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Letter

3D Imaging of Roadside and Underground Railway Particulate Matter Structures

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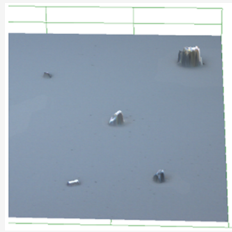
ABSTRACT: Air pollution in cities is associated with adverse health conditions; however, the underlying reasons are poorly understood. Previous particulate matter (PM) imaging studies have mostly used 2D microscopic techniques. In this study, the 3D appearances of roadside and underground railway station airborne PMs and the elemental characteristics of underground station PM were examined. Roadside and underground station PMs were collected using a cyclone particulate matter collection system. The 3D structures of the PMs were examined using an Olympus LEXT OLS4100 confocal microscope. Elemental characteristics of underground railway station PM were evaluated with a Zeiss EVO 50 scanning electron microscope (SEM) in backscattered electron (BSE) mode. Confocal microscopy of both PM sources indicated some particles can have sharp appearing edges. The longest axis length of roadside particles had a median (range) of 4.0 (1.3 to 6.6) μm and height of 1.4 (0.3 to 2.1) μm. SEM imaging of underground railway station PM was consistent in appearance and size with confocal imaging; BSE mode indicated compositions primarily of iron, calcium, and silicon. In conclusion, we observed that some roadside and underground railway station PMs can have sharp appearing surfaces from 3D confocal microscopy. SEM imaging of underground railway station PM was consistent with confocal microscopy and enabled elemental analysis.



Confocal microscope image of roadside Particulate Matter

KEYWORDS: *particulate matter, roadside, confocal microscopy, scanning electron microscopy, X-ray microanalysis, air pollution*

■ INTRODUCTION

Air pollution has been associated with increased risk of premature mortality.[1−3] Particulate matter (PM) is a key component of ambient pollution,[4] and elevated PM concentrations are associated with adverse health conditions such as asthma and lung cancer.[5−7] The effects of PM are not restricted to the lungs; for example, inhalable PM has recently been reported in both brain and heart tissues.[8,9] However, little is known about the mechanisms of action of PM that result in harmful effects on health leading to increased mortality and morbidity.

Previous studies of PM have mostly used 2D light microscopy, scanning electron microscopy (SEM), or transmission electron microscopy (TEM). However, 3D confocal microscopy can provide important additional structural details compared with the 2D techniques as it allows the combination of true color information with 3D visualization.[10,11] In a recent confocal imaging study, we observed that diesel particulate matter (DPM), also known as diesel exhaust particulate (DEP), can have a sharp jagged surface appearance.[11]

Roadside particles consist of combustion-derived PM as well as PM from other sources such as those associated with road construction properties, tire particles, and brake dust.[12] A recent study using SEM of particles deposited on leaves suggested that roadside PM can have a more spherical appearance compared with particles from other sources,[13] although that study did identify a range in particle shape and size. Another recent SEM-based study of DEP and brake dust particles from testing machinery observed that, following methanol extraction, larger brake dust particles can have a jagged appearance.[14]

2D light, scanning electron, and laser confocal microscopies provide structural details about the particles; however, health-related effects of PM may also be influenced by the elemental compositions of these particles. Previous studies have, for example, looked at the composition of PM in the London underground railway system and compared it with above ground measurements in London.[15]

Our study aimed to investigate structural characteristics of particulate matter collected by the side of a traffic-busy London road as well as 3D structure and elemental characteristics of particulate matter from a London underground railway station.

