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Recommendations for the interpretation of "black carbon" measurements

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Abstract

Although black carbon (BC) is one of the key atmospheric particulate components driving climate change and air quality, there is no agreement on the terminology that considers all aspects of specific properties, definitions, measurement methods, and related uncertainties. As a result, there is much ambiguity in the scientific literature of measurements and numerical models that refer to BC with different names and based on different properties of the particles, with no clear definition of the terms. The authors present here a recommended terminology to clarify the terms used for BC in atmospheric research, with the goal of establishing unambiguous links between terms, targeted material properties and associated measurement techniques.

1 Introduction

Within the discussion of global climate change, the international community recognized the importance of establishing inventories for sources and sinks of particulate, light absorbing carbon (UNEP/WMO, 2011; Bond et al., 2013). One of the major contributors to the carbon cycle is combustion of fossil fuel and biomass, with carbonaceous particulate matter being one of the most important combustion by-products besides CO_2. One fraction of the carbonaceous aerosol, commonly called black carbon (BC), is characterized by its strong absorption of visible light and by its resistance to chemical transformation (Ogren and Charlson, 1983; Goldberg, 1985; Heintzenberg and Winkler, 1991). These distinct properties give it relevance in various fields related to climate change, air chemistry, ambient air quality, biogeochemistry, and paleoclimatology.

The BC fraction of the carbonaceous aerosol has been included in the Strategic Plan of the Global Atmosphere Watch program (GAW) of the World Meteorological Organization (WMO) (Müller et al., 2007). It has also become one of the key targets for current research on the aerosol impact on climate and also on mitigation strategies. Relative to the long-lived greenhouse gases (particularly CO_2 and CH_4), the light-absorbing

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carbonaceous aerosol is referred to as a short-lived climate forcer and its emission control policies are being contemplated as one near-term mitigation strategy for the climate impacts of anthropogenic emissions; see e.g. the integrated assessment of black carbon and tropospheric ozone by UNEP/WMO (2011).

Despite its high relevance for climate change research (Ramanathan and Carmichael, 2008; Bond et al., 2013), there is no agreed clear and unambiguous terminology available for quantifying carbonaceous matter in atmospheric aerosols. In the end, all definitions used in the scientific literature refer to a specific property of the respective carbonaceous fraction, or to the method that is used for the measurement. As there is no consensus within the community for using a specific definition for a particular measuring technique, there are numerous publications in the scientific literature that refer to the same property but with different terms and, vice-versa, with publications referring to different properties but with similar names. To a minor extent, the same is true also for modeling exercises where different terms are used, not always in relation to properties that can be derived from direct measurements.

While data on light-absorbing carbonaceous aerosols are collected globally by different measurement techniques, global emission inventories and modeling studies (e.g., Bond et al., 2007; Junker and Liousse, 2008; Vignati et al., 2010; Granier et al., 2011; Lee et al., 2012), as well as scientific assessments (Solomon et al., 2007; Bond et al., 2013), require data sets that are independent of the measurement method. It is difficult to clearly distinguish these terminologies in atmospheric chemistry and climate model applications.

In particular, BC emission inventories are to a large extent based on emission factors derived using thermal methods based on the detection of evolved carbon, while data from atmospheric monitoring stations are mostly derived from optical absorption methods. Consequently, Vignati et al. (2010) investigated the sources of uncertainties in modeling BC at the global scale and requested an increased understanding of observational data and associated uncertainties. However, the uncertainties are difficult to establish because the reasons for the large discrepancies between methods are often

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not fully understood and are to a large extent dependent upon season and location of sampling and type of aerosol.

This publication proposes a definition of terms and recommendations for interpreting measurements of "black carbon", "elemental carbon", "light absorption", "refractory carbon" and other properties related to this distinct fraction of the carbonaceous aerosol. We start with a formal definition of black carbon and elemental carbon including the constituting properties of BC. An overview of available analytical methods will prepare the ground for a synopsis of historical and current operational definitions. Finally, the terminology recommended for future use is presented based on targeted particle properties. It will link considered properties to associated analytical methods in an unambiguous manner. These recommendations are a result of discussions carried out in the context of the Scientific Advisory Group for Aerosols of the WMO GAW program. However, the authors express their own views and do not act on behalf of, or commit, their institutions, ministries or WMO.

