Atmos. Chem. Phys. Discuss., 9, C11044–C11049, 2010
www.atmos-chem-phys-discuss.net/9/C11044/2010/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Sources of uncertainties in modelling Black Carbon at the global scale" *by* E. Vignati et al.

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Received and published: 22 February 2010

This is a good paper, and suitable for publication in ACP. Here are my suggestions for certain sections of the paper. We thank the reviewer for the valuable comments. We address the specific points below.

Major issues: – Page 5, line 2 I enjoyed the discussion about the BC/EC differences, but I disagree with this sentence: "Regrettably however, these discrepancies are usually disregarded in the literature and the terms elemental carbon and black carbon are used interchangeably as synonyms of soot."

The measurement community is very good about using the conventional operational definitions. The modeling community uses both terms interchangeably, but that is ok

since both measurement techniques are attempting to measure the same thing (i.e., the nearly graphitized portion of soot). – We have changed the text in the introduction accordingly to the suggestion: "Regrettably however, these discrepancies are usually disregarded in the modelling studies and the terms elemental carbon and black carbon are used interchangeably as synonyms of soot." The authors believe that thermal-optical measurements (EC) and light absorption measurements (BC) are not directly comparable since they both attempt to measure atmospheric "soot", however taking advantage of two different key properties, its refractivity and its ability to absorb light, respectively. More than 50 inter-method and inter-laboratory comparisons for the determination of EC and BC have been conducted, showing typical differences of a factor of 2 between methods but sometimes even of a factor of 7 (Watson et al., 2006). These discrepancies cannot be disregarded nor the terms EC and BC used interchangeably as synonymous of soot.

Watson, J.G. Chow, J.C. Chen, L.-W.A.: Summary of organic and elemental carbon/black carbon analysis methods and intercomparisons, Aeros. Air Qual. Res., 5, 65-102, 2005.

Page 5, line 6 The authors state: "Of the three, the term black carbon is the one most commonly used in the climate modeling community for soot/black carbon/elemental carbon, as it refers to the optical property, which is that relevant for climate." I disagree that BC refers to an optical property; BC is *derived* from an optical property, but it doesn't really refer to an optical property per se (i.e., units are still mass of BC per unit volume of air). Since the optical properties of BC changes as BC becomes internally mixed with other aerosols (and the instrument does not account for this because it uses a single specifice absorption for conversion), the optical information is essentially unknown.

We have changed the text in the introduction accordingly: "Of the three, the term black carbon is the one most commonly used in the climate modeling community for soot/black carbon/elemental carbon, as it derived from the optical property, which is

that relevant for climate."

I suspect (but do not know) that the modeling community has converged on the term BC because they often compare their results to aircraft measurements. Since thermal methods are generally too slow for aircraft measurements, modelers often use the term BC. – The term BC is preferred because modelling studies are driven more by radiative forcing evaluations rather than the use of aircraft measurements

Page 11, line 26: What and where are the long term campaign measurements of EC and BC? Some info about these campaigns should be included, as well as info about the instruments that are used to measure BC. –

We have added the information in the text, as it follows: "The long-term measurements were collected at the Arctic stations of Alert (Sharma et al. 2004, 2006 (aethalometer)) and Barrow (Bodhaine, 1995 (aethalometer)), in the Amazon basin (Echalar et al, 1998 (light reflectance technique)), and at Halley, Antarctica (Wolff and Cachier, 1998 (aethalometer))."

Page 14, lines 12-20: Regarding "It is difficult to point to the right reason for this underestimation..." Strictly speaking, organics can affect EC measurements if some of the organics are charred in the heating process. Organics don't affect BC measurements at 880 nm (the Aethalometer wavelength) though, and have little affect at 565 nm (PSAP wavelength), per Andreae and Gelencser (2006); hence, they should *not* affect BC measurements. However, there is some evidence of large carbon spheres with significant absorption throughout the visible and NIR wavelengths, but the prevalence of these highly absorb-ing spheres in the atmosphere is presently unknown (Alexander, 2008; Posfai, 2004). It seems that they are associated with biofuel and biomass burning.

More precisely, organics can affect thermal-optical measurements of EC essentially under two circumstances: i) a non-adequate correction for charring is applied, leading to an overestimation of EC, as stated by the referee or ii) high molecular weight organic

species with high degree of refractivity are present and are erroneously determined as EC and not as OC (i.e. organic compounds which do not evolve in the helium phase of the thermal-optical analysis and slip into the successive helium/oxygen phase) (Cavalli et al., 2010). Moreover, organics do not likely absorb light at IR wavelengths as 880 nm; however, absorption increases towards lower wavelengths (Kirchstetter and Novakov, 2004). Therefore organics can strongly influence light absorption measurements made with instruments using green light (e.g. the PSAP at 565 nm) or white light (e.g. the white-light aethalometer) (Andreae and Gelencsér, 2006).