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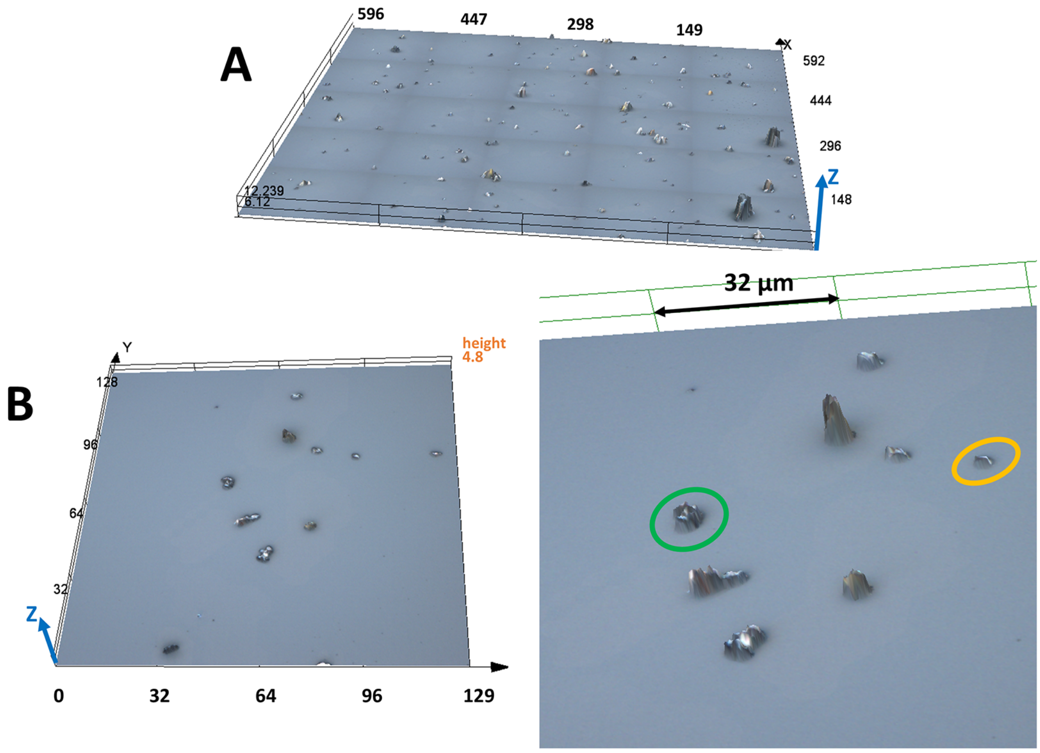

Figure 1. Example of confocal microscope imaging of roadside particles. Panel A shows a 3D representation image of particles on a glass slide consisting of 25 individual images stitched together to form a total area of 592 μm × 596 μm. Panel B shows a 128 μm × 128 μm 3D image of particles and panel C a close-up of some of the particles in panel B showing that some particles appear to have sharp or jagged edges. In panel C, a PM_{10} particle with the longest and perpendicular lengths of 4.9 μm × 4.6 μm and of height 1.3 μm is shown surrounded by a green ellipse, and a $PM_{2.5}$ particle with dimensions of 2.4 μm × 2.3 μm and of height 0.7 μm is shown surrounded by an orange ellipse (on right). In panels A and C, the z axis is magnified by 2 in order to show the 3D shape more clearly.

MATERIALS AND METHODS

Sample Collection. Air pollution-derived particulate matter (PM) less than 10 μm in size (PM_{10}) was collected as dry particles on a major London road (Marylebone Road) and a London underground railway station (Baker Street Station) using a high-volume cyclone[16] without a PM_{10} size separation inlet device. For the roadside assessment, the cyclone was placed on the pavement at ground level and approximately 1 m from the curbside; for both the roadside and the underground station measurements, the air intake height was 30 cm above ground level. PM was collected over a period of 5−8 h per day, over 10 days during summer 2019. PM_{10} samples were pooled and stored at room temperature in a sterile glass container.

Slide Preparation for Confocal Microscopy. A clean slide was placed in a Petri dish and covered. A few grains of the collected aggregated roadside particles were put in a small glass vial, and a few drops of propan-2-ol were added. The particles then separated immediately to form a temporary suspension. The vial was agitated and some of the suspension extracted using a 1 mL syringe with a needle. A droplet of the suspension was put on the glass slide which was then covered with a Petri dish lid. The droplet containing the particles in suspension dispersed across the surface of the glass slide, and the propan-2-ol was allowed to evaporate naturally at room temperature over a period of 20 min to leave just the particles on the glass slide.

In order to investigate whether the shapes of particles could be affected by the slide preparation technique, we also prepared slides for confocal microscopy using double-sided adhesive tabs used in scanning electron microscopy in a similar fashion to that we have previously described in a diesel particulate matter imaging study.[11] In essence, the steps were as follows: a piece of filter paper was placed in a plastic Petri dish with a lid. A sample of the underground railway PM was sprinkled on the filter paper using a small spatula and the lid replaced. The lid of the Petri dish was tapped to help reduce the size of the clumped charged particles which causes the finer particles to be attracted to the underside of the plastic lid. A double-sided self-adhesive circular carbon tab of a diameter of 12 mm was attached to a glass slide and placed in the center of the Petri dish and the lid replaced. Tapping the lid caused the particles, including $PM_{2.5}$ and PM_{10}, on the lid to fall onto the tab.