2 Definition of black carbon

From a formal standpoint and without referring to measurement methods or formation processes (Schwartz and Lewis, 2012), the technical term "black" describes ideally a completely light-absorbing object with reflectivity of zero, an absorptivity of unity and an emissivity of unity, although an object with an absorptivity of 0.95 would still be considered "black". The term "carbon" refers to the sixth element of the periodic system while "elemental carbon" is used to denote carbon that is not bonded to other elements. Combining these formal views provides a strict definition of the terms "black carbon" and "elemental carbon":

– Black carbon (BC) is carbon that is black. The formation process is excluded from this definition because of the variety of potential processes. While BC is mostly formed in incomplete combustion, it can be a product of pyrolysis of carbonaceous matter, i.e. the change of the chemical structure of carbonaceous

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compounds from loss of hydrogen and/or oxygen atoms at temperatures above approx. 250 °C (Chow et al., 2004), of dehydration of sugar, or of heating of wood under an oxygen-free atmosphere (Schwartz and Lewis, 2012). This fundamental definition of BC as carbon that is black agrees with the operationally-based definition by Moosmüller et al. (2009) who defined BC as "carbonaceous material with a deep black appearance, which is caused by a significant, non-zero imaginary part ... of the refractive index that is wavelength independent over the visible and near-visible spectral regions".

– Elemental carbon (EC) is formally defined as 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" (Schwartz and Lewis, 2012). Examples of elemental carbon are diamond, carbon nanotubes, graphite or fullerenes.

Hence, the formal terms "black carbon" and "elemental carbon" refer to a set of materials with different optical and physical properties instead of a given material with well-defined properties.

Unfortunately, these strict definitions are not particularly useful in practice, because carbonaceous matter appears in atmospheric aerosols under no circumstances as pure matter. Instead, it occurs as a highly variable mixture of different carbonaceous compounds with different material properties.

A more useful definition of BC takes into account the various properties of the particles that make them so relevant to climate change, air chemistry, ambient air quality, biogeochemistry, and paleoclimatology. These properties, compiled in Table 1, control the effects of the particles, as well as their atmospheric removal processes and hence spatial distributions. It is the combination of these properties that leads to the classification of BC as a unique substance, but unfortunately, none of the currently-available measurement methods quantifies all five of those properties simultaneously.

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3 Analytical methods

The terms used to identify the various fractions of carbonaceous aerosol are primarily associated with the corresponding measurement methods (Andreae and Gelencsér, 2006; Bond and Bergstrom, 2006; Kondo et al., 2011; Buseck et al., 2012). Commonly, the terms "black carbon", "soot", "elemental carbon", "equivalent black carbon" and "refractory black carbon" synonymously refer to the most refractory and light-absorbing component of carbonaceous combustion particles, even though the underlying definitions and measurement methods are different. Historical definitions and those used in the current literature will be summarized in Sect. 4, whereas this section introduces the families of available analytical methods.

3.1 Evolved carbon

Most common carbon-specific methods consist of combined thermal and gas-analytical approaches based on the analysis of gasification products evolving from a heated filter sample (Malissa et al., 1976; Puxbaum, 1979; Novakov, 1984). These methods make use of the thermal resistivity of the "elemental carbon" fraction of carbonaceous matter, which does not volatilize in an inert atmosphere at temperatures as high as 4000 K. It can only be gasified by oxidation starting at temperatures above 340 °C (Cachier et al., 1989; Jennings et al., 1994). The carbon contained in the analyzed aerosol sample is detected as CO_2 by non-dispersive infrared absorption or other CO_2 specific detection methods.

Currently, different analytical protocols are in use, e.g. IMPROVE (Chow et al., 1993), IMPROVE_A , NIOSH (Peterson and Richards, 2002; Chow et al., 2007a), and EUSAAR-2 (Cavalli et al., 2010). A recent review of evolved carbon methods is given by Chow et al. (2007b). The analytical protocol, however, is an essential part of the data and must be documented in metadata of the databases.

