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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Chemical composition and mixing-state of ice residuals sampled within mixed phase clouds" *by* M. Ebert et al.

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23866/line 3: instead of "Penner et al., 2001" we now cite "Lohmann and Feichter, 2005

U. Lohmann and J. Feichter, Global indirect aerosol effects: a review, Atmos. Chem. Phys., 5, 715–737, 2005.

23867 line 11: "is dominating" changed to "dominates"

23868 line 6: "7 to 9" was replaced by "6"

23868 line 19: We also agree that there is no evidence that soot is a good IN and we have not made such a statement in this manuscript. To make this point more clear we will delete the misleading soot references from 23868 line 19.

sentence from 23868 line 17-19 is replaced by:

Silicates emitted by volcanic eruptions were also considered as a possible source for IN (Isono et al., 1959; Hobbs et al., 1971; Durant, 2008) and indeed enhanced IN concentrations during a volcanic ash event were measured by Bingemer et al (2011).

There are three main points of reviewer 1

1.) There seems to be a misunderstanding in the composition of the C,O,S group. We have not summarized different particles in the "C,O,S" group. These "C,O,S" particles have all pretty much the same composition. They are all complex internal mixtures. Each particle consists of parts of organics, sulphates and nitrates and sometimes these particles have also soot inclusions. Our description was obviously not clear enough. Therefore, we have changed the description in the manuscript (23874) in order to clearify that firstly we are talking about one type of particle and secondly we have identified soot only in some of these particles – not all the carbon in these particles is EC.

Replaced text (23874 starting line 13):

C-O-S group (complex mixtures of organics, sulfates, and nitrates) Particles in the C-O-S group are internal mixtures of the secondary components: organics, sulfates, and nitrates. In some of these particles we have also identified primary soot inclusions. These complex mixtures are a very common particle type and are often found in the fine-mode of urban or urban influenced aerosol samples. These complex secondary mixtures often incorporate soot inclusions (e.g., Murphy et al., 2006; Vester et al., 2007, Adachi et al., 2010). Freshly emitted unaltered soot particles can be clearly identified by SEM analysis on base of their highly characteristic morphology. Even small soot agglomerates (< 100 nm) on the surface of other particles can be identified unambiguously. The soot inclusions, which were detected in this study within the C,O,S particles exhibit a complex mixing state and are highly compacted (Figure 2) indicating advanced atmospheric aging (Weingartner et al., 1997). The soot identification by SEM was verified by high resolution transmission electron microscopy which showed nano-

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crystalline graphitic layers. This typical soot microstructure was observed in several previous papers (e.g. Posfai et al., 1999; Wentzel et al., 2003). The occurence of soot in the IR fraction is in agreement with the findings of Cozic et al. (2008),

Also changed: 23880 line 1 “soot” to “carbonaceous”

2.) One major criticism of both reviewers is the missing explanation of the ICE-CVI as sampling instrument for IR respectively a discussion of possible sampling artefacts. In the manuscript up to now only the reference Mertes et al. (2007) was given, where the instrument is explained in detail. I will add a detailed explanation of the ICE-CVI and possible sampling artefacts in chapter 2.1.

Added text (starting 23869 line 25):

The sampling of IN respectively IR within mixed-phase clouds remains a difficult task with a variety of possible sampling artefacts. ICE-CVI sampling artifacts could have different reasons. Possible causes could be in general a) unintentional transmittance of few interstitial particles that could still bias the sampling due to the small amount of ice particle residues, b) unintentional transmittance of supercooled drops, c) ice particle break-up inside the Ice-CVI and subsequent sampling of their fragments and d) particle abrasion from the inner walls of the Ice-CVI inlet due to ice particle impacts. The pre-segregation of interstitial particles is solely carried out by a conventional CVI (Mertes et al. 2005), which is one part of the Ice-CVI setup. In many airborne applications of this CVI design in pure ice clouds with small ice crystal concentrations it was shown that interstitial particles were successfully suppressed. Moreover, out-of-cloud test samplings were carried out with the Ice-CVI system during the CLACE campaigns, revealing a factor of 100 to 1000 less counts than during ice particle residual samplings in mixed-phase cloud events (Kamphus et al. 2010). Supercooled drops were pre-segregated in an impactor upstream the CVI using cold impaction plates where the drops freeze and are thus removed from the sample flow. Flow rates, orifice diameters and thus particle velocities are adjusted to achieve a non-critical Weber number (ratio