Slide Preparation for Scanning Electron Microscopy. Samples of PM particles collected in the underground railway station were suspended in propan-2-ol and sonicated with ultrasound for about 5 min to separate the particles. A drop of the particles in suspension was placed on a glass coverslip, which was mounted on a specimen stub, and allowed to air-dry overnight; the sample was then carbon coated to make the surface conductive.

Confocal Microscopy. The separated particles deposited onto the glass slide were imaged with an Olympus LEXT OLS4100 confocal microscope (Olympus Corporation, Japan) with a 100× objective lens. The confocal microscope has a 405 nm laser, and images were collected with a resolution of 1024 × 1024 pixels and with the 3D scanning in fine mode. 3D surface scanning images of PM_{10} and $PM_{2.5}$ particles were assessed for size in three dimensions as well as shape and color

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using Olympus LEXT microscope software. In addition the aspect ratio was calculated as the fraction of (longest length)/(perpendicular length).

Elemental Analysis Using Scanning Electron Microscopy of Underground Railway Station PM. Slides of underground railway station PM were examined with a Zeiss EVO 50 scanning electron microscope (SEM) in both the secondary and backscattered modes. Secondary electron imaging (SEI) was used for examining texture, morphology, size, and overall appearance. Detection of backscattered electrons (BSE) was used to assess differences in chemistry; brighter phases identified via BSE imagery were selected and qualitatively examined using energy dispersive X-ray microanalysis.

■ RESULTS AND DISCUSSION

Particles with sizes corresponding to both PM_{10} and $PM_{2.5}$ were imaged with the confocal microscopy technique from both the roadside and underground railway station samples. The 3D appearance showed that particles from both sites had a variety of shapes as seen for roadside particles in Figure 1 showing predominately examples of both PM_{10} and $PM_{2.5}$. Figure 1A shows a 3D image of roadside-collected particles on a glass slide consisting of 25 individual images each of 128 μm × 128 μm stitched together to form an image with total area of 592 μm × 592 μm. The particles in the image vary in size, shape, and color. The particles are predominantly black, gray, or white in color which in part may be due to lighting effects; however, in addition, some particles appear to have a yellow or orange tinge. Figure 1B shows a 128 μm × 128 μm 3D image of particles; a close-up of some of the particles is seen in Figure 1C with some particles appearing to have sharp or jagged edges. Again the color of particles varies with some being dark gray and others light gray or white; other particles have an orange or yellow tinge. An example of a PM_{10} particle is shown in Figure 1C surrounded by a green ellipse and a $PM_{2.5}$ particle surrounded by an orange ellipse.

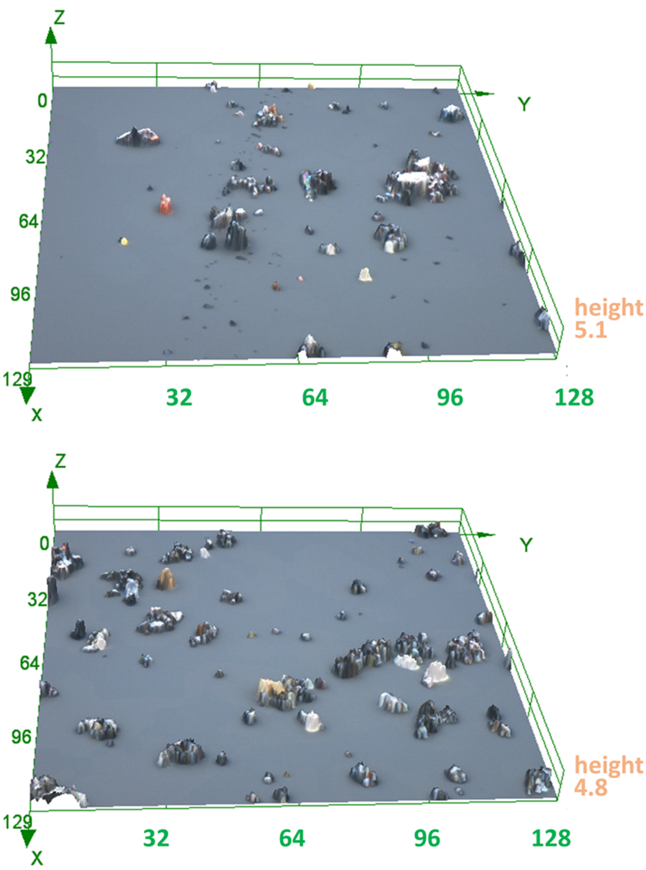

Figure 2. Examples of confocal microscope images of underground railway station particulate matter collected from two platforms. Each image shows a 128 μm × 128 μm 3D image of particles with different shapes and colors some of which appear to have sharp edges. The z axes of the images are magnified by 2 in order to show the 3D shape more clearly.

