Author's response letter

Thank you for your careful reviews and comments to improve the quality of the manuscript. We have provided point to point responses to all comments and questions. The original referee comments are in black and the authors' response are in blue. Please note that, there are two Figures included at the end of this file which is not included in the manuscript itself. The reason for adding those figures is to answer one of the reviewer's comments.

Anonymous Referee #1

In section 2.1.1 the authors comment on the use of TIR+NIR MOPITT data and the restriction of the data used to daylight only. They comment that the daylight data "has better information content". However the NIR channels operate by reflected sunlight and so at night the TIR+NIR product is identical to the TIR product that has no sensitivity near the surface. This should be mentioned as a stronger driver for working only with daylight data.

Thank you for your suggestion. The paper was revised based on the reviewer comment, as shown in lines 141-144.

In section 3.1 with reference to Figure 2 the winter variation DJF seems comparable with the MAM and JJA with SON showing the flattest distribution. It is hard to see the author's assertion that the spatial variations are less prominent in winter. It would be very useful to have the topography as well since the column amount is influenced by the topography – less atmosphere = less CO at constant average mixing ratio. The authors mention topography on p 31776 but only in the context of fewer sources – which is also true.

We agree and this was corrected, as shown in line 196.

Regarding topography effect on total column CO values, we created a SRTM 90 DEM map over Alberta (Figure 1) and it demonstrates mountain peaks along the western border, to lowland areas in northeastern Alberta as mentioned in line 223 of the revised manuscript. Then we converted the CO total column to volume mixing ratio (vmr) to find out the influence of topography on CO spatial variations as shown in Figure 2. It is obvious that total CO vmr has the same spatial pattern as CO total column which implies that the topography does not have a significant influence on CO spatial variations. Accordingly we decided to not include the effect of topography in this part of discussion.

In Figure 3 the SON season is said to be similar to JJA but it does not show the same maximum in the North East as JJA – it seems distinctly flatter.

Corrected, as shown in line 207.

The main difference between figures 2 and 3 seems to be that in DJF the CO is generally concentrated nearer the ground (possible influenced by subsidence).

In fact, to figure out if CO is concentrated near to the ground, CO profile should be analyzed and this is shown in Fig 5, however the main difference between figures 2 and 3 is that, the spatial variations in spring are less than total column variations (Fig. 2) as mentioned in line 208.

On p 31776 line 12 it should be noted that CO is a product of incomplete combustion processes and so although CO increases with increasing combustion, it decreases with increasing regulation of the combustion: forest fires produce a lot of CO, power stations comparatively little.

We agree; that is why we analyzed MODIS fire counts as well to help in the interpretation of CO variations. The manuscript was revised based on the above comment, as shown in line 231.

On P 31778 the authors say that "Calgary is expected to experience Chinook winds more frequently than Edmonton" I would assume that the data exist to verify this speculation. The referee wonders whether the fact that the Edmonton winter/spring profile seems less well mixed than Calgary or Fort McMurray is significant.

The fact that Edmonton winter/spring profile seems less well mixed than Fort McMurray is significant as it is verified by positive Omega averages at 700hPa from 2002 to 2013 especially from Dec-Mar.

On p 31780 The text talks about a "rate" but the specifies a unit of "%" whereas a rate should have a time unit as well. This is also true for P 31783.

It is changed accordingly to "decreasing percent for the whole period" as shown in lines 336 and 402.

Figure 8 and its explanation is somewhat confusing. If the annual average value is subtracted from each month, then the trend over years should be zero, but there is a trend which implies perhaps that the series annual average is being subtracted. This should be clarified.

We are sorry for the typo; actually the whole series average is being subtracted. The manscuript is revised accordingly, as shown in line 332.

The rates of decline of CO over the cities is clearly visible in Figure 10. More intriguing to the referee is the suppression of the seasonal cycle in all three regions which seems very pronounced. The authors attribute this to the improvement in vehicles and if so, this is remarkable.

Yes the rates of decline of CO over the cities are clearly visible in Figure 10. However there is no suppression of the seasonal cycle in Fig 10, just the y axis scale is large (up to 1000 ppb) enough to show the seasonality of MOPITT data as it is more suitable to plot ground data. The MOPITT seasonality is clearer in Fig 8 where the scale fits the data very well.

There is almost always a problem with relating satellite measurements of pollutants to specific surface sites, since the surface sites are often chosen to be where the signal is highest, not where it is "typical". Perhaps some comment on the location of the sites as typical of the region or to monitor specific hot spots could be made.

