Title

Evidence of Indoor Dust Acting as Carrier for Metal-Based Nanoparticles: A Study of Exposure and Oxidative Risks

Journal

Environmental Science & Technology Letters, 9(5)

ISSN

2328-8930

Authors

Liu, Ziyi
Peng, Rong
Lv, Shangsi
et al.

Publication Date

2022-05-10

DOI

10.1021/acs.estlett.2c00192

Peer reviewed

Environmental Science & Technology Letters

pubs.acs.org/journal/estlcu

Letter

Evidence of Indoor Dust Acting as Carrier for Metal-Based Nanoparticles: A Study of Exposure and Oxidative Risks

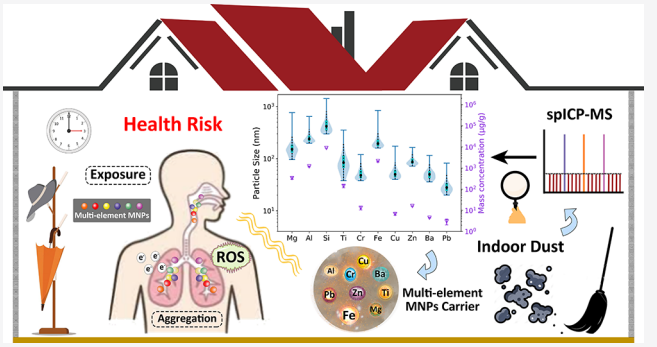
Ziyi Liu, Rong Peng, Shangsi Lv, Aodi Wang, Lijuan Zhao, Shuofei Dong, Dong Yan, Arturo A. Keller, and Yuxiong Huang*

Cite This: https://doi.org/10.1021/acs.estlett.2c00192

Read Online

ACCESS | Metrics & More | Article Recommendations | Supporting Information

ABSTRACT: The wide application of nanotechnology has led to the inevitable release and transport of metal-based nanoparticles (MNPs) into the environment. However, very few studies have examined the occurrence of MNPs in indoor dust. Here, we report the occurrence of 10 distinct types of MNPs in indoor dust, confirming its carrier role for MNPs. In particular, up to ∼4000 μg of MNPs/g of dust, including Fe-based MNPs (∼200 nm, equivalent spherical diameter), were found in indoor dust samples collected from both residential and public areas. Though the collected indoor dust exhibited a magnetic response, negligible differences were observed in the composition of MNPs, particle concentrations, and size distributions before and after magnetic separation, which suggested that MNPs in indoor dust were clusters containing multiple elements. Furthermore, indoor dust-associated MNPs easily aggregated when being exposed to lung fluid (e.g., the size of Fe-based MNPs increased ∼2.8-fold). Indoor dust with multielement MNPs can induce oxidative stress by generating more reactive oxygen species, and the estimated •OH concentration was increased by 1.5 times compared with the control. Long-term exposure to MNPs in indoor habitats may induce health risks, highlighting the need to better characterize these indoor contaminants.



KEYWORDS: *metal-based nanoparticles, indoor environment, dust, exposure, stability and bioavailability, oxidative potential*

INTRODUCTION

Growing public health concern has developed with respect to the indoor environment, as people spend more than three-quarters of their time indoors and are more likely to be exposed to indoor contaminants in the long term.[1] Ambient indoor contaminants, for example, particulate matter (e.g., $PM_{2.5}$), volatile organic compounds (VOCs), bioaerosols, and metals,[2−5] may pose considerable risks to humans.[6] In addition, indoor dust can serve as the carrier for hazardous substances (endotoxins, pathogens, metals, etc.),[7−9] leading to increased daily exposure to sorbed contaminants, which are detrimental to human health.[10]

Recent studies have addressed the potential risks of metal ions (e.g., Cu^{2+}, Pb^{2+}, and Zn^{2+}) in indoor dust.[8,11,12] In addition, metal-based nanoparticles (MNPs) have been widely embedded in consumer products with extensive applications,[13] which could be unintentionally released into different environmental compartments[14] (e.g., soil,[15] air via vehicle exhaust,[16] water via wastewater treatment plants,[17] etc.). However, very few studies have examined the occurrence of MNPs in indoor dust. The indoor dust-associated MNPs could enter and bioaccumulate in the human respiratory system (i.e., lung)[18,19] via inhalation. Because of their unique physicochemical properties (e.g., high reactivity[20] and oxidative potential[21]), MNPs can trigger extracellular and/or intracellular reactive oxygen species (ROS) formation,[14,22] which may damage the cell membrane and induce oxidation of cellular components (e.g., lipid peroxidation).[23,24] Long-term exposure to MNPs may cause cytotoxicity and even chronic alveolar inflammation of lung cells.[25,26] In addition, MNPs could be translocated to other organs (e.g., lymphatic system and liver)[27] in the human body. For example, magnetic MNPs, rather than their dissolved metal compounds, can bypass the blood−brain barrier, which poses a significant threat to human health (e.g., causing neurodegenerative diseases).[28] Thus, indoor dust can play a critical role in the indoor fate of MNPs. However, their occurrence, exposure, and risks have not been systematically investigated.

