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Atmospheric Chemistry and Physics Discussions

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

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First of all we want to thank referee 2 for the helpful comments on our manuscript. Please find below our point-to-point answers. For convenience, the referee questions are highlighted in bold letters.

1. The authors speculate, that troposphere to stratosphere exchange (TST) occurred during the travel of the air masses from the WCB to the measurement location rather than directly through the ascent. What are the reasons for this hypothesis, since the authors state, that most trajectories encountered PV > 2PVU just after the WCB. Is there any further evidence?

We agree that mixing processes between the tropospheric and stratospheric air

masses certainly took place also just at the end of the WCB lifting process. However, there are several arguments which indicate that the TST exchange proceeded during the next days during which the pollution plume was transported across the pole:

- The in-situ data clearly show that the stratospheric influence increases towards the boundaries of the encountered filament. This suggests that further TST exchange happened after the WCB up-lift, since the probed elongated filament was generated more recently, during the cross-polar transport in the jet, due to stirring and filamentation of the polluted tropospheric streamer (see Fig. 6 for the temporal evolution of the streamer).
- The quasi-ideal linear mixing line observed in the O3-CO scatter plot (Fig. 9a) indicates mixing between two air masses having well-defined tracer "end-members": Mixing processes at an earlier stage during the WCB uplift would rather be evident in deviations from this ideal mixing line (due to subsequent mixing and/or chemical processes). This in turn suggests that the major TST exchange happened more recently during the cross-polar transport.
- Our assumption is further supported by the forward trajectory calculation. As mentioned by the referee, the trajectories reach PV > 2 PVU within the WCB ascent or shortly later. However, PV further increases along the forward trajectories, finally reaching values between 3 and 10 PVU above Greenland. This additional gain in PV (and Theta) reflects further entrainment of PV-rich, warmer air from the stratosphere during the cross-polar transport.

We included parts of this discussion in the final manuscript.

2. What is the reason to split the emission data in so many sub-clusters, doesn′t this lead to inconsistencies?

It is well-known that emission inventories have large uncertainties. For this reason it is better to use the best available emission inventory for each region, i.e. those with the best spatial resolution and the latest update, rather than using one global inventory

which is consistent but out-dated. For example, for Europe the EMEP inventory has the better resolution and is more up-to-date than the EDGAR inventory.

3. What motivates a tracer removal after 20 days, since it strongly depends on the dynamics acting on the individual plume, how efficient the mixing with the background is?

The FLEXPART model is a particle dispersion model which however does not include any chemical removal processes, as for example the reaction of CO with OH, the main sink for CO in the atmosphere. Therefore, due to the on-going emission of the pollution tracers the aged emissions have to be removed at some point in time in order to avoid its artificial accumulation within the whole atmosphere. In addition, the CO mixing ratios calculated by the FLEXPART model are used to identify polluted air masses influenced by recent emissions. In order to exclude aged pollution from air masses influenced by fresh emissions, the tracers are removed after 20 days, which has been shown to be a reasonable time period (Sodemann et al., 2011).

4. Replace 'connected' by 'associated'

Ok.

5. Are the LAGRANTO trajectories initialized along the flight track?

No, as stated in the later section 3.3.2, the trajectories were initialized from a box centred at the location where the Asian pollution was sampled. To avoid misunderstanding, we now mention this earlier in section 2.3.

6. It is stated that 92% (of the 4.5%) intersected the 2 PVU tropopause. How long did they stay above 2 PVU? Was there a criterion to check the time, which the air parcels spent in the stratosphere (to account for frequent reversible undulations around the dynamical tropopause)? What is the maximum PV of the air parcels?

This is an interesting point which we did not discuss in section 3.4 since it is focused on the meteorological situation in the source region. Only a small number of trajectories

starting in the source region is transported over the pole and finally reaches Greenland (and northernmost Canada), which makes it difficult to infer a statistical statement from this specific trajectory set up. Nevertheless, we found that these trajectories spent 81% of the time in the stratosphere (above 2 PVU) and showed almost no undulations. These trajectories finally reach PV values ranging between 3 and 10 PVU above Greenland. We included these findings in the final version of the manuscript.

7. Isn't this a bit contradictive? The WCB is associated with the air streams ahead the surface cold front in the warm sector, which leads to the strongest uplift. This is generally not the cyclone center.

We agree that this formulation is not very precise. We wanted to state that the trajectories that reach the Arctic were located on the western side of the coherently ascending ensemble of trajectories, which indeed means closer to (and not within) the cyclone centre. We rephrased the sentence in the final manuscript.