Actually the stations in Edmonton and Calgary are part of the monitoring network in typical urban centers, however Fort McMurray station represents industrial region and this is clarified in line 167.

In section 3.5 – neither dry conditions nor sinking air "cause" fires – an ignition source is needed – but they do set conditions for fires to spread and persist.

Corrected as shown in line 451.

Figure 13 c,d shows the MOPITT CO distribution during the fire of 2012. It would be useful to compare that distribution to another year without fires – even at the expense of deleting one of c,d to make room. Technical Details.

This topic warrants further details, that can be more appropriately addressed in a separate paper. We are writing now a paper about the impact of biomass burning on air quality including a comparison with another year without fires. Hence we prefer to leave this part as is.

P 31768 Lines 18 and 22 - it may be just my pdf reader, but the "s" on "sites" and "exacerabates" is detached from the word itself. P 31768 Line 24 "..the declining trend..."

Done as shown in lines 26, and 30.

P 31769 Line 18 "..used to decrease the bitumen's...."

Done as shown in line 51.

P 31769 Line 20 "Large amounts of natural gas...."

Done as shown in line 53.

P 31770 Line 18 ".. there has been no research published using them...." (is that the intent?)

Done as shown in line 76.

P 31770 Line 27 428692.5 is surely stated far beyond the accuracy of the assessment.

Done as shown in line 85.

P 31771 Line 7 “CO can also be produced....”.

Done as shown in line 93.

P31772 Line 3 “analysed using MODIS fire counts.”

changed as shown in line 116.

P 31772 Line 9 “..represents an industrial...” .

Done as shown in line 122.

P31773 Line 15 “..cloud edges and coastlines.”

Done as shown in line 157.

P 31773 Line 20 “The CASA Data...”

Done as shown in line 162.

P 31774 Line 5 (and several figure legends) “The symbols F,... represent the cities of Fort....Calgary respectively.”

Done as shown in line 193.

P 31775 Line 12 “...seasons display minimum...”.

Done as shown in line 200.

P 31775 Line 17 “The summer season demonstrates...” .

Done as shown in line 204.

P 31775 Line 19 “..fall season illustrates a similar....” .

Done as shown in line 205.

P31775 Line 20 “...spring and summer indicate a...” .

Done as shown in line 208.

P 31776 Line 7 “..are less than 1.5 x 10ˆ?? molecular cm-2..” Needs an exponent, surely?

Done as shown in line 221.

P 31777 Line 5 “...temporal climatologies...”.

Done as shown in line 245.

P 31777 Line 17 “Calgary, the vertical...”.

Done as shown in line 256.

P 31778 Line 12 “..contribute to pollution....”.

Done as shown in line 276.

P 31778 Line 17 “...and declines rapidly.”

Done as shown in line 282.

P 31778 Line 26 “...attributed to other sporadic....fires. The forest fire...” .

Done as shown in line 289.

P 31779 Line 16 “..above the planetary...”.

Done as shown in line 307.

P 31783 Line 1 “..maximum rates of decline of 4.7...” .

Done as shown in line 402.

P 31786 Line 1 “..affected by the fire emissions...” .

Done as shown in line 482.

Figure 1: Personally I would have chosen a different colour than blue for the oil sand regions. My first thought was “what are those lakes doing in Alberta?”

The map is changed to another one as shown in line 812.

Figures 2,3 Legend “The symbols F,... represent the cities of Fort....Calgary respectively.”

Done as shown in lines 819 and 828.

Author's response letter

Thank you for your careful reviews and comments to improve the quality of the manuscript. We have provided point to point responses to all comments and questions. The original referee comments are in black and the answers are in blue.

Anonymous Referee #2

Page 31770, line 18: Change “no researches” to “no studies”

Done and it is shown in line 76.

2. Page 31770, line 27: Use a comma when you have a number with four or more digits, e.g. “428 692.5” should be 428,692.5”

Done and it is shown in line 85.

3. Page 31771, line 7: Remove “it can” so that the sentence reads “CO can also be produced...”

Done and it is shown in line 93.

4. Page 31771, line 11: CO is not a significant source of ozone. What are its “significant” impacts on air quality? I would suggest removing the word “significant” here.

Done and it is shown in line 96.

5. Page 31771, line 12: Should be “sufficient”

Done and it is shown in line 97.

6. Page 31774, lines 5-9: What is the range of altitudes for which you have aircraft data?

The range of altitudes of aircraft data is from surface till about 300-250 hPa.

7. Page 31775, line 13: It should be “especially in central and southern Alberta.”

Done and it is shown in line 200.

