxyl radicals. *Environ. Health Prospect.* **1997**, *105*, 1285−1289.
(330) Valavanidis, A.; Salika, A.; Theodoropoulou, A. Generation of hydroxyl radicals by urban suspended particulate air matter. The role of iron ions. *Atmos. Environ.* **2000**, *34*, 2379−2386.
(331) Aust, A. E.; Ball, J. C.; Hu, A. A.; Lighty, J. S.; Smith, K. R.; Straccia, A. M.; Veranth, J. M.; Young, W. C. Particle characteristics responsible for effects on human lung epithelial cells. *Res. Rep. Health Eff. Inst.* **2002**, *110*, 1−77.
(332) Ghio, A. J.; Cohen, M. D. Disruption of iron homeostasis as a mechanism of biologic effect by ambient air pollution particles. *Inhalation Toxicol.* **2005**, *17*, 709−716.
(333) Li, K.; Reichmann, H. Role of iron in neurodegenerative diseases. *J. Neural. Transm.* **2016**, *123*, 389−399.
(334) Pelclova, D.; Zdimal, V.; Kacer, P.; Fenclova, Z.; Vlckova, S.; Syslova, K.; et al. Oxidative stress markers are elevated in exhaled breath condensate of workers exposed to nanoparticles during iron oxide pigment production. *J. Breath Res.* **2016**, *10*, 016004.
(335) Moretti, S.; Smets, W.; Hofman, J.; Mubiana, K. V.; Oerlemans, E.; Vandenheuvel, D.; Samson, R.; Blust, R.; Lebeer, S. Human inflammatory response of endotoxin affected by particulate matter-bound transition metals. *Environ. Pollut.* **2019**, *244*, 118−126.
(336) Collingwood, J.; Dobson, J. Mapping and characterization of iron compounds in Alzheimer's tissue. *J. Alzheimer's Dis.* **2006**, *10*, 215−222.
(337) Quintana, C.; Bellefqih, S.; Laval, J. Y.; Guerquin-Kern, J. L.; Wu, T. D.; Avila, J.; Ferrer, I.; Arranz, R.; Patiño, C. Study of the localization of iron, ferritin, and hemosiderin in Alzheimer's disease hippocampus by analytical microscopy at the subcellular level. *J. Struct. Boil.* **2006**, *153*, 42−54.
(338) Plascencia-Villa, G.; Ponce, A.; Collingwood, J. F.; Arellano-Jiménez, M. J.; Zhu, X.; Rogers, J. T.; Betancourt, I.; José-Yacamán, M.; Perry, G. High-resolution analytical imaging and electron holography of magnetite particles in amyloid cores of Alzheimer's disease. *Sci. Rep.* **2016**, *6*, 24873.
(339) Hautot, D.; Pankhurst, Q. A.; Khan, N.; Dobson, J. Preliminary evaluation of nanoscale biogenic magnetite in Alzheimer's disease brain tissue. *Proc. R. Soc. London B* **2003**, *270*, S62−S64.
(340) Pankhurst, Q.; Hautot, D.; Khan, N.; Dobson, J. Increased levels of magnetic iron compounds in Alzheimer's disease. *J. Alzheimer's Dis.* **2008**, *13*, 49−52.
(341) Allsop, D.; Mayes, J.; Moore, S.; Masad, A.; Tabner, B. J. Metal-dependent generation of reactive oxygen species from amyloid proteins implicated in neurodegenerative disease. *Biochem. Soc. Trans.* **2008**, *36*, 1293−1298.
(342) Tabner, B. J.; Mayes, J.; Allsop, D. Hypothesis: Soluble Aβ oligomers in association with redox-active metal ions are the optimal generators of reactive oxygen species in Alzheimer's disease. *Int. J. Alzheimers Dis.* **2011**, *2011*, 546380.
(343) Teller, S.; Tahirbegi, I. B.; Mir, M.; Samitier, J.; Soriano, J. Magnetite-Amyloid-β deteriorates activity and functional organization in an *in vitro* model for Alzheimer's disease. *Sci. Rep.* **2015**, *5*, 17261.
(344) Kobayashi, A.; Yamamoto, N.; Kirschvink, J. Studies of inorganic crystals in biological tissue: Magnetite in human tumor. *Funtai oyobi Funmatsu Yakin* **1997**, *44*, 294−300.
(345) Brem, F.; Hirt, A. M.; Winklhofer, M.; Frei, K.; Yonekawa, Y.; Wieser, H.-G.; Dobson, J. Magnetic iron compounds in the human brain: a comparison of tumour and hippocampal tissue. *J. R. Soc., Interface* **2006**, *3*, 833−841.
(346) Dobson, J.; Grassi, P. Magnetic properties of human hippocampal tissue - evaluation of artefact and contamination sources. *Brain Res. Bull.* **1996**, *39* (4), 255−259.
(347) Könczöl, M.; Ebeling, S.; Goldenberg, E.; Treude, F.; Gminski, R.; Gieré, R.; Grobéty, B.; Rothen-Rutishauser, B.; Merfort, I.; Mersch-Sundermann, V. Cytotoxicity and genotoxicity of size-fractionated iron oxide (magnetite) in A549 human lung epithelial cells: Role of ROS, JNK, and NF-κB. *Chem. Res. Toxicol.* **2011**, *24*, 1460−1475.
(348) Könczöl, M.; Weiss, A.; Stangenberg, E.; Gminski, R.; Garcia-Käufer, M.; Gieré, R.; Merfort, I.; Mersch-Sundermann, V. Cell-cycle changes and oxidative stress response to magnetite in A549 human lung cells. *Chem. Res. Toxicol.* **2013**, *26*, 693−702.
(349) Apopa, P. L.; Qian, Y.; Shao, R.; Guo, N. L.; Schwegler-Berry, D.; Pacurari, M.; Porter, D.; Shi, X.; Vallyathan, V.; Castranova, V.

