

Interactive comment on “On the formation of radiation fogs under heavily polluted conditions” by H. Kokkola et al.

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Thank you for your thoughtful comments. The points you have addressed in your comments will be reflected in the revised version of the article.

We would first like to address the general nature of the paper. The formation of unactivated fogs has been speculated to result from the depression of the Köhler curve caused by hygroscopic gases. Also, measurements in Po Valley have indicated that many of the fogs observed there were in fact are unactivated (Frank et al., Contr. Atmos. Phys., 71, 65–85. 1998.). In this article we examine the effects of different gaseous pollutants, especially sulfate forming species, on the sizes of fog droplets. In a way, we have extended the monodisperse approach of the modified Köhler theory to describe a polydisperse droplet population. However, sulfate formation has to be dealt with in a time-dependent manner, and therefore we are combining equilibrium (gas absorption) and non-equilibrium (liquid phase chemistry) models. The approach

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is a very simple one, but we believe that it should indicate qualitatively what effects may and what effects may not take place under heavily polluted conditions. On the other hand, because of the simplicity of the model, we are not trying to specifically model any observed pollution fogs. We have basically just chosen gas concentrations which may exist (or may have existed) in polluted locations, although the specific combination chosen might not reflect the actual circumstances at any specific location. We will clarify these matters in the revised version.

(1) Also other fog model studies have used fixed temperature profiles as model input [Pandis and Seinfeld (1990), Frank et al. (1998)]. We agree that it would be useful to check what happens when temperature is decreased further. However, the further decrease of the minimum temperature would cause the air to reach supersaturation after which our equilibrium approach (with respect to gas absorption) can not be used since the largest droplets reach the maxima of their Köhler curves, and non-equilibrium growth follows.

(2) As pointed out above, the calculations were qualitative and the purpose of this study was not to simulate measured results but to qualify the effects of the hygroscopic gases on the droplet sizes.

(3) In eastern Europe (for example Czech Republic, where intense fog episodes occur), the concentrations of SO_2 significantly exceed 400 ppb [Brigman et al. (2002), Novak et al. (1995)]. We believe that this may also be the case with certain Chinese cities.

The given concentrations of NH_3 and HNO_3 are misleading in the article and we will address this point in the revised article. Actually, the 5 ppb of HNO_3 and 10 ppb of NH_3 are equilibrated between the liquid phase and the gas phase in the beginning of the model run, so that the the resulting (initial) gas phase concentrations are in fact much less than those. As figure 5 illustrates, the concentration product of NH_3 and HNO_3 is much less than 1 ppb². If we compare the model values to the measured values

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(for example in Po Valley, [Fuzzi et al. (1992)]) the initial concentration of HNO_3 used in the model could be even higher. Now its concentration drops to almost 10^{-4} ppb during the simulation. Also, the measured values for NH_3 given by Fuzzi et al. show high concentrations of NH_3 during fall-winter time in Po Valley.

(4) The deposition of the pollutants is an important factor of the fog characteristics and also something that will be included in the model in the future. For this study, the deposition can not be included. However, for example nitric acid is almost completely in the liquid phase during the simulation so a very small amount of constant emission of HNO_3 in the simulated domain could be able to maintain the calculated gas phase concentration and reach a steady state value.

(5) Our preliminary calculations using a model which includes an explicit description of gas-phase kinetics show that in the radiation fogs, the temperature decrease is so slow that the nitric acid and ammonia are still mainly partitioned in the larger droplets. Still, you are right in that in the equilibrium model the concentrations of nitric acid and ammonia in the large droplets will be over-estimated.

(6) We will discuss the mass concentrations and improve the figures for the revised version.

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