

Interactive comment on “Using measurements for evaluation of black carbon modeling” by S. Gilardoni et al.

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We thank both the referees for their suggestions that we believe improved the readability of the paper.

Referee #1

1. This paper addresses the difficult topic of validating modeled BC concentrations with measurements. This topic is difficult, in part, because we do not have great confidence in our ability to routinely measure BC at the present time. Additionally, most of the available BC measurements are obtained at the surface, so we are stuck with a spatial sampling mismatch (i.e., surface measurements are “points,” and models provide concentrations in a grid). Additionally, the surface is a difficult region for models,

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as the boundary layer dynamics are more difficult to parameterize than the rest of the atmosphere. Unfortunately, the authors weren't able to offer any significant improvements to the present situation.

We are grateful to the first referee for pointing out the need to clarify the goal of the present study. The authors do not want to validate the TM5/M7 global model, which was already evaluated in Vignati et al 2010b; this paper focuses on the methodologies to evaluate black carbon modeling in general. The TM5/M7 model was used in the discussion as an example and its choice was due to its availability. To avoid misunderstanding, the paper objectives (page 11319, line 14) are reformulated as:

- i. the optimization of a validation procedure that includes evaluation of observation representativeness,
- ii. the discussion of statistical tools to compare model and observations.

The paper describing TM5/M7 validation is added to the references and to the text at page 11325 line 6. The first paragraph of the conclusion is reworded as follow:

“Model evaluation studies are based on the comparison of model data and observations. The goal of this paper is to discuss, compare and evaluate a variety of statistical and analytical tools in order to better inform such evaluation.”

In addition, to avoid that the reader attention would focus on the TM5/M7 model validation, any reference to the performance of this specific model is removed from the conclusions.

2. The paper is not easy to follow, in general. It is often difficult to see where the author is leading the reader. I suggest “lead-in” paragraphs at the beginning of each section, and “lead-in” sentences at the beginning of each paragraph. It is very important for the reader to know where he/she is going, as this tremendously aids comprehension. Otherwise, the reader is forced to backtrack for clarification, and most customers are not patient enough to do this for the length of a paper. Passive voice adds to the

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confusion; it is better to use an active voice.

Following the referee suggestions the authors add several lead-in paragraphs and concluding paragraphs, as reported at point 5, 11, and 12 of answers to referee #2.

3. The authors spend quite a bit of time in the intro explaining the problems associated with measuring black carbon, but then they do not solve or even address these problems in the paper. The intro is supposed to launch the rest of the paper, and placing this info in the intro leads the reader to believe that this paper will address this issue. I would move the first few paragraphs of the intro to the Method section, and begin the paper with what is presently on line 19 of page 4 ("BC modeling is a crucial component of. . ."). This focuses the reader on the true topic of the paper, which is an attempt at validation of modeled BC.

We thank the referee for his suggestion and move the first paragraphs of the introduction to section 2.1.1.

4. I strongly recommend against the term "equivalent BC." If the authors insist on using this terminology, they need to define "equivalent." The term "BC" already denotes an optical measurement, so what value does the adjective "equivalent" add to the definition?

We clarify the difference between black carbon and equivalent black carbon adding the following sentence to section 2.1.1 Light attenuation and equivalent black carbon measurements.

To take into account this dependency and variability we refer to the light absorbing carbon as equivalent black carbon (EBC) and not BC. The term BC implies to have optical properties equivalent to those of soot, while EBC is operationally defined as the amount of light absorbing carbon that would give the same signal of soot in an optical instrument like the aethalometer (Andreae et al. 2006).

5. The 6x4 degree grid is very large for comparisons to surface sites, and the au-

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thors arguments to justify the comparisons are not very strong. For instance, the authors state that Fig 2a indicates that the EBC trend at Alert is similar to other polar sites (which is true), but the magnitude of the monthly means can vary by a factor of 2-3 between sites. Also, on page 17, line 23, the authors even attribute the model/measurement discrepancy to grid size. Since grid size is always a potential factor in these comparisons, how can one possibly validate the model in these large grid cells?

TM5/M7 global model was already evaluated by conventional methods in Vignati et al. (2010b), so this paper uses this specific model to illustrate the use of observations to discuss model outputs. As the referee points out, a large model spatial resolution might compromise the use of surface data during model validation. The authors want to discuss this issue, e.g. the difficulty to compare point observation to large surface model output; for this reason the discussion includes a spatial resolution of 6x4 degree. The discussion of sites with different features shows that for remote areas the model spatial resolution is not an issues (i.e. Alert), while in spatial heterogeneous areas it can be responsible for model/observation disagreement (i.e. Bondville). To clarify the objectives of the 6x4 degree grid cell use, the authors add the following sentence at page 11327 line 18:

The large grid cell is a non-optimal condition that we want to test in order to discuss model evaluation.

6. The authors talk about the variability of measured values for σ^* (Figure 3 even includes two assumptions for σ^*), but they do not talk about the modeled values for σ^* . Some comments about the modeled σ^* are important, as this directly effects the radiation field. The authors need to think about how internal mixing affects the specific absorption; internal mixtures with high BC fractions will have much lower specific absorption than internal mixtures with low BC fractions.

The model does not calculate optical properties of the particles. Black carbon concen-

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trations are calculated from emissions given as input to the model through BC emission inventory maps taking into account the atmospheric processes.

