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Atmospheric
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Interactive comment on "Pore structure 3-D imaging by synchrotron micro-tomography of graupel grains" *by* F. Enzmann et al.

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Reply to referee #2

We are grateful to this referee for his/her review and much appreciate for all comments elaborated. Our main replies and changes to be made to a revised manuscript are listed below.

Comment 1 – Quality of the writing

We agree that the difference between graupels and hailstones is ambiguous even in the literature. According to general agreement, ice particles larger than 5 mm in diameter are accounted as hailstones. In general it is also common to denote particles

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with higher density values near that of bulk ice as hailstones rather than as graupels. However, graupels and hailstones are not clearly divided on basis of densities only. The rather broad density range of 0.05 to 0.9 g cm-3 as given by Pruppacher and Klett (1997) for graupels was based on literature values, as for example, 0.13 g cm-3 (Magono, 1953), 0.05 to 0.45 g cm-3 (Locatelli and Hobbs, 1974), 0.25 to 0.45 g cm-3 for particle diameters from 1 to 2 mm, and 0.45 to 0.7 g cm-3 for particle diameters between 0.5 and 1 mm (Zikmunda and Vali, 1972), 0.45 g cm-3 for conical graupel (Heymsfield, 1978), and even as high as 0.5 to 0.7 g cm-3 (List, 1958) and 0.85 g cm-3 (Braham, 1963). Zikmunda and Vali (1972) claimed that their graupels had "intermediate" densities and that graupel densities vary widely in response to variations of cloud temperatures, cloud drop size distributions, and liquid water contents. On the other hand, low densities were also found for hailstones, e.g., 0.4 g cm-3 for hailstones with 1 mm diameter (Knight and Heymsfield, 1983), but more often lies around 0.8 g cm-3 (e.g., Prodi, 1970). An optical observation may give an additional clue as to whether an ice particle is a graupel or a hailstone. Furthermore, graupels look more opaque, hailstones more clear. Thin sections of hailstones show an onion-like structure and a germ in the centre.

Another objective is the growth regime which is affected by the temperature at the surface of the rimed ice particle. Generally the surface temperature is higher than the ambient temperature because latent heat is released when the riming droplets freeze. As long as this latent heat is efficiently transported to the environment the surface temperature remains clearly below 0 °C and the riming droplets freeze immediately when they collide with the ice surface (dry growth regime). On the other hand, when the amount of released latent heat increases (e.g., because of high mass accretion rates) so that it cannot be quickly dissipated into the environment the surface temperature increases towards 0°C. In this case riming droplets do not freeze right away when they are deposited on the ice surface (wet growth regime), but the accreted water changes phase depending on the heat transport from the graupel into the environment (Pruppacher and Klett, 1997). The density of graupel is dependent on the amount of air bubbles which are included and, therefore, on the regime which is dominant during its growth. Measurements of the number and size of air bubbles in graupel and hailstones by Carras and Macklin (1975) indicated that during the dry growth regime smaller but numerous air bubbles are included while during the wet growth regime fewer larger bubbles are found. However, estimating the total fraction of air bubbles per volume of ice indicates that generally during wet growth, lower net amounts of air are included than during dry growth. Thus, the density of graupel and hailstones would increase during wet growth.

We definitely consider the investigated ice particles from the wind tunnel as graupels. The samples from the Mainz vertical wind tunnel were grown while freely floating in the upstream of the wind tunnel in a cloud of supercooled droplets. The growth process began with a frozen drop of 350 μm radius. The subsequent riming process increased the radius by 200 μm. From measurements of the surface temperature of the rimed ice particles it was ascertained that for temperatures between -5 and -15 °C, and with rather low liquid water droplet contents (below 2 g m-3), that the graupels grow in the dry growth regime. This was the case for the rimed ice particles sampled for tomography. Growth in the wet growth regime can be expected only at temperatures above -4 °C and/or with extremely high liquid water contents (e.g. riming with large drops). This would be typical for hailstone formation (v. Blohn et al., 2009). Thus, the droplets colliding with the ice particle froze immediately forming a porous structure around the germ. However, the density of the entire graupel was rather high because of the target frozen drop of 700 μm diameter with a density near that of bulk ice. Probably the voids between the frozen droplets were too small (i.e., <1 um) to be seen by our tomography setup, a conclusion to be added in the revised manuscript.

The reviewer mentioned electron microscopic studies of graupel in the literature. The technique of cryo-SEM pioneered by the group of Wergin et al. is clearly superior to x-ray microtomography (XMT) when the focus is on the morphology of graupel. XMT, on the other hand, enables the internal structure of a graupel to be viewed nondestruc-

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tively in 3-D and thus enables its evolution to be traced both in space and time (4-D). Our focus was on quantification of the interior (closed) porosity for which XMT is the better choice. The pictures given in Rango et al. (2003) obviously show rimed ice crystals (as, e.g., needles, plates, columns) as the primordial structure still visible in most cases. According to another definition, graupel is called an ice particle whose original habit is no more visible (Pruppacher and Klett, 1997). The graupels from the wind tunnel have a frozen drop as germ and therefore they have a spherical shape. Even the graupels shown in Fig. 2 of Dominé at al. (2001) probably also originated from some sort of ice crystal and not from a frozen drop. In natural graupels and hailstones both ice crystals or frozen drops were found as germs but only the latter was of interest in this study.

