Response to Referee #2 Comments on egusphere-2024-1259, "Dominant role charged meteoric smoke particles in the polar mesospheric clouds"

We would like to thank the Referee #2 for the valuable comments, which are very helpful for improving this article. In the following the remarks are responded point by point.

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.

We are very grateful to the referee for supporting the importance of the CMN scheme in PMCs.

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.

Although the GS scheme is widely used and almost the only theory for the PMC formation, but we find that the "sedimentation" in GS scheme is unimportant and unnecessary. The logic of this manuscript is simple: if the GS scheme is proved to be (partly) incorrect, then the only alternative theory for PMC formation will be the CMN scheme. That's why we try to prove that CMN is correct but GS is incorrect throughout the manuscript.

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 lifecyle.

> We are in favor of the "nucleation" and "growth" processes, but we strongly doubt the importance of the "sedimentation" processes. Sedimentation of ice particles affected by gravity and upwelling is too slow to explain the rapid variability of PMCs.
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> The major difference between the CMN and the GS process is that the CMN can explain the observed PMC phenomenon in the absence of "sedimentation". In other words, there is no need to simulate particle trajectories in the CMN model, which significantly reduces the computational time.

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. NM", 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.

> We respect the referee's decision to reject this manuscript, and we are very grateful for the valuable advice. We strongly agree that a new PMC model should be developed to support our hypothesis. We will prepare and submit a better manuscript in the future.

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.

> Agreed. Thanks.

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.

By analyzing the SOFIE PMC data, we also found no signals of geomagnetic or lightning activity. However, we did find a weak link between IMF B_y and PMCs, and we are verifying the significance of this link by analyzing data from other satellites.

It should be noted that, according to the CMN scheme, the IWC of PMC is dominated by the temperature rather than the concentration of ice nuclei, as shown in Fig. 11. From the perspective of the CMN scenario, the occurrence of PMC is not expected to be influenced by the geomagnetic activity via the concentration of charged-MSPs.

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).

We are grateful for this comment. In the CMN scheme assumes that the ice nucleation can occur at any altitude. It is therefore very important to check whether ice nucleation can occur at the bottom of PMCs where the humidity is not large enough.

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. figues 2+3, 4+5 etc.

On the one hand, the variations in PMC height (δh) are small, as shown in Table 1, thus the variation in water vapor is also small and may not account for the large variations in the column mean of r and N.

On the other hand, Fig.9 has shown that the r and N are not correlated with the water vapor mixing ratio (column average between Z_{bot} and Z_{top}), similarly, it can be easily calculated that the r and N are also not determined by the absolute amount of water vapor.

When discussion 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 microphysics alone. (Still, a one-dimensional microphysical model may be sufficient to better quantify many of the ideas brought forward in the manuscript.)

Although the observed wave structure or multi-layer structure in PMCs has been explained by the GS scheme, however, they can also be well explained by the CMN scheme. Of course, we fully agree that a new PMC model is needed to explicitly demonstrate our claims.

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.

The "freeze-drying effect" can be derived directly from the GS scheme, and the observed dehydration/hydration of water vapor above/below PMCs is usually attributed to the growth, sedimentation, and sublimation of ice particles. However, it should be noted that almost all simulations based on the GS theory overestimate the freeze-drying effect.

According to the CMN scheme, the "sedimentation" is unimportant, so the redistribution of water vapor with altitudes should not result from the growth, sedimentation, and sublimation of ice particles.

We are preparing another manuscript to show that the traditional concept of "freeze-drying" based on "sedimentation" is (partly) incorrect, in which we will also argue again that the GS scheme is wrong.

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.

Assuming that the CMN scheme is correct, namely, the column mean r and N are determined by the mean PMC height h (the charged-MSPs increase rapidly with h), then the zero-day lag shown in Fig. 7 indicates that the ice particle growth is rapid, within one day. The r, N, and h in Fig. 7 are all observed from SOFIE. Of course, from the perspective of the GS scheme, the method in Fig. 7 is nonsense.

In Fig. 8, we compared the *r* from CIPS with the *h* from SOFIE, just to check that it is consistent with the result in Fig. 6. More interestingly, the IWC does not depend on *h* (shown in Fig. 11), but the *albedo* from CIPS is negatively correlated with *h*, which is possibly because the *albedo* is proportional to r^6. In short, the results in Fig. 8 support the results in Fig. 6 (i.e., the mean *r* depends on the mean *h*).

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 anti-correlation between *r* and IWC seems to be a common phenomenon in gravity wave region, as shown by Gao et al. (2018). Further work should be done to justify whether it is ill-defined. Both Rusch et al. (2017) and Gao et al. (2018) explained this phenomenon by the GS scheme, however, the CMN scheme may also provide an alternative explanation through the variation of the PMC height.

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.

Thanks a lot for this reference. The simulation by Berger and von Zahn (2007) is very interesting, clearly showing the latitudinal and vertical transport of ice particles, with a transport time of 36 hours. In the CMN scheme, the nucleation is assumed to occur in situ throughout the PMC altitude range in a much shorter time, and it is the electrons rather than the neutral MSPs that determine the distribution of ice nuclei (charged-MSPs).