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Interactive Comment

Interactive comment on "Aircraft measurements over Europe of an air pollution plume from Southeast Asia – aerosol and chemical characterization" *by* A. Stohl et al.

A. Stohl et al.

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We thank the reviewer for her/his positive comments on our paper. In the following, the reviewer's comments are repeated in italics, followed by our responses:

This article presents the clearest evidence to date of anthropogenic pollution transport directly from East Asia to the upper troposphere over Europe. A sound modelling strategy employing CO tracers is used to attribute elevations in CO concentrations observed near Europe to Asian emissions. No stone is left unturned, and I think that fewer figures would suffice to convince the reader that the pollution is indeed Asian (see specific comments). The paper moves on to discuss the characteristic composition of the Asian plume and to present clear evidence of mixing with stratospheric air as the *plume heads southeastwards over Europe. Richardson number is calculated from an aircraft profile to demonstrate that turbulence is likely to occur below the tropopause and a tropopause fold, mixing the Asian pollution with stratospheric air. Some salient observations are made on aerosol size spectra in the plumes. Although this is one transport event, there is some discussion of those features that might be typical of transport from Asia. Although beyond the scope of this paper, it would only be possible to move away from case specifics and make more concrete general conclusions by simulating chemical and aerosol transformation using a numerical model.*

The scientific approach is excellent, although not especially original, and the presentation is clear. However, the paper would benefit from discussing fewer figures.

We agree that this case would be ideal to be simulated with a chemistry transport model. Much could be learned about the ozone formation and aerosol tranformation from such a simulation. However, this is beyond the scope of the present paper and we hope that the case will be taken up by others capable of doing such simulations.

We do not agree that fewer figures would be better. The purpose of the figures is not only to convince the reader that the plume was of Asian origin but, in the case of Fig. 1 and 3 for instance, also to show how well the forecasts predicted the plume position, relative to post-mission analyses. Specific reasons for keeping the figures are given below.

1. Section 2.1: the second paragraph was not very clear. It would be better to discuss PSAP and FSSP after the instruments associated with the aerosol size spectrum, rather than mention everything in the first sentence.

Yes, we agree that this paragraph was not very clear. We will move the description of the PSAP to the end of the aerosol instrumentation description and also rephrase the remainder of this paragraph.

2. Section 3 and elsewhere: "backside of the trough" -> "rear of the trough"

Thanks. We will change this as suggested.

3. Omit Fig.1 since so similar to Fig.3g.

Fig. 1 is indeed very similar to Fig. 3g. However, Fig. 1 is the forecast that the entire flight planning was built upon, and by comparing it to Fig. 3g, we want to show how accurate this forecast was. This can only be done by showing both figures. Also, in the final format, Fig. 1 will not accupy much space compared to Fig. 3, and so we will likely keep it.

4. Omit Fig.6 and discussion in Section 4.1 since they only distract from the main story of the paper.

We disagree that Fig. 6 should be omitted. We think it is important to show the spatial variability in middle to upper tropospheric humidity in order to place the flight in a meteorological context. We also come back to this figure in section 4.3.2, where we discuss the mixing with stratospheric air seen on flight B. The description should not distract much from the main story, since it is given at the end of the section on the meteorology and transport.

5. Section 4.2.1: Burma -> Myanmar

Thanks. We will correct this mistake.

6. Section 4.2.2: Why do the simulations using GFS data perform worse. Is the resolved ascent too slow associated with lower resolution of the parent NWP model?

We do not want to open up a discussion of the relative performance of the model versions driven with ECMWF and GFS data in the paper, since the particular reasons for the perhaps somewhat poorer performance using the GFS data are not obvious. In the past, we have also seen cases where the GFS-driven model version performed superior. Please also note that given the long transport distance in the present case both simulations were surprisingly accurate.