Received: March 19, 2022
Revised: April 12, 2022
Accepted: April 14, 2022

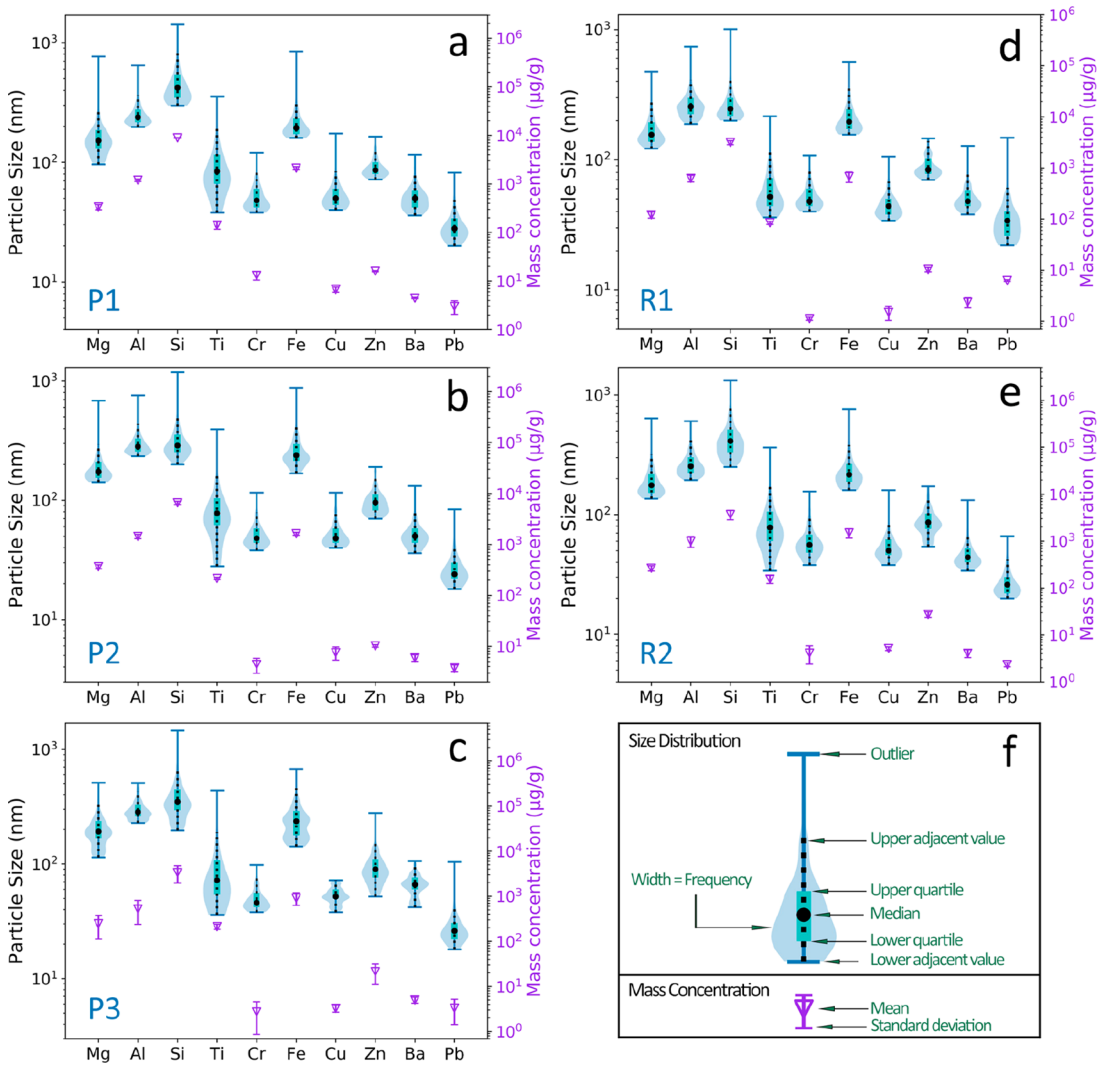


Figure 1. Particle size distribution and mass concentration of MNPs in indoor dust determined by spICP-MS. Panels a–e represent different indoor dust samples (Table S1): (a) P1, (b) P2, (c) P3, (d) R1, and (e) R2. The violin plot and scatter point are shown in panel f. The violin plot displays the median diameter (black dots) and upper and lower quartile (small rectangle) for the size distribution. The upper or lower quartile is the top 25% of numbers in the data set or the 75th percentile. The dotted line represents the upper and lower adjacent value, and the cap of the solid line represents the outlier value. The body of the violin shows different widths, which represent different diameter frequencies. The scatter plot shows the average mass concentration (micrograms of MNPs per gram of dust) and its standard deviation. The corresponding data are listed in Table S7.

A major challenge for assessing their risk is the quantitative analysis of low-abundance MNPs in indoor dust.[29] Recently, single-particle inductively coupled plasma mass spectrometry (spICP-MS) emerged as a powerful technique for nanoparticle quantification[30] (e.g., MNPs in various environmental samples, including wastewater and plant tissues),[31−34] with a low detection limit of particle mass concentration (down to 50 ng/L).[35] Furthermore, spICP-MS can provide fast identification and quantification of multielement MNPs in environmental samples,[31] which is essential for the thorough evaluation of the risks induced by MNPs. Thus, spICP-MS exhibits the capability to quantitatively study the environmental occurrence of MNPs in indoor dust.

In this study, the presence of MNPs in indoor dust was determined with spICP-MS. In addition, the exposure and bioavailability of MNPs in lung fluid as well as the oxidative potential of indoor dust-associated MNPs were evaluated. As emerging indoor contaminants require urgent and timely investigation, our findings serve to assess the occurrence and risks of MNPs in indoor dust.

MATERIALS AND METHODS

Indoor dust samples were collected from five representative sites (Table S1) with physicochemical characteristics listed in Table S2. MNPs in indoor dust were analyzed with spICP-MS (8900 ICP-MS, Agilent Technologies), and the experimental details are presented in Tables S3−S5 and eq S1. The stability of MNPs exposed in lung interstitial fluids (Table S6) was investigated,[36] and the oxidative potential of MNPs was evaluated with an ascorbic acid (AA) assay.[37] Additional details are supplied in section S1 (Materials and methods details).

RESULTS AND DISCUSSION

Identification of MNPs in Indoor Dust. The spICP-MS investigation provided evidence of the existence of multiele-

https://doi.org/10.1021/acs.estlett.2c00192
Environ. Sci. Technol. Lett. XXXX, XXX, XXX−XXX

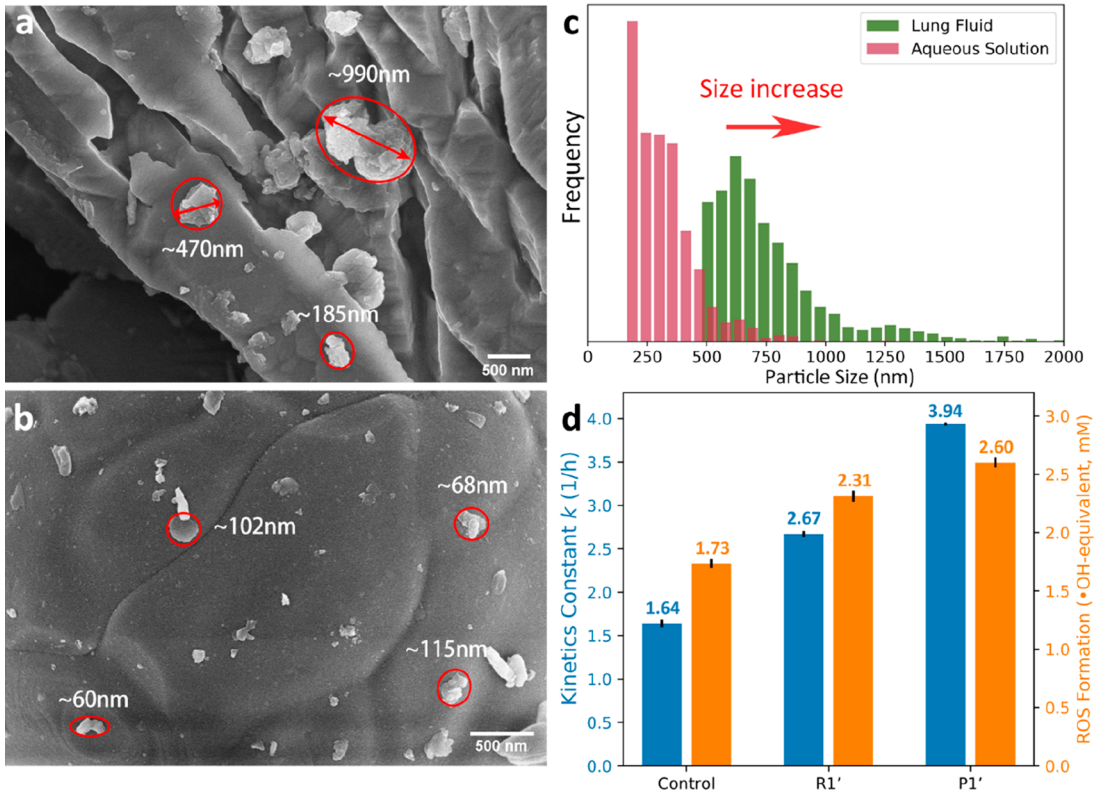


Figure 2. (a and b) SEM images of MNPs on indoor dust. (c) Particle size distribution of MNPs in lung fluid and aqueous solution determined by spICP-MS (for Fe-based MNPs in dust samples collected from the residential area). (d) Oxidative potential assessment of indoor dust. Detailed information about peroxidase-like activity kinetic constants is presented in Table S13, and the amount of •OH formation was calculated with eq S4.

