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Comment on acp-2021-728

Anonymous Referee #4

Referee comment on "Formation of ice particles through nucleation in the mesosphere" by Kyoko K. Tanaka et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-728-RC2, 2022

This manuscript addresses the microphysics of noctilucent cloud formation in the mesosphere. The authors make an important case that a semi-phenomenological model is better suited for describing ice particle nucleation under mesospheric conditions than classical (or modified classical) nucleation theory. They apply the model to investigate whether homogeneous or heterogeneous nucleation dominates the formation of noctilucent clouds. The idea is good of directly comparing the nucleation rates resulting from homogeneous and heterogeneous nucleation under given mesospheric conditions. The conclusion that heterogeneous nucleation is expected to dominate under most conditions is reasonable and in line with earlier studies.

A major problem of the approach is the nucleation scenario that the authors adopt. They assume a process of continuous temperature decrease down to a very low temperature at that the nucleation rate reaches a maximum. They call this temperature "nucleation temperature" and use it as a characteristic parameter describing the cloud nucleation process. I argue that this scenario is not relevant for mesospheric conditions. Sufficient homogeneous nucleation can occur at temperatures substantially above the authors' "nucleation temperature". In fact, it would be very much counter-productive for the formation of noctilucent clouds if these (unrealistically) low "nucleation temperatures" were reached in the mesosphere.

This can be illustrated using Figure 2. Here, the semi-phenomenological model has been used to describe a very slow cooling process, starting out from a typical polar summer mesopause temperature (135 K) and then extending over more than 30 hours. After this time, a homogeneous nucleation rate of about 1 cm-3 s-1 is reached at a "nucleation temperature" Tp of about 65 K. However, it is not necessary to reach this maximum nucleation rate in order to form noctilucent clouds. Already a nucleation rate of e.g. 0.01 cm-3 s-1 leads to an ice particle concentration of about 100 cm-3 after few hours. This is sufficient for noctilucent clouds, and it is achieved at significantly higher temperature. Moreover, at 65 K the nucleation of rate 1 cm-3 s-1 will lead to so many nucleation events that competition for the available water vapour will prevent the individual particles from growing large. This will make it impossible to form visible noctilucent clouds (that typically

require particle radii exceeding 20 nm). I thus argue that Tp is not a meaningful parameter to describe homogeneous nucleation of noctilucent clouds in the mesosphere. It follows that cooling rates slower than 1e-5 K s-1 are not a requirement for the occurrence of homogeneous nucleation in the mesosphere (lines 238-239). Also, the very strong statement "there is no particle formation via homogeneous nucleation on Earth" in the Conclusions (lines 295-296) does not hold based on the Tp analysis.

There are more inconsistent statements about the cooling rates. In section 3.1 it is argued that very slow cooling rates (< 1e-5 K s-1) are necessary for homogeneous nucleation (lines 238-239). In section 3.2, on the other hand, it is concluded that high cooling rates (> 1e-2 K s-1) are needed for homogeneous nucleation to be important (figures 6 and 7). This contradiction needs to be discussed.

Cooling rates in the manuscript are expressed in units K s-1 (e.g. figures 4 and 5). Using the unit K h-1 would be much more instructive and would provide the reader with a better feeling for the mesospheric processes. The authors should discuss what cooling rates can typically be expected in the mesosphere. One could e.g. consider the cooling rate connected to a typical gravity wave of amplitude 10 K and period of a few hours.

Some comments concerning the heterogeneous nucleations:

- Line 157-158: It is stated "As indicated above, the radius of the critical cluster is very small

(i = 2−10), making this assumption reasonable." I do not find where "above" this is indicated. Please provide a justification why the critical radius is so small (i = 2-10).

- Line 180-182: The condition given by equation 18 states that at least 50% of the initial molecules n1(0) in the water vapour are consumed by heterogeneous ice particle growth. In contrast to this, lines 181-182 state that the number density of water vapour is largely unchanged during the nucleation process, i.e. water vapour is largely not consumed. The latter statement is the basis for the linear growth of the ice partile radus with time described by equation 19. This seems to be a contradiction that would make equations 20-24 invalid.

Some comments concerning the equations:

- equations 12 and 13: r1 in these equations should be r0 in order to be consistent with the radius of the monomer defined in line 103.

- equation 18: "r" should be replaced by "a", the radius of the dust grain.

Some comments concerning the Introduction:

- Line 15: "mesosphere" should be replaced by "mesopause region".

- Line 19: The sentence "Ice particles, also known as polar mesospheric clouds, have recently been observed by satellites (Hervig et al., 2012)" should be rephrased. "Noctilucent clouds" are also known as "polar mesospheric clouds". Polar mesospheric clouds have been observed by satellites not only "recently" but as early as in the 1970s.

- Line 23: The authors seem to imply that noctilucent clouds "were considered to exist" before their discovery by observations. This is not the case.

- Line 23: What is meant by "[noctilucent clouds] were difficult to observe visually before the twentieth century"?

- Line 39: It is stated "Meteoric smoke particles consist of sodium bicarbonate, sodium hydroxide, soot, sulfuric acid, and proton hydrates". Soot, sulfuric acid and proton hydrates are indeed considered to be part of the middle atmospheric aerosol. However, they are not expected to be ingredients of meteoric smoke particles in the mesosphere.

- Line 49: Please make clear that by "solid particles" in this sentence you mean meteoric smoke particles, not ice particles.

- Line 73: Avoid the term "measured" when referring to molecular dynamics simulations. More suitable terms may be "studied" or "derived".

Some editorial comments:

- Line 287: “Section 4” should be “Section 3.2”.

- Line 331: Please provide a complete reference.

- Line 332: “Merner” should be “Megner”.

- Line 350: “Hoffner” should be “Höffner”. “Rottger” should be “Röttger”.