Reply to Reviewer II

1. *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.

We thank for the reviewer's valuable comments. As the reviewer pointed out, we defined the nucleation temperature in our paper. Although our method is a simplification, we believe that it is applicable to mesospheric conditions. The reviewer pointed out that temperatures higher than the nucleation temperature would be sufficient for homogeneous nucleation to occur, but we think this is difficult, because the nucleation rate is very low in the temperature range from the nucleation temperature to around 100K. In the mesosphere, the minimum temperature is never lower than 100K, so we concluded that the homogeneous nucleation is very unlikely to occur.
The explanation was inadequate, so we added the description in the revised paper, as stated in the reply to the comment 2.

2. *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.*

Figure 2 shows the nucleation rate in the cooling process. The reviewer pointed out that the nucleation temperature does not have to be 65 K, and that a larger nucleation rate of 0.01 cm^{-3} s^{-1} **is sufficient. However, the temperature, at which such a large value as 0.01**cm^{-3} s^{-1} **is realized, is very low less than 100K. As shown in Figure 2, for example, the nucleation rate is only** 10^{-5} cm^{-3} s^{-1} **at 85K. This indicates that only about 1** cm^{-3} **of nuclei is formed in one day. Considering that the temperature in the mesosphere is larger than 100 K, we concluded that the homogeneous nucleation is difficult.**

We showed the nucleation temperature at which the nucleation rate peaks in the cooling process, but did not the monomer consumption. The temperature at which the nucleation rate peaks and the temperature at which the number density decreases steeply are very near, so we consider Tp is meaningful. Since Figure 2 did not show the variation of monomer consumption due to condensation, we added the number density change of vapor in Figure 2. The wording has been slightly changed for the strong statement in the conclusions pointed out by the reviewers.

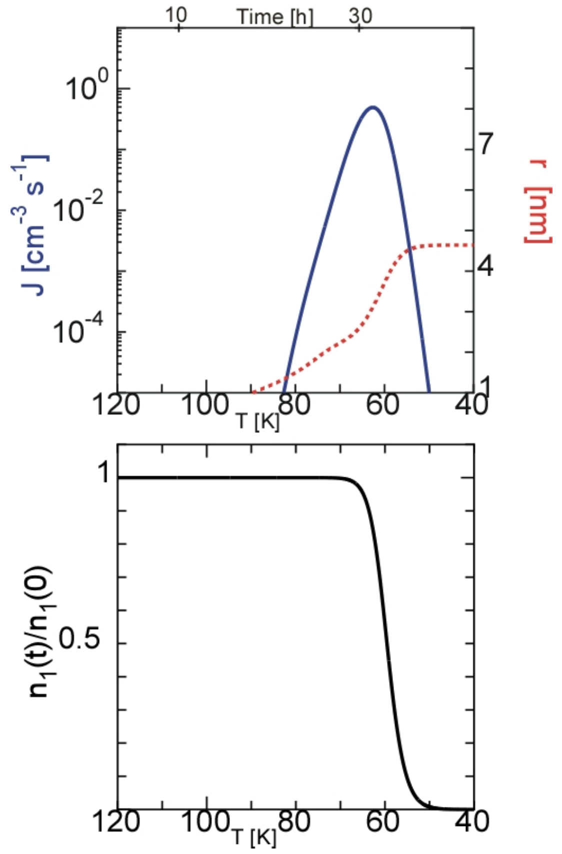

Fig.2 Time evolution of the nucleation rate and mean particle radius during homogeneous nucleation (upper panel), and the ratio of the number density of water molecule to the initial value (bottom panel) calculated using the SP model. The initial temperature was 135 K and the cooling rate is 3.6 K h^{-1}.

3. *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.*

We are investigating two different conditions for homogeneous nucleation. The first condition is not a requirement for homogeneous nucleation to occur, but a limitation on the temperature, that is, the condition required for condensation temperature realized in mesosphere when homogeneous nucleation occurs. From this condition, we found that a slow cooling rate (< 10^{-5} K s^{-1}) is required. The second condition is that the

homogeneous nucleation is dominant in the presence of dusts. For this condition, a high cooling rate (> 10^{-2} K s^{-1}) is required. The fact that the two conditions do not have an overlapped region of the cooling rate indicates that the homogeneous nucleation is difficult to achieve. We should include this description, so we included it in the revised paper.

4. *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.*

According to the reviewer's comment, we changed the unit (K h^{-1}) for all descriptions and graphs in the revised paper. Typical values of the cooling rate obtained from previous studies have been added in the new section 3.1.

5. 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).

The calculation of homogeneous nucleation gives information about the size of critical nuclei. As mentioned in the paper, the sizes of the critical clusters are very small in both models, e.g., two and four molecules for the SP and MCNT models for the case of Fig.3. The cause of small critical cluster is due to extremely high supersaturation ratios:

From the nucleation theory, the size of critical cluster, i_*, is given by $\left(\frac{2\eta}{3lnS}\right)^3$ for the CNT and MCNT, and $i_* = \left(\frac{\eta+\sqrt{2\eta+3\xi lnS}}{3lnS}\right)^3$ for the SP model. In our calculation ranges, the size of critical nuclei ranged from 2 to 10. As the reviewer pointed out, the description was insufficient. We have therefore added the description of how the critical nucleus is determined in Section 2 and explained the size of critical cluster in more detail.

- Line 180-182: The condition given by equation 18 states that at least 50% of the initial molecules $n_1(0)$ 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.

As the reviewers pointed out, the estimates were rough. So we corrected the condition within a reasonable range and changed the threshold from 50% to 10%. The figure and description were revised to reflect this change. With this condition, we obtain the number density of dust particles for the significant start of the heterogeneous nucleation, rather than the predominance. We have rewritten the expression in the paper. Although our estimates are rough, we consider them useful because it is a straightforward formulation of how the number density of smoke dusts necessary for the heterogeneous nucleation depends on the dust size and water vapor content.

6. 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.

We corrected it. The monomer radius was unified to be r_1 in the revised paper.

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

We corrected it.

Some comments concerning the Introduction:

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

We corrected it.

- 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.

We corrected it.

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

As the reviewer said, this sentence was not explanatory enough and did not need to be between the preceding and following sentences, so we removed it.

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

As mentioned in the above comment, we corrected it.

- 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.

According to the comment, the description of meteor smoke was not accurate, so we revised the description.

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

We corrected it.

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

We changed the word.

7. Some editorial comments:

- Line 287: "Section 4" should be "Section 3.2".

We corrected it. We revised to Section3.3, since Section 3.1 has been added in the revised paper.

- Line 331: Please provide a complete reference.

We corrected it.

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

We corrected it.

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

We corrected it.