In terms of variation in size and shape, from 10 roadside particles, the median (range) longest length was 4.0 (1.3 to 6.6) μm, perpendicular length 2.1(1.2 to 5.6) μm, and height 1.4 (0.3 to 2.1) μm. The median (range) aspect ratio was 1.3 (1.0 to 2.3).

Examples of confocal microscopy imaging of underground railway station PM collected on two different platforms are shown in Figure 2 using slides prepared with propan-2-ol. As in the roadside PM images, there is a range of size with some particles appearing to have sharp edges. The sharp appearance of some particles was not obvious to discern from 2D images alone as seen in the example in Figure 3A when compared with the equivalent 3D image in Figure 3B. Similar types of particles' appearances were seen in slides prepared with the double-sided adhesive circular tabs to the propan-2-ol method. However, there was more particle agglomeration apparent in all directions x, y, and z with the tab approach, and so fewer PM_{10} and below particles were seen as shown in the example in Figure 3C and D.

SEM secondary electron imaging of underground railway station PM also showed a variety of shapes in particles of similar dimensions to that found on confocal imaging as seen in the example in Figure 4. Backscattered electron spectra from ten railway PM_{10} particles selected at random showed the presence predominantly of silicon, calcium, and iron as seen in the example spectra in Figure 4. Some particles also showed the presence of carbon, although this element can be difficult to detect using this approach because of the coating of the sample with carbon. Although it is possible that the ultrasound sonication used in preparing particles for SEM imaging and backscattered electron spectra could have affected their shapes, the variety of shapes seen were consistent with the confocal microscopy images; furthermore, it seems unlikely that the SEM particle preparation method would affect the shapes of particles with metallic elements identified using energy dispersive X-ray microanalysis.

In this study, confocal microscopy was successfully applied to investigate the structure, size, and appearance of particulate matter (PM) collected beside a busy London road as well as two platforms at a London underground railway station. It has been previously shown that the composition of PM changes between types of workplace.[17] The jagged nature of some particles may not be clear on 2D imaging alone which may explain why some SEM studies have suggested roadside particles may have a more spherical appearance;[13] nevertheless, our SEM imaging of underground railway station PM was consistent with confocal microscopy in showing that some particles can have sharp appearing edges. The geometries of microparticles have previously been shown to affect interaction with cancer cells;[18] in that study, microparticles were found not to enter cells if the surface in contact was round, whereas

