

Interactive comment on “Influences on the fraction of hydrophobic and hydrophilic black carbon in the atmosphere” by G. R. McMeeking et al.

Anonymous Referee #1

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The manuscript presents a comprehensive exercise on BC hygroscopicity and activation. The authors use state-of-the-art instrumentation to study water uptake and all relevant BC properties simultaneously, which is a great advantage over conventional monitoring techniques. The study extends from laboratory experiments with BC proxies to ambient measurements at an urban location. The approach the authors rely on is purely a modelling one, namely that BC is getting hygroscopic and becoming CCN in cloud formation. While this is clearly an important question and relevant for global aerosol climatology, the instrumentation used by the authors would allow more detailed answers, far beyond that required by the modellers.

The simple view is that BC particles acquire some soluble coating which eventually makes them available as CCN at specific supersaturations as a result of the combined

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effects of increased size and higher water-soluble fraction. The coating likely consists of sulphate and perhaps some oxidized organic compounds (e.g. malonic acid) which deposit gradually on the solid BC core. The authors focus their discussion on the case of nucleation scavenging only. A more realistic scenario is that hydrophobic BC particles do not necessarily need to reach the fully developed hygroscopic state to be activated in clouds. It is enough for them to coagulate with hygroscopic particles or get effectively scavenged into existing cloud droplets (in-cloud scavenging) which then evaporate leaving a larger and more hygroscopic residue (in-cloud processing). It is a fundamental question how to include such complex processes into global models. On the other hand, it can be misleading to use an overly simplified approach to calculate BC distribution in the global troposphere.

The results of this study also confirm that there is no continuous transition in the hygroscopic behaviour of BC particles which could have been expected if there were only gas-phase coating processes. Instead, there are virtually two well-separated sub-modes: one with growth factor around 1.0, and another around 1.6. A BC particle can escape removal from the atmosphere only if it is sufficiently large to be effectively incorporated into cloud droplets (by in-cloud scavenging) (around 300 nm, see left curves on Fig. 3 f) and g)). Smaller particles (blue lines, same figure) which do get into cloud droplets acquire a soluble coating and in case they do not precipitate they become susceptible for subsequent wet scavenging processes.

Therefore the aging of BC particles is not a straightforward chemical process in which a soluble coating is getting gradually deposited on particles, but rather a probability-driven on-off process which determines which fraction of BC gets cloud-processed. In this respect, in-cloud scavenging efficiency of a given BC particle would probably be more relevant than its ability to form CCN at any specific supersaturation. The two processes have different requirements for BC properties.

I would have expected the authors to elaborate on these mechanisms and to define an explicit 'accumulation mode' hydrophobic BC distribution that would effectively escape

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both in-cloud and nucleation scavenging and get transported into the global troposphere. From Figure 9, it turns out that the mean diameter of this fraction is around 0.2 micron, but no size distribution is provided. From the modelling perspective, it is therefore misleading to assume that all BC particles that are not efficient CCNs would not be scavenged from the atmosphere. In fact they will be if they are sufficiently small enough to be scavenged by already existing cloud droplets.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 917, 2011.

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