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Are black carbon and soot the same?

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Are black carbon and soot the same?

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Abstract

The climate change and environmental literature, including that on aerosols, is replete with mention of black carbon (BC), but neither reliable samples nor standards exist. Thus, there is uncertainty about its exact nature. That confusion can be avoided if terms are defined and widely understood. Here we discuss an ambiguity between BC and soot and propose a more precise definition for soot as a specific material, which we call ns-soot, where “ns” refers to carbon nanospheres. We define ns-soot as particles that consist of nanospheres, typically with diameters < 100 nm, that possess distinct structures of concentrically wrapped, graphene-like layers of carbon and with grape-like (acinoform) morphologies. We additionally propose that, because of their importance for climate modeling and health issues, distinctions are made among bare, coated, and embedded ns-soot. BC, on the other hand, is not a well-defined material. We propose that the term should be restricted to light-absorbing refractory carbonaceous matter of uncertain character and that the uncertainty is stated explicitly.

1 Introduction

Carbonaceous aerosol particles are important for climate, visibility, and health. They are major contributors to light absorption and thus radiative heating of the atmosphere. Many papers discuss the various types of such particles, but the terminology is confusing. Here we propose that a clearer definition of carbonaceous particle types and improved terminology can be established based on the results of transmission electron microscopy (TEM) analyses of individual aerosol particles.

We previously identified spherical, amorphous, carbonaceous particles in the smoke of biofuel and biomass fires and termed them “tar balls” (Pósfai et al., 2004). The identification and characterization of this widespread particle type led to subsequent studies that determined their compositions (Adachi and Buseck, 2011; Tivanski et al., 2007), hygroscopic properties (Semeniuk et al., 2007), and complex refractive index

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(Alexander et al., 2008; Chakrabarty et al., 2010), illustrating the utility of TEM-based categorization of particle types. In the present study, we focus on the major forms of light-absorbing carbonaceous aerosol particles, which are commonly called soot and black carbon (BC). They have uneven usage and different meanings (Andreae and Gelencsér, 2006; Bond and Bergstrom, 2006).

IPCC (2007), the definitive work on climate change, uses the definitions of soot and BC of Charlson and Heintzenberg (1995), which are, respectively, “Particles formed during the quenching of gases at the outer edge of flames of organic vapours, consisting predominantly of carbon, with lesser amounts of oxygen and hydrogen present as carboxyl and phenolic groups and exhibiting an imperfect graphitic structure” and “Operationally defined aerosol species based on measurement of light absorption and chemical reactivity and/or thermal stability; consists of soot, charcoal and/or possible light absorbing refractory organic matter”.

These definitions are based largely on origin and measurement type, respectively, rather than on material properties. More specifically, the definition of BC is unacceptably vague as a material description. In the section Changes in Atmospheric Constituents and Radiative Forcing, IPCC (2007) uses “BC” instead of “soot”, e.g., “Electron microscope images of BC particles show that they are emitted as complex chain structures (e.g., Pósfai et al., 2003) . . . ,” even though the paper by Pósfai et al. (2003) is mainly about soot and only mentions BC parenthetically and in passing. Thus, although soot and BC are used interchangeably by IPCC and others, we question the validity of that usage and conclude there is neither a clear consensus nor justification for they way they are used in much of the current literature.

BC in the atmospheric sciences is the term used for materials that are strongly light absorbing, commonly determined by the attenuation of light by particles typically collected onto a filter and measured by an aethalometer (Hansen et al., 1984), although it is also measured using other methods (Table 1). A widely used approach for measuring BC is, contrary to its name, the Single Particle Soot Photometer (SP2) (Schwarz et al., 2008). There are references in the SP2 literature to “refractory BC” or “rBC,” but even

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though the latter excludes non-refractory phases it is still not a clearly identifiable material. In the absence of an accepted BC standard, surrogate soot-like materials such as fullerene, graphite, and Aquadag are used as standards (e.g., Kondo et al., 2011; Gysel et al., 2011; Laborde et al., 2012; Baumgardner et al., 2012). Although BC brings to mind a material and is commonly used in the aerosol literature as if it were a specific material, laboratory samples of atmospheric BC that can be measured do not exist.