ment MNPs in indoor dust, as MNPs containing up to 10 distinct elements were detected in all indoor dust samples (Figure 1 and Table S7), with the MNP size limit of detection (LOD_{size}) calculated with eq S2 and shown in Table S8. According to the violin plots of the particle size distribution of MNPs (Figure 1), Mg-, Al-, Si-, and Fe-based MNPs were presented in larger particle sizes and wider size distributions with median sizes ranging from 100 to 500 nm (equivalent spherical diameter), while Ti-, Cr-, Cu-, Zn-, Ba-, and Pb-based MNPs were smaller (10−100 nm). Scanning electron microscopy (SEM) images of indoor dust (Figure 2a,b) also evidenced that MNPs were mostly evenly distributed on the dust surface, ranging from ∼60 to ∼990 nm.

As shown by the scatter plots in Figure 1, the mass concentration of MNPs exhibited very large discrepancies across different elements. The mass concentrations of Cr-, Cu-, Zn-, Ba-, and Pb-based MNPs were around 2−30 μg of MNPs/g of dust, while the mass concentrations of Al-, Fe-, and Si-based MNPs ranged from 500 to ∼9000 μg of MNPs/g of dust. Al, Fe, and Si are the major elements on Earth and usually present abundantly in dust and soil samples, which may lead to unsurprisingly high mass concentrations of MNPs in the dust samples. However, Ti-based MNPs also showed relatively high mass concentrations [up to 215 μg of MNPs/g of dust (Figure 1b and Table S7)] in indoor dust samples, which may be attributed to the extensive application of Ti-based MNPs in industry and consumable products.[38] While Cu-based MNPs are used in paints and textiles,[39] Zn-based MNPs frequently applied in sunscreens would also be introduced into the indoor environment and thus be bioavailable and potentially toxic.[39,40]

Notably, indoor dust collected from public areas (P1, P2 and P3) showed higher concentrations of MNPs, compared to those of indoor dust collected from residential ones [R1 and R2 (Figure 1 and Table S7)], which may be due to more sources (e.g., urban construction and vehicular traffic),[41] resuspension, and ventilation/air mixing in public areas. In addition, a sample collected from a near-road site [P1 (Table S7)] exhibited higher concentrations of MNPs (e.g., Fe- and Cr-based MNPs) compared with another source farther from a road (P2), suggesting that particle pollution that originated from vehicles played a significant role,[2,42] being transferred to and accumulating in indoor dust.

The results presented above evidenced that indoor dust can act as typical carriers and collectors of various MNPs (Figure 2a,b). Notably, compared to some minor MNPs [e.g., ∼10 μg of MNPs/g of dust for Cu-based ones (Table S7)], a high abundance of Fe-based MNPs (up to 2100 μg of MNPs/g of dust) was detected in indoor dust. Fe-based MNPs can exhibit magnetic characteristics that have raised exposure concerns because magnetic MNPs can penetrate the blood−brain barrier.[28,43] Thus, magnetic separation was conducted to investigate the characteristics of magnetic MNPs in indoor dust.

Magnetic MNPs in Indoor Dust. The indoor dust samples were separated with a magnetic field, and the treated indoor dust exhibited a magnetic response (Figure S1) as well as a flocculent and elongated shape (Figure S2c,d) due to activation of magnetism,[44] which was different from that of the original indoor dust samples (Figure S2a,b). Interestingly, spICP-MS results (Figure S3 and Table S9) indicated that the magnetically recovered indoor dust contained multielement

https://doi.org/10.1021/acs.estlett.2c00192
Environ. Sci. Technol. Lett. XXXX, XXX, XXX−XXX

MNPs, which had not been reported in previous studies. Though the concentration of Fe-, Cu-, Zn-, and Pb-based MNPs slightly increased after the magnetic separation, indiscernible differences were shown for Mg-, Ti-, and Ba-based MNPs (Figure 1 and Figure S3). We thus postulated that these MNPs in indoor dust were aggregates and/or clusters containing multiple elements rather than single-element MNPs,[45] as the magnetic separation process could not isolate Fe-based MNPs. Similarly, the size distributions of the MNPs in magnetic indoor dust showed little difference, with Mg-, Al-, Si-, Ti-, and Fe-based MNPs exhibiting slightly wider size distributions than the MNPs in the original dust (Figure 1 and Figure S3). Notably, Fe-based MNPs exhibited dual peaks in the particle size distribution (Figure S3a,d), suggesting the formation of agglomerates under the magnetic field.

In addition, according to the elemental mapping by energy dispersive spectroscopy (EDS), Fe-based MNPs were distributed rather evenly in the indoor dust (Figure S4), further confirming the carrier role of indoor dust for MNPs. The proportions of different elements determined by EDS indicated insignificant differences in indoor dust before and after magnetic separation (Table S10), which were aligned with the spICP-MS results (Table S9). Furthermore, the elemental composition and chemical speciation of Fe-based MNPs were quite similar before and after magnetic separation (Figure S5).

Magnetic multielement MNPs (Figure S3) clustered in indoor dust may generate synergistic effects and induce higher risks.[14,45] Specifically, Fe-based MNPs [commonly as Fe(II) and Fe(III) oxide MNPs] would act as an electron shuttle to trigger Fenton-like reactions, and the generated ROS would induce cellular oxidative stress, resulting in inflammation and genotoxicity.[46] Ti-based MNPs (up to 215 μg of MNPs/g of dust) may cause cardiovascular disease,[47] while Cu- and Zn-based MNPs may result in pulmonary inflammation and metal fume fever.[48] The transition metal MNPs (e.g., Fe-, Ti-, Cu-, and Zn-based MNPs) could easily enter the human respiratory system (e.g., lung) and generate ROS to cause oxidative stress.[28,49] Consequently, it is essential to evaluate indoor dust-associated MNPs' exposure, stability, and oxidative potential in the biological environment.