While evolved carbon methods agree within $< 10\,\%$ (Schmid et al., 2001) or 0.22 (± 0.12) $\mu g\,m^{-3}$ (ten Brink et al., 2004), respectively, in in determining the total mass

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concentration of particulate carbonaceous material, the selectivity of separating "elemental carbon" from the bulk of carbonaceous matter varies strongly with the analytical protocol (Schmid et al., 2001; Cavalli et al., 2010; Chow et al., 2011; Pio et al., 2011) and with impurities that may strongly modify the oxidation behavior of the carbonaceous fraction (Schmid et al., 2011). It has also to be mentioned that a correction for pyrolysis or charring, respectively, of carbonaceous matter, i.e. for the transformation of any carbonaceous matter into EC during the analytical process, is required depending on the analytical technique used (Huntzicker et al., 1982; Chow et al., 1993, 2004; Petzold and Niessner, 1995; Boparai et al., 2008).

3.2 Light absorption

The volumetric cross-section for light absorption, commonly called the light absorption coefficient (σ_{ap}), is the principal measure of any optical technique for measuring light-absorbing particles. It is typically reported with units of $m^2\ m^{-3}$, i.e., m^{-1}, or Mm^{-1}, where 1 $Mm^{-1} = 10^{-6}\ m^{-1}$. There is no overall agreed reference method for measurement of the aerosol light absorption coefficient, because all available methods suffer from cross-sensitivity to light-scattering particles and other potential measurement artifacts. However, photoacoustic spectroscopy is a candidate reference method for atmospheric observations and analytical applications (Arnott et al., 2003), while in the laboratory the measurement of light extinction minus light scattering may offer another possibility (Schnaiter et al., 2005b; Sheridan et al., 2005). An in-depth review of light absorption measurement methods is provided by Moosmüller et al. (2009).

The conversion of aerosol light absorption coefficient into light-absorbing carbon mass concentration [BC] is based on the relationship [BC] = $\sigma_{ap} \times MAC^{-1}$. It therefore requires precise knowledge of the mass-specific absorption coefficient (MAC) often reported in units of $m^2\ g^{-1}$. This coefficient, however, varies significantly from time and space depending upon source emissions, transformation during transport, etc. (Bond and Bergstrom, 2006; Chan et al., 2011).

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As long as particles are fractal-like agglomerates with diameters D_{ps} of primary spherules falling into the Rayleigh regime, i.e., $D_{ps} \ll \lambda$, the MAC value of primary spheres is independent of D_{ps}, because for fractal-like aggregates particle absorption depends on the size of the primary spherules and not on the size of the aggregates (Berry and Percival, 1986; Petzold et al., 1997). If this condition is not met, then the MAC of the individual particles may depend on their sizes and the MAC of an aerosol composed of such particles will depend on the size distribution of those particles.

The application of this conversion also assumes that BC is the only light-absorbing particulate species present. Contributions to absorption from non-carbonaceous light-absorbing aerosol components like mineral dust (see e.g. Petzold et al. 2009, 2011), or by non-BC light absorbing carbonaceous matter (= brown carbon; see Andreae and Gelencsér (2006) and next section for a definition) must be excluded or corrected.

The most promising method for excluding measurement artifacts by non-BC light absorbing species is based on the spectral dependence of light absorption properties for different aerosol compounds, which is characterized by the absorption Ångström exponent $\mathring{a}_{ap} = -\ln(\sigma_{ap}(\lambda_1)/\sigma_{ap}(\lambda_2))/\ln(\lambda_1/\lambda_2)$ for a certain wavelength interval $[\lambda_1, \lambda_2]$. While BC is characterized by a low value of $\mathring{a}_{ap}$ between 1.0 and approx. 1.5 (Kirchstetter et al., 2004; Schnaiter et al., 2006; Kim et al., 2012), organic carbon containing aerosol may show strong light absorption in the blue to ultraviolet spectral range (Kirchstetter et al., 2004; Graber and Rudich, 2006; Adler et al., 2010; Kim et al., 2012) associated with $\mathring{a}_{ap}$ values as high as 7 and beyond for the visible range (Chen and Bond, 2010). Mineral dust as another important light absorbing aerosol compound is characterized by strong absorption in the blue and green visible range and low absorption in the red spectral range which results in $\mathring{a}_{ap}$ values of 3 and larger at visible wavelengths (Petzold et al., 2009). Summarizing, over-determination of light absorption associated with BC by non-BC light-absorbing aerosol compounds can be minimized by choosing a wavelength in the red spectral region ($\lambda > 600$ nm) where cross-sensitivities to mineral dust and organic carbon compounds are lowest.

