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Interactive comment on "Are black carbon and soot the same?" *by* P. R. Buseck et al.

Anonymous Referee #2

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In their manuscript entitled "Are black carbon and soot the same?" Buseck, Adachi, Gelencsér, Tompa, and Pófsai (referred to as "BAGTP" in the following) make the laudable effort of clarifying previous definitions and adding a new material definition "ns-soot", where "ns" refers to carbon nanospheres. To review this effort, it needs to be put into context with previously used definitions to evaluate its usefulness.

The common usage of "soot" is, for example defined by Merriam-Webster's dictionary, "a black substance formed by combustion or separated from fuel during combustion, rising in fine particles, and adhering to the sides of the chimney or pipe conveying the smoke; especially : the fine powder consisting chiefly of carbon that colors smoke." From a scientific point of view, one may want to add that the combustion process involves a hydrocarbon-based fuel and that soot mass is dominated by elemental carbon (EC) and organic carbon (OC) with additional common contributions by sulfur, metals, etc. EC occurs in solid form and OC can be a mixture of solid and liquid, in contrast with BAGTP's assertion (p. 24824 l. 7-12) that soot is a solid material. Here BAGTP also state that "a source of confusion is that one term [i.e., soot] applies to origin and the other [i.e., black carbon] applies to optical measurements." I don't think this is a source of confusion but just two different, complementary, and appropriate definitions.

There seems to be general agreement as to what OC is, however, not necessarily as to how to quantify it. OC is generally seen as a mixture of very many (thousands?) of organic substances. On the other hand, EC has a nice simple ring to it, elemental carbon or carbon in its elemental form, that is diamond, graphite, graphene, carbon nanotubes or fullerenes. However, transferring this simple straightforward definition to a use in the field of aerosol and atmospheric sciences has resulted in multiple related terms as follows:

1. Elemental Carbon (EC) is generally defined by thermal-optical methods as refractory carbonaceous component evolving to carbon dioxide when oxygen is introduced at high temperature. While this seems to be a nice definition, complications arise when, upon heating in an inert gas, OC is pyrolyzed into an EC-like compound often called pyrolyzed carbon (PC). This makes the thermal-optical definition of EC an operational one, greatly depending on temperature protocol and instrument used. In addition, as discussed in Prather's comment, the single particle mass spectrometry community uses EC to describes particles "which produce a characteristic mass spectrum with mainly C cluster ion peaks."

2. Black Carbon (BC) is defined as carbonaceous material with a deep black appearance, which is caused by a significant, non-zero imaginary part of its refractive index that is wavelength independent over the visible and near-visible spectral regions. The constant imaginary part of the refractive index results in an absorption coefficient that is inversely proportional to the wavelength for both for bulk BC and for small BC particles in the Rayleigh regime. However, light absorption measurements only quantify the absorption coefficient. While quantifying amounts of BC by its total absorption cross-section is feasible, virtually all applications quantify BC by mass, related to the

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absorption cross-section through the mass absorption efficiency. The mass absorption efficiency can only be determined using additionally a non-optical, mass-based measurement such as the thermal-optical method for quantifying EC.

3. Graphitic Carbon (GC) is defined by the graphitic lattice structure, which can be operationally quantified through Raman spectroscopy or x-ray diffraction.

4. Insoluble carbon (IC) is defined as carbonaceous material insoluble in all polar and non-polar solvents. This definition is not commonly used as most researchers tire of testing solubility in “all” solvents.

5. Refractory Black Carbon (rBC) is defined as the fraction of strongly light absorbing carbonaceous material (BC as defined above) that is additionally thermally refractory and operationally quantified by laser-induced incandescence such as measured by the single particle soot photometer (SP2).

All these definitions are tied to an operational component, an instrument or method used to quantify the defined term, albeit generally the measurement procedure is not well defined. BAGTP now “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 quantitative analysis for ns-soot seems to be tied to TEM tomography coupled with human judgment for assessing “grape-like morphologies” and “concentrically wrapped, graphene-like layers of carbon” within nanospheres to obtain ns-soot volume within a sample. This ns-soot volume would then be multiplied with an unspecified ns-soot density to obtain ns-soot mass. At least, this is how I imagine the operational procedure. It would be highly beneficial if BAGTP could outline their vision for the operational procedure of determining ns-soot mass in their manuscript and comment on the feasibility and costliness of this procedure for individual samples, field campaigns, and routine monitoring networks such as IMPROVE.

BAGTP also propose to discard volume mixing of soot with other materials from models and state that “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.” However, it is unclear to me how this procedure for determining scattering cross sections of ns-soot would actually implemented, especially if ns-soot is embedded in other materials. Additional guidance would be very helpful.

Furthermore, I totally disagree with BAGTP on their suggestion for BC. In my view, BC is an optically defined material along the lines given above, having optical properties in general agreement with GC and the term BC should not be restricted to “to light-absorbing refractory carbonaceous matter of uncertain character”.

In summary, the suggestion of introducing the term of “ns-soot” in connection with TEM analysis is well intended but needs additional clarifications and definitions to result in a practical definition and to avoid future confusion. I also suggest that this term should be connected with the combustion origin commonly implied by the term “soot”.

The final question for a reviewer is “to publish or not to publish”. I believe that BAGTP make a potentially valid addition to the 30-plus years of discussions of definitions, methods, and terms related to carbonaceous aerosols with their TEM-based definition of ns-soot and their added discussion of other terms. However, I also suggest that before publication in ACP, their manuscript needs major revisions along the lines suggested above.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 24821, 2012.