

Short comment by M. Gysel to the manuscript “Are black carbon and soot the same”

The terminology for the light-absorbing and/or refractory carbonaceous matter in atmospheric aerosols was, is and will remain a topic of scientific dispute. It is worthwhile and needed to lead this discussion. Any new contribution to this topic should primarily aim at identifying and reducing existing ambiguities and misunderstandings. This can only be achieved with a very clear concept. This manuscript and the associated public discussion indicate two alternative concepts: The terminology can either be based on specific materials or material groups (identified by their chemical and/or physical properties) or it can be based on instruments or measurement techniques. A mixture of these two concepts is certainly not wanted.

Choosing an instrument based terminology has the advantage that the actual measurement with its strengths and caveats is reflected, however, it will lead to an increasing number of terms with each new method and it does not a priori contribute to the understanding of how the results from different methods can/should be compared in between themselves and with models.

Personally I favour the material (or material groups) based approach, as it has been put forward in Stephen Schwartz’s short comment, though it is unfortunately not so easy to define the materials in a clear and useful manner. The experimental scientists then get the duty to optimize their experimental and data analysis methods for specific sensitivity to a certain material, to explain which material they (try to) quantify and to state how well they (believe) to achieve this.

The author reply to Kim Prather’s short comment is an example how clarity is not achieved. The key question is not whether the carbon fragments are the “real building blocks” of the material under investigation, instead it is all about the question whether they can be uniquely attributed to a certain material (or material group) and how well the mass of this material can be quantified.

Table 1 provides an important overview over different methods for the measurement of light-absorbing and/or refractory carbonaceous matter including some of their specific features. However it needs some improvement. The soot particle aerosol mass spectrometer (SP-AMS; Onasch et al., 2012), another instrument with a disputable name, should be included. The question about the appropriate method group for the SP-AMS (with ATOFMS, with SP2 or its own group) is an interesting one and a detailed discussion on similarities and differences between ATOFMS, SP-AMS and SP2, all of which use a strong laser to evaporate the carbonaceous particles, would give quite some insight into the terminology problematic (though it would probably go too far to lead this discussion for all instruments).

The SP2 is attributed to the absorption measurements in Table 1. This is a misclassification, which is unfortunately quite common. It is true that light-absorption plays a role in the applied method, as a strong laser is used to heat the “rBC” to its

vaporisation temperature. However, the “rBC” mass is quantified via the thermal radiation emitted by the incandescent particle rather than any kind of light-absorption measurement. The incandescence signal measured by the SP2 for e.g. a “graphitic” particle does not depend on the intensity of the laser (if it is above a critical threshold) nor on the wavelength of the laser. Furthermore, strong light absorption is not a necessary condition to make a material quantifiable by the SP2. Instead, the material must primarily be highly refractory for two reasons. First, the thermal emission of single nanoparticles becomes only detectable at very high temperatures. Second, the material must have the highest vaporisation temperature of all components in the particle, such that it is present in pure form when the particle reaches its maximal temperature, i.e. the vaporisation temperature of the most refractory component. Moderate light absorption is sufficient (depending on available laser power and particle size) to make highly refractory materials detectable by the SP2. Some examples are metals, volcanic ash or hematite. In atmospheric aerosols the incandescence signal of the SP2 can typically be attributed to essentially “elemental carbon” with graphitic, amorphous and/or fullerenic nanostructure (the relative contribution of these structures in a particle actually determines the calibration factor for the conversion between thermal radiation and carbon mass). Thus “EC” (or “rEC” if the instrumental qualifier “r” for refractory needs to be added) might be a better name for the material measured by the SP2 than the term “BC” (or “rBC”) commonly used for SP2 measurements, particularly to avoid the confusion with absorption measurements. Diamond is of course an allotrope of carbon which remains undetected by the SP2, while it would in principle be detected by thermal methods if operated at sufficiently high temperature, however, it is not expected to be a major component of atmospheric aerosols.

Some more specific comments:

p. 24830, l. 21ff: “Volume mixing of soot with other materials should be discarded from models...” – It is true that soot (or EC) is not homogeneously mixed with other aerosol components even if present in the same particles. However, this does not mean that assuming homogeneous mixture is per se a bad model assumption, as other approximations on the morphology are made at the same time. Kahnert et al. (2012) actually showed that assuming homogeneously mixed spheres can in some cases outperform the assumption of spherical core-shell morphology.

p. 24831, l.3ff: “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.” – “BC” never means “light absorption”, instead the measured light absorption coefficient is often used to infer the “BC mass”, i.e. the mass of the light absorbing matter. Above statement is just one example of several in this manuscript of the unclear distinction between measured quantity (absorption coefficient in this case) and mass of the matter of interest that is inferred from it. Such impreciseness does certainly not contribute to present a clear and consistent terminology concept.

References:

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Onasch, T. B., Trimborn, A., Fortner, E. C., Jayne, J. T., Kok, G. L., Williams, L. R., Davidovits, P., and Worsnop, D. R.: Soot particle aerosol mass spectrometer: development, validation, and initial application. *Aerosol Sci. Technol.*, **46**, 804-817, doi:10.1080/02786826.2012.663948, 2012.