Atmospheric
Chemistry
and Physics

Open Access

Recommendations for reporting "black carbon" measurements

A. Petzold[1], **J. A. Ogren**[2], **M. Fiebig**[3], **P. Laj**[4], **S.-M. Li**[5], **U. Baltensperger**[6], **T. Holzer-Popp**[7], **S. Kinne**[8], **G. Pappalardo**[9], **N. Sugimoto**[10], **C. Wehrli**[11], **A. Wiedensohler**[12], and **X.-Y. Zhang**[13]

[1]Forschungszentrum Jülich GmbH, Institut für Energie- und Klimaforschung IEK-8, 52425 Jülich, Germany
[2]NOAA/ESRL Global Monitoring Division, Boulder, CO 80305, USA
[3]Norwegian Institute for Air Research (NILU), 2027 Kjeller, Norway
[4]Laboratoire de Glaciologie et Géophysique de l'Environnement, Université de Grenoble I – CNRS, 38402 Saint Martin d'Hères cedex, France
[5]Environment Canada, Processes Research Section, Toronto, ON M3H 5T4, Canada
[6]Laboratory of Atmospheric Chemistry, Paul Scherrer Institute, 5232 Villigen PSI, Switzerland
[7]Deutsches Fernerkundungsdatenzentrum, DLR, 82234 Oberpfaffenhofen, Germany
[8]Max Planck Institute for Meteorology, Bundesstrasse 53, 20146 Hamburg, Germany
[9]Istituto di Metodologie per l'Analisi Ambientale (CNR-IMAA), Potenza, 85050, Italy
[10]National Institute for Environmental Studies, 16-2 Onogawa, Tsukuba, Ibaraki 305-8506, Japan
[11]Physikalisch-Meteorologisches Observatorium Davos (PMOD/WRC), 7260 Davos, Switzerland
[12]Leibniz Institute for Tropospheric Research, Permoserstr. 15, 04318 Leipzig, Germany
[13]Chinese Academy of Meteorological Sciences, 46 Zhong-Guan-Cun S. Av., Beijing 100081, China

Correspondence to: A. Petzold (a.petzold@fz-juelich.de)

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). These distinct properties give it relevance in various research 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 related mitigation strategies. Relative to the long-lived greenhouse gases (particularly CO_2 and CH_4), the light-absorbing 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 (Heintzenberg and Winkler, 1991; Pöschl, 2003; Bond et al., 2013). 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, 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 mainly based on emission factors derived from thermal-optical methods that detect the carbon evolving from a heated filter sample, 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 not fully understood and are to a large extent dependent upon season and location of sampling, and type of aerosol.

Furthermore, BC is a highly relevant topic with respect to research on adverse health effects of airborne particulate matter, much of which relies on air quality monitoring. An overview of current knowledge on the impacts of the atmospheric aerosol particle burden on human health is given in the recent integrated assessment of black carbon and tropospheric ozone by UNEP/WMO (2011). Since most epidemiological studies correlate particulate-matter-related health effects to aerosol (including BC) mass concentrations measured by air quality monitoring networks, the recommended terminology may also apply to this research area.

This publication proposes definitions of terms and recommendations for reporting 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, 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 close to unity would still be considered "black" (Schwartz and Lewis, 2012). 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 formally defined as an ideally light-absorbing substance composed of carbon. The formation process is excluded from this definition because of the variety of potential processes. While BC is mostly formed in incomplete combustion of carbonaceous matter, it can also be a product of pyrolysis of carbonaceous matter, i.e., the change of the chemical structure of carbonaceous compounds from loss of hydrogen and/or oxygen atoms at temperatures above approximately 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 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, nonzero 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.

3 Analytical methods

The terms used to identify the various fractions of carbonaceous aerosol are primarily associated with the corresponding measurement methods (e.g., Pöschl, 2003; 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; Gundel et al., 1981; 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 nondispersive infrared absorption or other CO_2 specific detection methods or as CH_4 by a flame-ionization detector.