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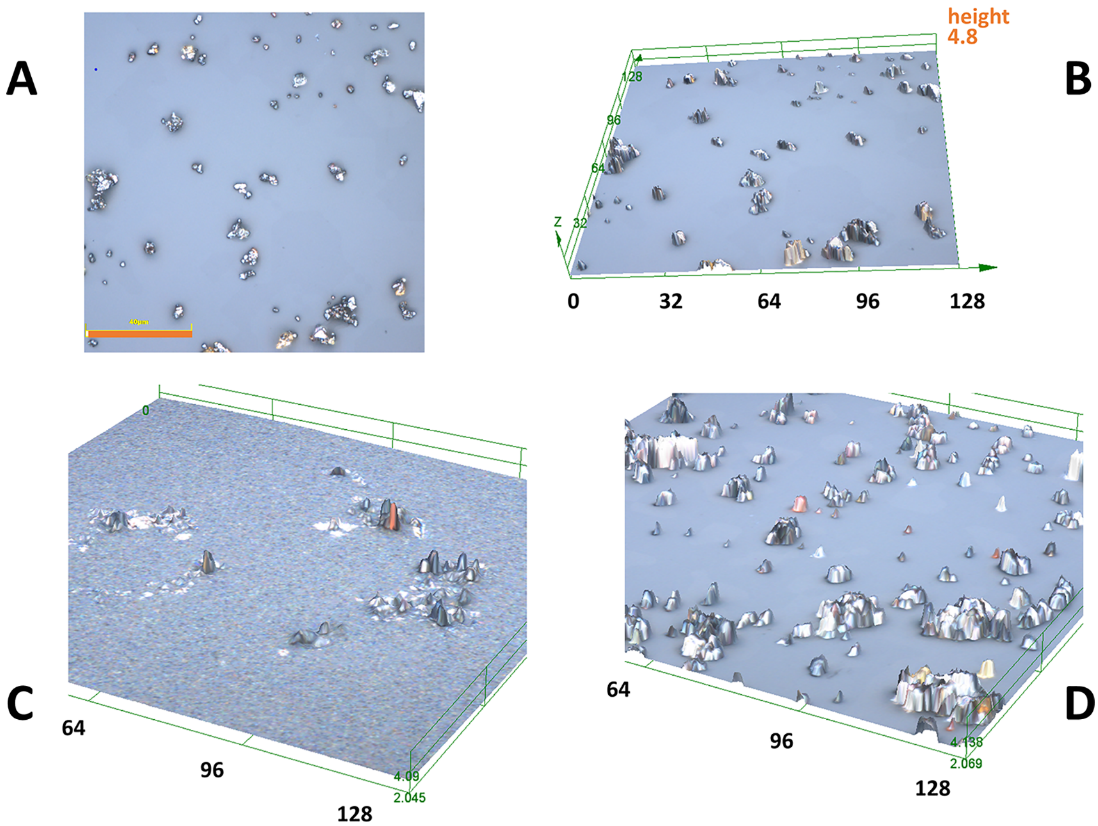

Figure 3. Top row shows an example of a 2D image (A) and a corresponding 3D confocal microscope image (B) each of size 128 μm × 128 μm using a 100× objective lens showing particles of underground railway station particulate matter with different shapes and colors some of which appear to have sharp edges in the 3D image, but this cannot clearly be discerned from the 2D image. The lower part of the figure shows examples of confocal microscope images of underground railway station particulate matter using the double-sided adhesive tab method (C) and the propan-2-ol slide preparation method (D). The z axes of the 3D images are magnified by 2 in order to show the 3D shape more clearly.

sharp particles could attach to cells. Furthermore, the particle appearance on SEM seen in our study is consistent with images previously reported.[19,20]

The type of PM can vary according to height from the ground as seen in a study of PM at pram and adult heights.[21] As roadside particulate matter is heterogeneous and may originate from a number of sources,[12] methods of characterizing the particles may help in identifying their origin. Size and shape may give some indication, but the technique applied in this study also allows their color to be studied. It is not clear whether color of the PM could be reliably used to help characterize particles or identify the likely origin. In addition, without true color imaging, it may be difficult to appreciate that the origin of individual particles could be different.

Unlike regular particle collections that use a filter to catch the particulate matter, the high-volume cyclone collects particulate matter as dry respirable powder directly into a sterile container. As a result, particles are ready to be utilized without the need for extraction, and so we believe that particle morphology is unlikely to change as a result of the air sampling technique. As the sampled air is passed over a Teflon cup, it is possible that some smaller particles may be lost in the PM acquisition procedure due to electrostatic forces. We observed apparent agglomeration of particles which may be related to electrostatic effects. The confocal microscope slides prepared with propan-2-ol appeared to show well-dispersed small particles, whereas those using the double-sided pads showed more particle agglomeration. We assume that the propan-2-ol technique is unlikely to affect the morphologies of particles, and this is supported by the consistent small particle range of appearance seen with the slides prepared using double-sided adhesive tabs. We thus assume propan-2-ol is unlikely to affect particle appearance as well as based on its typical usage; for example, it can be used in preparing carbon-based paints for conductive films.[22,23]

In accordance with earlier studies,[15,24] we observed the presence of particles with elemental characteristics suggestive of iron on BSE analysis of underground railway station dust; this perhaps is not surprising given the proximity to rails particularly as PM acquisition was close to station floor level. Furthermore, the magnetic property of iron containing PM opens up the possibility of applying magnetic filters to remove such particles.[25] The roadside PM was collected near the curbside of a major London road with approximately 80,000 vehicles daily.[26] It is possible that the elemental characteristics of PM will be affected by the height from the ground primarily because of density, and this needs further investigation.