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Furthermore, absorption enhancement by coated particles (Schnaiter et al., 2005a; Lack et al., 2009a; Lack and Cappa, 2010) and by relative humidity effects (Arnott et al., 2003; Lack et al., 2009b) has to be considered in the data analysis.

Another challenge for applying this conversion is the absence of an overall agreed reference material which links light absorption to BC mass concentration. Instead, different methods use different reference materials; see Baumgardner et al. (2012) for a state-of-the-art overview. From a large number of method intercomparison studies on chemical and optical methods in the past decade (e.g., Schmid et al., 2001; ten Brink et al., 2004; Hitzenberger et al., 2006; Park et al., 2006; Reisinger et al., 2008; Chow et al., 2009; Cavalli et al., 2010; Kondo et al., 2011), we know that mass concentrations of BC derived from chemical methods and those derived from optical methods may differ substantially, by up to a factor of 7, even though BC mass concentrations determined by both types of methods are usually correlated at a statistical significance level $P \leq 0.05$.

3.3 Laser incandescence

More recent methods for measuring the mass concentration of light-absorbing carbonaceous aerosol by means of laser heating of light-absorbing aerosol particles and subsequent analysis of emitted radiation (Melton, 1984) have developed from applications in flame diagnostics to atmospheric observation. These techniques are implemented as laser-induced incandescence method (LII) (Snelling et al., 2005; Chan et al., 2011) or as single-particle soot photometer method (SP2) (Stephens et al., 2003; Schwarz et al., 2006). Particularly the SP2 instrument was extensively compared in studies reported by Slowik et al. (2007), Cross et al. (2010), and Kondo et al. (2011). In a recent development the SP2 technology of laser vaporization was coupled to an aerosol mass spectrometer (SP-AMS) for analyzing charged clusters of vaporized carbon particles (Onasch et al., 2012); see further discussion in Sect. 3.5.

Incandescence methods detect carbon-containing particles by absorption of intense radiative energy which is transformed into heat and results in the re-emission of thermal

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radiation (Melton, 1984; Stephens et al., 2003; Schwarz et al., 2006; Chan et al., 2011). While the primary signal is generated by absorption of radiation, i.e., by an optical process, the method response is due to the thermal emission from heated matter. Therefore, incandescence methods are mass-based, but, as for absorption methods, the instrument response depends on the type of carbonaceous particle (Gysel et al., 2012; Laborde et al., 2012) and the conversion of thermal radiation to carbon mass has to be established by proper calibration. Furthermore, the lower limit of detectable particle sizes has to be considered. This limitation is a serious constraint especially for the single-particle SP2 method (Schwarz et al., 2010), which only detects particles larger than 70–80 nm diameter. The calibration of incandescence instruments must be performed using reference carbon material such as fullerene or recommendations from Baumgartner et al. (2012).

3.4 Raman spectroscopy

Methods sensitive to the structural order of carbon atoms in aerosol particles, such as Raman spectroscopy (Sze et al., 2001; Sadezky et al., 2005; Ivleva et al., 2007), are well suited for unambiguously identifying carbonaceous particles with an inherent graphite-like structure. They have shown the direct link between graphite-like carbon structure and strong light absorption properties (Rosen and Novakov, 1977). Combined with suitable calibration methods, this relationship can be used for the measurement of graphite-like carbon in atmospheric particle samples (Mertes et al., 2004). Whereas this method has its strengths in identifying characteristics of the carbon structure, its applicability for a quantitative measurement of carbon mass is limited for today's technology.

3.5 Aerosol mass spectroscopy

Aerosol mass spectrometry methods utilize single particle laser ablation systems based on laser induced plasma or multi-photon ionization, or laser vaporization

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methods under incandescent conditions combined with heated filaments, and subsequent mass-spectrometry techniques for analyzing the chemical composition of individual aerosol particles. The actual measurements are ions of carbon clusters (e.g., C+, C2+, C3+, etc.) in the mass spectra. These methods thus target the elemental chemical composition of the particles. Soot particle aerosol mass spectrometry (SP-AMS) (Cross et al., 2010; Onasch et al., 2012) and aerosol time-of-flight mass spectrometry (ATOFMS) (Noble and Prather, 1996; Spencer and Prather, 2006; Spencer et al., 2007) are the most advanced representatives of this family of methods.