Stability and Bioavailability of MNPs in Lung Fluids. The exposure, stability, and bioavailability assessments of MNPs in the respiratory system were conducted in lung interstitial fluids (detailed composition in Table S6). MNPs exhibited larger particles after exposure to lung interstitial fluids (Table S11), compared to that in the aqueous solution (Table S9). For example, Fe-based MNPs showed a 2.8-fold increase in size from ~250 to ~700 nm (Figure 2c), and similar tendencies were observed for Ti-, Cu-, and Cr-based MNPs (Tables S9 and S11). The high ionic strength of lung interstitial fluids (Table S6) promoted the aggregation of MNPs,[50] which decreased the stability of MNPs. In addition, on average the MNPs exhibited almost electric neutrality, with a ζ-potential ranging from −2.5 to 0 mV in the lung interstitial fluids (Table S12), which also decreased their stability. The agglomeration of MNPs resulted in lower mobility (e.g., deposition) in lung interstitial fluids, supported by the downward trend in mass concentrations (Tables S9 and S11). Larger MNPs (~500 nm) that are less stable and less mobile would easily deposit in the bronchioles and alveolar region.[14] Meanwhile, the smaller MNPs [down to ~100 nm (Table S11)] reported in this study would be more mobile and bioavailable, potentially penetrating through epithelium cells and translocating via the blood circulation to secondary organs.[27]

On the contrary, multielement MNPs in indoor dust can release metal ions when exposed to lung fluid. In particular, dissolved Fe concentrations reached 1900 μg of Fe/g of dust, dissolved Zn concentrations increased from 190 to 240 μg of Zn/g of dust, and dissolved Ti concentrations reached 52 μg of Ti/g of dust (Table S11). The dissolved Fe ions may form complexes [e.g., ferric EDTA and monoiron(III) dicitrate], which may bind to the cell surfaces,[51] to induce intracellular iron accumulation and further ferroptosis and cytotoxicity.[52] Released Zn ions have been reported to exert growth reduction, exhibiting cytoderm damage and intracellular degradation.[53] Furthermore, as indoor dust could be exposed and retained within the respiratory system, the associated multielement MNPs and their dissolved ions (e.g., Fe, Ti, Zn, etc.) can form ROS (e.g., •OH),[14] which may cause cellular oxidative stress,[54] lung injuries, and other health risks.[14,55] Therefore, assessing the oxidative potential of indoor dust is urgent.

Oxidative Potential Assessment of Indoor Dust. The oxidative potential of MNPs carried by indoor dust was evaluated considering their peroxidase-like activity. The depletion of AA was measured as an important biomarker of oxidative stress,[56] and the formation of ROS was calculated. As shown in Figure 2d and Figure S6, the presence of indoor dust significantly accelerated the depletion of AA. The peroxidase-like activity kinetic constants [k (1/h) (Table S13)] were calculated using eq S3, increasing from 1.64 (control) to 2.67 (R1′) and 3.94 (P1′) (Figure 2d). These results revealed that exposure to MNPs in indoor dust can generate ROS, which in turn can induce oxidative stress. On the basis of eq S4, the concentration of generated ROS (•OH equivalent) is shown in Figure 2d, increasing from 1.73 mM (control) to 2.31 mM (R1′) and 2.60 mM (P1′), which is up to 1.5 times greater than that of the control group, though the subcellular effect of indoor dust-associated MNPs generated needs to be investigated further *in vivo*.

Forming ROS can adversely generate respiratory health problems (e.g., inflammation) due to the high reactivity with tissues and cell components,[57] oxidizing extracellular and/or intercellular biomacromolecules[58] (e.g., lipid peroxidation and DNA damage).[59] Some chronic pathologies (e.g., asthma) caused by respirable fine particles may result in transient electron exchange reactions between MNPs and cells.[46] Furthermore, Fe-based MNPs were reported to translocate through human organs (e.g., lung) and blood circulation[60] and bypass the blood−brain barrier,[28] which would induce oxidative stress to the human brain and may be associated with neuropathologic diseases.[57] Furthermore, the co-occurrence of multielement MNPs (e.g., Fe and Cu) in indoor dust can generate a greater abundance of ROS via the synergistically peroxidase-like activity.[56] The indoor dust-associated transition MNPs (e.g., Fe-, Cu-, and Zn-based MNPs) may also interact with amyloid proteins to form ROS, leading to neuro-degenerative disease.[61]

Environmental Implications. The indoor environment is the habitat where people spend most of their time, and indoor dust may act as an unsuspected carrier of emerging MNP pollutants. Several studies have reported the presence of airborne fine particles (e.g., MNPs) inside the human body,[62−64] but the exposure route and fate, as well as the

https://doi.org/10.1021/acs.estlett.2c00192
Environ. Sci. Technol. Lett. XXXX, XXX, XXX−XXX

health impact, still require clear identification.[60] This study for the first time comprehensively investigates the occurrence of MNPs in indoor dust collected from representative public and residential indoor areas. Compelling evidence that indoor dust is a carrier for multielement MNPs (e.g., Fe-, Ti-, Cu-, and Zn-based MNPs) was investigated via spICP-MS, while previous studies merely assessed the total metal content and metal ions.[8,11,12] Though sources in the outdoor environment (e.g., factories and vehicles)[65,66] have been considered to be responsible for relatively high-concentration MNP releases,[67] significant amounts of MNPs (up to 4400 μg of MNPs/g of dust for Fe-based ones), which accumulate and gather in indoor dust (considered as receptors) through outdoor/indoor penetration, may pose potential risks to human health after exposure to the respiratory system. In lung interstitial fluids, dust-borne MNPs exhibited instability and aggregation (e.g., ~2.8-fold increase in the size of Fe-based MNPs), which may lead to MNPs being deposited in bronchioles and posing potential pulmonary health risks. Smaller bioavailable MNPs (down to 100 nm) reported in this study can be considered as the missing puzzle in the indoor environment assessment as previous studies mostly focused on dissolved metals.[68,69] Additionally, the oxidative assessment further revealed the peroxidase-like activity of MNPs in indoor dust, generating 1.5 times more ROS than the control, which correlates with some nasal and respiratory diseases, as well as neuropsychiatric outcomes.[70] Our comprehensive assessment raises the flag on the presence of multielement MNPs in indoor dust and their potential health risks, particularly because people stay in indoor habitats for a long period (even longer during the COVID-19 epidemic[71]) without intentional protection.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.estlett.2c00192.