The natural specimens sampled at Mainz University campus were small white ice particles showing spheroidal graupel habit. The temperature on that day was indeed as high as +9°C, indeed. When exposed to higher atmospheric temperatures during descend from the cloud to the ground some surface melting might have taken place. When such particles plunge into liquid nitrogen, melt water which crept into open voids on the ice surface refroze. This might have increased the density of the graupels artificially to some extent during the sampling procedure, and might have also obscured the outer morphology of the graupel particle. However, most of the air bubbles inside the ice were not really affected by this artifact. On the other hand, these samples might be more typical for lower tropospheric conditions than the spectacular lump graupel particles sampled at -12 °C in Colorado and depicted by Rango et al. (2003).

We agree that in first version of our manuscript it has not been clearly explained what we consider as graupel or hailstone and why. This will be added in the revised manuscript. The generation of the graupel in the wind tunnel will be explained in more detail and the reasons for the rather high densities of the graupels sampled from wind tunnel and from the Mainz campus will be discussed. Furthermore, the literature will be checked and the new 2010 references mentioned by the reviewers will be added. With regard to the terms metamorphosis and annealing, both terms have been replaced consistently by the term "metamorphism" throughout the text as used consistently in the more recent literature. Please note, however, that the term metamorphosis is still used if we speak of "metamorphosing graupel". The style and wording will be checked by a native English speaking colleague.

Comment 2 – Experimental protocol

We are aware of the paper by Erbe et al. (2003) and agree that the state-of-the-art approach is to keep samples in LN all the time which is simple in case the samples do not leave the laboratory. For high-resolution synchrotron XMT, however, the tiny specimen has to be fixed in the holder to protect it from any submicron movement on the rotation stage during transportation and measurement. For the latter constraint we have experimented with alternative sample fixation and related cooling approaches which turned out to work and hence were documented as well. The um spatial resolution was possible at the time the work only at synchrotron facilities, but is currently possible also with state-of-the-art XMT lab machines which merit a report of our initial non-LN sample fixation approaches. Clearly, for sub-micrometer spatial resolutions currently available on synchrotron facilities these fixation approaches will no longer work as has been described in detail in the paper.

Comment 3 – Results

Close-up images will be provided for all graupel samples up to a scale where the individual voxels can be recognized. Note, however, that the main problem is to reproduce the b/w ice-air contrast calculated by the software for a 3-D image on a printed paper which is not a trivial task and quite different from the gray-scale images produced by, e.g., SEM imaging. The exercise to determine the SSA by XMT has already been published by some of our co-authors. However, we agree that this is no longer possible if the porosity becomes closed, and we will add this conclusion to our related discussion.

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Modeling the optical properties of the ice particles from XMT data was in fact the original aim of the project when started, and hence is mentioned in the introduction. However, it became clear during the progress of the work as reported here, that this is not a trivial task. There is some more recent literature how to model multiple light backscattering on external and internal (porosity) surfaces, e.g. by Kaempfer et al., 2005; Randrianalisoa et al., 2006; Bänninger et al., 2008; and Lipinski et al., 2010. The approaches reported, however, rely on the domain of parameters where the scattering of radiation can be considered in the approximation of geometrical optics. Such a ray-tracing approach will work therefore only in the case where the size of the individual pores is at least two orders of magnitude larger than the wavelength of the scattered light, but not when the diameter of the dispersed pore medium scatterers becomes inferior to the wavelength. However, the mean sizes of the pores we found with our graupel samples were in the same order of magnitude as the wavelength (few micrometers) and hence beyond the geometrical optics limit. For the latter case, a reliable radiative transfer model has to be derived from the Maxwell equations and Lorenz-Mie theory directly, neglecting at least the effects of dependent scattering for a first approximation. We are not aware yet of any literature report on such an approach to model light backscattering by multiple pores, and couldn't do it either. We will consider using an iterative solver based on the discrete dipole approximation once our new 600 node HPC will be available next year, with a dipole grid identical to the XMT voxel grid.

Comment 5 – Recommendation

Clarification on the sample characteristics (graupel or hail?) will be done along the arguments detailed above. Additional experiments will not be carried out until we find somebody who can provide us with a reliable model to calculate backscattering from the 3-D porosity microstructure at the micrometer scale which may help for a resumption of the project. However, this is not an argument for us to refrain publishing our success with synchrotron tomography, in particular as we hope by this way to find somebody who could help us with the appropriate albedo model development and with whom we may share our old (and new if necessary) tomography data for a model verification run.

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