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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Effect of photochemical aging on the ice nucleation properties of diesel and wood burning particles" *by* C. Chou et al.

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We thank Reviewer 2 for the comments on the manuscript and present below the responses

comment) 1. The literature on ice nucleation experiments cited on the introduction should include more recent work, such as (Koehler et al., 2009;DeMott et al., 2009), including papers reporting on the effect of photochemical aging on the ice formation ability of particles, e.g. (Moehler et al., 2008;Sullivan et al., 2010).

The missing literature has been included and discussed in the new version of the manuscript.

comment) 2. The description of the applied methodology is incomplete. The operation

conditions for the diesel engines are stated as idle. Does that mean that the engine was just sitting there and idling without any load on the engine? If this is the case then I am surprised by the extremely low ratio of organic to black carbon, because for an idle condition with it's extremely inefficient combustion the exhaust aerosol is usually dominated by organic matter. Please add OC-BC ratios for fresh exhaust to Tables 1 and 2. Furthermore, a short description is needed how OC and BC were measured. In the present version, the reader has to consult several publications from the same experiment in order to get a full overview over the applied methods.

Idle experiments can be separated into cold idle and warm idle (which will now be clarified in table 1 of the revised manuscript). Cold idle refers to the injection of the exhaust from engine cold start. Warm idle refers to the injection of the exhaust from warm engine at idle mode. No load was applied to the engine during injection. As discussed in Chirico et al., 2011, the organic to black carbon content is mainly depending on the specification of the vehicle (type of engine, temperature, oxidation catalyst, etc...). It also depends on the dilution of the exhaust as it was already shown in Lipsky and Robinson (2006). The OC/BC ratios for fresh exhaust as well as the instrumentation for OC and BC measurements have been added to the revised manuscript on page 5, line 145 to 153.

comment) 3. The authors state that the PINC instrument cannot separate drops from ice above the droplet survival line by size alone (P 14704, line4). Are there other instrumental options for separating ice particles from water drops? If not then this should be stated clearly that for all cases where PINC sees particles above the droplet survival line these data cannot be interpreted as ice nucleation and have to be excluded from the data interpretation. If I understand the results and discussion section correctly, then this was done in the presented data analysis. However, if there are other means available to separate drops from ice (e.g., depolarisation of scattered light), then this method should be mentioned and applied. In particular, a much clearer discussion of results presented in Figures 2-6 is needed. It should be made clear that all data above this survival line are excluded from interpretation. Such a statement is made for Figure 4 in the first paragraph of section 3.1, but not for the other Figures presented.

Instruments like the Ice Optical DEpolarization detector (IODE, Nicolet et al., 2010) would have been a possible way to differentiate ice from water droplets above the droplet survival line. Note that the instrument is not integrated in PINC, but can be attached to it. Two reasons for not using the detector are: 1) we wanted to focus on deposition nucleation, 2) previous laboratory work using IODE (Lüönd et al., 2010 and Ladino et al., 2011) showed that particle concentration above 1000 particles.cm-3 led to the inability to clearly distinguish ice from droplets. The captions of figures 2-6 have been changed in the revised manuscript to clarify that data above the droplet survival line are not interpretable.

comment) 4. The sequence of figures is not straightforward. Fig. 7 is referenced prior to Fig.5.

The sequence has been corrected.

comment) 5. Particle size appears to be a crucial parameter in the presented data interpretation. However, data shown for particle size are very sparse. Why not presenting a kind of mass closure using OC-BC data (Figures 8 and 9) and particles size distributions from SMPS measurements? Performing such an analysis would also indicate whether all bulk components were recovered by the applied analysis, and it would show the relevance of particle shape for this kind of mass closure. Fresh combustion particles are of highly irregular and fractal-like shape while particles showing a larger OC coating may exhibit a more spherical shape. Furthermore, I would like to see a Figure which investigates a potential link between particle size and ice fraction. Figure 7 is the only graph linking particle size to activated fraction, but this link is difficult to get from the figure. Hence, the main conclusion of the paper that size governs ice nucleation ability is not convincingly presented.

Figures 1 and 2 show the following ratio: aerosol mass concentration (V)/ initial aerosol

mass concentration before lights on (V0), as a function of the OC:BC evolution for diesel and wood burning experiments, respectively. In all the cases except for experiment 15w, there is a loss of aerosol mass to the condensation of OC. For experiment 15w, it is possible that the shape of the particles were affected which led to an increase in the mass, followed by a decrease whereas the OC:BC ratio evolution is increasing through time (Figure 9 of the manuscript).