In summary, the results of this study suggest that some roadside and underground railway station PM_{10} and $PM_{2.5}$ particles collected 30 cm above ground level have a sharp or jagged appearance which may help explain their ability to tightly adhere to and infiltrate the respiratory system and thus affect health. Further studies are required to examine whether such particle characteristics vary with height from ground level and particularly at heights consistent with breathing for children and adults.

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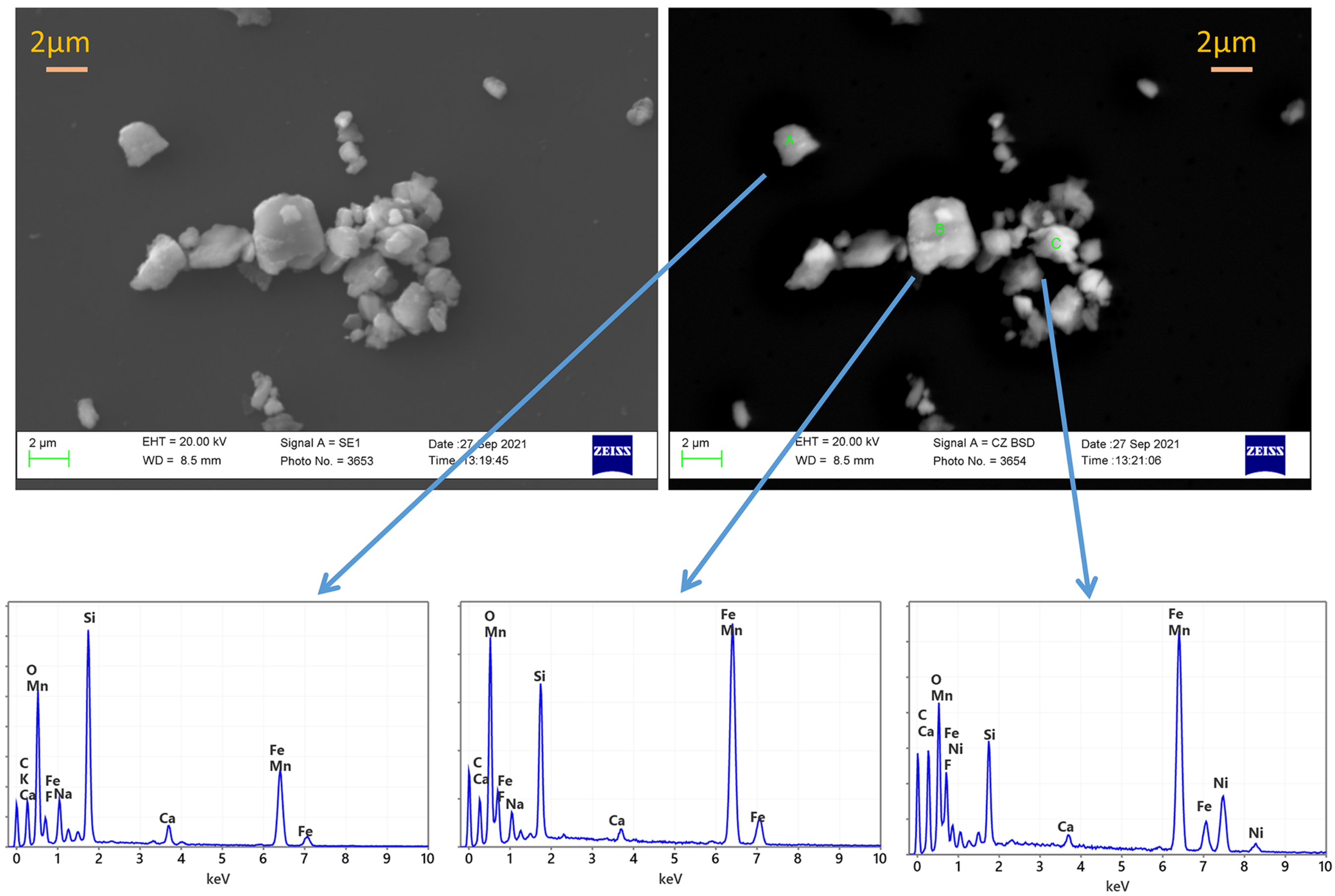

Figure 4. Scanning electron microscopy images and spectra from three separate particles (image on left is a secondary electron image; image on right is a backscattered electron image). The spectra show peaks predominantly consistent with iron (Fe), calcium (Ca), and silicon (Si).

