

Review of egosphere-2024-1259 “Dominant role of charged meteoric smoke particles in the polar mesospheric clouds” by Liang Zhang, Zhongfang Liu, and Brian Tinsley

This manuscript is about the potential role of charged meteoric smoke particles (MSP) as condensation nuclei for polar mesospheric clouds (PMC). This is an important issue. Current models of PMC mostly consider nucleation on neutral MSP as nucleation process. As compared to nucleation on neutral MSP, nucleation on charged MSP can indeed be expected to lead to rather different nucleation rates and to a rather different altitude distribution of nucleation events. And indeed, there is today substantial observational and theoretical evidence that a substantial fraction of MSP in the mesosphere is charged. I consider the relative important of nucleation on neutral MSP and on charged MSP as an important open question for understanding PMC. So I fully agree with the authors that the importance and the consequences of “charged MSP nucleation (CMN)” is an important topic to investigate.

Then, unfortunately, I find that the authors put this topic in the wrong perspective. Throughout the manuscript, they contrast PMC nucleation on charged smoke to the “growth and sedimentation” scheme that is commonly used when describing the growth of PMC ice particles. This entire starting point for the manuscript is not meaningful.

We cannot really doubt that there are basically three distinct stages in the evolution of a PMC ice particle: (1) an initial ice particle nucleation, (2) ice particle growth in a supersaturated environment, and (3) sublimation of the ice particle when it encounters an unsaturated environment. The question of the nucleation mechanism e.g. by charged MSPs belongs to point (1). Once nucleation has occurred, stage (2) takes over and PMC ice particles will grow through deposition of water vapour as long they are in a supersaturated environment. The growth rate is essentially determined by the surrounding number density of water vapour (e.g., Hesstvedt, J. Geophys. Res., 1961). While they exist, the ice particles will of course be subject to gravity, and hence they will sediment relative to the surrounding air. Both the growth and sedimentation are basic physics, not mere assumptions introduced by a “growth and sedimentation model”. They do take place in stage (2), independent of the question whether the nucleation in stage (1) occurred because of charged MSP, neutral MSP or any other nucleation process.

So contrasting “charged MSP nucleation (CMN)” and “growth-sedimentation (GS)” does not make sense. The manuscript would have made perfect sense if it instead contrasted “charged MSP nucleation (CMN)” and “neutral MSP nucleation (NMN)” within stage (1). As opposed to growth and sedimentation, the nature of the nucleation process can indeed be regarded as an “assumption” in current PMC models, as long as these models do not explicitly simulate what fraction of the MSPs is charged and what fraction of the nucleation events takes place on charged MSPs. So, I would consider this a very interesting manuscript if it contrasted CMN and NMN, and if it investigated the consequences that either scenario has on the resulting PMC properties and PMC lifecycle.

With its current focus on “CMN vs. GS”, I do not consider this manuscript to be publishable. Now a question to me as reviewer is: Do I think that the current manuscript can be revised to a form that is acceptable for publication. Unfortunately, my answer is no. In order to make this manuscript publishable, it would not only be necessary to give it a new focus on “CMN vs. NMN”, as outlined above. It would also be necessary to handle things much more rigorously. The manuscript contains good ideas concerning the properties of PMC resulting from nucleation on charged MSPs. And the manuscript also contains a valuable collection of statistical studies of AIM/SOFIE and AIM/CIPS satellite data. However, neither the connections made between charged MSP nucleation and resulting PMC properties, nor the subsequent connections to the AIM data are rigorous enough. There is too much hand-waving. I think that in order to make this work rigorous, one really needs to

involve a microphysical PMC model of suitable complexity. Model simulations are needed to show how charged MSP nucleation really would lead to modified PMC properties as suggested by the authors.

However, this is much work and beyond the scope of a revised manuscript. The ideas I have outlined above essentially refer to a different paper. Hence, from an ACP perspective, I suggest to reject the current manuscript, and then possibly to hope for a new one.

Below, I add some more comments that may be useful for the continued work.

When referring to PMC trends in section 1.1, it would be good to refer to the latest publications (DeLand and Thomas, *Atmos. Chem. Phys.*, 19, 7913–7925, doi: 10.5194/acp-19-7913-2019, 2019). Also, “global warming” (or global cooling) is the wrong term when referring to the effect of a methane trend on PMC. Methane primarily affects PMCs in terms of water vapour.