8. Page 31778, line 11: Should this be “fresh air from the mountains”?

Done and it is shown in line 276.

9. Page 31778, line 12: Can you substantiate this claim that Calgary has less development than Edmonton? Are there bottom-up inventories for emissions from Calgary and Edmonton that you can compare?

Deleted and it is shown in line 271.

10. Page 31778, lines 14-16: I don’t understand this sentence “The seasonality in the Fort McMurray area is progressively less pronounced...” Please clarify or rephrase.

Done and it is shown in line 279.

11. Page 31778, line 17: Change “in summer than the springtime” to “in summer than in spring”

Done and it is shown in line 281.

12. Page 31778, line 22: Change “on a clean air area” to “to a clean air area”.

Done and it is shown in line 280.

13. Page 31778, line 24: Change “transported at the aircraft’s” to “transported to the aircraft’s”
Done and it is shown in line 287.

14. Page 31779, line 4: Please see comment #2 above about numbers of four or more digits.
Done and it is shown in line 296.

15. Page 31779, line 7-9: It is not clear if the authors are referring to “deep convection” over North America or elsewhere. I would be surprised if deep convection is important over North America in spring.
Changed to “vertical motion or mixing” and it is shown in line 300.

16. Page 31779, line 16: Change “where cities such as” to “whereas cities such as”
Done and it is shown in line 307.

17. Page 31779, line 18: Add “the” between “over” and “Edmonton area”
Done and it is shown in line 309.

18. Page 31779, lines 25-26: I don’t understand what the authors mean by “the degree of repeatable seasonal variability varies by year and region” in the context of Fig 7.
Changed to: "the magnitude of seasonal variability is not the same for all years and regions" as shown line 316.

19. Page 31780, lines 1-3: The claim that emissions are confined to the PBL due to subsidence is unsubstantiated. The plots of the vertical motions in Fig 4 do not prove that the emissions are confined to the PBL. For example, the surface CO peaks in April over Calgary (Fig 7), but the mean omega in the region is negative in April (Fig 6), suggesting ascent. Anyway, it is not clear to me why the authors are concerned about confinement here. The vertical gradient in CO in Fig 7 is expected if there are emissions at the surface.
We agree that emissions at the surface affect vertical gradient, as stated in line 319-320 of the original manuscript. However we analyzed omega variations at 700 hPa to understand the strength of vertical mixing. For example, the mean omega values are negative in April which means there is an ascent, and this is seen in Fig. 7 (CO profile) as high CO values are extended to higher altitudes especially over Calgary.

20. Page 31780, lines 7-9: This statement about transport emissions relative to industrial emissions is redundant. It was already stated on page 31779, lines 1-3.
It is deleted.

21. Page 31780, lines 15-17: Why remove the annual mean for each year instead of the 12-year mean? If there is a long-term trend, removing the annual means should remove the trend, but that is not the case here.
Actually this was incorrectly stated in the original manuscript. The calculation is based on the 12-year mean. This issue is fixed as shown in line 332.

22. Page 31780, line 20: I assume that this is -1%/year?
Actually it is -1 % for the whole period. This is clarified in the revised manuscript.

23. Page 31781, line 8: “the” should be “The”.
Done and it is shown in line 353.

24. Page 31781, lines 14-19: I assume that the aircraft profiles do not extend much above the upper troposphere. In applying Eq (1), what do the authors do for the upper part of the profiles? For example, Worden et al. (2010) used a MOZART climatology at altitudes above 250 hPa when they transformed the MOZAIC data.
The missing profile part above the highest altitude where MOZAIC/IAGOS measures is estimated from the MOPITT a priori profile and it is added in line 361.

25. Page 31781, lines 23-25: Deeter et al. (2013), which is cited in the manuscript, showed that there is a 14% high bias in MOPITT V5J data at 200 hPa. This should be mentioned here.
Done and it is shown in line 371.

26. Page 31782, line 5: I don’t understand what is meant by the statement that “winter surface emissions are more entrained in the boundary layer”. Do the authors mean confined to the boundary layer? What is the evidence that this is the case?
Yes we meant confined in the boundary layer as this is indicated by the omega monthly variations and the seasonal spatial variations of mega (positive values). The sentence was revised accordingly, as shown in Line 380.