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However, averaged over a large number of cases, one must expect the GFS-driven version to be inferior. First of all, a better overall accuracy of ECMWF analyses has been demonstrated in several comparisons of meteorological parameters from the two centers (e.g., comparisons of their respective re-analysis data sets against independent observations). ECMWF's superior four-dimensional data assimilation system is likely a key factor for this. Another reason might be the higher resolution at which the ECMWF model is run. Furthermore, we employ the ECMWF data almost optimally: We use all 91 ECMWF model levels; we use a 0.36 degree high-resolution nest, which is close to the original ECMWF model resolution; we retrieve the ECMWF directly from the ECMWF archives with our own routines, which give an accurate vertical wind and also ensure mass consistency with a high degree of accuracy. In contrast, we use 26 pressure levels (not model levels) for the GFS data; we have no high-resolution nest available; and mass consistency in these data is not as good as with the ECMWF data (which is partly a consequence of using pressure-level data). These latter factors could be improved by using also GFS model-level data and employing these data at a better resolution (both in the horizontal and in the vertical) but these data are currently not available to us.

7. Fig.8 (10): On left axis would be better to replace 100 (120) with 0 at the bottom of the top three panels.

The reviewer is right and we will change this.

8. Fig.9: At this size it is almost impossible to distiguish the black and red dots for fire counts.

Yes but this is a consequence of the ACPD format, which only allows half an A4 page for a figure (including the caption). Figure 9 has a long caption and as a consequence the figure is far too small. We expect this to be much improved in the eventual ACP version of the paper, where the figure will be blown up.

9. Section 4.3.1: It seems dangerous to state that dO3/dCO "measures" the number of

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O3 molecules formed per CO molecule emitted when mixing clearly has such a major influence near the tropopause. These conjectures are too speculative without running a photochemical model.

We agree with the reviewer that this statement is probably misleading. We will replace it with: "Under the assumptions that both CO and O_3 are conserved during transport and if mixing with surrounding air can be neglected, the $\Delta O_3/\Delta CO$ slopes give the number of O_3 molecules formed per CO molecule emitted." This avoids the rather strong word "measures", and also points towards the complicating factor of mixing.

10. Section 4.4: Some of the conjectures in this section were rather sketchy. In particular, do you have any further evidence for new particle formation in the "cloud-free FT" air mass and suppression in the Asian plumes (as opposed to different but unexplained origins)? Although the correlation coefficient between accumulation mode number concentration and CO was lower for air mass II, is this really to do with cirrus cloud encounters? Strong linear correlations are typically associated with regions of mixing between air masses. The cluster of points near CO 170ppbv is associated with the centre of the plume where concentrations are rather homogeneous and the edges of this feature are extremely sharp. These features would reduce the correlation. The only "mechanism" required to explain the isolation of the high CO points (even more obvious in Fig.12) is a very weak mixing rate relative to the horizontal shear on the flanks of the jet carrying the plume.

We do not have any further evidence for new particle formation, other than the particle size spectra themselves. A similar observation over Europe has been made by Petzold et al. (2007, paper in preparation) in biomass burning plumes from North America. There were also few small particles in the dense biomass burning plumes with large numbers of pre-existing accumulation mode particles. Thus, we think that suppression of new particle formation due to the pre-existing large particles is a very likely explanation for the fewer small particles in the plume.

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We admit that it is not completely clear whether the somewhat lower correlation for period II can be explained by cirrus cloud encounters or other factors (though cirrus clouds were present during period II, as opposed to the other periods). Therefore, we will remove the two sentences discussing the somewhat lower correlation during period II, which is a minor point of the paper anyway and probably distracting from the main story.

The reviewer is right about the effect of mixing on the isolation of data points. This may also explain lower correlations during period II.

11. Fig.18: Show the size spectra side by side or use only the volume density.

Number and volume density plots convey different messages, so we will keep both plots. We are not entirely sure what the reviewer means with "side by side" but we realize that it might be better to show the two panels stacked vertically. We will change this accordingly.

12. Section 5: "Trace gas correlations between CO, NOy and O3 were all positive" for flight A. Only air mass III on flight B was similar. I suggest removing speculation about reduced small particles in Asian plumes due to high concentrations in the accumulation mode. I would omit the last two sentences concerning ozone of "stratospheric origin" at Zugspitze. There are many candidate processes to account for this and it is too speculative to relate to mixing of pollution into the stratosphere.

We have added "flight A" in the section on trace gas correlations, as suggested. We also added the word "probably" to the statement on reduced concentrations of small particles being due to enhanced accumulation mode aerosols, as this is only a likely explanation but wasn't proven in this paper. We agree that the reference to the Zugspitze observations are probably too speculative (at least for the conclusions section) and, thus, we will remove them.

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 12611, 2006.