Soot, on the other hand, is the solid, carbon-rich material that condenses from the vapor phase during combustion (Fig. 1) (Ishiguro et al., 1997; Richter and Howard, 2000). Much aerosol usage has this as equivalent to BC (Jacobson, 2001; Penner and Novakov, 1996), with the implicit assumption that the sole source of the optical absorption is soot, although a cause of confusion is that one term applies to origin and the other to optical measurements.

No consensus exists about whether soot is a part of BC, BC is a part of soot, or BC is equivalent to soot. The definition used by IPCC indicates that soot is a part of BC. The usage of soot in electron microscopy studies commonly agrees with that of the IPCC (Adachi et al., 2010). In contrast, in other papers (e.g., Bond and Bergstrom, 2006; Seinfeld and Pandis, 2006; Salamanca et al., 2012), soot consists of BC and other combustion-generated carbonaceous materials, and Ramanathan and Carmichael (2008) wrote that “BC refers to the absorbing components of soot, often defined using elemental carbon and some condensed organics”.

Elemental carbon (EC) is another term used in the aerosol community. It is essentially a high-T residue of thermal-optical measurements. EC is commonly measured together with organic carbon (OC), with the latter being the low-T fraction (Table 1).

An additional complication is that these terms are used differently in various scientific disciplines. BC is widely used in the environmental communities dealing with sediments and soils for carbonaceous substances of pyrogenic origin that are resistant to thermal or chemical degradation. Such BC includes char, charcoal, and soot (International Steering Committee for Black Carbon Reference Materials; Preston and Schmidt, 2006). This BC definition is unrelated to instrumental measurements of optical

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properties, i.e., the BC of atmospheric scientists and the BC of sedimentologists and soil scientists are not the same (Masiello, 2004). The combustion community mainly uses soot instead of BC to describe carbonaceous combustion products that condense from aromatic hydrocarbons and have a characteristic microstructure and morphology (Maricq, 2007; Ishiguro et al., 1997; Lapuerta et al., 2012). Watson and Valberg (2001) report that several morphologically distinct types of particulate carbon occur in soot, and Palotás et al. (1996) refer to different types of soots.

Bond and Bergstrom (2006) discussed the terminological issue and proposed grouping carbonaceous materials that “strongly” absorb light in the visible range as light-absorbing carbon, LAC for short. LAC was also used by Andreae and Gelencsér (2006) but with a different meaning, since their LAC includes all absorbing carbonaceous particles, even “weakly” absorbing brown carbon (BrC). We favor the use of LAC as intentionally being purely operational and thus neutral to both material type and origin. However, a material descriptor would be highly useful for atmospheric science in general, as well as uses such as source apportionment, assessment of particle health effects, and modeling radiative effects. Using transmission electron microscopy (TEM), we morphologically and structurally define nanosphere-soot (ns-soot), a major carbonaceous aerosol type.

2 Proposed ns-soot terminology

As pointed out above, the term soot is used for both combustion products as a group and for a material with specific, defined properties. TEM has been widely used for studying this material. Although we have referred to soot extensively in our papers (Buseck and Pósfai, 1999; Pósfai et al., 1999, 2003, 2004; Li et al., 2003; Buseck and Schwartz, 2003; van Poppel et al., 2005; Adachi et al., 2007, 2010; Adachi and Buseck, 2008; Pósfai and Buseck, 2010; Li et al., 2010), we believe it is appropriate to draw a clear distinction based on physical features rather than origin or optical properties for at least this one important type of aerosol particle.

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We propose the term “ns-soot” for particles with grape-like (acinoform) morphologies that consist of nanospheres that possess distinct internal structures of concentrically wrapped, graphene-like layers of carbon. The nanosphere diameters are typically between 10 and 100 nm (e.g., ~ 44 nm for ambient ns-soot particles from Mexico City; Adachi and Buseck, 2008). Regardless of their source, from fewer than 10 to roughly 100 nanospheres typically occur in ns-soot particles in the atmosphere.