Currently, different 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) µg m^{-3} (ten Brink et al., 2004) in determining the total mass 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 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). Pyrolysis correction is performed by measuring filter transmission or reflectance during the thermal-optical analysis step. Yet the correction differs significantly between transmission measurement (TOT, thermal-optical transmission) or reflectance measurement (TOR, thermal-optical reflectance) and temperature protocol (Schmid et al., 2001). The EC fraction formed by OC conversion during pyrolysis is referred to as pyrolyzed carbon (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 $1\,\text{Mm}^{-1} = 10^{-6}\,\text{m}^{-1}$. There is no overall agreed reference method for measurement of the aerosol light absorption coefficient, because many of the 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 (e.g., Petzold and Niessner, 1996; Arnott et al., 1999, 2003; Lack et al., 2006), while the measurement of light extinction minus light scattering may offer another possibility in the laboratory (Schnaiter et al., 2005b; Sheridan et al., 2005) or in atmospheric plumes with very high aerosol mass concentrations (Weiss and Hobbs, 1992). An in-depth review of light absorption measurement methods is provided by Moosmüller et al. (2009).

The conversion of the aerosol light absorption coefficient into a light-absorbing carbon mass concentration [BC] is based on the relationship $[\text{BC}] = \sigma_{ap} \times \text{MAC}^{-1}$. It therefore

Table 1. Properties defining black carbon and their consequences for effects and removal.

Property	Characteristics	Consequences
Microstructure	Graphite-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	Fractal-like chain aggregates consisting of small carbon spherules of $< 10\,nm$ to approximately $50\,nm$ in diameter; fractal dimension ranges from ≤ 2.0 for fresh combustion particles to $\cong 3.0$ for aged aerosol; specific surface area typically larger than $10\,m^2\,g^{-1}$ and may exceed $100\,m^2\,g^{-1}$.	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 any solvent including water.	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, nonzero and 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.

requires precise knowledge of the mass-specific absorption cross section (MAC; also referred to as mass absorption efficiency, MAE) often reported in units of $m^2\,g^{-1}$. This coefficient, however, varies significantly in time and space depending upon source emissions, transformation during transport, etc. (Bond and Bergstrom, 2006; Chan et al., 2011).

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 effective MAC of an aerosol composed of such particles will depend on their size distribution.

The application of this conversion also assumes that BC is the only light-absorbing particulate species present. Contributions to absorption from noncarbonaceous light-absorbing aerosol components like mineral dust (see, e.g., Petzold et al., 2009, 2011), or by non-BC light-absorbing carbonaceous matter (i.e., 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 can be characterized by the absorption Ångström exponent $å_{ap} = -\ln(\sigma_{ap}(\lambda_1)/\sigma_{ap}(\lambda_2))/\ln(\lambda_1/\lambda_2)$ for a certain wavelength interval $[\lambda_1, \lambda_2]$. While BC is characterized by a low value of $å_{ap}$ between 1.0 and approximately 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; Chen and Bond, 2010; Kim et al., 2012) associated with $å_{ap}$ values as high as 7 and beyond for the visible range. However, in a recent paper, Lack and Langridge (2013) investigate the uncertainties of using the method of separating BC and organic non-BC light-absorbing species by $å_{ap}$ values. The main conclusion is that unless the non-BC absorbers contribute more than 40 % of absorption, a quantitative attribution of the various absorbers cannot be derived.

Mineral dust as another important light-absorbing aerosol component is characterized by strong absorption in the blue and green visible range and low absorption in the red spectral range, which results in $å_{ap}$ values of 3 and larger at visible wavelengths (Petzold et al., 2009). Summarizing, overdetermination of light absorption associated with BC by non-BC light-absorbing aerosol compounds can be minimized by choosing a detection wavelength in the red spectral region (600 nm $< \lambda \leq$ 700 nm) where cross-sensitivities to mineral dust and organic carbon compounds are lowest.

Furthermore, absorption enhancements 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) must be considered in the interpretation of light absorption measurements.

