

Interactive comment on “In-situ observation of Asian pollution transported into the Arctic lowermost stratosphere” by A. Roiger et al.

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We want to thank reviewer 2 for the positive assessment of our manuscript. Please find below our answers. For convenience, the referee questions are highlighted in bold letters.

Although there is ample evidence about the transport process (warm conveyor belt) and the source region (eastern China), I wonder if biomass burning can be completely excluded as an explanation for the pollution. In particular the very high CO₂ value in Fig 9c (approx. 385 ppmv at 140 ppbv CO), which is higher than any tropospheric value, indicates a very strong combustion source, e.g. biomass burning. From Fig. 9c I estimate a delta CO per delta CO₂ ratio of about 4-5 ppbv/ppbm, which is similar to emission ratios in crown fires, that can

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inject biomass burning debris high into the TP region. Are there other measurements of biomass burning tracers, e.g. CH₃CN, so that biomass burning can be definitely excluded as a source? Also, it might be worth checking whether the highest CO₂ values observed are consistent with surface measurements over the source region, i.e. eastern China.

This is an interesting discussion point which has been addressed in the manuscript, but perhaps not detailed enough. Since the Falcon measured no biomass burning tracers such as CH₃CN or HCN, we cannot definitely exclude influence from forest fires emissions based on our in-situ observations. However, we suggest that the high CO₂/CO mixing ratio even supports the conclusion that the sampled pollution originates from anthropogenic sources.

As mentioned also in the final response to referee 1 (question 11), the $\Delta\text{CO}_2/\Delta\text{CO}$ ratio is difficult to quantify due to the absence of a well-defined background value not affected by pollution. However, as correctly stated by the referee, the CO₂ value of 385 $\mu\text{mol/mol}$ is much higher than other tropospheric CO₂ values observed during the GRACE campaign. As already discussed in section 3.6.1, this indicates that we sampled a quite different type of pollution in comparison to all other GRACE flights. The whole GRACE data set was highly influenced by biomass burning (e.g. Sodemann et al., 2011) and typically showed lower CO₂/CO ratios than in the case discussed herein (CO₂ = $380.8 \pm 1.5 \mu\text{mol/mol}$ for CO > 120 nmol/mol, please compare also with Figure 9c). Low CO₂/CO ratios result from low combustion efficiencies of forest fires, which are especially low for surface fires as typically observed in Siberia (e.g. Wooster and Zhang, 2004). The high CO₂/CO ratio observed in our case thus points to a different pollution source region than biomass burning emissions from Siberia, and a more efficient combustion source as for example industrial pollution: In anthropogenic emissions, initial CO₂/CO slopes may be up to a magnitude higher than in biomass burning pollution (Andreae and Merlet, 2001; Suntharalingam et al., 2004).

A direct comparison with surface observations is difficult since the polluted air mass certainly was diluted with tropospheric background air before it reached the

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tropopause. However, they support our assumption: From measurements conducted at a rural site near Beijing in summer 2008, Wang et al. (2010) report CO₂ mixing ratios typically greater than 385 $\mu\text{mol/mol}$. In contrast, significant lower CO₂ mixing ratios of ~ 380 $\mu\text{mol/mol}$ on average were observed in 6 distinct fire plumes sampled in July 2008 over Siberia (Paris et al., 2009).

Finally we want to note that special observations during POLARCAT 2008 gave no indication for the occurrence of pyro-convection events over Siberia, which could have lifted fire emissions directly into the upper troposphere/lowermost stratosphere (M. Fromm, personal communication).

All these results support our conclusion that Asian anthropogenic pollution and not biomass burning emissions from Siberia was the dominant source of the observed pollution plume.

We included part of the above discussion in the final manuscript.

In the conclusions the authors state that the mixing is irreversible and thus affects the chemical composition of the mixing layer above the local TP. In order to validate this statement, the authors should calculate forward trajectories from the flight track to investigate the future development of the streamer.

Our statement is already based on the results of a forward trajectory calculation. As mentioned in the last paragraph of section 3.6.2, forward trajectories have been started along the flight track and show a steady increase in PV during the following days (on average from 5.1 PVU to 9.4 PVU after 5 days). This is pointed out more clearly in the final manuscript.

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