<u>Reviewer #2</u>

We thank the reviewer for the careful reading of the manuscript and helpful comments. We have revised the manuscript following the suggestions, as described below. The revisions are shown in purple colour in the new manuscript.

<u>General comments:</u>

First of all, following your recommendations, the article has been rewritten by considering the altitude relative to the thermal tropopause and consequently all the conclusions relative to the altitude of the pollution layer have been reorganized (CO vmr profile, FLEXTRA trajectories, correlations and REPROBUS simulations results). A complete section about the altitude of the tropopause during August 2009 has been added in the final manuscript in Section 3.1.

(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.

This section (4.2.2) has been rewritten in order to guide more the reader. Indeed the difference in CO vmr between SPF07 and SPF24 for all simulations have been reconsidered by using the relative altitude compared to the thermal tropopause (Tp). We added a new simulation taking into account the fire pollution from Alaska-Canada and Siberia. We looked at the contribution of the different regions for two different altitudes, one in the stratosphere (+250 m above the Tp of SPF07) and the other one in the troposphere (500 m below the Tp). The conclusion for this section has been rewritten as:

"According to the simulations, the air masses sampled in the L1 mainly come from anthropogenic emissions. The stratospheric part of L1 (250 m above the Tp) is more influenced by East Asia than North America pollution, while the tropospheric part (500 m below the Tp) is influenced less by East Asia than by North America pollutions. The fires in Alaska, Canada and Siberia have a negligible influence on the CO vmr in the intrusion. This is also in agreement with the FLEXTRA trajectories (section 3.4.2 and Fig. 5), which show that the Alaska-Canada fire emissions influence equally SPF07 as SPF24."

The warning concerning the interpretation of the data has been put after the conclusion, but no uncertainty can be extracted from these simulations.

(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 O_3). 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.

The part 4.1 has been rewritten by following the correction and advice:

The CO-O_3 correlations have been plotted again for both flights with the altitude range for the correlation points corresponding to the relative altitude from the thermal tropopause observed in the new Fig. 3, (2.5 km and 0.6 km below the thermal for the 07 and 24 August, respectively). Using the definition from Hegglin et al. (2009), we found for SPF07 the stratospheric and tropospheric branches and cut the correlation into three parts: the stratosphere, the mixing layer and the troposphere. For SPF24, only the stratospheric branch has been defined because the CO measurements begin only 0.6 km below the thermal tropopause and the O_3 concentration was not found lower than 100 ppbv in this altitude range (value used by Hegglin et al. (2009) to find the tropospheric branch).

In the manuscript, the part with the mixing lines was deleted and the position of the mixing layer and the origin of the air masses of this layer have been discussed, instead the tracer-tracer correlations, the relative position of the tropopause and the probability distribution function for both flights have been used.

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:

The first sentence of our introduction has been completed by "In addition the SPURT campaign (Engel et al., 2006) focussed on the transport in the UT/LMS (upper troposphere / lowermost stratosphere) during the four seasons between 35°N and 75°N over Europe."

(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.

Production, sink and chemistry of CO in the stratosphere have been added in the introduction: "At higher altitude, in the stratosphere, CO is principally produced by methane oxidation. Reaction with OH is the main sink of CO in the troposphere and the lower stratosphere. CO has a chemical lifetime of around 2 months in the troposphere, allowing for possible direct transport into the stratosphere where its lifetime is much longer. CO is a good chemical tracer and, correlated with O3, is a powerful tool to study the exchange between troposphere and stratosphere (Fischer et al., 2000; Hoor et al., 2002; Brioude et al., 2006). CO plays an important role in the atmosphere oxidation capacity and in the production of tropospheric ozone. Ozone chemistry, enhanced in spring and summer, is closely depending on the CO and NOx emissions and on the season (Hegglin et al., 2006)."

(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.
AND
(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.
The vorticity potential is only available by using the ECMWF era interim data.
We calculated for the three flights the dynamical tropopause corresponding to the 1-3 PVU surface by using the ECMWF data. It appears for SPF07, the altitude corresponding to the three PV surfaces is relatively close (dynamical tropopause between 11.5 km and 11.9) but for SPF24, the corresponding altitude is really different (dynamical tropopause between 5.7 km and 8.7 km). For more convenient, we choose to use the thermal tropopause (WMO, 1957) which is directly accessible by the SPIRALE data. So, we choose to plot the Fig.2 (new Fig.3) in altitude relative to the thermal tropopause. All the discussion in the final manuscript is now reorganized by use the altitude relative to the thermal tropopause.

(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.
SWIR data points have been put in black contour for more clarify as recommended.