■ ASSOCIATED CONTENT

Data Availability Statement

The data sets used and/or analyzed during the current study are available from the corresponding author on reasonable request.

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Author Contributions

All authors contributed to devising the study as well as design. Ian Gill, Richard Giddens, Lisa Miyashita, and Gary Foley prepared slides used in this study. Lisa Miyashita, Gary Foley, Richard Giddens, Gavin Gillmore, and David Wertheim collected microscope image data used in this study. Ana Rule helped design the cyclone used for particulate matter sample collection. All authors read and approved the final version of the manuscript.

Notes

The authors declare the following competing financial interest(s): L.M., G.F., I.G., R.G., G.G., R.D., S.S., A.R., and

D.W. have no competing interests. Professor Grigg received personal fees from AstraZeneca, GSK, Novartis, Vifor Pharma, OM Pharma, and Omron outside the submitted work.

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■ REFERENCES

(1) Cohen, A. J.; Brauer, M.; Burnett, R.; Anderson, H. R.; Frostad, J.; Estep, K.; Balakrishnan, K.; Brunekreef, B.; Dandona, L.; Dandona, R.; Feigin, V.; Freedman, G.; Hubbell, B.; Jobling, A.; Kan, H.; Knibbs, L.; Liu, Y.; Martin, R.; Morawska, L.; Pope, C. A., 3rd; Shin, H.; Straif, K.; Shaddick, G.; Thomas, M.; van Dingenen, R.; van Donkelaar, A.; Vos, T.; Murray, C. J. L.; Forouzanfar, M. H. Estimates and 25-year trends of the global burden of disease attributable to ambient air pollution: an analysis of data from the Global Burden of Diseases Study 2015. *Lancet.* **2017**, *389* (10082), 1907−1918.

(2) Goodkind, A. L.; Tessum, C. W.; Coggins, J. S.; Hill, J. D.; Marshall, J. D. Fine-scale damage estimates of particulate matter air pollution reveal opportunities for location-specific mitigation of emissions. *Proc. Natl. Acad. Sci. U. S. A.* **2019**, *116*, 8775−8780.

(3) Khomenko, S.; Cirach, M.; Pereira-Barboza, E.; Mueller, N.; Barrera-Gómez, J.; Rojas-Rueda, D.; de Hoogh, K.; Hoek, G.; Nieuwenhuijsen, M. Premature mortality due to air pollution in European cities: a health impact assessment. *Lancet Planet Health.* **2021**, *5* (3), e121−e134.

(4) Abraham, S.; Brugha, R.; Grigg, J. Particulate air pollution exposure using modes of urban transport. *Archives of Disease in Childhood* **2014**, *99*, A40−A41.

(5) Gehring, U.; Wijga, A. H.; Koppelman, G. H.; Vonk, J. M.; Smit, H. A.; Brunekreef, B. Air pollution and the development of asthma from birth until young adulthood. *Eur. Respir. J.* **2020**, *56* (1), 2000147.

(6) Holst, G. J.; Pedersen, C. B.; Thygesen, M.; Brandt, J.; Geels, C.; Bønløkke, J. H.; Sigsgaard, T. Air pollution and family related determinants of asthma onset and persistent wheezing in children: nationwide case-control study. *BMJ.* **2020**, *370*, m2791.

(7) Zhang, Z.; Zhu, D.; Cui, B.; Ding, R.; Shi, X.; He, P. Association between particulate matter air pollution and lung cancer. *Thorax* **2020**, *75*, 85−87.

(8) Maher, B. A.; Ahmed, I. A.; Karloukovski, V.; MacLaren, D. A.; Foulds, P. G.; Allsop, D.; Mann, D. M.; Torres-Jardón, R.; Calderon-Garciduenas, L. Magnetite pollution nanoparticles in the human brain. *Proc. Natl. Acad. Sci. U. S. A.* **2016**, *113* (39), 10797−801.

(9) Calderón-Garcidueñas, L.; González-Maciel, A.; Mukherjee, P. S.; Reynoso-Robles, R.; Pérez-Guillé, B.; Gayosso-Chávez, C.; Torres-Jardón, R.; Cross, J. V.; Ahmed, I. A. M.; Karloukovski, V. V.; Maher, B. A. Combustion- and friction-derived magnetic air pollution nanoparticles in human hearts. *Environ. Res.* **2019**, *176*, 108567.