Terms such as “primary particles” or “monomers” have been used for carbon nanospheres (e.g., Vander Wal et al., 1999; Liu et al., 2008), but these terms depend on the aggregate form of the particles and do not indicate their chemical and structural features. Using the term carbon nanosphere, we can discuss the fundamental optical, chemical, and thermal properties of atmospheric ns-soot regardless of its aggregated shapes and coatings. Once such properties are known, the influence of aggregate morphology and internal mixing on optical and hygroscopic properties should be taken into account in order to model the atmospheric effects of ns-soot.

High-resolution TEM images of carbon nanospheres show the poorly ordered, concentrically wrapped, wavy or curved graphene-like layers that resemble onion-like structures (Fig. 1c). The irregular stacking of these layers has been called turbostratic (Palotás et al., 1996), consistent with such nonuniform stacking in sheet silicates. Such carbon structures, and their electron energy-loss spectrometry (EELS) signals (Fig. 2a), indicate that carbon nanospheres consist largely of layered, π -bonded carbon that absorb visible light.

The lack of distinct reflections in selected-area electron diffraction (SAED) patterns indicates the poorly ordered character of the carbon nanospheres. However, three blurred rings typically occur in SAED patterns as a result of short-range order within and between the graphene-like layers (Fig. 2b). The ~ 0.36 -nm lattice spacing that corresponds to the first ring can be matched to but is larger than the (002) crystal spacing in graphite, the distance between two neighboring graphene layers. First-neighbor atomic distances calculated from SAED patterns of carbon nanospheres indicate that

the carbon atoms within the graphene-like layers are closer to one another than in graphite (Kis et al., 2006).

The basic structural features of carbon nanospheres appear to be independent of their sizes and are not affected by aggregation or mixing with other atmospheric constituents. Although both the interpretation of fine structure in EELS spectra and the quantitative evaluation of electron-diffraction patterns are perhaps beyond the scope of standard TEM users, SAED patterns and HRTEM images (e.g., Figs. 2b and 1, respectively) can be obtained routinely and provide enough information for the unambiguous identification of ns-soot. Tar balls, which are also spherical, lack distinctive diffraction features and the wavy, graphene-like layers that are characteristic of ns-soot. Thus, the two can be readily distinguished.

3 Associated materials and coatings

The term soot has been used inconsistently in the atmospheric and combustion literature in the sense that in some instances it includes the embedding material (Salamanca et al., 2012) and in other instances it does not (Virtanen et al., 2004). Many ns-soot particles are coated with or embedded within other materials (e.g., organic matter, sulfate), forming internally mixed particles (Adachi et al., 2010) (Fig. 3).

Consistent with our definition of ns-soot above, we propose that only the “backbone” of internally mixed particles, i.e., the aggregates of carbon nanospheres, should be termed ns-soot. We find this restrictive definition practical, since it does not depend on whether the mixing of ns-soot with organics, sulfate, or other materials occurred at the time of particle formation or during the aging of ns-soot in the atmosphere.

Much ns-soot is either coated or embedded within other materials. The distinctions are somewhat arbitrary, but we propose that coated ns-soot has a thin (< 1 to ~ 10 nm) outer layer of a different material, whereas embedded ns-soot is thickly “coated” (Figs. 4, 5). Outlines of embedded ns-soot tend to be smooth and round, whereas those of coated ns-soot tend to be irregular and similar to bare ns-soot.

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TEM studies are typically performed under high-vacuum conditions that may result in the loss of volatile components. Thus, one could argue that ns-soot may be a definition for particles that do not even exist. However, atomic force microscopy (AFM) images obtained from vehicle emissions under ambient conditions clearly show that externally mixed, pure ns-soot particles do occur in the atmosphere, and both uncoated and coated ns-soot particles are emitted, depending on the combustion conditions (Fig. 6).

4 ns-soot vs. BC

Although not practical for monitoring purposes, TEM can be used for rapid spot checks of morphologically distinct carbonaceous particles, as is done with asbestos measurements. We suggest that if one assumes that BC even exists as a distinct material, which we question (Buseck et al., 2012), one should only speak of a “BC particle” if its material properties have been determined and it has been measured optically, and that combination has not yet been done. We further suggest that it is not acceptable to speak of “BC particles” unless the light absorption of the specific particles is known. In most instances, BC is applicable to an ensemble of carbonaceous materials that strongly absorb visible light (Fig. 4c). BC can contain ns-soot, commonly as a major constituent, but it can also include other light-absorbing carbonaceous materials. In contrast, all ns-soot is included within the BC (or rBC) category.

