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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Detection in the summer polar stratosphere of air plume pollution from East Asia and North America by balloon-borne in situ CO measurements" *by* G. Krysztofiak et al.

Anonymous Referee #2

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This manuscript presents a comprehensive case study of balloon-borne in-situ measurements of CO from the SWIR and SPIRALE instruments during summer near Kiruna. Different tools (satellite measurement comparisons, trajectory studies, and CTM modelling) have been used to support the conclusion that polluted air masses from the East Asian and Northern American troposphere have been transported up to the polar lower stratosphere. The topic is well suited for publication in ACP. However, the conclusions in my eyes remain rather inconclusive given the complex tools at hand. These have the potential to shed more light onto the question of air mass origin raised in this study. Source attribution of the pollution seems still unclear to me, as outlined in further detail below. Also, while the manuscript is generally well and clearly struc-

tured, some sections need to be improved considerably in language and logic in order to make the manuscript more easily comprehensible. I ask for minor revisions before I can recommend this manuscript for publication.

General/major comments:

(1) My first concern about the presented study is that while it explores different sources of the observed pollution event in the lowermost stratosphere using comprehensive tools, it fails at pinpointing down the quantitative information on the source regions in a conclusive way. It is for example interesting to use masked regions to infer the contributions of a certain region, however, this method seems insofar flawed to me that the total of the individual contributions from the different regions exceed the value derived in the whole-world simulation. I know that you point out a problem in the use of REPROBUS, but you need to guide the reader in the conclusion section as well and in a better way on how much we can learn from your results, what are the relative contributions of the different pollution sources, and what uncertainty these numbers are associated with. At present, the conclusions are not very contenting since it seems to highlight the role of the jet-stream as major pathway of pollutant transport between the continents, a result well known from many earlier studies.

(2) Another concern I have is about the particular use of the CO-O3 correlation tool to argue that August 7 represents a special event. I suspect that the comparison to August 24 looks so extraordinary mostly due to the fact that the latter profile is cut off well above the tropopause or in other words that the profile had been sampled in a very strong stratospheric intrusion event, in which the relations between the tropopause and potential temperature are highly different. Tracer-tracer correlations cannot be used to argue for mixing events without discussing the position of the tropopause. You further extrapolate some to me rather arbitrary mixing lines between a theoretical L-shaped correlation expected in an atmosphere without any mixing, a mid-latitudinal and a polar reference point taken from mid-latitudinal observations and your measurements, respectively. This to me is not a scientifically sound method, since there is no argument

for using the data point of stratospheric background value you have chosen. You basically could use any point on the stratospheric branch to draw your mixing lines in any way to fit your data points. We know that the chemical transition region between the troposphere and the stratosphere is dependent on latitude and if you instead compare your results to Figure 7 of Hegglin et al. (2009), you'll see that your balloon measurements do not show any unexpected values, but nicely fall in between the envelope for the NH summer polar CO-O3 correlation provided by the satellite measurements (e.g., CO values around 70 ppbv at about 300 ppbv O3). This is not to say that we are not interested in the origin of the air masses that lead to this observed mixing layer, rather the contrary, but that the motivation of and approach taken in this discussion section in particular needs to be cleaned up.

Minor and technical comments:

(1) Introduction, P15506 L5: The SPURT campaign (Engel et al., 2006) has been sampling the polar lowermost stratosphere in all seasons. Please add a reference to these results.

(2) Introduction, P15506 L20 onward: You write about chemistry and lifetimes in the troposphere, however, you study pollutant transport into the lowermost stratosphere where CO lifetimes are somewhat longer and ozone chemistry is expected to be different from that in the troposphere. Here ozone chemistry is dependent on season (Hegglin et al., 2006). Increased CO (and NOx) levels lead to ozone production mainly during spring and summer and depend on background ozone values.

(3) The interpretation of the elevated CO values in L2 is rather minimal. Could it be that due to the drift in longitude-latitude along the balloon is experiencing during its travel, it is getting into a region of lower PV again? I suggest to plot both PV (interpolated onto the balloon pathway) and/or the distance to the dynamical (2 PVU) tropopause into Figure 2.

(4) I don't understand how you derive the tropopause in your study shown in Figure

2. The 'cold point' tropopause (as you call it and I would understand the term) is used in the tropics, in the extratropics either the thermal tropopause (WMO, 1957) or the dynamical tropopause (WMO, 1987) are used. In tropopause folds, the dynamical PV is often more meaningful. Please add the PV tropopause and explain in more detail how you derive the tropopause you use.