(10) Wertheim, D.; Gillmore, G.; Gill, I.; Petford, N. High resolution 3D confocal microscope imaging of volcanic ash particles. *Sci. Total Environ.* **2017**, *590−591*, 838−842.

(11) Miyashita, L.; Foley, G.; Gill, I.; Gillmore, G.; Grigg, J.; Wertheim, D. Confocal microscopy 3D imaging of diesel particulate matter. *Environ. Sci. Pollut Res. Int.* **2021**, *28*, 30384−30389.

(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) Castanheiro, A.; Wuyts, K.; Hofman, J.; Nuyts, G.; De Wael, K.; Samson, R. Morphological and elemental characterization of leaf-deposited particulate matter from different source types: a microscopic investigation. *Environ. Sci. Pollut Res. Int.* **2021**, *28*, 25716.

(14) Selley, L.; Schuster, L.; Marbach, H.; Forsthuber, T.; Forbes, B.; Gant, T. W.; Sandström, T.; Camiña, N.; Athersuch, T. J.; Mudway, I.; Kumar, A. Brake dust exposure exacerbates inflammation and transiently compromises phagocytosis in macrophages. *Metallomics.* **2020**, *12* (3), 371−386.

(15) Smith, J. D.; Barratt, B. M.; Fuller, G. W.; Kelly, F. J.; Loxham, M.; Nicolosi, E.; Priestman, M.; Tremper, A. H.; Green, D. C. PM2.5 on the London Underground. *Environ. Int.* **2020**, *134*, 105188.

(16) Rule, A. M.; Geyh, A. S.; Ramos-Bonilla, J. P.; Mihalic, J. N.; Margulies, J. D.; Polyak, L. M.; Kesavan, J.; Breysse, P. N. Design and characterization of a sequential cyclone system for the collection of bulk particulate matter. *J. Environ. Monit.* **2010**, *12* (10), 1807−14.

(17) da Silveira Fleck, A.; Catto, C.; L'Espérance, G.; Masse, J. P.; Roberge, B.; Debia, M. Characterization and Quantification of Ultrafine Particles and Carbonaceous Components from Occupational Exposures to Diesel Particulate Matter in Selected Workplaces. *Ann. Work Expo Health.* **2020**, *64*, 490−502.

(18) He, Y.; Park, K. Effects of the Microparticle Shape on Cellular Uptake. *Mol. Pharmaceutics* **2016**, *13*, 2164−71.

(19) Seaton, A.; Cherrie, J.; Dennekamp, M.; Donaldson, K.; Hurley, J. F.; Tran, C. L. The London Underground: dust and hazards to health. *Occupational and Environmental Medicine* **2005**, *62*, 355−362.

(20) Loxham, M.; Cooper, M. J.; Gerlofs-Nijland, M. E.; Cassee, F. R.; Davies, D. E.; Palmer, M. R.; Teagle, D. A. Physicochemical characterization of airborne particulate matter at a mainline underground railway station. *Environ. Sci. Technol.* **2013**, *47*, 3614−22.

(21) Sharma, A.; Kumar, P. Quantification of air pollution exposure to in-pram babies and mitigation strategies. *Environ. Int.* **2020**, *139*, 105671.

(22) Karuppannan, S. K.; Neoh, E. H. L.; Vilan, A.; Nijhuis, C. A. Protective Layers Based on Carbon Paint To Yield High-Quality Large-Area Molecular Junctions with Low Contact Resistance. *J. Am. Chem. Soc.* **2020**, *142* (7), 3513−3524.

(23) Shukla, S.; Bhattacharjee, S.; Weber, A. Z.; Secanell, M. Experimental and Theoretical Analysis of Ink Dispersion Stability for Polymer Electrolyte Fuel Cell Applications. *Journal of The Electrochemical Society.* **2017**, *164*, F600−F609.

(24) Luglio, D. G.; Katsigeorgis, M.; Hess, J.; Kim, R.; Adragna, J.; Raja, A.; Gordon, C.; Fine, J.; Thurston, G.; Gordon, T.; Vilcassim, M. J. R. PM2.5 Concentration and Composition in Subway Systems in the Northeastern United States. *Environ. Health Perspect.* **2021**, *129*, 027001.

(25) 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*, 331−40.

(26) 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.