5 Issues related to ns-soot

Chemical analyses of combustion products are typically of bulk material, whether collected from flames or vehicle tailpipes, rather than the pure ns-soot considered here. Such analyses create a potential source of confusion. Although the dominant component of such bulk material is carbon, it also contains variable amounts of hydrogen, oxygen, and perhaps other elements. It is clear that ns-soot is a major component of

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this material, but it is less clear whether the elements other than carbon occur (a) within the structure of the ns-soot, (b) as coatings on its surfaces, or (c) in interstitial areas.

There seems to be a broad consensus in the literature that hydrogen is a component of carbon nanospheres, and both spectroscopic methods and electron diffraction indicate the presence of aromatics (Salamanca et al., 2012; Kis et al., 2006). However, it is unclear how much of the hydrogen, and perhaps small amounts of other non-carbon elements, are essential parts of the nanospheres or simply adventitious.

A related unresolved question is the origin of the curvature of the nanospheres. We assume that the fringes that are evident in high-resolution TEM images (Figs. 1c, 5b, 7a) are edge-on views of graphene-like layers containing mostly hexagonal rings of C. Pentagonal rings cause the curvature of fullerenes and similar materials and presumably occur in the carbon nanospheres in ns-soot. Chemical inhomogeneities can also contribute to the curvature. Hydrogen-bearing aromatics can disrupt the periodicity of graphene, resulting in layers that consist of non-periodically attached, misaligned islands of a few hexagonal rings of carbon (Fig. 7b). In the absence of a perfectly periodic crystalline structure, there is no constraint on the morphology, so that its layers can readily assume a spherical shape, resulting in the characteristic structure of carbon nanospheres.

The refractive index of ns-soot is not well known. Reasons for the difficulty of measurements are (1) ns-soot commonly coexists with other materials and (2) aggregates of carbon nanospheres contain many voids and interstices (van Poppel et al., 2005; Adachi et al., 2007). Bond and Bergstrom (2006) reviewed the issues related to the optical properties of strongly absorbing carbon (LAC) and showed a range of its refractive index. We believe that ns-soot is the main component of LAC, and therefore as a first approximation the refractive index determined for LAC can be used for ns-soot.

The detailed morphology of aggregated carbon nanospheres, i.e., ns-soot, varies from source to source and changes upon aging during its transport (e.g., Abel et al., 2003). Such aggregated shapes, which can be characterized using fractal dimensions,

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largely influence the radiative scattering of ns-soot in the atmosphere (Adachi et al., 2007; Mackowski, 1995).

6 Recommendations

We propose that strongly absorbing carbonaceous species in atmospheric aerosols should be considered, and treated in models, as follows:

6.1 ns-soot

We define ns-soot as particles that consist of nanospheres that possess distinct internal structures of concentrically wrapped, graphene-like layers of carbon and with grape-like (acinoform) morphologies.

Until there are better data, we recommend using the current best estimates for the refractive index and mass absorption cross section of LAC, i.e., $1.95-0.79i$ and $7.5 \pm 1.2 \text{ m}^2 \text{ g}^{-1}$ at 550 nm, respectively (Bond and Bergstrom, 2006), for externally mixed ns-soot particles.

The scattering cross sections of ns-soot depend strongly on morphology, type and extent of internal mixing, or embedding, all of which can be determined using TEM, with the limitation that volatile components remain undetected. Visual scanning is rapid and can be used to select a relatively smaller number that appear to be representative.

We propose that for climate modeling and health issues distinctions be made between bare, coated, and embedded ns-soot. Each has distinct optical and chemical properties.

Volume mixing of soot with other materials should be discarded from models, in agreement with suggestions of others (e.g., Jacobson, 2001; Bond and Bergstrom, 2006; Bond et al., 2006). Ns-soot is never mixed with sulfate or organics as a homogeneous particle (Adachi and Buseck, 2010; Pósfai and Buseck, 2010).

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6.2 Black carbon (BC)

Although a practical term for inferring atmospheric light absorption in atmospheric measurement and modeling, BC is not a well-defined material. The term should be restricted to light-absorbing refractory carbonaceous matter of uncertain character and should be used with a definition to explain what is meant, e.g., the total absorption resulting from ns-soot + organic carbon + other absorbing particle types.