Materials and methods details (section S1), magnetic response of indoor dust (Figure S1), morphological characteristics of indoor dust (Figure S2), particle size distribution and mass concentration of MNPs in magnetic indoor dust determined by spICP-MS (Figure S3), SEM with EDS element mapping of indoor dust (Figure S4), XPS spectra for Fe 2p (Figure S5), oxidative potential assessment of indoor dust (Figure S6), sampling collection details (Table S1), physico-chemical characteristics and recovery of indoor dust (Table S2), spICP-MS instrumental details (Table S3), nominal and determined values of Au NPs by spICP-MS (Table S4), physicochemical properties of the target MNPs (Table S5), components of the simulated lung interstitial fluids (Table S6), mass concentration and particle size of MNPs in untreated indoor dust dispersed in UPW (Table S7), MNP LOD_{size} determined by spICP-MS (Table S8), mass concentration and particle size of MNPs in magnetic indoor dust dispersed in UPW (Table S9), element concentration of the indoor dust obtained by EDS (Table S10), mass concentration, ionic concentration, and particle size of MNPs in magnetic indoor dust in lung fluid (Table S11), ζ potential and pH of the suspension of MNPs (Table S12), peroxidase-like activity kinetics for the pseudo-first-order reaction in different oxidative potential assessment systems (Table S13), transport efficiency determined with the particle size method (eq S1), size limit of detection of MNPs by spICP-MS (eq S2), pseudo-first-order reaction kinetics (eq S3), and correlation of AA consumption and •OH formation (eq S4) (PDF)

■ AUTHOR INFORMATION

Corresponding Author

Yuxiong Huang − *Tsinghua-Berkeley Shenzhen Institute (TBSI), Shenzhen International Graduate School (SIGS), Tsinghua University, Shenzhen 518055, China;* orcid.org/0000-0001-8124-643X; Phone: +86 0755 2640 3079; Email: huang_yuxiong@sz.tsinghua.edu.cn

Authors

Ziyi Liu − *Tsinghua-Berkeley Shenzhen Institute (TBSI), Shenzhen International Graduate School (SIGS), Tsinghua University, Shenzhen 518055, China*

Rong Peng − *Tsinghua-Berkeley Shenzhen Institute (TBSI), Shenzhen International Graduate School (SIGS), Tsinghua University, Shenzhen 518055, China*

Shangsi Lv − *Tsinghua-Berkeley Shenzhen Institute (TBSI), Shenzhen International Graduate School (SIGS), Tsinghua University, Shenzhen 518055, China*

Aodi Wang − *State Key Laboratory of Pollution Control and Resource Reuse, School of Environment, Nanjing University, Nanjing 210023, China*

Lijuan Zhao − *State Key Laboratory of Pollution Control and Resource Reuse, School of Environment, Nanjing University, Nanjing 210023, China;* orcid.org/0000-0002-8481-0435

Shuofei Dong − *Agilent Technologies Company, Ltd. (China), Beijing 100102, China*

Dong Yan − *Agilent Technologies Company, Ltd. (China), Beijing 100102, China*

Arturo A. Keller − *Bren School of Environmental Science and Management, University of California, Santa Barbara, California 93106, United States;* orcid.org/0000-0002-7638-662X

Complete contact information is available at: https://pubs.acs.org/10.1021/acs.estlett.2c00192

Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

This research was financially supported by National Natural Science Foundation of China (42077293 and 22006088), Natural Science Foundation of Guangdong Province (2019A1515011692 and 2019QN01L797), and Shenzhen Municipal Science and Technology Innovation Committee (RCYX20210609104448111). Y.H. is also thankful for the financial support from Tsinghua Shenzhen International Graduate School (QD2021010N and HW2020002).

■ REFERENCES

(1) Barrio-Parra, F.; De Miguel, E.; Lázaro-Navas, S.; Gómez, A.; Izquierdo, M. Indoor Dust Metal Loadings: A Human Health Risk Assessment. *Expo Health* **2018**, *10* (1), 41−50.

(2) Karjalainen, P.; Saari, S.; Kuuluvainen, H.; Kalliohaka, T.; Taipale, A.; Rönkkö, T. Performance of Ventilation Filtration Technologies on Characteristic Traffic Related Aerosol down to Nanocluster Size. *Aerosol Sci. Technol.* **2017**, *51* (12), 1398−1408.