DOI: 10.1021/acs.est.9b01505
Environ. Sci. Technol. 2019, 53, 9970−9991

Iron oxide nanoparticles induce human microvascular endothelial cell permeability through reactive oxygen species production and microtubule remodeling. *Part. Fibre Toxicol.* **2009**, *6* (1), 1.

(350) Naqvi, S.; Samim, M.; Abdin, M. Z.; Ahmed, F. J.; Maita, A. N.; Prashant, C. K.; Dinda, A. K. Concentration-dependent toxicity of iron oxide nanoparticles mediated by increased oxidative stress. *Int. J. Nanomedicine* **2010**, *5*, 983−989.

(351) Singh, N.; Jenkins, G. J. S.; Asadi, R.; Doak, S. H. Potential toxicity of superparamagnetic iron oxide nanoparticles (SPION). *Nano Rev.* **2010**, *1*, 5358.

(352) Feng, Q.; Liu, Y.; Huang, J.; Chen, K.; Huang, J.; Xiao, K. Uptake, distribution, clearance, and toxicity of iron oxide nanoparticles with different sizes and coatings. *Sci. Rep.* **2018**, *8* (1), 2082.

(353) Kirschvink, J. L.; Kobayashi-Kirschvink, A.; Woodford, B. J. Magnetite biomineralization in the human brain. *Proc. Natl. Acad. Sci. U. S. A.* **1992**, *89*, 7683−7687.

(354) Schultheiss-Grassi, P. P.; Wessiken, R.; Dobson, J. TEM investigations of biogenic magnetite extracted from the human hippocampus. *Biochim. Biophys. Acta, Gen. Subj.* **1999**, *1426*, 212−216.

(355) Beelen, R.; Raaschou-Nielsen, O.; Stafoggia, M.; Jovanovic Andersen, Z.; Weinmayr, G.; Hoffmann, B.; Wolf, K.; Samoli, E.; Fischer, P.; Nieuwenhuijsen, M.; et al. Effects of long-term exposure to air pollution on natural-cause mortality: an analysis of 22 European cohorts within the multicentre ESCAPE project. *Lancet* **2014**, *383*, 785−795.