(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.
The tracer profiles to the ground are not available for SPIRALE. The in-situ measurements start for CO at 8.5 km on average corresponding to the moment when the SPIRALE beam is deployed. The profile for SPF24 has been extended to 8.5 km in Fig.2 (new Fig.3). Figure 2 (new Fig.3) has been plotted with altitude relative to the thermal tropopause (as explained in question (3)). Moreover, the variability of the potential vorticity during August 2009 has been discussed in a new section (section 3.1). A new figure has been created showing the potential vorticity against the time and pressure altitude.

(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.
The latitude-longitude box has been added in the Fig. 3 (new Fig.4). The IASI CO observation on 24 August coincides approximately with the strong PV gradient on 24 August at 18 UTC but much better at 00 UTC (Figure above). Indeed, the IASI CO data concern all the day of the 24 August 2009.

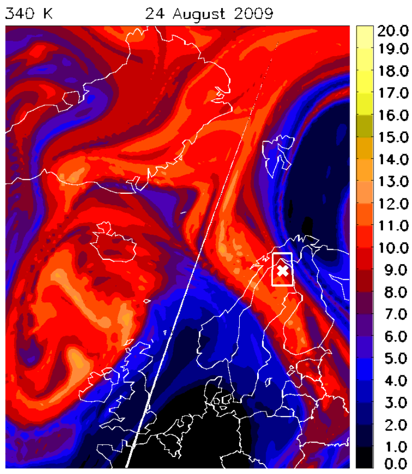

Figure: Potential vorticity from MIMOSA model at 340 K level on 24 August 2009 at 00 UTC. The white cross sign shows the location of the balloon flights

(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.

Figure 5 (new Fig.6) has been plotted in function of the relative altitude to the tropopause and the discussion about it has been totally rewrite (see section 4.1 in the manuscript and our response in the General comments)

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

REPROBUS allows using passive tracer related to one region in particular to follow the air parcel coming from this region. Different passive tracers are used and multiplied by number of region of origin wanted. We use approximately the same approach for the section 4.2.2, for the corrections, we added a new mask for the two regions of fires emission (North America and Asia).

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

Corrected

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

Corrected

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

Corrected

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

Corrected

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

Corrected

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

'molecule cm^{-2}' have been replaced by 'molecules cm^{-2}' in the text and also in the Figures**.**

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

(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.

The sentence has been changed by "Three models, namely FLEXTRA (Stohl et al., 1995), MIMOSA (Modèle Isentrope de transport Méso-échelle de l'Ozone Stratosphérique par Advection; Hauchecorne et al., 2002), and REPROBUS (REactive PRocesses ruling the Ozone BUdget in the Stratosphere; Lefèvre et al., 1994) have been used to calculate backward trajectories to track air mass origin, to calculate potential vorticity maps to study dynamical conditions, and to determine the CO emission regional distribution, respectively."

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

(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.
Thank you for this advice, we change the title to "3. Measurement description and interpretation"

(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 cm^{-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

The difference in the total column between 7 and 24 August is 0.18*1E18 molecules cm^{-2} and between the 7 and 14 August is 0.33*1E18 molecules cm^{-2} with only IASI measurement and 0.24 *1E18 molecules cm^{-2} using SWIR total column. And, we agree with the difference of 0.15*1E18 molecules cm^{-2} in the partial column. So the conclusion, according your advice, has been rewrite as follow: "To conclude, the difference in the total column between the 7 August and the 14 or 24 August is on average 0.2×10^{18} molecules cm^{-2} and the difference in the partial column (between 9 and 34 km) is about 0.15×10^{18} molecules cm^{-2}. Consequently, the increase in the total column is mainly (around 75%) explained by the change in the CO concentration in the altitude range [9, 34] km. In the following, we focus on this part of the atmosphere."

(21) P15514, L23: change 'This later' to 'This latter', because you don't mean later in time, but later in the discussion.
This sentence has been changed and "This latter" deleted.

(22) P15515, L16: change to '. . .typical for polar latitudes.'
Corrected

(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.'

The section has been rewritten.

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

We change the sentence by "Unlike CO, O_3 is mainly produced in the stratosphere and has thus a low vmr in the troposphere (below 100 ppb, 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).

The section 4.1 has been totally rewritten.

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

Corrected

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

We cannot estimate the uncertainties from these simulations.

(28) Figure 7: There is cluster of high NH_3 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?

The high NH_3 cluster on the West coast of America is due to the numerous crops in California. There is no incidence in the data interpretation; we can only suppose if pollutants with long lifetime are emitted from these crops, they could possibly reach also the polar stratosphere.