(3) Isaxon, C.; Gudmundsson, A.; Nordin, E. Z.; Lönnblad, L.; Dahl, A.; Wieslander, G.; Bohgard, M.; Wierzbicka, A. Contribution of Indoor-Generated Particles to Residential Exposure. *Atmos. Environ.* **2015**, *106*, 458−466.
(4) Xie, W.; Li, Y.; Bai, W.; Hou, J.; Ma, T.; Zeng, X.; Zhang, L.; An, T. The Source and Transport of Bioaerosols in the Air: A Review. *Front. Environ. Sci. Eng.* **2021**, *15* (3), 44.
(5) Cahill, T. M. Ambient Acrolein Concentrations in Coastal, Remote, and Urban Regions in California. *Environ. Sci. Technol.* **2014**, *48* (15), 8507−8513.
(6) Goldstein, A. H.; Nazaroff, W. W.; Weschler, C. J.; Williams, J. How Do Indoor Environments Affect Air Pollution Exposure? *Environ. Sci. Technol.* **2021**, *55* (1), 100−108.
(7) Schlosser, O.; Robert, S.; Debeaupuis, C.; Huyard, A. Inhalable Dust as a Marker of Exposure to Airborne Biological Agents in Composting Facilities. *Waste Management* **2018**, *81*, 78−87.
(8) Zhou, L.; Liu, G.; Shen, M.; Hu, R.; Sun, M.; Liu, Y. Characteristics and Health Risk Assessment of Heavy Metals in Indoor Dust from Different Functional Areas in Hefei, China. *Environ. Pollut.* **2019**, *251*, 839−849.
(9) Doyi, I. N. Y.; Isley, C. F.; Soltani, N. S.; Taylor, M. P. Human Exposure and Risk Associated with Trace Element Concentrations in Indoor Dust from Australian Homes. *Environ. Int.* **2019**, *133*, 105125.
(10) Whitehead, T.; Metayer, C.; Buffler, P.; Rappaport, S. M. Estimating Exposures to Indoor Contaminants Using Residential Dust. *J. Expo Sci. Environ. Epidemiol* **2011**, *21* (6), 549−564.
(11) Isley, C. F.; Fry, K. L.; Liu, X.; Filippelli, G. M.; Entwistle, J. A.; Martin, A. P.; Kah, M.; Meza-Figueroa, D.; Shukle, J. T.; Jabeen, K.; Famuyiwa, A. O.; Wu, L.; Sharifi-Soltani, N.; Doyi, I. N. Y.; Argyraki, A.; Ho, K. F.; Dong, C.; Gunkel-Grillon, P.; Aelion, C. M.; Taylor, M. P. International Analysis of Sources and Human Health Risk Associated with Trace Metal Contaminants in Residential Indoor Dust. *Environ. Sci. Technol.* **2022**, *56*, 1053.
(12) Han, Q.; Liu, Y.; Feng, X.; Mao, P.; Sun, A.; Wang, M.; Wang, M. Pollution Effect Assessment of Industrial Activities on Potentially Toxic Metal Distribution in Windowsill Dust and Surface Soil in Central China. *Science of The Total Environment* **2021**, *759*, 144023.
(13) Keller, A. A.; Lazareva, A. Predicted Releases of Engineered Nanomaterials: From Global to Regional to Local. *Environ. Sci. Technol. Lett.* **2014**, *1* (1), 65−70.
(14) Rabajczyk, A.; Zielecka, M.; Porowski, R.; Hopke, P. K. Metal Nanoparticles in the Air: State of the Art and Future Perspectives. *Environ. Sci.: Nano* **2020**, *7* (11), 3233−3254.
(15) Garner, K. L.; Keller, A. A. Emerging Patterns for Engineered Nanomaterials in the Environment: A Review of Fate and Toxicity Studies. *J. Nanopart. Res.* **2014**, *16* (8), 2503.
(16) Rönkkö, T.; Pirjola, L.; Ntziachristos, L.; Heikkilä, J.; Karjalainen, P.; Hillamo, R.; Keskinen, J. Vehicle Engines Produce Exhaust Nanoparticles Even When Not Fueled. *Environ. Sci. Technol.* **2014**, *48* (3), 2043−2050.
(17) Lazareva, A.; Keller, A. A. Estimating Potential Life Cycle Releases of Engineered Nanomaterials from Wastewater Treatment Plants. *ACS Sustainable Chem. Eng.* **2014**, *2* (7), 1656−1665.
(18) Salthammer, T. Emerging Indoor Pollutants. *International Journal of Hygiene and Environmental Health* **2020**, *224*, 113423.
(19) *Exposure Factors Handbook*; Office of Health and Environmental Assessment, U.S. Environmental Protection Agency, 1989.
(20) Adegboyega, N. F.; Sharma, V. K.; Siskova, K. M.; Vecerova, R.; Kolar, M.; Zbořil, R.; Gardea-Torresdey, J. L. Enhanced Formation of Silver Nanoparticles in Ag + -NOM-Iron(II, III) Systems and Antibacterial Activity Studies. *Environ. Sci. Technol.* **2014**, *48* (6), 3228−3235.
(21) Gawda, A.; Majka, G.; Nowak, B.; Marcinkiewicz, J. Air Pollution, Oxidative Stress, and Exacerbation of Autoimmune Diseases. *Cent. Eur. J. Immunol.* **2017**, *3*, 305−312.
(22) Li, X.; Kuang, X. M.; Yan, C.; Ma, S.; Paulson, S. E.; Zhu, T.; Zhang, Y.; Zheng, M. Oxidative Potential by PM2.5 in the North China Plain: Generation of Hydroxyl Radical. *Environ. Sci. Technol.* **2019**, *53* (1), 512−520.
(23) 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* (4), 565−572.
(24) Huang, Y.; Gao, M.; Wang, W.; Liu, Z.; Qian, W.; Chen, C. C.; Zhu, X.; Cai, Z. Effects of Manufactured Nanomaterials on Algae: Implications and Applications. *Front. Environ. Sci. Eng.* **2022**, *16* (9), 122.
(25) 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* (5), 4542−4552.
(26) Sung, J. H.; Ji, J. H.; Park, J. D.; Yoon, J. U.; Kim, D. S.; Jeon, K. S.; Song, M. Y.; Jeong, J.; Han, B. S.; Han, J. H.; Chung, Y. H.; Chang, H. K.; Lee, J. H.; Cho, M. H.; Kelman, B. J.; Yu, I. J. Subchronic Inhalation Toxicity of Silver Nanoparticles. *Toxicol. Sci.* **2009**, *108* (2), 452−461.
(27) Kolanjiyil, A. V.; Kleinstreuer, C. Nanoparticle Mass Transfer From Lung Airways to Systemic Regions—Part II: Multi-Compartmental Modeling. *Journal of Biomechanical Engineering* **2013**, *135* (12), 121004.
(28) Maher, B. A.; Ahmed, I. A. M.; Karloukovski, V.; MacLaren, D. A.; Foulds, P. G.; Allsop, D.; Mann, D. M. A.; Torres-Jardón, R.; Calderon-Garciduenas, L. Magnetite Pollution Nanoparticles in the Human Brain. *Proc. Natl. Acad. Sci. U.S.A.* **2016**, *113* (39), 10797−10801.
(29) Koehler, K.; Good, N.; Wilson, A.; Mölter, A.; Moore, B. F.; Carpenter, T.; Peel, J. L.; Volckens, J. The Fort Collins Commuter Study: Variability in Personal Exposure to Air Pollutants by Microenvironment. *Indoor Air* **2019**, *29* (2), 231−241.
(30) Liu, Z.; Zhu, Y.; Lv, S.; Shi, Y.; Dong, S.; Yan, D.; Zhu, X.; Peng, R.; Keller, A. A.; Huang, Y. Quantifying the Dynamics of Polystyrene Microplastics UV-Aging Process. *Environ. Sci. Technol. Lett.* **2022**, *9* (1), 50−56.
(31) Huang, Y.; Keller, A. A.; Cervantes-Avilés, P.; Nelson, J. Fast Multielement Quantification of Nanoparticles in Wastewater and Sludge Using Single-Particle ICP-MS. *ACS ES&T Water* **2021**, *1*, 205.
(32) Keller, A. A.; Huang, Y.; Nelson, J. Detection of Nanoparticles in Edible Plant Tissues Exposed to Nano-Copper Using Single-Particle ICP-MS. *J. Nanopart. Res.* **2018**, *20* (4), 101.
(33) Cervantes-Avilés, P.; Huang, Y.; Keller, A. A. Incidence and Persistence of Silver Nanoparticles throughout the Wastewater Treatment Process. *Water Res.* **2019**, *156*, 188−198.
(34) Minelli, C.; Wywijas, M.; Bartczak, D.; Cuello-Nuñez, S.; Infante, H. G.; Deumer, J.; Gollwitzer, C.; Krumrey, M.; Murphy, K. E.; Johnson, M. E.; Montoro Bustos, A. R.; Strenge, I. H.; Faure, B.; Høghøj, P.; Tong, V.; Burr, L.; Norling, K.; Höök, F.; Roesslein, M.; Kocic, J.; Hendriks, L.; Kestens, V.; Ramaye, Y.; Contreras Lopez, M. C.; Auclair, G.; Mehn, D.; Gilliland, D.; Potthoff, A.; Oelschlägel, K.; Tentschert, J.; Jungnickel, H.; Krause, B. C.; Hachenberger, Y. U.; Reichardt, P.; Luch, A.; Whittaker, T. E.; Stevens, M. M.; Gupta, S.; Singh, A.; Lin, F.; Liu, Y.-H.; Costa, A. L.; Baldisserri, C.; Jawad, R.; Andaloussi, S. E. L.; Holme, M. N.; Lee, T. G.; Kwak, M.; Kim, J.; Ziebel, J.; Guignard, C.; Cambier, S.; Contal, S.; Gutleb, A. C.; Kuba Tatarkiewicz, J.; Jankiewicz, B. J.; Bartosewicz, B.; Wu, X.; Fagan, J. A.; Elje, E.; Rundén-Pran, E.; Dusinska, M.; Kaur, I. P.; Price, D.; Nesbitt, I.; O'Reilly, S.; Peters, R. J. B.; Bucher, G.; Coleman, D.; Harrison, A. J.; Ghanem, A.; Gering, A.; McCarron, E.; Fitzgerald, N.; Cornelis, G.; Tuoriniemi, J.; Sakai, M.; Tsuchida, H.; Maguire, C.; Prina-Mello, A.; Lawlor, A. J.; Adams, J.; Schultz, C. L.; Constantin, D.; Thanh, N. T. K.; Tung, L. D.; Panariello, L.; Damilos, S.; Gavriilidis, A.; Lynch, I.; Fryer, B.; Carrazco Quevedo, A.; Guggenheim, E.; Briffa, S.; Valsami-Jones, E.; Huang, Y.; Keller, A. A.; Kinnunen, V.-T.; Perämäki, S.; Krpetic, Z.; Greenwood, M.; Shard, A. G. Versailles Project on Advanced Materials and Standards (VAMAS) Interlaboratory Study on Measuring the Number

https://doi.org/10.1021/acs.estlett.2c00192
Environ. Sci. Technol. Lett. XXXX, XXX, XXX−XXX