Figure 3 show that diesel particles with an average diameter below 160 nm do not form ice at -30 or -35°C. Larger particles (around 180 nm) would trigger ice nucleation at -35°C when diesel particles are coated with alpha-pinene (much thicker coating). However it would be difficult to conclude as no other experiments have been performed on diesel particles of size equal or larger than 180 nm average mobility diameter. Wood particles do not show any difference in the RH value for a defined activated fraction. The size parameter does not seem to affect wood burning particles ice nucleation ability but we believe that this is due to the fact that ash from the combustion is nucleating ice. Size governing ice nucleation could be partly the conclusion for diesel particles but we believe that other parameters like the shape and chemical composition play an important role in ice nucleation.

Specific comment) P 14703, line 2: should read: “neither ozone nor propene”.

P 14704, line 24: should read, e.g., “photochemically aged particles due to : : :”.

Fig. 7: please add the unit to the RHwater – axis

Figures 8 and 9: please add the unit to the time - axis.

The corrections suggested by the reviewer have been applied.

References:

Chirico, R., DeCarlo, P. F., Heringa, M. F., Tritscher, T., Richter, R., Prevot, A. S. H., Dommen, J., Weingartner, E., Wehrle, G., Gysel, M., Laborde, M., and Baltensperger, U.: Impact of aftertreatment devices on primary emissions and secondary organic aerosol formation potential from in-use diesel vehicles: results from smog chamber experiments, Atmos. Chem. Phys., 10, 11545–11563, doi:10.5194/acp-10-11545-2010, 2010.

Ladino, L., O. Stetzer, F. Lüönd, A. Welti, and U. Lohmann, 2011: Contact freezing experiments of kaolinite particles with cloud droplets. J. Geophys. Res., D22202, doi:10.1029/2011JD015727.

Lipsky, E. M. and Robinson, A. L.: Effects of dilution on fine particle mass and partitioning of semivolatile organics in diesel exhaust and wood smoke, Environ. Sci. Technol., 40, 155–162, 2006.

Lüönd, F., O. Stetzer, A. Welti, and U. Lohmann (2010), Experimental study on the ice nucleation ability of size selected kaolinite particles in the immersion mode, J. Geophys. Res., 115, D14201, doi:10.1029/2009JD012959.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 14697, 2012.

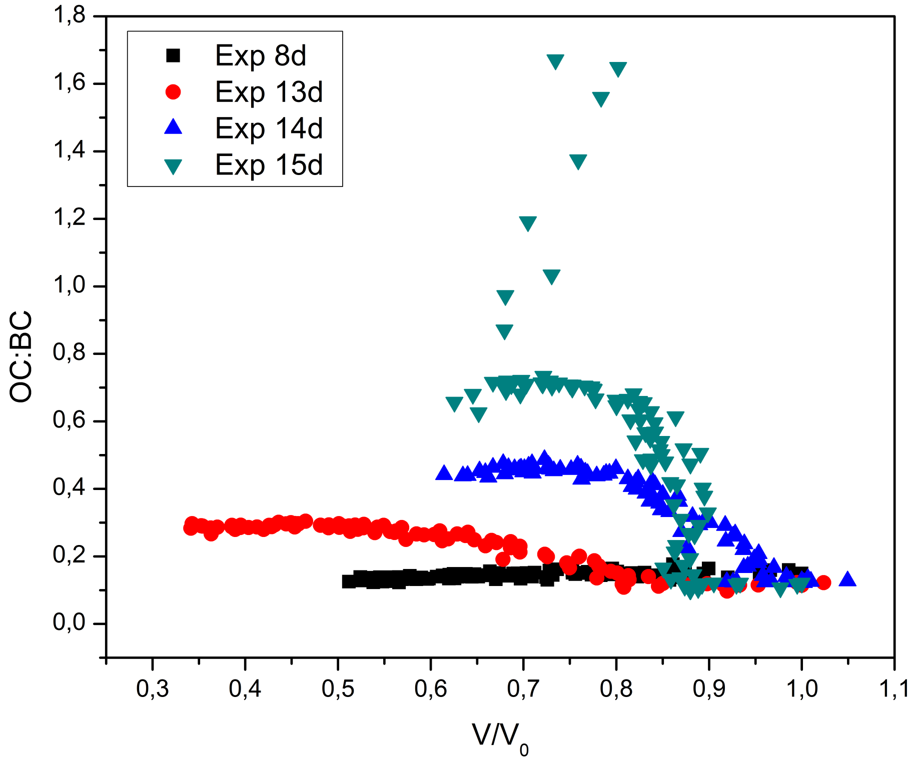

Fig. 1. Aerosol mass concentration evolution as a function of OC:BC ratio for diesel experiments.