As a distinct feature, the SP-AMS technique represents a hybrid of laser incandescence and mass spectrometry methods. It combines a laser incandescence approach for heating and vaporizing the sampled particles with mass spectrometry techniques for the detection of resulting charged carbon clusters. With respect to the detected property, SP-AMS measurements are more similar to the single particle mass spectrometers (i.e., carbon cluster ion detection) than the incandescence signal (intensity of thermal radiation) measured by the SP2. In contrast, the carbon ions measured by an SP-AMS are related to the carbon that is evaporating under incandescent conditions (i.e., refractory), and not a product of a laser induced plasma or multi-photon ionization events which may control the ions observed by single particle laser ablation systems. Thus, it is a not yet fully answered question whether the SP-AMS measurements should be classified with SP2 measurements or single particle laser ablation measurements.

3.6 Electron microscopy

Particle morphology and microstructure are commonly addressed by means of electron microscopy, either in its transmission (TEM) or scanning (SEM) mode (Fruhstorfer and Niessner, 1994; Posfai et al., 2003, 2004; Adachi et al., 2007; Tumolva et al., 2010). In particular, electron tomography (van Poppel et al., 2005) is a promising technique for identifying three-dimensional structures of nanoparticles. Although microscopy techniques are the only available methods that directly target particle morphology, their

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application for routine monitoring purposes is strongly limited due to labor-intensive sample preparation and data analysis.

4 Historic and current terminology

As stated in the WMO/GAW Report 153 on Aerosol Measurement Procedures (Baltensperger et al., 2003), carbonaceous species are the least understood and most difficult to characterize of all aerosol chemical components. As a first step, total aerosol carbon mass (TC) can be divided into three fractions: inorganic carbonates (IC), organic carbon (OC), and a third fraction called variously elemental carbon, black carbon, soot, or refractory carbon. In climate change and air quality research, the latter fraction of the carbonaceous aerosol is commonly addressed as black carbon (BC), but is often assumed to be elemental carbon (EC). It is also loosely termed soot even though soot denotes the ensemble of the particles emitted during incomplete combustion, i.e., the sum of black carbon and organic carbon (see below).

4.1 Historic definitions

Starting from the pioneering work of Novakov (1984), Goldberg (1985) and Shah and Rau (1990) the following analytically-based definitions have been introduced:

- Total carbon (TC): total particulate carbonaceous material (Novakov, 1984); commonly assumed as TC = EC + OC (Shah and Rau, 1990), often neglecting inorganic carbon.
- Organic carbon (OC): any of the vast number of compounds where carbon is chemically combined with hydrogen and other elements like O, S, N, P, Cl, etc. (Shah and Rau, 1990).

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- Elemental carbon (EC): a form of carbon that is essentially pure carbon rather than being chemically combined with hydrogen and/or oxygen. It can exist either in an amorphous or crystalline structure (Shah and Rau, 1990).
- Carbonate carbon (CC) or inorganic carbon (IC): inorganic carbonate salts (Shah and Rau, 1990).
- Black carbon (BC): combustion-produced black particulate carbon having a graphitic-like microstructure (Novakov, 1984), or "... an impure form of the element [carbon] produced by the incomplete combustion of fossil fuels and biomass. It contains over 60 % carbon with the major accessory elements hydrogen, oxygen, nitrogen, and sulfur" (Goldberg, 1985).

From a source-based approach the following definitions were made:

- Primary carbon: particulate carbon produced in sources, rather than in the atmosphere, being the sum of primary organic species and black carbon (Novakov, 1984).
- Secondary carbon: organic particulate carbon formed by atmospheric reactions from gaseous precursors (Novakov, 1984). In current literature this fraction is referred to as secondary organic aerosol (SOA).
- Soot: synonymous with primary carbon derived from combustion (Novakov, 1984), or a common name for elemental carbon (Shah and Rau, 1990).

From these historic definitions it is evident that there is no unambiguous separation line between the definitions for elemental carbon, black carbon and soot. Rather, these terms are commonly, but incorrectly, used synonymously.