(356) Donaldson, K.; Tran, L.; Jimenez, L. A.; Duffin, R.; Newby, D. E.; Mills, N.; MacNee, W.; Stone, V. Combustion-derived nanoparticles: A review of their toxicology following inhalation exposure. *Part. Fibre Toxicol.* **2005**, *2* (10), 10.

(357) Hoek, G.; Krishnan, R. M.; Beelen, R.; Peters, A.; Ostro, A.; Brunekreef, B.; Kaufman, J. D. Long-term air pollution exposure and cardio-respiratory mortality: a review. *Environ. Health* **2013**, *12* (1), 43.

(358) Hoek, G.; Beelen, R.; Kos, G.; Dijkema, M.; van der Zee, S.; Fischer, P. H.; Brunekreef, B. Land use regression model for ultrafine particles in Amsterdam. *Environ. Sci. Technol.* **2011**, *45*, 622−628.

(359) Weichenthal, S.; van Ryswyk, K.; Goldstein, A.; Bagg, S.; Shekkarizfard, M.; Hatzopoulou, M. A land use regression model for ambient ultrafine particles in Montreal, Canada: A comparison of linear regression and a machine learning approach. *Environ. Res.* **2016**, *146*, 65−72.

(360) Weichenthal, S.; van Ryswyk, K.; Goldstein, A.; Shekarrizfard, M.; Hatzopoulou, M. Characterizing the spatial distribution of ambient ultrafine particles in Toronto, Canada: A land use regression model. *Environ. Pollut.* **2016**, *208*, 241−248.

(361) Weichenthal, S.; Lavigne, E.; Valois, M.-F.; Hatzopoulou, M.; van Ryswyk, K.; Shekarrizfard, M.; Villeneuve, P. J.; Goldberg, M. S.; Parent, M.-E. Spatial variations in ambient ultrafine particle concentrations and the risk of incident prostate cancer: A case-control study. *Environ. Res.* **2017**, *156*, 374−380.

(362) Weichenthal, S.; Bai, L.; Hatzopoulou, M.; van Ryswyk, K.; Kwong, J. C.; Jerrett, M.; van Donkelaar, A.; Martin, R. V.; Burnett, R. T.; Lu, H.; Chen, H. Long-term exposure to ambient ultrafine particles and respiratory disease incidence in Toronto, Canada: a cohort study. *Environ. Health* **2017**, *16* (64), DOI: 10.1186/s12940-017-0276-7.

(363) Van Nunen, E.; Vermeulen, R.; Tsai, M.-Y.; Probst-Hensch, N.; Ineichen, A.; Davey, M.; et al. Land use regression models for ultrafine particles in six European areas. *Environ. Sci. Technol.* **2017**, *51*, 3336−3345.

(364) Ostro, B.; Hu, J.; Goldberg, D.; Reynolds, P.; Hertz, A.; Bernstein, L.; Kleeman, M. J. Associations of mortality with long-term exposures to fine and ultrafine particles, species and sources: Results from the California teacher study cohort. *Environ. Health Perspect.* **2015**, *123* (6), 549−556.

(365) Bai, L.; Hatzopoulou, M.; Jerrett, M.; Kwong, J. C.; Burnett, R. T.; van Donkelaar, A.; Copes, R.; Martin, R. V.; van Ryswyk, K.; Lu, H.; et al. Exposure to ambient ultrafine particles and nitrogen dioxide and incident hypertension and diabetes. *Epidemiology* **2018**, *29* (3), 323−332.

(366) Campbell, A.; Becaria, A.; Lahiri, D. K.; Sharman, K.; Bondy, S. C. Chronic exposure to aluminum in drinking water increases inflammatory parameters selectively in the brain. *J. Neurosci. Res.* **2004**, *75*, 565−572.

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Environ. Sci. Technol. 2019, 53, 9970−9991