

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

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We are gratified by the reactions and discussion stimulated by our paper. Here we combine replies to the Comments of Anonymous Referee #2 and to Schwartz and Lewis (respectively shortened to “AR2” and “SL” for this Reply). We thank both for their thoughtful Comments.

AR2 and SL agree on some issues and disagree on others. They agree that the effort to clarify the terminology is “laudable” (AR2, p. C9054) and “we applaud them for raising an important issue. We wholly concur with their (Buseck et al.) statement that ‘confusion can be avoided if terms are defined and widely understood’” (SL, p. C9100).

Elsewhere they are in direct conflict. For example, AR2 “totally disagrees” with us regarding BC because it “is an optically defined material” and “BC should not be restricted to light-absorbing refractory material of uncertain character” (C9057). In contrast, SL

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write: “They (Buseck et al. 2012) observe that BC is not a well defined material and propose that the term should be restricted to light-absorbing refractory carbonaceous matter of uncertain character, an assessment with which we concur” (C9101).

Clearly, the questions are far from solved; our paper should not be judged on that account unless one prefers the judgment of one referee over the other (we have a favorite in this case). We also conclude from the above disagreement plus the other Comments that terminology is of interest. Now to some specifics.

SL identify the problem: BC, EC, and soot are, respectively, a “category,” “set,” and “broad set of substances” that “refer, respectively, to optical properties, composition, and formation process, which are independent concepts” (C9102). Thus, in their view none are pure materials nor are they necessarily equivalent to one another.

As pointed out in our paper, but disputed by AR2 and SL, there is no consensus on the meaning of “soot” and whether it is a specific substance, as is widely used in many papers from the combustion literature that show TEM images of particles labeled soot (e.g., Abid et al., 2008; Alfè et al., 2010; C.H. Kim et al., 2008; K.B. Kim et al., 2008; Miller et al., 2007; Öktem et al., 2005; Santamaria et al., 2010; Vander Wal and Tomasek, 2004; Vander Wal et al., 2007). AR2, SL, and others prefer to use soot as a term for a mixture of many products emitted through combustion, for example as in the citations in the SL Comments.

We disagree that “soot” should be used for all the stuff resulting from combustion. It is a specific material, much as we defined it. As a material, it has a specific formation mechanism, which is basically high-temperature nucleation. In our opinion, all other (even blackish) products of combustion, such as unburned coal chunks from coal-fired power plants, pieces of partially burned biomass, condensed pyrolysis products, silica-containing stuff, and the like should have names such as char, fly ash, or even tar balls. Even if most EC/BC determinations do measure these as EC/BC (unless discarded by size-selective inlets), they are not soot particles. Introduction of “ns-soot” is a proposed

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way of sidestepping the issue of uneven usage of “soot.”

AR2 and SL agree that BC has a deep black appearance, and we happily concur with SL's observation that “Black carbon is carbon that is black” (C9103). The issue is that these statements, and the examples provided by SL, apply to macroscopic samples where appearance can be observed and composition determined. However, both conditions are not met by spectral measurements of aerosol BC, so even the truism is not useful when applied to aerosol particles.

In the instance of the SP2, which measures thermal radiation during incandescence of individual particles, BC (or refractory BC) is determined by incandescence during laser heating. That could be single materials, mixed particles, clumps of particles, or any combination thereof. Details of its color, identity, and purity can only be estimated, although graphitic stuff is a highly likely component. SL point out assumptions that make accurate mass determination from such measurements challenging. Nonetheless, there is general agreement that BC is a convenient term for atmospheric studies despite the imprecision regarding its exact identity. That situation could change if and when there is a well-understood standard for aerosol BC, but we do not see that on the horizon.

AR2 is asking for information (C9056 lines 22 to 26) beyond that in our paper. Although it is indeed possible to accurately determine the volume of individual ns-soot particles by using electron tomography (e.g., Adachi et al., 2010), routine monitoring networks are outside the scope of our purview. Nevertheless, we believe that it is important to have a realistic and thorough understanding of what is being monitored, even if it does not result in the desired routine mass determinations for field campaigns and monitoring networks.

AR2 overlooks the important use of the standard technique of selected-area electron diffraction (SAED) in observing that “Graphitic Carbon (GC) is defined by the graphitic lattice structure, which can be operationally quantified through Raman spectroscopy

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or x-ray diffraction” (C9056). Actually, ns-soot produces three distinct rings in SAED patterns, two of which differ from graphite spacings and one that is larger than graphite (002).

AR2 quotes our ms.: “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” (C9057).

AR2 makes a valuable point. We will revise the ms. to state “TEM can be used to determine morphology and the type and extent of internal mixing, or embedding of ns-soot, 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 if additional detailed study, as with tomography, is appropriate. These variables strongly influence the scattering cross sections of ns-soot, although additional steps are required to determine those cross sections. Also, this information improves the evaluation of particle mass from its scattering and absorption measurements (e.g., Cappa et al. 2012; Adachi et al., 2010).”

In the submitted acpd ms., we limited the discussion of EC because, as shown by the title, our goal was to distinguish between BC and ns-soot. However, as suggested by reviewers, EC needs discussion. In general, we agree with the descriptions of EC that it “is generally defined by thermal-optical methods as refractory carbonaceous components evolving to carbon dioxide when oxygen is introduced at high temperature” (AR2, C9055) and “is a substance containing only carbon, carbon that is not bound to other elements, but which may be present in one or more of multiple allotropic forms” (SL, C9103). “In addition . . . the single particle mass spectrometry community uses EC

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to describes (sic) particles ‘which produce a characteristic mass spectrum with mainly C cluster ion peaks’” (AR2, C9055). Since EC is defined by its thermal-optical property or mass spectroscopy, EC should be distinguished from optically defined BC, although there could be overlap because the definitions are operational.

We also agree that “it is essential that investigators report precisely the measurement that is being made and the conversion from the measured quantity to the reported quantity. This would allow the measurements to be used by other investigators with confidence in understanding of the reported quantity, irrespective of its name.” (SL C9108)

Finally, both AR2 and SL dislike the term “ns-soot” because to them it implies an origin. We describe a material with specific physical and chemical characteristics. We tried many alternative names but found none better that are compact, descriptive, and representative of what we observe.

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