

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

Anonymous Referee #1

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This is one of the first modeling studies of a fog forming when the atmosphere is not supersaturated (RH less than 100 percent). The authors perform this Sunactivated fog simulation by assigning the appropriate temperature profile and the corresponding initial conditions (e.g., high nitric acid vapor concentration).

(1) The present model does not perform the energy balance of the lower atmosphere so the below-saturation fog is, in a sense, an input to the model. Given the fact that such fogs are not predicted by models that simulate both the fog dynamics and chemistry (for example the work of Bott and collaborators), one is left to wonder about the applicability of the present results to real fogs. A sensitivity analysis of the results to the temperature profile used (e.g. decreasing the minimum temperature by a fraction of a degree) could help address this point.

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(2) The results of the model need to be compared with the available measurements from radiation fogs in polluted environments. For example there have been multiple field studies of radiation fogs in the San Joaquin Valley of California (Collett et al., 1999; 2002 and references therein), an area characterized by high HNO_3 vapor concentrations. The results of the model (volume concentration of the fog, size distribution of the fog droplets) appear to be inconsistent with the observations in that area.

(3) The conclusions of the paper may depend critically on the rather unique set of initial conditions used for the simulations.

The authors have chosen an extremely high SO_2 concentration (400 ppb) which is not applicable to most current fog events in polluted areas. The sensitivity of the results to this concentration should be discussed. If the model is used to simulate the 1952 London fog a much higher particle number concentration should be used and the title of the paper should be changed to reflect that focus.

The second problem concerns the initial NH_3 and HNO_3 gas-phase concentration. At the initial temperature the equilibrium constant of ammonium nitrate is much less than 1 ppb^2 however the authors start with a concentration product of 50 ppb^2 . Starting the system so far from equilibrium (coupled the lack of a dynamic treatment discussed below) could bias the results.

Finally, the results concern scenarios where the gas-phase ammonia concentration exceeds that of the nitric acid vapor. The areas where this is the case, are probably limited to the neighborhood of significant ammonia sources. Some discussion is necessary about the relative importance of these two concentrations.

(4) It appears that the model does not simulate the dry or wet removal of pollutants from the simulated domain. Dry removal of HNO_3 before the start of the fog should be quite rapid. The effect (or lack of an effect) of the wet removal of the fog droplets should be discussed. It is difficult to tell how important wet removal is in this case, because only number (and not volume) distributions are presented in the paper.

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(5) The model assumes thermodynamic equilibrium between the gas and aerosol phases. The dissolution of nitric acid in fog droplets of different sizes is probably governed by mass transfer and not thermodynamics. I would expect that most of the available nitric acid vapor will dissolve in the smallest droplets. A future extension of the model to capture these effects is recommended.

(6) Some discussion of the mass distributions of water and the major ions is necessary. Some of the existing figures could be combined (2b, 2c, 2d) or eliminated (Figure 8) if necessary.

References Bott A. and Carmichael G. R. (1993) Multiphase chemistry in a microphysical radiation fog model—a numerical study, *Atmos. Environ.*, 27A, 503-522.

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