T.W. Kirchstetter, T. Novakov, and P.V. Hobbs (2004). Evidence that the spectral dependence of light absorption by aerosols is affected by organic carbon. J. Geophys. Res. 109, D21208, doi: 10.1029/2004JD004999.

F. Cavalli, M. Viana, K.E. Yttri, J. Genberg, and J.-P. Putaud (2010). Toward a standardised thermal-optical protocol for measuring atmospheric organic and elemental carbon: The EUSAAR protocol. Atmos.Meas. Tech., 3, 79-89.

I have another hypothesis for model underestimation in biomass burning regions: BC is typically measured with an Aethalometer, which converts an absorption measurement to a mass retrieval via a single conversion factor. This conversion factor is related to the absorption efficiency (m2/g) of BC, which varies with EC mass fraction for internally mixed EC (see Petzold, 1997;Neusub, 2002). Since biomass burning aerosols have larger than average EC mass fractions (and consequently, lower absorption efficiencies), the "standard" Aethalometer conversion factor may be too high for those aerosols. – The authors compare BC measurements with model output and discuss discrepancies for regions strongly influenced by biomass burning episodes. The model always underestimates the observations. A series of hypotheses are discussed in the manuscript. The one proposed by referee could obviously be an additional explanation for the observed differences. The standard specific cross section used to derive EBC concentrations in aethalometer measurements might not be appropriate for biomass burning aerosol.

Accordingly to the reviewer comment the following text has been changed in the introduction: “Furthermore, observations of both EC and BC, which are heavily influenced by biomass burning sources, are known to be modified by the presence in the sample of light-absorbing organic material that is not black, the so-called brown carbon (e.g. highly refractive organics determined as EC in thermal-optical methods and standard specific cross section used to derive BC concentrations in aethalometer measurements not appropriate for biomass burning aerosol) (Andreae and Gelencsér, 2006).”

Page 17, line 25: The absorption of OC at the Aethalometer and PSAP wavelengths is essentially nil, according to Andreae and Gelencser (2006). Dust absorption at these longer wavelengths is negligible as well. However, Andreae and Gelencser (2006) discuss some issues associated with all filter measurements that are relative to this paper. –

As stated above, light absorption measurements at 880 nm, generally used to derive EBC, are likely to be not affected by organics; however, interferences by organics increase towards lower wavelengths and therefore most strongly influence light absorption measurements made with instruments using green light (e.g. the PSAP at 565 nm) or white light (e.g. white-light aethalometer). Furthermore, measurements have shown that dust appreciably absorbs at wavelengths below 600 nm (Sokolik and Toon, 1999) due to the presence of iron oxide, as hematite; however, non negligible interferences have been also reported at larger wavelengths including 880 nm (Fialho et al. 2005). Generally speaking, the equivalent specific attenuation of dust is 2 to 3 orders of magnitude smaller than that of EBC. Thus, a dust storm concentration of 100 μg m-3 of actual dust might produce an incorrect contribution to the Aethalometer "as if" there was 0.1 to 1 μg m-3 of EBC. In general, single-wavelength light absorption measurements may not be adequate for assessing absorption of solar radiation in the troposphere.

Minor issues: + There are several run-on sentences that make it difficult for the reader to follow (p10, line 9-11; p10 line 25 through p11 line 3; p12, line 11-14; p17, line 10-14;

p18, line 19-22, to name a few). The readability of the paper could benefit with some light technical editing.

The whole paper has been revised for technical editings

+ Page 14, line 8: does "other" mean "other than IMPROVE and EMEP"? This should be stated explicitely.

Yes. This has been specified in the text

References: Alexander, D., P. Crozier, and J. Anderson (2008), Brown carbon spheres in east asian outflow and their optical properties, Science, 321, 833-836. Neusub, C., T. Gnauk, A. Plewka, H. Herrmann, and P. Quinn (2002), Carbonaceous aerosol over the Indian Ocean: OC/EC fractions and selected specifications from size-segregated onboard samples, J. Geophys. Res., 107 (D19), 8031, doi:10.1029/2001JD000327. Petzold, A., C. Kopp, and R. Niessner (1997), The dependence of the specific attenuation cross-section on black carbon mass fraction and particle size, Atmos. Environ., 31 (5), 661-672. Posfai, M., A. Gelencser, R. Simonics, K. Arato, J. Li, P. Hobbs, and P. Buseck (2004), Atmospheric tar balls: Particles from biomass and biofuel burning, J. Geophys. Res., 109, D06213, doi:10.1029/2003JD004169.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 24317, 2009.