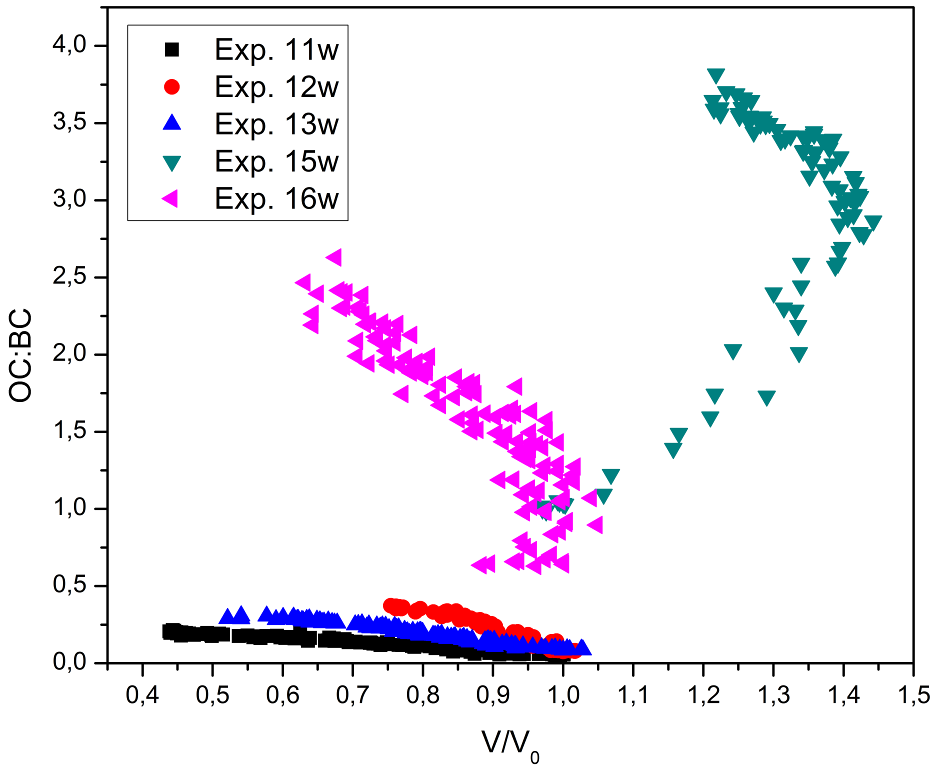

Fig. 2. Aerosol mass concentration evolution as a function of OC:BC ratio for wood burning experiment

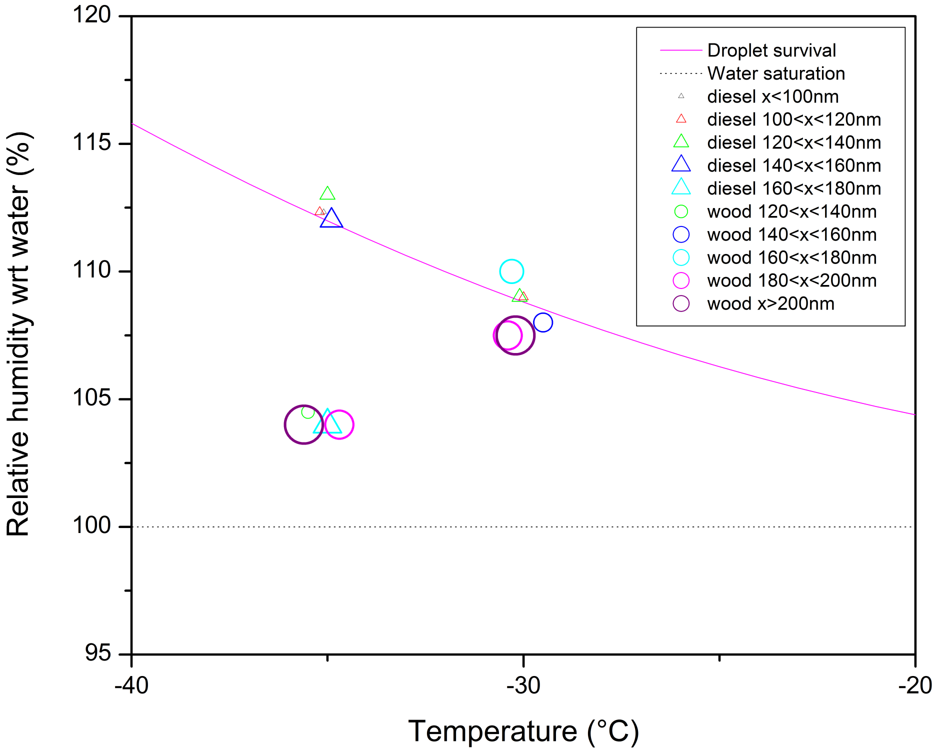

Fig. 3. Ice nucleation efficiency of different size classes of diesel and wood burning particles. The reported values are for a 0.1% activated fraction.