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Interactive comment on "Chinese SO_2 pollution over Europe – Part 2: Simulation of aerosol and cloud condensation nuclei formation" *by* V. Fiedler et al.

V. Fiedler et al.

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Author response to referee #1

We feel that the comments made by referee #1 are constructive and extremely helpful. We have considered all comments and in response to the comments we have made major revisions of the paper draft.

We not only considered the referees comments and changed the paper according to them, but we also extended the paper by including: (a) new AEROFOR and FLEXPART model simulations; (b) soot particle coating by H2SO4/H2O; (c) aerosol particle

activation including also soot particles; (d) light scattering and absorption by modeled aerosol particles,

Since the paper text has changed a lot new sections of the draft are explicitly indicated in the following author comment in brackets.

General comments:

General comment 1: back-trajectory simulations.

FLEXPART forward-simulations have now been added (new Figure 4 and a movie given in the supplementary material).

General comment 2: box-model
(a) Plume dilution due to mixing is now included in the AEROFOR model. Plume dilution was taken from the FLEXPART forward-simulations. (All model results)
(b) The AEROFOR model is used to simulate the plume only during its high altitude-travel when there were no clouds inside and above the plume. (sections 4 and 5)
(c) Simplifications and uncertainties of the AEROFOR model simulations are now critically discussed in detail. (section 4)
(d) AEROFOR model sensitivity studies (SO2, OH, dilution, UV-flux, nucleation rate) have now been included. (section 5, last paragraph)

Specific and technical comments:

1. The abstract has been changed in consideration of the referee comments and extended considering the new parts in the paper (abstract).

2. The occurrence of clouds along the plume trajectory has been investigated and is now explicitly discussed. It has been obtained from satellite data and meteorological analyses using the LAGRANTO model. After lifting, an analyses of cloud top temperatures suggests that, in the upper troposphere on its way from East Asia to Europe, the air parcel traveled always with exception of very short periods above clouds and

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not in clouds the whole way from East Asia to Europe (Figures 6 and 7, section 3, fourth paragraph ff.). Our argumentation therefore is that we neglect all cloud related effects on purpose, as we start the simulation after the air parcel was already lifted to 9 km. AEROFOR model simulations of the conversion of SO2 to sulfuric acid and sulfuric acid induced aerosol formation and growth, and coagulation are used only for the high-altitude travel of the plume, when clouds were absent inside and above the plume. Increased OH (comment 2c), due to UV-backscattering by underlying clouds, has been included (see sensitivity study). We also do not pretend to simulate reality with our model, but we think that it is a helpful demonstration of "textbook knowledge" with rare measurement data. The model is now also described more clearly and the processes that are included are all mentioned in detail (section 4, paragraph 1 ff.).

2a. It is right that we only considered what happened after SO2 has been lifted up to 8 km. This was the idea of the model study. We now clarified in the text, that we choose cloud free conditions, because of the analyses of cloud top pressure satellite pictures, which suggest, that the air parcel under consideration mainly traveled above clouds (section 3, fourth paragraph ff.). A figure with the cloud top pressures and the trajectory pressure is added to the new paper version (Figure 6). The cloud top pressures show that on all days the trajectory traveled above clouds with exception of 2 very short periods ($\approx$ 5 h). There the cloud top pressure was similar to that of the trajectory (Figure 7).

2b. Liquid water clouds were not encountered along the trajectory, and therefore SO2 removal by cloud processes should not have occurred during the upper troposphere travel. Of course, AEROFOR includes particle dry deposition according to Schack et al. (1985); processes such as Brownian diffusion, interception and gravitational settling are taken into account; and also dry deposition of gases (section 4, paragraph 1 ff.). Usually the height of the box model is from the earth surface up to the boundary layer height (varies during a day from 200 m - 2 km) and inside the box the air is well mixed. Now we assume that the air parcel is transported in the FT and is never in contact with

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the boundary layer. Entrainment of ambient air from the surrounding FT air is included (section 4, paragraph 1 ff.).

2c. The OH concentration (depending on altitude and latitude) was taken from Logan et al. In the sensitivity studies the OH concentration has been changed by a factor of 2 also according to the referees suggestions (section 5, paragraph 5).

2d. Mixing with ambient tropospheric air is now included in the model. Vertical entrainment velocity was prescribed so that the total dilution ratio was 10 (5 in the sensitivity study) (section 3, paragraph 2).

3. The discussion and caption of Figure 1 have been extended and now contain the relevant information (Figure 1).

4. The source identification changed and additional source regions, not only China, are taken into account. We also clarified that the trajectory data used as input data for the model simulations in this paper was not FLEXPART data, but ECMWF data prepared with LAGRANTO. However, FLEXPART forward-simulations (Figure 4) are now also included in the paper under discussion, which provide information on plume dilution - Plume dilution has now been included in the AEROFOR model. In the supplementary material of this paper you now find the whole FLEXPART forward simulation of the plume, which shows, that the plume was at least diluted by a factor of 6.7 (supplementary material).

5. Dilution is now included. The considered total dilution over 8.5 day time span is 10. See also the Flexpart forward model simulations in the supplementary material, which show this approximate dilution by a factor of 6.7. It has to be taken into account that the Flexpart figures are column densities. This means that the actual dilution is a bit higher, since also vertical entrainment has to be taken into account (section 3, paragraph 2). Moreover in the sensitivity runs, the total dilution over 8.5 days has also been reduced to a factor of 5 (section 5, paragraph 5). Reconstruction of SO2 has now been carried out by consideration of plume dilution and OH-reaction. The SO2

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reconstruction covers only the period 26 April - 3 May when there were no clouds in and above the plume parcel. The reconstructed SO2 was then used to calculate the OH-induced formation of gas-phase sulfuric and sulfuric acid induced formation and growth of aerosols (section 4, paragraph 2).

6. The required graph is added to the figures in our revised version (Figure 7).

7. T, RHW and RHI in the plume parcel have been determined using ECMWF meteorological data and the LAGRANTO model. The data are shown in figure 7 of the revised paper draft and are also discussed (section 3, paragraph 5 ff.).

8. The relevant text passages have been changed completely, the term secant-like daytime diurnal variation is used (Section 4, paragraph 5 ff.).

9. The new figures show hourly values instead of 10 min values in order to smoothen the curves. The irregularities in the nucleation rate and therefore in Ntot were due to the changes in RH, T (every 6 hours) and OH. This has been corrected now (Figure 8 and Figures in supplementary material).

10. The classical binary nucleation theory really overestimates nucleation rates in the FT (see Vehkamäki et al., 2002) compared to the observations (eg. Hanson and Lovejoy, 2006), and predicts higher nucleation rates than ion-induced nucleation. A sensitivity study with nucleation rate divided by 100 has been added to the sensitivity studies (Section 5, last paragraph). The response of the concentrations of grown particles (which have the ability to substantially scatter light and act as water vapor condensation nuclei) to the lowering of J is relatively small (3%).

11. All aerosol diameters in the old figures were dry diameters as well as in the figure showing the activation CCN diameter. Now, more information is given in the revised paper draft for the new figures and in the text. It is always clearly mentioned, if dry or wet diameters are considered.

12. Figure captions have been improved (All Figures).

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