8. The reference to Plumb and Ko is only correct when looking at really long-lived tracers (long-lived with respect to the underlying transport processes). This is not the case here -especially CO is not covered by the prerequirements in Plumb and Ko.

Since this reference indeed might be misleading, we removed it.

9. Ozone = 100 nmol/mol was selected as criterion and not determined from the data, I guess?

Actually it was determined from the data. Smallest ozone mixing ratios of about 100 nmol/mol have been measured in the centre of the "MR", meaning that the polluted tropospheric parent air mass in this part contains the smallest fraction of stratospheric air.

10. The H2O ozone correlation - as stated correctly by the authors - is heavily affected by temperature. They interpret the deviating tropospheric correlation

as dehydrated. Does this fit to the Lagrangian Cold point, which can be easily deduced from the trajectories?

Using the saturation mixing ratio at the Lagrangian cold point derived from the backward trajectories, we would expect a H2O mixing ratio of about 90 µmol/mol. Taking the uncertainty of this calculation into account, this is in quite good agreement with the measured H2O mixing ratio of ∼70 to 80 µmol/mol.

11. Please specify: I guess the enhancements ratio (Delta CO2)/Delta(CO) is meant. Are there any specific values for the CO/CO2 ratio around? The simple occurrence of positive slopes does not necessarily indicate combustion, since positive slopes of both tracers can occur in normal 'background' conditions. Since this is most likely not the case here, a number would help here. Further, the NO and NOy data are correlated with ozone, but anti correlated with CO (see Fig.2). This does not support the conclusion, that strong pollution sources have affected the observations. How do correlations of CO versus NO (or NOz versus CO or ozone) look like?

We do not use enhancement ratios ΔCO2/ΔCO: The pollution plume was not sampled in the troposphere, therefore the CO2 background value (of the tropospheric parent air mass) is not well defined which in turn makes it difficult to derive enhancement ratios ΔCO2/ΔCO. We can use however the absolute CO2 value of the air sample having the highest fraction of tropospheric air (i.e. CO2 ∼385 µmol/mol at CO ∼140 nmol/mol). As already addressed in section 3.6.1, a comparison with other CO2 mixing ratios measured in the biomass burning polluted troposphere during GRACE reveals that those are much lower (CO2 = 380.8 ± 1.5 µmol/mol for CO > 120 nmol/mol, as evident in Figure 9c), which points to a different source region of the pollution plume. For a further discussion on this, please see also answer to question 1 of referee 2.
Concerning the comment on the correlation between NO and NOy with O3: The positive correlation of NO and NOy with ozone is clearly dominated by the typically increasing values of NO and NOy within the stratosphere. We agree that one could

expect higher NO and NOy values in fresh polluted tropospheric air than observed in this case. However, as already discussed in section 3.6.1, we have to keep in mind that 1) the polluted air was already aged (∼6-10 days old) and thus chemically processed, meaning that NO was already cycled into higher NOy reservoirs such as e.g. HNO3, and 2) The pollution subsequently was lifted within a WCB, and it is well-known that the NOy species HNO3 is efficiently removed during export from the boundary layer to the free troposphere within precipitation areas (e.g. Stohl et al., 2002, Myiazaki et al., 2003). Finally, we do not expect to see very strong and distinct pollution signatures since we penetrated the upper boundary of an aged pollution plume which was chemically processed and diluted both in the troposphere and stratosphere.
Based on the above answer, we extended the discussion in section 3.6.1.

12. The authors suggest that most of the mixing happened during the travel of the air masses due to stirring and filamentation. How is the evolution of PV along the trajectories? Do they show a PV increase during their travel (e.g. a plot as in Figure 4, but for PV only for TST trajectories).

As stated in the last paragraph of section 3.6.2, the polar-crossing WCB trajectories show a continuous increase during their journey to Greenland. They reach PV values ranging between 3 to 10 PVU by the time they arrive in our measurement area (see also answer to question 1).

13. The general statement that CO in the LMS exceeds 100 nmol/mol north of the polar jet is misleading. This can happen, but is a strong function of altitude relative to the tropopause - please rephrase.

The statement is possibly misleading. We rephrased it in the final manuscript: "Furthermore, the lowermost stratosphere north of the jet stream contains a wide range of CO mixing ratios (Cooper et al., 2005). In polar regions, CO exceeds 100 nmol/mol about 25% of the time in the 1 km thick layer above the summertime tropopause (Tilmes et al., 2010), indicating frequent entrainment of polluted air masses."

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