4.2 Current terminology

More precise and operational definitions have been developed with progressing understanding and measurement capabilities. An in-depth discussion of these issues can

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be found in the papers by Bond and co-authors (2006, 2013), Andreae and Gelencsér (2006), and in interactive comments to Buseck et al. (2012); see Schwartz and Lewis (2012), Prather (2012), Gysel (2012) and published reviews:

- "Soot carbon" or "Soot" (C_{soot}): particles containing carbon with the morphological and chemical properties typical of soot particles from fossil fuel combustion. Soot carbon particles are formed from agglomerates of spherules composed of graphitic-like micro-crystallites. They consist almost exclusively of carbon, with minor amounts of hydrogen and oxygen (Ogren and Charlson, 1983; Andreae and Gelencsér, 2006) and are characterized by a specific surface area ≥ 100 m^2 g^{-1} (Gilot et al., 1993; Kandas et al., 2005). Note that this definition excludes any organic species that might be present as a coating on the spherules.

- Graphitic carbon: particulate carbon having a graphitic-like microstructure characterized by sp^2 – bonded carbon atoms (Ogren and Charlson, 1983). Graphitic carbon is often used as another term for EC (Shah and Rau, 1990).

- ns-soot: from the standpoint of particle morphology, Buseck et al. (2012) introduced the term "ns-soot", which refers to the carbon nanospheres as the constituting element of typical combustion particle aggregates. This definition is linked to the various methods of electron microscopy.

- Elemental carbon (EC): carbonaceous fraction of particulate matter that is thermally stable in an inert atmosphere to high temperatures near 4000 K and can only be gasified by oxidation starting at temperatures above 340 °C. It is assumed to be inert and non-volatile under atmospheric conditions and insoluble in any solvent (Ogren and Charlson, 1983).

- Black carbon (BC): Following Bond et al. (2013), who deserve credit for synthesizing BC definitions for the first time, BC is characterized by the following distinct properties: (1) it strongly absorbs visible light with a mass absorption cross section (MAC) at a wavelength $\lambda = 550$ nm above 5 m^2 g^{-1} for freshly produced

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particles; (2) it is refractory with a volatilization temperature near 4000 K; (3) it is insoluble in water, in organic solvents including methanol and acetone, and in the other components of the atmospheric aerosol; and (4) it consists of aggregates of small carbon spherules of < 10 to approx. 50 nm in diameter. In order to include a distinct microstructural feature, we add a fifth property saying that (5) it contains a high fraction of graphite-like sp^2 – bonded carbon atoms; see Table 1 for a compilation of properties.

With respect to its light-absorbing properties the following definitions have been introduced:

- Light-absorbing carbon (LAC): carbon fraction of the atmospheric aerosol that strongly absorbs light in the visible spectral region (Andreae and Gelencsér, 2006; Bond and Bergstrom, 2006).

- Brown carbon (C_{brown}): light-absorbing organic matter in atmospheric aerosols of various origins, e.g., soil humic substances, humic-like substances (HULIS) (Graber and Rudich, 2006), tarry materials from combustion, bioaerosols, etc. (Posfai et al., 2004; Andreae and Gelencsér, 2006), which tend to appear brown rather than black. The brownish appearance is associated with a non-uniform absorption over the entire visible wavelength range, i.e., increasing absorption with decreasing wavelength in the visible range of the solar spectrum.

4.3 Limitations of current terminology

Currently used terminology exhibits distinct ambiguities and limitations. The term "black carbon" implies optical properties and composition similar to soot carbon or light-absorbing carbon (LAC, which includes C_{soot} and C_{brown}), and particle morphology similar to ns-soot. The word "black" has also come to be associated with measurements by filter-based optical methods, which frequently assume a particular wavelength dependence and absorption per unit mass (Liousse et al., 1993; Petzold et al., 1997; Jeong et

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al., 2004). Moreover, the term "black" is associated with the almost uniform absorption of light over the entire visible wavelength range, with the imaginary part of the refractive index being almost wavelength-independent over the visible and near-infrared spectral range. However, in the climate-science community, BC is the most commonly used term, without consideration of its unclear definition.

The term "elemental carbon" is rated as not necessarily provided by the measurements (Andreae and Gelencsér, 2006; Bond and Bergstrom, 2006) because the name implies a near-elemental composition of the carbon. Instead, EC determined by evolved carbon methods from atmospheric aerosol samples still contains some carbon with functional groups (e.g., C-O) and the molar H/C ratio determined for black carbon in ash is about 0.20 (Kuhlbusch, 1995). Following this concern, Andreae and Gelencsér (2006) proposed the use of "Apparent Elemental Carbon" (EC_a) as the proper terminology for the fraction of carbon that is oxidized above a certain temperature threshold in the presence of an oxygen containing atmosphere. However, the term "elemental carbon" is well established in a wide range of literature focusing on combustion methods and emission inventories. In addition, it is widely used within official bodies as CEN, ISO, as well as NIOSH and operationally defined in all the thermal protocols included in respective standards. Finally, the term "elemental carbon" is used in legislation related to ambient air quality and workplace safety.