Another challenge for applying this conversion is the absence of an overall agreed reference material which links light absorption to BC mass. 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-induced 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 of an ensemble of particles (LII) (Snelling et al., 2005; Chan et al., 2011) or of single particles, e.g., the single-particle soot photometer (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.

Laser-induced 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 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. The calibration of incandescence instruments must be performed using reference carbon material such as fullerene or recommendations from Baumgardner 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; Potgieter-Vermaak and Van Grieken, 2006; 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 the 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. Limitations are mainly related to variations in the parameters of the Raman spectra, i.e., bandwidths and band intensities, for different types of carbonaceous reference materials and the carbonaceous fraction of the atmospheric aerosol. Yet, the method of Raman mapping (Ivleva et al., 2007) offers a promising approach towards a quantitative application of Raman spectroscopy for carbon mass concentration measurements.

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 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^+, C_2^+, C_3^+, 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.

The SP-AMS technique represents a hybrid of laser-induced incandescence and mass spectrometry methods because it combines a laser-induced incandescence approach for heating and vaporizing the sampled particles with mass spectrometry techniques for the detection of resulting charged carbon clusters. As a result of this combination of techniques, SP-AMS measurements are, from the standpoint of the detection scheme, more similar to single particle mass spectrometers (i.e., carbon cluster ion detection) than the incandescence signal (intensity of thermal radiation) measured by the SP2. However, the carbon ions measured by an SP-AMS come from carbon that is evaporating under incandescent conditions (i.e., refractory), and are not a product of a laser-induced plasma or multi-photon ionization events that 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 (e.g., Fruhstorfer and Niessner, 1994; Pósfai 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 application for routine monitoring purposes is strongly limited due to labor-intensive sample preparation and data analysis. However, these limitations are reduced to a large extent by recent computer-controlled image processing approaches, enabling the automatic characterization of the morphology of thousands of particles deposited on a filter.

3.7 Limitations due to particle size

For many methods, the lower limit of detectable particle size must be considered since carbonaceous particles may be as small as 10 nm diameter or less, depending on their origin. Methods measuring volume properties, such as in situ light absorption techniques or LII, are not affected by a lower limit of detectable size because they do not refer to single-particle properties. For techniques based on the analysis of particle-loaded filters, the lower size limit is determined by the sampling efficiency of the selected filter material; see, e.g., Hinds (1999) for details. For single-particle methods like SP2, SP-AMS or electron microscopy methods, however, this limitation can become a serious constraint. In particular, the single-particle SP2 method can only detect particles larger than 70–80 nm diameter (Schwarz et al., 2010), a limitation that may also be an issue for the related SP-AMS method (Onasch et al., 2012), while microscopy-based methods can characterize particles as small as 10 nm diameter (Tumolva et al., 2010).

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 (OC, see below).

4.1 Historic definition

In 1978, the first Conference on Carbonaceous Particles in the Atmosphere was held in Berkeley, California, USA. At this conference, the first methods for the measurement of carbonaceous aerosols were presented and the link between strong light absorption by aerosol particles in the visible spectral range and their carbonaceous matter content was established (Rosen et al., 1978a, b; Weiss et al., 1978). The former article (Rosen et al., 1978a), which led to the development of the continuous, filter-based absorption photometers, compared Raman spectra and the optical "absorption" measurement of "graphitic soot". The first mention of the optical determination of "black carbon" came in an article that compared thermal methods and the optical transmission method (Gundel et al., 1981). It states in the first paragraph "The term 'black carbon' is used in this paper to refer to the optically absorbing carbonaceous component of ambient and source aerosols."

Based on the pioneering work of Novakov (1984), Goldberg (1985) and Shah and Rau (1990), the following analytically based definitions have been introduced since then:

–　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).