27. Page31782, lines 8-11: The statement that here is more lofting of the emissions in spring is unsubstantiated. Either demonstrate this or remove this statement. This shift in the seasonality could be due to the sensitivity of the surface level retrievals, which should increase in spring and summer as the thermal contrast increases. However, because the CO burden is decreasing from winter to spring, the surface maximum will be shifted into spring. I would recommend that the authors look at the seasonal variation of the surface layer degrees of freedom for signal (DFS), as shown in Fig 3 of Worden et al. (2010). That might provide greater insight into what the MOPITT data are showing.
We agree that there is a seasonal variation of degrees of freedom for signal (DFS) and it affects CO monthly variations, but also Fig.6 shows negative omega for all regions in April and May which implies better emissions lofting as mentioned in the manuscript. We believe this demonstrate that that here is more lofting of the emissions in spring. Additionally the highest MOPITT sensitivity in general is still to CO in the middle of troposphere (as shown by averaging kernel), so when there is better vertical mixing (negative omega), this will help MOPITT to capture the surface emissions.

28. Page 31782, line 28: Remove “cities” after Calgary.
Done and it is shown in line 402.

29. Page 31783, line 1: Declining rate on what time scale? Are these annual decreases?
The declining rate is over the whole time. This is clarified in the revised manuscript.

30. Page 31783, line 14: Remove “the” between “where” and “central”
Done and it is shown in line 415.

31. Page 31783, line 21: “MOPPIT” should be “MOPITT”.
Done and it is shown in line 421.

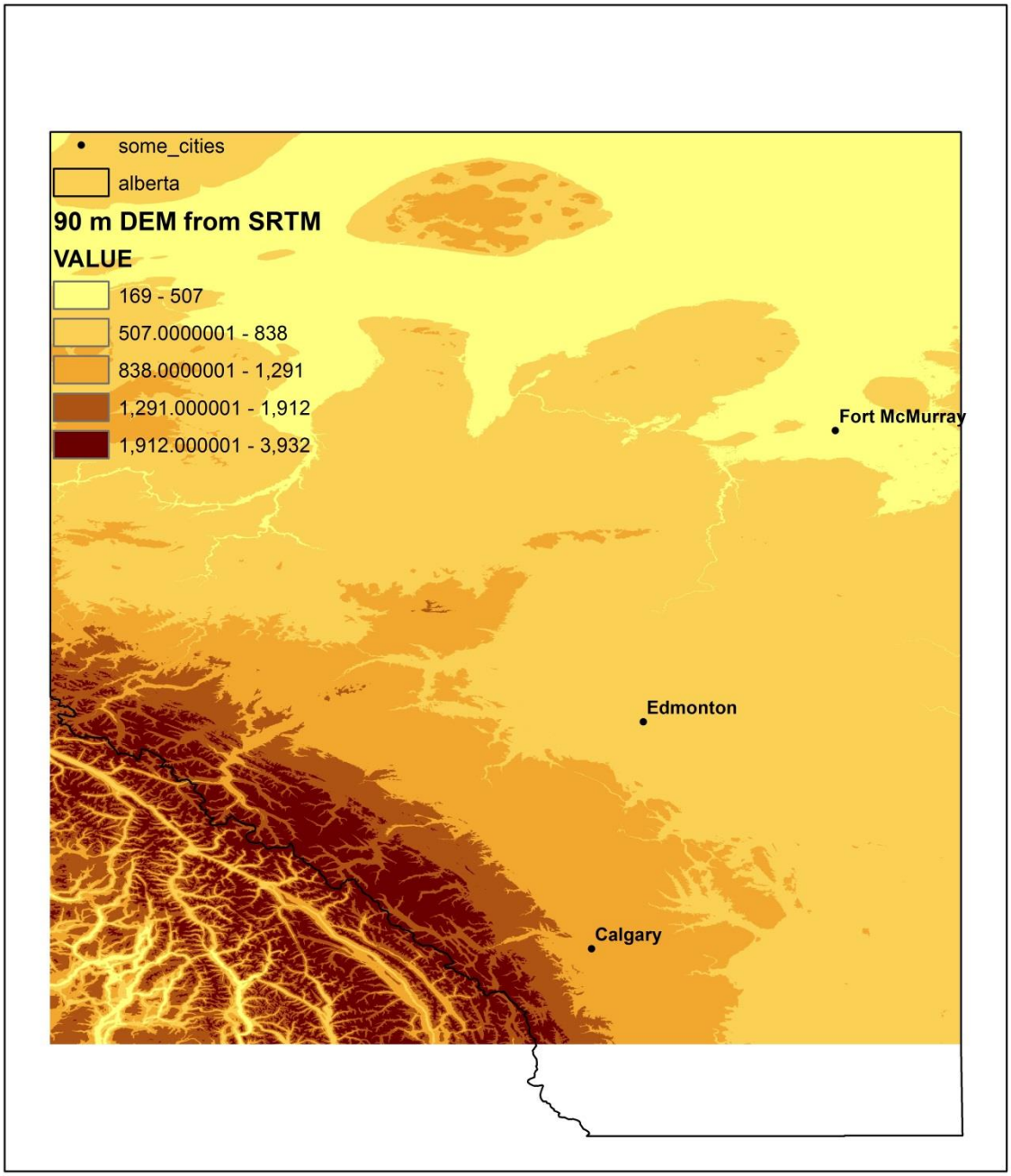

Figure 1 SRTM 90 DEM over Alberta.