We recommend the use of BC_{equiv} (equivalent black carbon) to indicate atmospheric light absorption by assuming BC consists of 50-nm monodisperse spheres with a refractive index of $1.95-0.79i$. This convention would define optical properties for model calculations and clearly imply that BC_{equiv} is a virtual rather than actual reference material. For SP2 measurements, the assumption that $rBC = ns\text{-soot}$ should be made explicit, if that is indeed the assumption.

6.3 Elemental carbon (EC)

The term “elemental carbon” should be restricted to describing the high-T component of thermal-optical analysis. Because EC does not exist in the atmosphere as such, it should never be used as a synonym of either ns-soot or BC.

6.4 Final thoughts

To avoid ambiguity, we recommend using the term ns-soot if it is either known or there is reason to believe that the material consists of aggregated carbon nanospheres. BC should be restricted to mixtures of ns-soot plus additional material.

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Table 1. Methods for measuring the concentrations or properties of ns-soot and BC.

| Method (or instrument) | Acronym | Operating principle | State of measured aerosol | What it purportedly measures | Problems, artifacts | Reference | |
|-------------------------|---|-------------------------|---|------------------------------|--|--|--------------------------------|
| Microscopy | Transmission electron microscopy (coupled with electron diffraction, energy-dispersive X-ray spectroscopy, electron energy-loss spectroscopy and electron tomography) | TEM (ED, EDS, EELS, ET) | Interaction of electron beam with particle (signals for imaging, diffraction, and elemental composition are measured) | single particle ^a | Soot + OC (particle size, shape, crystal structure, elemental composition) | Labor-intensive, limited statistics, highly volatile species are lost | Pósfai and Buseck (2010) |
| | Atomic force microscopy | AFM | Interaction between sample surface and a sharp tip mounted on a cantilever while scanning the surface | Single particle ^b | Particle size, 3-D shape | Labor-intensive, no chemical information, ambiguous interpretation, scanning artifacts | Pósfai and Buseck (2010) |
| | Scanning transmission X-ray microscopy (coupled with near-edge X-ray absorption fine-structure spectroscopy) | STXM (NEXAFS) | Interaction of X-rays with particles | Single particle ^b | Composition and bonding character of C | Labor-intensive, synchrotron radiation needed, no quantitative information | Tivanski et al. (2007) |
| Absorption measurements | Integrating plate (sphere, sandwich) | | Optical attenuation | Bulk ^b | BC | | Horvath (1997) |
| | Aethalometer | | Optical attenuation, measured continuously at several wavelengths | Bulk ^b | BC | Mass effect (multiple scattering), absorption enhancement by non-absorbing coatings, uncertainty in parameters used for conversion between absorption and BC concentration, presence of other absorbing particles (including BrC), wavelength dependence of absorption | Hansen et al. (1984) |
| | Particle soot absorption photometer | PSAP | Optical attenuation, measured continuously at several wavelengths | Bulk ^b | BC | | Arnott et al. (2005) |
| | Multi-angle absorption photometer | MAAP | Optical attenuation, measured continuously at multiple angles | Bulk ^b | BC | | Petzold and Schönlinner (2004) |
| | Photo-acoustic spectrometer | PAS | Energy from absorbed light converted to pressure | Bulk ^c | BC | Same as above except mass effect | Moosmüller et al. (1997) |
| | Extinction cell + nephelometer ("difference method") | | Difference between extinction and scattering | Bulk ^c | BC | Uses a small difference between two large numbers, needs large sample volumes, includes other absorbing species; affected by internal mixing | Schnaiter et al. (2005) |
| | Single particle soot photometer | SP2 | Laser-induced incandescence | Single particle ^c | rBC | Uncertainty about nature of coatings | Schwarz et al. (2008) |

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Table 1. Continued.

