I. Response to Referee #1 comments

We thank anonymous Referee #1 for his careful and explicit review of our paper and for his valuable comments and for endorsing our scientific methodology and results.

The suggested figures have been included in the updated supplementary materials (figures S2 to S7) and are suitably mentioned in the body of the text (sec. 4.1, lines 265 to 268).

II. Response to Referee #2 comments

We thank anonymous Referee #2 for his careful and explicit review of our paper and for his valuable comments and for endorsing our scientific methodology and results.

Main comments

1. A brief description of the parameterization is now included in the Abstract. A detailed description is also now included, in a separate paragraph, under the Model description section.

2. A list describe the simulation setup is now shown in Table 1 in the revised manuscript and is suitably mentioned in the model description section.

3. **a)** We respectfully do not agree with Referee #2 that "the hope for the parameterization is that it would improve wintertime sulphate concentrations".

   The main objective of this study is to investigate the impact of realistically simulating HONO levels (which was, by default, about one order of magnitude underestimated) on the aerosol composition, that is, not to improve the model-measurement of aerosol, though it happened in some regions.

   We showed clearly that realistically simulating HONO levels does significantly enhance sulphates, nitrates and ammonium as well as aerosol number concentration in highly polluted continental regions, especially during the winter season (see secs. 4.1), thus generally increasing their model/measured ratio, most apparently in North America (see Table 5 in the revised manuscript), improving model/measured sulphates in this region.

   However, whether the model predictions (compared to measurements) would be improved or not is a different question and depends on several other parameters, among which also the measurement uncertainties (e.g., as the case with nitrate measurements, see text).

   What we achieved in this study is to remedy one important parameter (i.e., HONO realistic simulation), which was found to improve the model/measurement agreement in some regions (e.g., sulphate in north America and nitrates in easten Asia) but not in other regions such as Europe.

   Sulphates, nitrates and ammonium are generally significantly enhanced and improved compared to measurements in all regions, hence HONO photolysis is certainly an important process in all these geographic locations. That model aerosol predictions are not improved (compared to measurements) in other regions does not mean that this process (HONO photolysis) is not important. It only means that other factors may cause this bias and have to be further investigated, which is what aerosol modeling community are currently working on towards better representation of aerosol chemistry and physics.

b) Though the model comparison to aerosol mass spectrometers (AMS) observations in Pringle et al. (2010) revealed a general trend of underestimation of all species, it worth also mentioning that this comparison may also be biased since the simulated period (2000-2001) is different than the measurement short periods over much larger time scale (between 2000 and 2006) which is also mentioned in Pringle et al. (2010). Therefore, we do not think that it worth repeating the comparison given that:

1) Our simulation results are valid for only one year, 2001, thus comparison with simulation results of Pringle et al. are not straightforward.

2) Pringle et al. (2010) calculated for only aerosol data <1μm.

3) Owing to the different measurement-simulation time periods and the different simulation periods between our study and that of Pringle et al. we do not think the comparison will give a cutting-edge answer of the model-measurement comparison and therefore does not justify to repeat the same comparison, which was already mentioned to be possibly biased due to the different simulation-measurements periods (Pringle et al., 2010). We therefore prefer to keep the current text, which is included in the "comparison with other large scale measurements" section and thus is also not confusing for the reader with our direct results.

c) The simulated seasonal cycle over eastern US and China is shown in Fig. 10. As shown in this figure, simulated aerosol number concentrations are significantly enhanced during winter months, especially over eastern China. These results show also that the new parameterization does also affect the aerosol seasonal cycle. Comparison with measurement requires simulating the same measurement period or long simulation (for several years) to minimize the meteorological effects on the results, which is out of the scope of this study.

4. The mentioned Paragraph about particle growth rates is now deleted as Referee #2 suggested.

Minor comments

1. Figures 1 and 2 are now improved in the revised manuscript.

2. For this figure, reducing the color gradient lead to almost one-color fields (i.e., changes in red will not be differentiable). It also gets clearer if slightly zoomed in (it is clearly seen for us by 125% instead of 100%). Therefore, We prefer to keep the figure as it is.

   Caption is now updated.

3. Now absolute enhancements are shown instead of the relative enhancement in Figure 9. Furthermore, Because we also realized that the relative enhancements in figure 9 are not very clear, we already included detailed figures for the hot spots in eastern US and Asia (Figure 11). As shown in Figure 11 and also mentioned in the text, the absolute enhancement in eastern Asia reaches about one order of magnitude higher that in eastern US (see se c, 4.1.4). It is also mentioned in sec. 4.1.4, that "The total enhancement in the aerosol number concentrations can thus be attributed to the strong enhancement in the nucleation mode, especially over eastern China, related mainly to the enhancement in the gas phase sulphuric acid concentration (i.e., nucleation of new particles is calculated in the model as a function of the temperature, relative humidity and the concentration of gas phase H_2SO_4, see Pringle et al., 2010)".