5 Recommended terminology and related measurement methods

In consideration of the inadequate definitions available in the literature, and in order to overcome this unsatisfying situation, the authors propose the following consistent terminology which is built along the line of targeted material properties. Table 2 summarizes the recommended terminology and includes related measurement methods and specific instruments. Reporting procedures for the World Data Centre for Aerosols are found at http://www.gaw-wdca.org/.

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Total carbon (TC) mass is used to characterize the mass of all carbonaceous matter in airborne particles.

Total carbon mass is a well-defined property that can be measured with precision better than 10 % by evolved carbon methods.

Black carbon (BC) is a useful qualitative description when referring to light-absorbing carbonaceous substances in atmospheric aerosol; however, for quantitative applications the term requires clarification of the underlying determination.

In the absence of a method for uniquely determining the mass of BC, the authors recommend that the term "BC" should be used as a qualitative and descriptive term when referring generally to material that shares some of the characteristics of BC (see Table 1), in particular its carbonaceous composition combined with its light-absorbing properties. In this manner, BC is already used in atmospheric modeling and assessment studies. For quantitative applications like reporting data from observations or building inventories, the authors suggest using more specific terminology that refers to the particular measurement method as defined in the following. One strong recommendation, however, is to avoid using the term "BC" for evolved carbon methods.

Equivalent black carbon (EBC) should be used instead of black carbon for data derived from optical absorption methods, together with a suitable MAC for the conversion of light absorption coefficient into mass concentration.

In the absence of a standard reference material, it is recommended to report such measurements as aerosol light absorption coefficient, thus avoiding the additional uncertainty introduced by assuming a specific MAC value. When reporting EBC, i.e. mass concentration, it is crucial to identify the MAC value used for the conversion and to specify the approach used for separating potential contributions of BrC or mineral dust to the aerosol light absorption coefficient.

Elemental carbon (EC) should be used instead of black carbon for data derived from methods that are specific to the carbon content of carbonaceous matter.

It is recommended to report data from evolved carbon methods and aerosol mass spectrometry methods as EC. Additionally, data from Raman spectroscopy, which

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addresses the graphite-like structure of carbon atoms, should be reported as EC. Data from any future methods that address the amount of carbon atoms contained in the analyzed sample of particulate matter should also be reported as EC.

Refractory black carbon (rBC) should be used instead of black carbon for measurements derived from incandescence methods.

For incandescence-based methods like LII, SP2 and SP-AMS it is recommended to report data as refractory black carbon, rBC, since these methods mainly address the thermal stability of the carbonaceous matter and require light-absorbing efficiency, i.e., some "blackness" of the analyzed particulate matter. Terminology used so far (e.g. refractory BC, rBC, equivalent refractory BC, erBC, and similar terms containing EC or refractory carbon, RC) should be replaced by the term rBC.

Soot is a useful qualitative description when referring to carbonaceous particles formed from incomplete combustion.

The term soot generally refers to the source mechanism of incomplete combustion (Glassman and Yetter, 2008) rather than to a material property. It is widely used in research on the formation of carbonaceous particles in combustion processes and on the emission of particulate matter from combustion sources. Since atmospheric research usually addresses mixed and aged particles that can no longer be associated with a combustion source process, the recommendation is to avoid using this term for atmospheric aerosol.

With the above recommendations almost all currently known needs for unambiguous terminology of black carbon related research should be covered. As a consequence we recommend terminating the use of other terms that have been applied in the past. In order to support the efforts towards consistent reporting of BC-related measurements the authors of future research papers are requested to clearly state means of calibration and conversion as metadata with any published values.

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6 Conclusions

Despite the huge efforts undertaken in the research field of carbonaceous particles in the atmosphere, the research community is still not and may never be in a position to offer unambiguous conversion relationships between BC data originating from different methods and different aerosol types. Methods are associated with distinct particle properties, which may depend not only on particle chemical composition but also on physical properties like particle size or mixing state. These complex interdependencies very likely inhibit universal quantitative one-to-one conversion relationships between properties.