7. There is something wrong with Eq 3 or Eq 4; these equations are nearly identical, with the exception of the ' σ^* '. Accepting both of these equations to be true requires $\sigma = \sigma^*$ and $C \times R = 1$. Also, I have never seen a form of the Lambert-Beer (or Bouguer) law that looks like Eq 3, where is the attenuation? Is Babs dimensionless, like BATN? No, that won't work. Is EBC a mass concentration or a number concentration? What are the dimensions of C and R? I suppose I could sort all of this out if I worked hard enough, but most readers don't want to work that hard (especially with the inconsistency of Eqs 3 & 4). This muddling of the equations causes additional confusion in the next paragraph. Eq 3 seems to indicate that σ is the intrinsic specific absorption of BC (i.e., without instrument artifacts), but the remainder of the paper discusses σ^* .

We are very grateful to the referee for pointing out the need to clarify equation 3 and 4 and the typos of equation 4. Equation 3 is derived from Weingartner et al (2003) and text is modified with the addition of this reference, while equation 4 is corrected as:

$$\text{BATN} = \text{EBC} \times \sigma^*$$

To clarify the equations, the dimensions of BATN, Babs, C, and R are specified in the text.

8. Are the aethalometer measurements "wet" or "dry?"

The measurement protocols vary with data provider. According to the metadata provided to the WDCA with the data, Alert, and Ispra make ambient air measurements, Mace Head has temperature control, but no specified temperature, Jungfraujoch heats to 25 C, while Bondville and Trinidad Head are both dried by heating, to less than 40% RH. Table 1 has now been expanded to include this information.

9. Starting on page 9, line 24, the authors state that the measured value of σ^* is 20 m²g⁻¹, but that they follow the lead of Malm et al. (1994) and use 10 m²g⁻¹. Why?

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How is this justified?

The authors use σ^* equal to 10 m²g⁻¹ for the site of Bondville in agreement with Malm et al. (1994). For clarification, the paragraph at page 11323 line 24-29 is worded as:

The comparison of EBC from light integrating plate measurements (LIPM) and EC from thermo-optical measurements at rural sites of the United States (Huffman et al. 1996, Malm et al. 1994) suggests that σ^* is equal to 20 m²g⁻¹; nevertheless Malm et al. (1994) used 10 m²g⁻¹ during their discussion of the IMPROVE network data, as suggested by theoretical calculations; in fact, the value 20 m²g⁻¹ would result from the misidentification of OC/EC split during the thermo-optical measurements and the consequent underestimation of EC. EBC concentrations presented here for Bondville and Trinidad Head are calculated assuming σ^* equal to 10 m²g⁻¹

10. Trinidad Head should be a much cleaner location than Bondville, and is likely to have smaller fractions of BC in the internally mixed component. Lower BC fractions result in higher specific absorptions, which should also be mentioned in this paper. Also, the authors state that "the uncertainty of black carbon optical properties compromises the use of EBC measurements for model evaluation" at Trinidad Head (page 15, line 23), yet they continue to use this site for the remainder of the paper.

Line 20 at page 11329 is rephrased to clarify that lower BC fractions lead to higher specific absorptions:

... aged aerosol is mixed with sulfate and other organic species that enhance ability of BC to absorb visible light through the lens effect (Liousse et al. 1993).

The authors want to make the reader aware that EBC measurement uncertainty can compromise the use of measurements for model evaluation, nevertheless the authors continue using this site because they intend to test the model evaluation tools and not the model itself; thus, non-optimal sites are of particular interest.

11. Table 1 indicates that the authors are using different wavelengths for determining

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the BC at the different surface sites, sometimes as low as 370 nm; why not use the 880 nm wavelength for all of the analyses? This would minimize or eliminate the influence of dust and OC. Also, the authors indicate an aethalometer wavelength of 820 nm at Alert, but this is not one of the standard or alternative wavelengths listed in the aethalometer manual. Is this a custom-built instrument, or a typo?

Table 1 reports the measurements made available for model evaluation by the Global Atmospheric Watch (GAW) network and by the European Monitoring and Evaluation Programme (EMEP), and not measurements performed by the authors, as illustrated in section 2.1. The measurement conditions at some sites might not be optimal to distinguish EBC from dust and OC interference, nevertheless, this is the typical situation encountered during model validation and the authors found important to discuss this issue.

As regards the wavelength of the measurements from Alert, the AE-6 aethalometer was a broadband instrument, as was the AE-9 model used at Mace Head, and Table 1 has been modified accordingly. Both 820 (Allen et al, 2000) & 880 nm (Lavanchy et al, 2003 & Sharma et al. 2004) have been suggested as the effective wavelength for the broadband signal. Curiously in the original metadata as supplied by Sharma the wavelength is given as 820 nm, whereas the 2004 paper refers to 880 nm.

Vignati, E., Karl, M., Krol, M., Wilson, J., Stier, P., and Cavalli, F., Sources of uncertainties in modeling black carbon at the global scale, *Atmospheric Chemistry and Physics*, 10, 2010b

Weingartner, E., Saathoff, H., Schnaiter, M., Streit, N., Bitnar, B., and Baltensperger, U., Absorption of light by soot particles: Determination of the absorption coefficient by means of aethalometers, *J. Aerosol Sci*, 34, 1445–1463, 2003.

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