

***Interactive comment on “Chinese SO₂ pollution
over Europe –
Part 1: Airborne trace gas measurements and
source identification by particle dispersion model
simulations” by V. Fiedler et al.***

V. Fiedler et al.

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Author response to referee #1

We thank referee #1 for the helpful recommendations and comments. Our point by point answers are as follows:

1. It is right that the dry deposition lifetime of SO₂ is about 1 day, but only if the air mass does not ascend and has contact with the surface. The possible lifting

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processes will be addressed further in the new paper version. Cloud and precipitation processes AFTER lifting were not considered as the analysis of cloud top temperature satellite figures suggests that the air mass traveled above clouds during the entire period from China to Europe. This argument will be pointed out clearer in the paper. We will also add, what a consideration of such cloud related effects would change.

2. We did not intend to infer the potential source of the measured SO₂ by using the SO₂/NO_y ratio. For that purpose, we used FLEXPART simulations. We just intended to show, that the SO₂/NO_y ratio nicely supports the FLEXPART results and also fits into the finding of East Asia as source region. OMI satellite data are of no help in this case, as boundary layer sources are often not detected. "Due to reduced OMI sensitivity to SO₂ in the PBL this product should be used only under optimal viewing conditions: radiative cloud fraction < 0.2, solar zenith angle < 40° and near-nadir viewing angles (cross track positions 20 to 40)" (OMSO2 README File v1.1.1).
3. The analysis of satellite figures suggests a lifting of the polluted air mass in a cyclone on the east Chinese border on 25 April 2006, which moved further to the ocean south east from Japan on 26-27 April. Convection in the cold air over China on 25 April is less probable and could also not explain the Japanese source contributions. The most probable scenario is the lifting in the warm conveyor belt first over China on 25-26 April and later also over Japan.
4. We think that this point is exactly addressed by FLEXPART. FLEXPART has been run to identify the main source regions. The identification of the main source regions will be clearer now by inspection of the new Figure 10. A panel has been added, which presents additionally the continental contributions to the predicted SO₂ plume.
5. The flight path of the Falcon in Figure 1 has been changed. Now the path is color
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coded with the SO₂ mole fraction. This figure now shows that the two plumes were measured in totally different regions. The East Asian plume has been detected during the middle of the flight off the West Coast of Ireland, whereas the American plume has been detected at the end of the flight just off the coast of Northern France. So it is not very likely that the Asian Plume resulted from the American one by regional convective mixing. Furthermore, it is right that plume A is maybe also slightly influenced by air that is younger than 8-9 days, if one considers a possible shift of the time axis of FLEXPART. We will clarify this in the next paper version. But, the plume which we discuss as the East Asian plume is ALWAYS the plume at 10:40 UTC, so plume B in Figure 10. And this plume clearly shows only contributions of air masses which are older than 8 days.

6. This point is more or less the same as point 3. A better description of the possible lifting scenarios will be added to the next paper version.
7. For trajectory models, the initialization of the starting altitude is critical and we agree that sensitivity studies would be needed for such a model. However, FLEXPART is no trajectory model. The initial lifting is described by the daily variations of boundary layer height, which is diagnosed from the meteorological analysis data. Thus, "injection height" is of no importance in our case, unless there is reason to believe that high-altitude sources are important (which is not the case here).

Regarding FLEXPART's ability to represent vertical motions, there is a large number of studies showing that lifting in warm conveyor belts (which is primarily responsible for the lifting in this case) is well represented by the model (Stohl and Trickl, 1999; Stohl et al., 2003, and many others). Regarding vertical motion unresolved by the ECMWF data, FLEXPART is equipped with a convection scheme that has been carefully validated with a large number of different data sets (Forster et al., 2007).

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Forster, C., A. Stohl, and P. Seibert (2007): Parametrization of convective transport in a Lagrangian particle dispersion model and its evaluation. *J. Appl. Met. Clim.* 46, 403-422.

Stohl, A., and T. Trickl (1999): A textbook example of long-range transport: Simultaneous observation of ozone maxima of stratospheric and North American origin in the free troposphere over Europe. *J. Geophys. Res.* 104, 30445-30462.

Stohl, A., C. Forster, S. Eckhardt, N. Spichtinger, H. Huntrieser, J. Heland, H. Schlager, S. Wilhelm, F. Arnold, and O. Cooper (2003): A backward modeling study of intercontinental pollution transport using aircraft measurements. *J. Geophys. Res.* 108, 4370, doi:10.1029/2002JD002862.

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