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of kinetic to surface energy), confirming that the drops up to a diameter of 20 μm did not splash during impaction. The same argument holds for avoiding ice particle shattering during impaction of ice particles up to the same size of 20 μm in the drop pre-impactor. Therefore, ice crystals larger than 20 μm are removed by a 90° sampling inlet and a virtual impactor upstream the drop pre-impactor (Mertes et al., 2007), even though this restricts the upper size range of sampled ice particles. Air velocities in front and inside the Ice-CVI inlet system are much smaller compared to airborne measurements, thus particle abrasion is unlikely. The only exception is the wind tunnel where the CVI is situated and the first part of the CVI inlet itself where particle velocities above 100 m sec-1 needs to be reached for a proper size separation by the CVI. But due to the vertical orientation no opposing surfaces exist in this part of the sampling system, where abrasion could occur. This was also checked by additional measurements that did not detect any substantial contribution of particles from the inner walls of the Ice-CVI.

3.) We have added a discussion of scavenging as a potential source of material in the IR fraction at two places in the manuscript.

New text –scavenging 1: (starting 23872 line 12)

Furtheron, ice crystals can scavenge other particles, which will be present in the IR fraction besides the original IN. To minimize this effect only small ice crystals are sampled. Nevertheless, because of all these reasons and the fact that within a mixed-phase cloud the IN fraction is very small (< 10-5 during CLACE5; Mertes et al., 2007) sampling artefacts cannot be excluded.

New text – scavenging 2: (starting 23876 line 7)

Another reason for the complex mixing state of the ice residuals could be scavenging of the small ice crystals, sampled by the ICE-CVI. In this case the complex mixing state of the IR would be an artefact and no characteristic of the original IN. Because of the small size of the sampled ice crystals we assume that scavenging plays only a minor

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role. Nevertheless, taking the results of this study and ignoring minor compounds of each IR as possible scavenged material (i.e. considering only the main components) the same particle groups (“C,O,S”/ “Pb-containing” and the main components of the group “complex mixtures”- silicates and metal oxides) are identified as original IN.

Erased references: Gorbunov et al., 2001; De Mott et al., 1990; Penner et al., 2001

Added references:

Adachi, K., Chung, S.H., and Buseck, P.R.: Shapes of soot aerosol particles and implications for their effects on climate, J. Geophys. Res., 115, D15206, doi: 10.1029/2009JD012868, 2010.

Heinz G. Bingemer, H.G., Klein, H., Ebert, M., Haunold, W., Bundke, U., Herrmann, T., Kandler, K., Müller-Ebert, D., Weinbruch, S. and Curtius, J.: Enhanced atmospheric ice nuclei in the Eyjafjallajökull volcanic ash plume over central Europe, submitted to ACP.

Durant, A. J., Shaw, R. A., Rose, W. I., Mi, Y. and Ernst, G. G. J.: Ice nucleation and overseeding of ice in volcanic clouds, J. Geophys. Res., 113, doi:10.1029/2007JD009064, 2008.

Lohmann, U. and Feichter, J.: Global indirect aerosol effects: a review, Atmos. Chem. Phys., 5, 715–737, 2005.

Posfai, M., Anderson, J.R., and Buseck, P.R.: Soot and sulphate aerosol particles in the remote marine atmosphere, J. Geophys. Res., 104, 21685-21693, 1999.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 23865, 2010.