Interactive comment on "Are black carbon and soot the same?"

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Reply to Comment of Anonymous Referee #1

1. General

We agree that it is important to know how well various instrumental techniques quantify the mass of the particulate products of combustion as well as what these products are. The black, oily material produced by combustion was originally called "soot" on the basis of visual observations when there was neither need nor adequate instruments to quantify it. In the 19th century, when black smoke became a problem in industrialized areas, the "smoke stain" method based on the blackening of filter paper was used for its quantification. Numerous analytical methods evolved from this simple technique. In the latter part of the 20th century some analysts created their own operational terms to correspond to specific measurement methods. EC and BC are among the resulting terms. However, since all methods have inherent limitations that require corrections (e.g., charring in thermal-optical transmittance/reflectance methods, filter effects in aethalometers, particle fragmentation in mass spectrometry), it became clear that different instruments were measuring different entities.

The results using different methods are in relatively good agreement for curbside aerosol dominated by diesel soot. However, they can differ by up to an order of magnitude for rural aerosol dominated by biomass-burning aerosol or secondary aerosol (brown carbon or "humic-like substances" – HULIS, e.g., Reisinger et al., 2008). Therefore, it is clear that ns-soot ≠ BC ≠ EC, and that these terms highlight the current measurement problems in the global atmosphere, particularly where the contribution of biomass burning is significant.

Optical measurements are cheaper and have better time resolution than thermal techniques. Thus, measurements of atmospheric light absorption have become an increasingly widespread component of climate change estimates. BC has evolved as the term of choice for such measurements, and we acknowledge the convenience of having a single term such as BC. Nevertheless, it is important to remember that it remains an operational term. Although embedded in the technical literature and public understanding, especially with the growing interest in climate change, we wish to clarify the nomenclature and point out the pitfalls of the incorrectly assumed BC = EC = soot (or ns-soot) parity. In the next sections we provide usage recommendations and then replies to specific comments of Referee #1 (shown in ***blue italics***).

2. Opening sentence of Referee comment

"Flags ambiguous usage, but doesn't provide an acceptable solution"

We propose the following recommendations; some of which are already in our ms.:

Recommendations for ns-soot, BC, and EC:

a. **ns-soot**: a subset of what is commonly called soot, has specific physical properties, and is a distinct and recognizable material.

Recommended usage: use for carbonaceous particles consisting of nanospheres possessing concentrically wrapped, graphene-like layers of carbon and with grape-like (acinoform) morphologies.

We recommend distinguishing, when possible, between bare, coated, and embedded ns-soot for climate modeling and health issues since each has distinct optical and chemical properties.

b. **Black carbon (BC)**: absorbs shortwave radiation and thus has distinct optical properties. However, it lacks a specific material as a standard and is a term for specific optical features.

Recommended usage: BC is an operational term rather than a well-defined material. Use of BC should be restricted to light-absorbing refractory carbonaceous matter of uncertain character.

We recommend using BC with an explicit definition to explain what is meant, e.g., the total absorption resulting from ns-soot + organic carbon + other absorbing particle types, or ns-soot + other unspecified refractory species, or whatever else is appropriate.

We recommend using BC_{equiv} 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 defines optical properties for model calculations and clearly implies that BC_{equiv} is a virtual rather than actual reference material.

c. **Elemental carbon (EC)**: an operational term used for both a) the high-T component of thermal-optical analysis and b) the carbon fragments measured using aerosol mass spectrometry.

Recommended usage: since EC does not exist in the atmosphere as such, it should not be used as a synonym of ns-soot or BC.

We recommend restricting EC to the high-T component of thermal-optical analysis.

We recommend using **"carbon clusters"** for the signal that is observed using aerosol mass spectrometry.

3. Replies to specific comments by Referee #1:

a. ***"it does not provide a complete analysis of nomenclature beyond revealing some of the inconstancies; it does not present a complete analysis of the issues, as exemplified by K. Prather's comment; and does not present a convincing solution to the problem."***

A review of the relevant literature is a component of our paper, but it is not the main goal nor is there a single solution to the entire problem of carbonaceous aerosol particle types. Instead, our aim is to point out ambiguities with current terminology, define the major aerosol component having identifiable physical properties, and provide recommendations for terminology going forward.

b. "For the first, often the experimentalists and modelers who use these different words believe that they are referring to the same unique substance."

Not consistently. Our manuscript points out that "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)." Other examples (Chin et al., 2009; Vander Wal et al., 2010) are cited the companion Comment by Schwartz and Lewis.

c. ***"Indeed, the authors maintain this basic framework of a single unique material of BC = EC = ns-soot in their section on coatings, in which they identify ns-soot as the "backbone" of a particle containing other materials."***

The ms. says, "We propose that only the "backbone" of internally mixed particles, i.e., the aggregates of carbon nanospheres, should be termed ns-soot." That statement does not suggest that BC = EC = ns-soot.

d. ***"Hence I recommend that the authors, instead of concentrating on supporting a new definition of BC, attempt to apply their insights within the existing framework of a general term, "soot", made up specifically of BC=EC(=ns-soot) and other materials."***

"The authors state that externally mixed "ns-soot" exists in nature, so I imagine they would have no problem with this usage if "BC" is used (first ambiguity) as an equivalent to ns-soot."

We disagree. The whole point of our ms. is that ns-soot is not the same as BC, which is not the same as EC. The referee seems to first recognize that point and then ignore it where writing "I will continue this review using "BC" to refer to this material" (p. C8424) as if it is indeed one material. That statement in the review begs the question and undermines the whole thrust of our ms.

The situation might be different if there were acceptable, agreed-on samples and standards of BC that could be studied by other than optical measurements, but there are none.

e. ***"TEM is not a measurement that can generally be applied to ambient measurements."***

This sentence is only qualifiedly true. TEM has been applied to ambient measurements for more than 30 years in many studies of, for example, asbestos contamination, combustion, and human health.

f. ***"It is time consuming, off line, expensive, complicated, and, without full 3-D analysis, ambiguous as to total BC volume in individual particles."***

TEM is indeed an off line technique, but it is neither time consuming nor complicated if the operator has adequate knowledge and experience, as is also required for the other techniques. Although electron microscopes are expensive, so are SP2 and AMS instruments. Moreover, far more institutions have electron microscopes than the more specialized aerosol instruments.

Our point is not to disparage these specialized instruments but to point out the utility of techniques that to some extent have been overlooked by the atmospheric community. Thus, we believe the challenge of using an instrument that is standard in many disciplines should not be reasons to avoid or even minimize TEM analysis of atmospheric aerosol particles. With electron tomography, TEM is a uniquely useful way to quantitatively determine the volume of individual ns-soot particles (Adachi et al., 2010).

g. ***"I point out that absorption measurements are not the same as BC mass measurements, an issue that goes beyond the questions of the material/optical properties of BC externally mixed, as even absolutely correct absorption measurements may contributions from non-BC materials."***

We agree with the first part of this statement, which provides one of the motivations for our manuscript. A major reason is that "BC absorption" depends on the shape, mixing states, and refractive index of ns-soot as well as the absorption of "non-BC" materials. We are uncertain about the second part of the statement.

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