(5) Figure 1: please consider putting a black thin contour around the SWIR data points. It is very hard to distinguish the circles from the squares.

(6) Figure 2: It would be helpful for the interpretation of the profiles to add PV vertical profiles to this plot and extend the tracer profiles to the ground if available.

(7) Figure 3: It would be helpful to overplot the position of the latitude-longitude box shown in Figure 1 onto the PV maps, in order to obtain a better picture of whether the strong gradient seen in the IASI CO observations on the 24 August coincides with the strong PV gradient at the edge of the streamer.

(8) Figure 5, caption. As mentioned in the major comments, I think it is not correct to draw the mixing line the way it is done here. Point (1) on the stratospheric branch is chosen in a rather arbitrary way.

(9) Does REPROBUS not allow for tagging the origin of air parcels? This would yield a less ambiguous quantification of the sources.

(10) Introduction, P15506 L17: change 'Polar stratosphere' to 'The polar stratosphere'.

(11) Introduction, p15508 L6: change sentence to 'we analyze the origin of the air masses sampled.'

(12) P15509, L28: spelling out 'short-wave infrared' here, with 'SWIR' in brackets

(13) P15510, L2: say 'In nadir-looking, the SWIR-balloon. . .'

(14) P15510, L19: rewrite first sentence to 'Satellite data are used to enhance the interpretation of the balloon measurements.'

(15) Throughout manuscript: Use ‘molecules per cm-2’ instead of ‘molecule per cm-2’.

(16) P15511, L13: change to ‘a full line-by-line radiative transfer model.’

(17) P15511, L25: suggest explaining shortly for what these approaches are used. E.g., ‘. . .have been used to calculate backward trajectories to track air mass origin, potential vorticity maps to study the dynamical situation, and . . .’. I don’t understand what you want to say with ‘chemistry scheme’, please improve language.

(18) P15512, L1: change to ‘We used 3-hourly ERA-interim reanalysis fields. . . vertical levels. Clusters. . .’

(19) P15513: Suggest using another title for Section 3 than just ‘Measurements’. A lot of people tend to skip the Measurement description section, and that what it currently sounds like.

(20) P15514, L15: I don’t think this is a well-founded conclusion. The difference in the total column between 7 and 24 August is 0.33*1E18 molecules cmÂň-2, but the difference in the partial column above 9 km only 0.15*1E18 (from table 1). So in my eyes only half of the enhancement is explained. The important thing is that it enhances the stratospheric column by 50

(21) P15514, L23: change ‘This later’ to ‘This latter’, because you don’t mean later in time, but later in the discussion.

(22) P15515, L16: change to ‘. . .typical for polar latitudes.’

(23) Section 3.3.2: This section is very badly written, and I can’t understand the approach too well. Please improve. Some things to change, but not an exhaustive list: First paragraph: The wind module cannot be denoted by the 30 ms-1, it is the results of the wind module that can be denoted. Please improve language. It is ‘10-day backward trajectories’ or ‘backward trajectories along 10 days’. L15: This first sentence doesn’t make sense. L24: it is ‘air mass trajectories’ not ‘air masses trajectories’. P15517, L7: ‘subject to’ not ‘subjected to’. L18: ‘Both revealed a crucial. . .’ is too obvious. May

say ‘Both clusters of air parcels experience fast inter-continental transport along the jet-stream.’

(24) P15518, L7: 200 ppbv ozone is still stratospheric, tropospheric ozone is mostly defined as values below 100 ppbv (Bethan et al., 1996).

(25) P15518, L15: As mentioned before, this may be because the profile is taken in strong stratospheric intrusion. You normally don’t find 15 ppbv below 18 km, especially during summer (cf., Hegglin et al., 2009).

(26) P15520, L9: change to ‘over Western Europe’.

(27) P15522, L4 onwards: elaborate more on what a uncertainty estimation is for your results given these short-comings.

(28) Figure 7: There is cluster of high NH3 on the West Coast of America (California) that is not correlated with number of fires detected. What does this mean for the interpretation. Also, you con’t include Northern Canada/Alaska into your masked area. Do you know how it would change if you were to use a box of the West-Coast only instead of including the East coast?

References:

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