The observed connection between IMF and PMC is intriguing. However, people have looked for a connection between PMC and mesospheric charging conditions in many different ways. Most importantly, there is no real evidence that PMC occurrence would be affected by geomagnetic activity. This argues against a very direct connection between charging and nucleation. For the manuscript, it would be good to look deeper into different aspects of this.

In line 76, the authors state “it seems unreasonable that ice nucleation takes place primarily at the top of the PMC, since the saturation levels throughout the PMC altitude range all favour PMC formation”. This statement ignores the fact that relative humidities much larger than 100% are needed for ice nucleation on tiny MSPs (curvature effect, Kelvin equation).

The negative dependence of particle radius on PMC height in figures 2+3 is not at all surprising. It is consistent with the growth/sedimentation scheme. What is missing in the manuscript is a clear statement that the particle growth rate essentially is controlled by the absolute number density of water vapour (not the water vapour mixing ratio). The absolute water vapour number density decreases quickly with altitude, both because of the total atmospheric density decrease and because of the efficient photolysis of water vapour in the upper summer mesosphere. Hence, PMC particle growth rates decrease quickly with altitude. This fact removes many of the “growth/sedimentation inconsistencies” claimed by the authors, e.g. figures 2+3, 4+5 etc.

When discussing the effect of (wave) dynamics on PMCs, one really needs to consider the time scales. As the authors state, most current models suggest that it takes an ice particle many hours to grow to visible size. However, once a particle enters an unsaturated region (e.g. because of wave activity) sublimation to sub-visible sizes can happen on much shorter time scales than an hour. So seeing wave structures in PMCs does not contradict current descriptions of the PMC physics. Again, the growth process and the time scales involved are all basic physics (e.g. Hesstvedt, 1961). As pointed out above, Invoking an appropriate growth description (in a microphysical model) will be needed as an important step towards making the authors ideas more quantitative.

Indeed, there are many observations of dual layer or multi-layer PMCs. These do not contradict the basic idea of the growth/sedimentation process. The PMC lifecycle is not a one-dimensional process. There are strong horizontal winds near the mesopause and, more importantly, there are substantial wind shears. Hence, structures in PMC can never be understood based on microphysics alone. (Still, a one-dimensional microphysical model may be sufficient to better quantify many of the ideas brought forward in the manuscript.)

The manuscript refers much to correlations and anti-correlations of various PMC properties with water vapour. The manuscript also correctly refers to the importance of “freeze-drying”. The effect of freeze-drying really makes any correlation discussions tricky. You very often run into a chicken-and-egg problem, depending on the concrete atmospheric situation and history.

Maybe I misunderstand the time-lag analysis in figure 7. For me, the method does not make sense. The authors correlate particle radius and particle concentration inferred from AIM/CIPS with the cloud height inferred from AIM/SOFIE. As expected, there is correlation/anticorrelation. But of course, these correlations completely disappear when correlating the AIM data with SOFIE data from a different day. On different days, the instruments observe different clouds, so of course there will not be any correlation.

An anti-correlation of PMC brightness and cloud radius as suggested by Rusch et al. (2017) (line281) is problematic. CIPS infers a kind of “column-averaged” mean particle radius. This quantity is usually useful, but becomes ill-defined in cases you have an aged PMC at the end of its lifecycle with large particles near the cloud bottom and nothing left above. The conclusions of Rusch et al. (2017) are likely affected by this, as discussed e.g. by Hultgren and Gumbel (2014).

The idea that nucleation not necessarily happens at the mesopause temperature minimum is not new. A nice reference is “Berger, U., and U. von Zahn (2007), Three-dimensional modeling of the trajectories of visible noctilucent cloud particles: An indication of particle nucleation well below the mesopause, *J. Geophys. Res.*, 112, D16204, doi:10.1029/2006JD008106.” These authors do not start out from a nucleation process. Vice versa, they trace the particles in the visible part of the cloud back to where the nucleation must have taken place. They suggest that nucleation in average takes place 3 km above the visible cloud.