

Interactive comment on “Different characteristics of char and soot in the atmosphere and their ratio as an indicator for source identification in Xi’an, China” by Y. M. Han et al.

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Received and published: 24 July 2009

We appreciate these valuable comments and suggestions for helping us to improve our manuscript. These comments will improve the clarity of the method, as well as the definition for EC, BC, brown carbon, light-absorbing carbon, char and soot.

1. The primary result of this study shows that comparing char-EC to soot-EC is a better indicator of EC combustion sources than the often used OC/EC ratios. The authors successfully explain the serious inadequacy in using OC/EC as a marker for combustion source and provide a strong case for the use of char-EC/soot-EC ratios instead. The primary weakness in the method is the complete reliance on the TOR method to

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separate the EC into char and soot components. In this method, EC is separated into 3 categories based entirely on the refractive properties of the sample (i.e. the temperature at which the EC is evolved). This assumes that the refractive properties of EC don't change after initial formation, which is a faulty assumption. The presence of salts and other catalysts in the sample can drastically alter the temperature at which optically active EC evolves (Novakov and Corrigan, 1995). Treating the samples with water and acetone removed different species from the samples and changed the temperature at which soot evolves by over 100 in some cases. There is a strong potential for soot EC in the presence of catalysts to be mistaken for char-EC using the TOR method alone. While I agree that the authors present strong evidence for their conclusions, relying solely on the TOR may lead to errors and this should be discussed. In the conclusions the authors also mention that char-EC absorption has a much stronger spectral dependence than soot-EC. Combining this observation with the TOR method may provide more robust results.

Reply: We agree with the reviewer about these comments completely. Indeed, some of salts and catalysts can influence the separation between OC and EC, as well as char-EC and soot-EC. This has also been presented in our recent paper (Han et al., 2009, Chemopshere), which suggests that the presence of ions such as Cl⁻, F⁻, and NO₃⁻ would decrease the activation energy of soot. Many studies have shown that there are still lots of problems in EC determination after long term experience in carbonaceous aerosol studies and no universally accepted method for EC determination exists. Similarly the method for the separation between char and soot in this paper seems also not "perfect". However, we agree with the reviewer that "Combining this observation with the TOR method may provide more robust results". We will discuss the potential influence of the presence of catalysts in our revised paper.

2. Other minor issues: Page13273 "EC is generally subdivided into char and soot." EC is certainly divided that way in this paper, but char is not generally referred to as EC, but rather "brown carbon" or a component of light absorbing carbon (LAC).

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Reply: The separation of EC into char and soot originates from the monograph Black Carbon in the Environment by E. D. Goldberg (1985), as well as from some review papers by Kuhlbusch (1997) and Masiello (2004), etc. There are still some confusions about the definition of EC or BC; many studies suggest that char is categorized into brown carbon, and in the aerosol field some acknowledged that only soot is EC. However, in soil and sediment fields, char was also incorporated into EC. In our opinion, BC or EC themselves just indicate the mixed materials with no universally accepted definition, it's better to use terms, char and soot, in carbonaceous aerosol studies. The two kind of EC have different chemical and physical characteristics, as well as the different optical properties.

3. Page 13276, line 17 “Comparing EC concentrations in Xi’an: : are well correlated with EC in different Chinese cities.” This sentence is awkwardly written and I am unclear as to what is meant by “rank highly”.

Reply: We changed it to “indicates that EC is very high in Xi’an, and this may also imply that char-EC is high since char-EC is well correlated with EC in different Chinese cities (Han et al., submitted).”

4. Page 13277, line 3, “: : which confirms the very small difference in soot-EC concentrations at large.” I’m not sure what the authors mean by this, but I think what they are trying to say is that atmospheric soot-EC concentrations do not vary much across China. Either way, it should be clarified.

Reply: We changed it to “. . . which confirms the similarity in soot-EC values across China.”

5. Line 24 -26, Why does neglecting wet scavenging imply that soot may have a longer lifetime? This doesn't really make sense, as wet scavenging is such a strong factor in soot lifetime that it should not be neglected. I agree with the first reviewer that there seems to be a discrepancy here. Early in section 4.4, the authors state that soot is very susceptible to wet removal, however at the end of section 4.4, make the statement that

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char is larger and therefore more easily removed by wet deposition and therefore soot remains longer in the atmosphere. This really needs to be clarified as the different removal rates of char and EC strongly affect the usefulness of using their ratios as a source identifier.

Reply: Sure, wet removal is a very important factor for soot lifetime. However, compared with the influence of wet scavenging on char, it seems that soot is less susceptible. So we deleted the first part of the sentence and just say “Soot-EC showed a narrower seasonal variation, with a maximum level in spring ($1.85\mu\text{g m}^{-3}$) and minimum level in summer ($1.15\mu\text{g m}^{-3}$). This implies that soot may have an even longer lifetime than previously estimated for total EC (Orgen and Charlson, 1983).” The corresponding modification has been done (see reply to the first reviewer).

6. Page 13278, line 8, “: :and soot from gas-particle re-condensation.” I think this should just be condensation. Page 13281, line 12 “: : neglected in summer.” Remove “in summer” from that sentence.

Reply: Corrected.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 13271, 2009.

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