| | Method (or instrument) | Acronym | Operating principle | State of measured aerosol | What it purportedly measures | Problems, artifacts | Reference |
|-----------------------------------|---|--------------------|---|------------------------------|--|--|---|
| Thermal analysis | Evolved gas analysis | EGA | Conversion (volatilization) of C into CO ₂ over a temperature range | Bulk ^b | TC – OC = EC | Interference with BrC and bioaerosol, interpretation of volatilization temperature ranges ambiguous, charring, filter effects | Cachier et al. (1989) |
| | Chemical extraction + EGA | | Extraction with water or organic solvent before volatilization to remove OC | Bulk ^b | EC | Same problems as above but somewhat reduced | Novakov and Corrigan (1995) |
| | Thermal-optical transmittance or reflectance analysis | TOT, TOR | Transmittance or reflectance of filter monitored during heating and carbon volatilization | Bulk ^b | EC | Uncertainty about the cut-point between OC and EC, charring within filter | Chow et al. (1993) |
| Single-particle mass spectrometry | Aerosol mass spectrometry, particle analysis by laser mass spectrometry, aerosol time-of-flight mass spectrometry | AMS, PALMS, ATOFMS | Laser- or electron-impact-induced ionization of particles; fragments measured with a time-of-flight mass spectrometer | Single particle ^c | EC + OC compounds | Ionization efficiencies and matrix effects affect quantitative results, fragment recombination is needed to infer particle types | Murphy et al. (2006); Sullivan and Prather (2005); Wexler and Johnston (2011) |
| Optical spectroscopy | Infrared spectroscopy | IR | Interaction of IR light with particles | Bulk ^b | Composition and bonding character of C | | Smith and Chughtai (1995) |
| | Raman spectroscopy | RS | Interaction of IR light with particles | Bulk ^b | Composition and bonding character of C | Bulk method with relatively poor spatial resolution | Mertes et al. (2004) |
| | UV-visible spectroscopy | UV-VIS | Interaction of light with particles | Bulk ^d | | | Apicella et al. (2004) |

^a On substrate, in vacuum. ^b On substrate. ^c Airborne. ^d In suspension or solution.



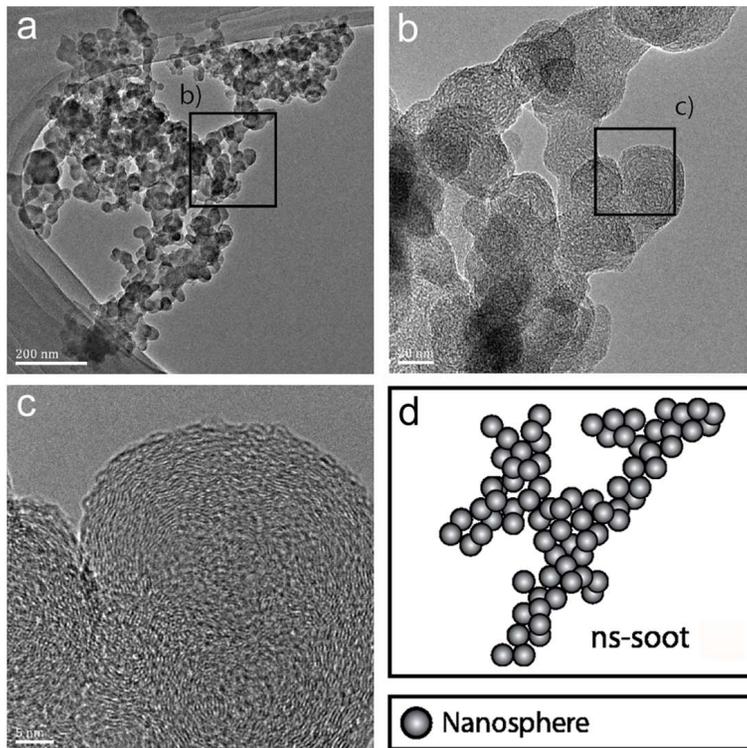


Fig. 1. TEM and schematic images of soot and carbon nanospheres. **(a)** An ns-soot particle from NIST Standard Reference Material (SRM) 1650b (diesel particulate matter). **(b)** Enlarged image of **(a)**. **(c)** High-resolution image of ns-soot particle from **(b)**. The lattice fringes of the ns-soot are evident. **(d)** Schematic image of carbonaceous nanospheres aggregated to form ns-soot.