After having critically reviewed the currently used terminology and after having considered the use of terms not only in the research area of atmospheric composition, air quality and climate change but also in legislation on air quality control and work place safety we propose a terminology that reflects the widespread origin of BC data and permits a consistent reporting of data in the scientific literature that were generated by similar methods.

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Table 1. Properties defining Black Carbon and their consequences for effects and removal.

Property	Characteristics	Consequences
Microstructure	graphitic-like structure containing a large fraction of sp^2-bonded carbon atoms	Low chemical reactivity in the atmosphere; slow removal by chemical processes; strong optical absorption
Morphology	aggregates consisting of small carbon spherules of < 10 to approx. 50 nm in diameter; specific surface area typically greater than 100 $m^2\ g^{-1}$	High capacity for sorption of other species
Thermal stability	refractory material with a volatilization temperature near 4000 K; gasification is possible only by oxidation, which starts at temperatures above 340 °C	High stability in the atmosphere; longer atmospheric residence time
Solubility	insoluble in organic solvents including methanol and acetone, in water, and in the other components of the atmospheric aerosol	Slow removal by clouds and precipitation, unless coated with water-soluble compounds; longer atmospheric residence time
Light absorption	strong light absorption in the spectral range of visible light with mass-specific absorption coefficient typically greater than 5 $m^2\ g^{-1}$ (at $\lambda = 550$ nm) for freshly-produced particles; weak wavelength dependence of light absorption with absorption Ångström exponent typically 1.0–1.5; characterized by a significant, non-zero and almost wavelength-independent imaginary part of the refractive index over the visible and near-visible spectral regions	Reduction of the albedo of clouds, snow, and ice; atmospheric heating; surface cooling – all of which lead to effects on solar radiation and climate

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Table 2. Recommended terminology and related measurement techniques and instruments.

Property	Technique	Instrument	Reference	Reported value	Recommendation
Light absorption	Light absorption measurement	various in-situ and filter-based methods	(Sheridan et al., 2005; Moosmüller et al., 2009)	Light absorption coefficient σ_{ap}; mass concentration computed from σ_{ap} by applying a specific mass absorption cross-section MAC	report as σ_{ap}; if reported as EBC, specify MAC value used for the conversion from light absorption into mass concentration
		Photoacoustic Spec.	(Arnott et al., 2003)		
		Aethalometer	(Hansen et al., 1984)		
		MAAP	(Petzold and Schönlinner, 2004; Petzold et al., 2005)		
		PSAP	(Bond et al., 1999; Virkkula et al., 2005)		
		COSMOS	(Miyazaki et al., 2008)		
Refractory	Measurement of thermal radiation	SP2	(Stephens et al., 2003; Schwarz et al., 2006; Kondo et al., 2011)	Mass concentration	report as rBC specify means of calibration, conversion factor from thermal radiation to carbon mass, and the size-cut of rBC particles
		LII	(Snelling et al., 2005; Chan et al., 2011)		
	Soot Particle Aerosol Mass Spectrometry	SP-AMS	(Onasch et al., 2012)	Mass concentration OC/rBC mass fraction	report as rBC
Chemical composition, carbon content	Evolved carbon methods, thermal evolution of carbon, with optical correction for pyrolysis	various temperature protocols	IMPROVE (Chow et al., 1993), IMPROVE_A , NIOSH (Peterson and Richards, 2002; Chow et al., 2007a), EUSAAR-2 (Cavalli et al., 2010)	Mass concentration OC/EC mass fraction	report as EC; specify temperature protocol used for the sample analysis
	Aerosol Time-of Flight Mass Spectrometry	ATOFMS	(Spencer and Prather, 2006)	Mass concentration OC/EC mass fraction	report as EC
	Soot Particle Aerosol Mass Spectrometry	SP-AMS	(Onasch et al., 2012)	Mass concentration OC/rBC mass fraction	report as rBC, because technique detects carbon that is evaporating under incandescent conditions
Graphite-like microstructure	Raman spectroscopy		(Sze et al., 2001; Mertes et al., 2004; Sadezky et al., 2005; Ivleva et al., 2007)	Mass concentration	report as EC, specify means of calibration
Particle morphology	Electron microscopy	TEM	(van Poppel et al., 2005; Tumolva et al., 2010)	Structural information	not applicable

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