–　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 graphite-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 [by mass] 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 improvements in understanding and measurement capabilities. An in-depth discussion of these issues can be found in the papers by Bond et al. (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 graphite-like microcrystallites. 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 surface area well above $10\,m^2\,g^{-1}$ with maximum values $\geq 100\,m^2\,g^{-1}$, depending on the combustion source (e.g., Gilot et al., 1993; Popovitcheva et al., 2000; 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 graphite-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 viewpoint of particle morphology, the term "ns-soot" was introduced, which refers to the carbon nanospheres as the constituting element of typical combustion particle aggregates (Buseck et al., 2012; Adachi and Buseck, 2013). 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 nonvolatile 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 MAC value above $5\,m^2\,g^{-1}$ at a wavelength $\lambda = 550\,nm$ for freshly produced 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 $< 10\,nm$ to approximately $50\,nm$ 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.

– Refractory black carbon (rBC): carbonaceous fraction of particulate matter that is insoluble and vaporizes only at temperatures near $4000\,K$ (Schwarz et al., 2010).

With respect to light-absorbing properties of carbonaceous aerosols, 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 (BrC): 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. (Pósfai et al., 2004; Andreae and Gelencsér, 2006), which tend to appear brown rather than black. The brownish appearance is associated with a nonuniform 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 BrC), 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 al., 2004). Moreover, the term "black" is associated with the efficient 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 not necessarily an accurate description of what is actually measured (Andreae and Gelencsér, 2006; Bond and Bergstrom, 2006) because the name implies a near-elemental composition of the carbon. In reality, 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, we propose the following consistent terminology that 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/.

Total carbon (TC) mass is used to describe 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. "BC" is already used this way in atmospheric modeling and assessment studies. For quantitative applications like reporting data from observations or building inventories, we suggest using more specific terminology that refers to the particular measurement method as defined in the following. One

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 Photoacoustic Spec. Aethalometer MAAP PSAP COSMOS	Sheridan et al. (2005); Moosmüller et al. (2009) Arnott et al. (2003) Hansen et al. (1984) Petzold and Schönlinner (2004); Petzold et al. (2005) Bond et al. (1999); Virkkula et al. (2005) Miyazaki et al. (2008)	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
Refractory	Measurement of thermal radiation Soot Particle Aerosol Mass Spectrometry	SP2 LII SP-AMS	Stephens et al. (2003); Schwarz et al. (2006); Kondo et al. (2011) Snelling et al. (2005); Chan et al. (2011) Onasch et al. (2012)	Mass concentration Mass concentration OC/rBC mass fraction	report as rBC specify means of calibration, conversion factor from thermal radiation to carbon mass, and the size-cut of rBC particles report as rBC
Chemical composition, carbon content	Evolved carbon methods, thermal evolution of carbon, with optical correction for pyrolysis Aerosol Time-of Flight Mass Spectrometry Soot Particle Aerosol Mass Spectrometry	various temperature protocols ATOFMS SP-AMS	IMPROVE Chow et al. (1993) IMPROVE_A, NIOSH Peterson and Richards (2002); Chow et al. (2007a) EUSAAR-2 Cavalli et al. (2010) Spencer and Prather (2006) Onasch et al. (2012)	Mass concentration OC/EC mass fraction Mass concentration OC/EC mass fraction Mass concentration OC/rBC mass fraction	report as EC; specify temperature protocol used for the sample analysis report as EC 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

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 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 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 methods based on laser-induced incandescence, 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 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 of hydrocarbon fuels (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 as well as in the field of particulate matter-related health effects. Thus, terming particles emitted from a combustion source as soot particles is in agreement with the recommended terminology.

Mixed particles containing a BC fraction should be termed BC-containing particles instead of BC particles or soot particles.

Since atmospheric research usually addresses mixed and aged particles that can no longer be associated with any combustion source process, the recommendation is to avoid using the terms soot or BC particle for atmospheric aerosol. It is also recommended to refer to the BC components of individual particles as the BC cores without any confusion about non-BC mass either internally or externally mixed with the BC. Reported BC fractions of particle mass should be consistently referred to as rBC, EC, or EBC fractions, depending on the measurement technique.

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.

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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