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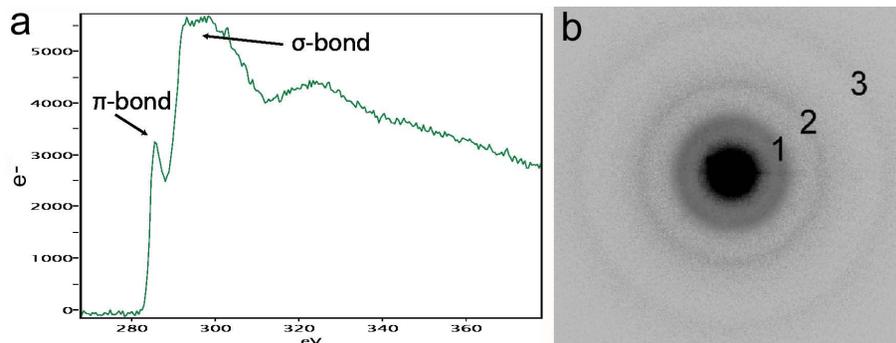


Fig. 2. (a) EELS of ns-soot from Mexico City showing peaks arising from carbon π - and σ -bonding. (b) Typical SAED pattern obtained from ns-soot from the emission of a diesel engine. The numbered rings correspond to lattice spacings of (1) ~ 3.6 , (2) 2.05, and (3) 1.2 Å.

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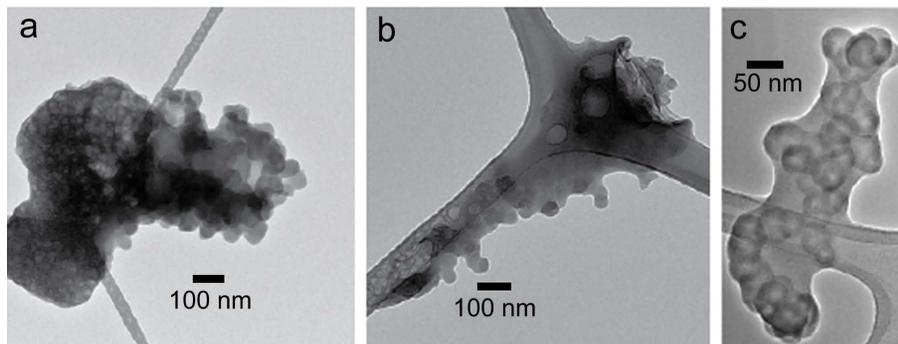


Fig. 3. TEM images of internally mixed ns-soot particles from Mexico showing **(a)**, **(b)** coating, and **(c)** embedding, although **(b)** shows that the distinction is gradational. The ribbon-like features are from the lacey carbon substrate. Samples **(a)** and **(b)** were collected from Mexico City and **(c)** was collected from biomass burning in Mexico. All samples were collected during the MILAGRO campaign (Adachi and Buseck, 2008).

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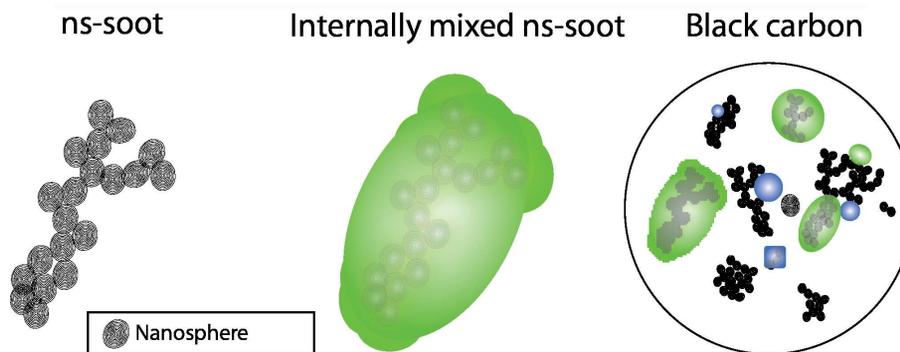


Fig. 4. Schematic images of **(a)** ns-soot, **(b)** internally mixed ns-soot particle, and **(c)** an ensemble of particles, the total absorption of which would be referred to as from BC when measured with an aethalometer. The aggregated, black layered spherules indicate ns-soot. The green and blue colors indicate organic matter and sulfate, respectively.