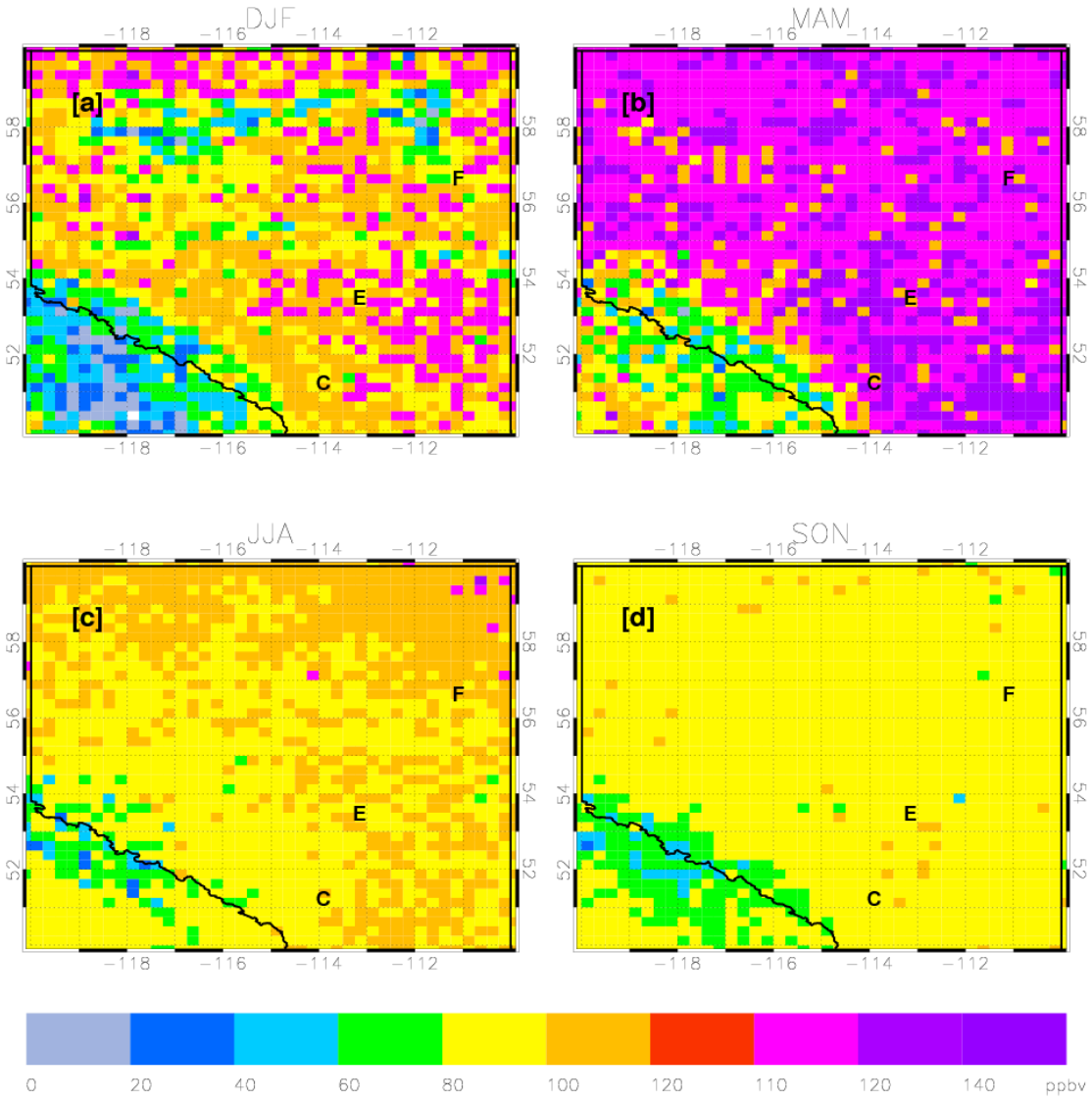

Figure 2 MOPITT CO total column in vmr (volume mixing ratio) for the period from 2003-2013