Concentration of Colloidal Gold Nanoparticles. *Nanoscale* **2022**, *14*, 4690−4704.
(35) Pace, H. E.; Rogers, N. J.; Jarolimek, C.; Coleman, V. A.; Higgins, C. P.; Ranville, J. F. Determining Transport Efficiency for the Purpose of Counting and Sizing Nanoparticles via Single Particle Inductively Coupled Plasma Mass Spectrometry. *Anal. Chem.* **2011**, *83* (24), 9361−9369.
(36) Boisa, N.; Elom, N.; Dean, J. R.; Deary, M. E.; Bird, G.; Entwistle, J. A. Development and Application of an Inhalation Bioaccessibility Method (IBM) for Lead in the PM10 Size Fraction of Soil. *Environ. Int.* **2014**, *70*, 132−142.
(37) Fang, T.; Verma, V.; Bates, J. T.; Abrams, J.; Klein, M.; Strickland, M. J.; Sarnat, S. E.; Chang, H. H.; Mulholland, J. A.; Tolbert, P. E.; Russell, A. G.; Weber, R. J. Oxidative Potential of Ambient Water-Soluble $PM_{2.5}$ in the Southeastern United States: Contrasts in Sources and Health Associations between Ascorbic Acid (AA) and Dithiothreitol (DTT) Assays. *Atmos. Chem. Phys.* **2016**, *16* (6), 3865−3879.
(38) Tan, W.; Peralta-Videa, J. R.; Gardea-Torresdey, J. L. Interaction of Titanium Dioxide Nanoparticles with Soil Components and Plants: Current Knowledge and Future Research Needs - a Critical Review. *Environ. Sci.: Nano* **2018**, *5* (2), 257−278.
(39) Zhao, J.; Ren, W.; Dai, Y.; Liu, L.; Wang, Z.; Yu, X.; Zhang, J.; Wang, X.; Xing, B. Uptake, Distribution, and Transformation of CuO NPs in a Floating Plant *Eichhornia Crassipes* and Related Stomatal Responses. *Environ. Sci. Technol.* **2017**, *51* (13), 7686−7695.
(40) Li, L.-Z.; Zhou, D.-M.; Peijnenburg, W. J. G. M.; van Gestel, C. A. M.; Jin, S.-Y.; Wang, Y.-J.; Wang, P. Toxicity of Zinc Oxide Nanoparticles in the Earthworm, Eisenia Fetida and Subcellular Fractionation of Zn. *Environ. Int.* **2011**, *37* (6), 1098−1104.
(41) Xia, W.; Wang, Y.; Chen, S.; Huang, J.; Wang, B.; Zhang, G. J.; Zhang, Y.; Liu, X.; Ma, J.; Gong, P.; Jiang, Y.; Wu, M.; Xue, J.; Wei, L.; Zhang, T. Double Trouble of Air Pollution by Anthropogenic Dust. *Environ. Sci. Technol.* **2022**, *56*, 761.
(42) Quang, T. N.; He, C.; Morawska, L.; Knibbs, L. D. Influence of Ventilation and Filtration on Indoor Particle Concentrations in Urban Office Buildings. *Atmos. Environ.* **2013**, *79*, 41−52.
(43) Li, S.; Zhang, B.; Wu, D.; Li, Z.; Chu, S.-Q.; Ding, X.; Tang, X.; Chen, J.; Li, Q. Magnetic Particles Unintentionally Emitted from Anthropogenic Sources: Iron and Steel Plants. *Environ. Sci. Technol. Lett.* **2021**, *8* (4), 295−300.
(44) 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. *Atmospheric Pollution Research* **2017**, *8* (4), 754−766.
(45) Yang, Y.; Vance, M.; Tou, F.; Tiwari, A.; Liu, M.; Hochella, M. F. Nanoparticles in Road Dust from Impervious Urban Surfaces: Distribution, Identification, and Environmental Implications. *Environ. Sci.: Nano* **2016**, *3* (3), 534−544.
(46) O'Day, P. A.; Pattammattel, A.; Aronstein, P.; Leppert, V. J.; Forman, H. J. Iron Speciation in Respirable Particulate Matter and Implications for Human Health. *Environ. Sci. Technol.* **2022**, DOI: 10.1021/acs.est.1c06962.
(47) Rossi, S.; Savi, M.; Mazzola, M.; Pinelli, S.; Alinovi, R.; Gennaccaro, L.; Pagliaro, A.; Meraviglia, V.; Galetti, M.; Lozano-Garcia, O.; Rossini, A.; Frati, C.; Falco, A.; Quaini, F.; Bocchi, L.; Stilli, D.; Lucas, S.; Goldoni, M.; Macchi, E.; Mutti, A.; Miragoli, M. Subchronic Exposure to Titanium Dioxide Nanoparticles Modifies Cardiac Structure and Performance in Spontaneously Hypertensive Rats. *Particle and Fibre Toxicology* **2019**, *16* (1), 25.
(48) Schraufnagel, D. E. The Health Effects of Ultrafine Particles. *Exp Mol. Med.* **2020**, *52* (3), 311−317.
(49) Zhang, Q.; Lu, D.; Wang, D.; Yang, X.; Zuo, P.; Yang, H.; Fu, Q.; Liu, Q.; Jiang, G. Separation and Tracing of Anthropogenic Magnetite Nanoparticles in the Urban Atmosphere. *Environ. Sci. Technol.* **2020**, *54* (15), 9274−9284.
(50) Conway, J. R.; Adeleye, A. S.; Gardea-Torresdey, J.; Keller, A. A. Aggregation, Dissolution, and Transformation of Copper Nanoparticles in Natural Waters. *Environ. Sci. Technol.* **2015**, *49* (5), 2749−2756.
(51) Hettiarachchi, E.; Ivanov, S.; Kieft, T.; Goldstein, H. L.; Moskowitz, B. M.; Reynolds, R. L.; Rubasinghege, G. Atmospheric Processing of Iron-Bearing Mineral Dust Aerosol and Its Effect on Growth of a Marine Diatom, Cyclotella Meneghiniana. *Environ. Sci. Technol.* **2021**, *55*, 871.
(52) Fang, S.; Yu, X.; Ding, H.; Han, J.; Feng, J. Effects of Intracellular Iron Overload on Cell Death and Identification of Potent Cell Death Inhibitors. *Biochem. Biophys. Res. Commun.* **2018**, *503* (1), 297−303.
(53) Bielmyer-Fraser, G. K.; Jarvis, T. A.; Lenihan, H. S.; Miller, R. J. Cellular Partitioning of Nanoparticulate versus Dissolved Metals in Marine Phytoplankton. *Environ. Sci. Technol.* **2014**, *48* (22), 13443−13450.
(54) Zhang, J.; Guo, W.; Li, Q.; Wang, Z.; Liu, S. The Effects and the Potential Mechanism of Environmental Transformation of Metal Nanoparticles on Their Toxicity in Organisms. *Environ. Sci.: Nano* **2018**, *5* (11), 2482−2499.
(55) Vance, M. E.; Marr, L. C. Exposure to Airborne Engineered Nanoparticles in the Indoor Environment. *Atmos. Environ.* **2015**, *106*, 503−509.
(56) Long, X.; Luo, Y.; Zhang, Z.; Zheng, C.; Zeng, C.; Bi, Y.; Zhou, C.; Rittmann, B. E.; Waite, T. D.; Herckes, P.; Westerhoff, P. The Nature and Oxidative Reactivity of Urban Magnetic Nanoparticle Dust Provide New Insights into Potential Neurotoxicity Studies. *Environ. Sci. Technol.* **2020**, *54* (17), 10599−10609.
(57) Gonet, T.; Maher, B. A. Airborne, Vehicle-Derived Fe-Bearing Nanoparticles in the Urban Environment: A Review. *Environ. Sci. Technol.* **2019**, *53* (17), 9970−9991.
(58) Valko, M.; Leibfritz, D.; Moncol, J.; Cronin, M. T. D.; Mazur, M.; Telser, J. Free Radicals and Antioxidants in Normal Physiological Functions and Human Disease. *International Journal of Biochemistry & Cell Biology* **2007**, *39* (1), 44−84.
(59) Zhang, Y.; Demokritou, P.; Ryan, D. K.; Bello, D. Comprehensive Assessment of Short-Lived ROS and H2O2 in Laser Printer Emissions: Assessing the Relative Contribution of Metal Oxides and Organic Constituents. *Environ. Sci. Technol.* **2019**, *53* (13), 7574−7583.
(60) Wang, W.; Lin, Y.; Yang, H.; Ling, W.; Liu, L.; Zhang, W.; Lu, D.; Liu, Q.; Jiang, G. Internal Exposure and Distribution of Airborne Fine Particles in the Human Body: Methodology, Current Understandings, and Research Needs. *Environ. Sci. Technol.* **2022**, DOI: 10.1021/acs.est.1c07051.
(61) 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* (6), 1293−1298.
(62) Liu, N. M.; Miyashita, L.; Maher, B. A.; McPhail, G.; Jones, C. J. P.; Barratt, B.; Thangaratinam, S.; Karloukovski, V.; Ahmed, I. A.; Aslam, Z.; Grigg, J. Evidence for the Presence of Air Pollution Nanoparticles in Placental Tissue Cells. *Science of The Total Environment* **2021**, *751*, 142235.
(63) Maher, B. A.; González-Maciel, A.; Reynoso-Robles, R.; Torres-Jardón, R.; Calderón-Garcidueñas, L. Iron-Rich Air Pollution Nanoparticles: An Unrecognised Environmental Risk Factor for Myocardial Mitochondrial Dysfunction and Cardiac Oxidative Stress. *Environmental Research* **2020**, *188*, 109816.
(64) Calderón-Garcidueñas, L.; González-Maciel, A.; Reynoso-Robles, R.; Hammond, J.; Kulesza, R.; Lachmann, I.; Torres-Jardón, R.; Mukherjee, P. S.; Maher, B. A. Quadruple Abnormal Protein Aggregates in Brainstem Pathology and Exogenous Metal-Rich Magnetic Nanoparticles (and Engineered Ti-Rich Nanorods). The Substantia Nigrae Is a Very Early Target in Young Urbanites and the Gastrointestinal Tract a Key Brainstem Portal. *Environmental Research* **2020**, *191*, 110139.
(65) Gonet, T.; Maher, B. A.; Kukutschová, J. Source Apportionment of Magnetite Particles in Roadside Airborne Particulate Matter. *Science of The Total Environment* **2021**, *752*, 141828.