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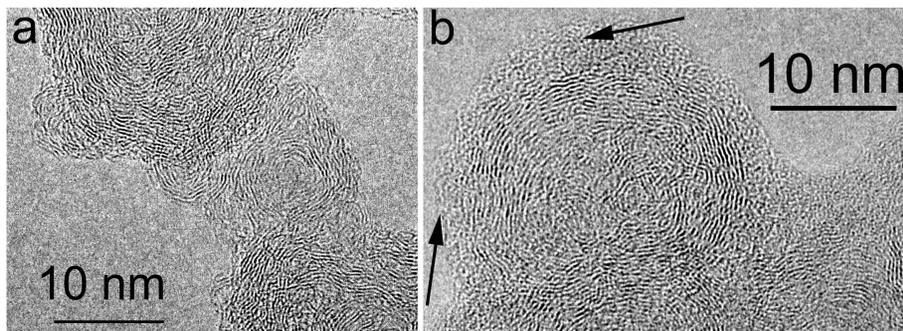


Fig. 5. High-resolution TEM images of (a) bare and (b) coated ns-soot particles from diesel emissions from buses. The arrows in (b) indicate an amorphous coating on carbon nanospheres.

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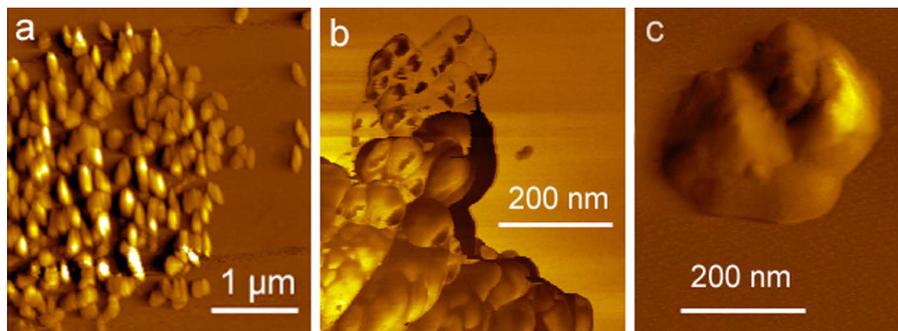


Fig. 6. Atomic force microscope images of **(a)** bare, **(b)** partly coated, and **(c)** embedded soot particles, all collected from the emissions of vehicles with internal combustion engines.

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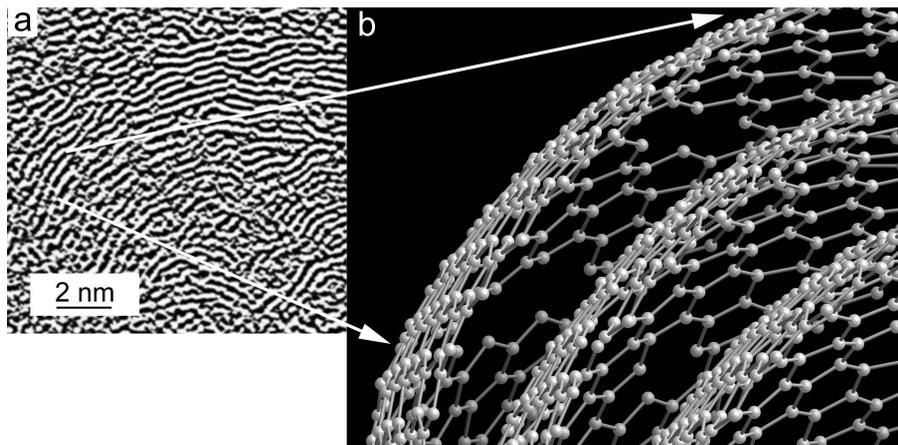


Fig. 7. (a) Part of Fig. 5b, with the contrast of graphene-like layers in ns-soot enhanced digitally. (b) A schematic model for three layers. They are graphene-like, but some C atoms are missing from the hexagonal nets. The missing atoms break the periodic order and possibly cause the misalignment of layer segments relative to one another, resulting in the layer curvature. H in ns-soot can be accommodated around the sites of missing C atoms, where some C atoms can be bonded to two C and one H.

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