(66) Li, S.; Zhang, B.; Wu, D.; Li, Z.; Chu, S.-Q.; Ding, X.; Tang, X.; Chen, J.; Li, Q. Magnetic Particles Unintentionally Emitted from Anthropogenic Sources: Iron and Steel Plants. *Environ. Sci. Technol. Lett.* **2021**, *8*, 295.
(67) McWhinney, R. D.; Badali, K.; Liggio, J.; Li, S.-M.; Abbatt, J. P. D. Filterable Redox Cycling Activity: A Comparison between Diesel Exhaust Particles and Secondary Organic Aerosol Constituents. *Environ. Sci. Technol.* **2013**, *47* (7), 3362−3369.
(68) Huang, H.; Jiang, Y.; Xu, X.; Cao, X. In Vitro Bioaccessibility and Health Risk Assessment of Heavy Metals in Atmospheric Particulate Matters from Three Different Functional Areas of Shanghai, China. *Science of The Total Environment* **2018**, *610−611*, 546−554.
(69) He, A.; Li, X.; Ai, Y.; Li, X.; Li, X.; Zhang, Y.; Gao, Y.; Liu, B.; Zhang, X.; Zhang, M.; Peng, L.; Zhou, M.; Yu, H. Potentially Toxic Metals and the Risk to Children's Health in a Coal Mining City: An Investigation of Soil and Dust Levels, Bioaccessibility and Blood Lead Levels. *Environ. Int.* **2020**, *141*, 105788.
(70) Calderón-Garcidueñas, L.; Ayala, A. Air Pollution, Ultrafine Particles, and Your Brain: Are Combustion Nanoparticle Emissions and Engineered Nanoparticles Causing Preventable Fatal Neurodegenerative Diseases and Common Neuropsychiatric Outcomes? *Environ. Sci. Technol.* **2022**, DOI: 10.1021/acs.est.1c04706.
(71) Jiang, G.; Wang, C.; Song, L.; Wang, X.; Zhou, Y.; Fei, C.; Liu, H. Aerosol Transmission, an Indispensable Route of COVID-19 Spread: Case Study of a Department-Store Cluster. *Front. Environ. Sci. Eng.* **2021**, *15* (3), 46.

https://doi.org/10.1021/acs.estlett.2c00192
Environ. Sci. Technol. Lett. XXXX, XXX, XXX−XXX