Interactive comment on “Trans-Pacific transport of reactive nitrogen and ozone to Canada during spring” by T. W. Walker et al.

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Received and published: 31 August 2010

The revised manuscript is attached as a supplement.

Response to Reviewer 1

0.0.1

The study uses a global chemical transport model (GEOS-Chem) in combination with aircraft measurements during the INTEX-B campaign in spring 2006 to analyze source contributions of ozone in the free troposphere over the remote Pacific and identify transported pollution entering North America, with a focus on Canada. A model base simulation and a set of sensitivity runs are conducted to examine the roles of Asian anthropogenic and lightning emissions and chemical processing to the ozone background. The paper also describes a scaling method of using multi-year satellite retrievals of NO2 to create timely NOx emissions for the period of interest. The study addresses an important issue in atmospheric sciences, especially since transpacific pollution transport might become an even bigger concern in light of the rapid economical growth in Asia. The work is well carried out and the methods generally sound. However, I have a few more specific comments that I would prefer if being addressed in more detail before publication.

0.0.2

We thank the reviewer for their comments. Specific reviewer comments are addressed individually below.

0.0.3

Specific comments:

0.0.4

Throughout the paper I find the way the results are presented often disconnected and hampering the flow of reading. For example, in the introduction it is talked about lightning emissions and their importance (page 8721), then the emissions modeling is discussed, then the focus gets back to discussing long-range transport, followed by stating a way for evaluating a model, and then the modeling part is discussed. Another example is Page 8730 (Line 3ff) where the model sensitivity simulations are discussed in Section "3.1 Estimates of Emissions" The emissions modeling itself, while a nice
method, seems to me as being outside of the objectives of the paper, at least the way it is presented.

0.0.5

The introduction has been rewritten to make the point of the paper more plain. The paragraph at the end of Section 3.1 has been moved to the model description, where it is more germane to the discussion.

0.0.6

Abstract, line 19: Does the 2% global increase refer to a sensitivity study considering PAN from all sources or only sources in Asia?

0.0.7

In the sensitivity study that produces this result, we disable the conversion of PAN to and from NO\(_x\) globally. The text has been updated to clarify this point.

0.0.8

Page 8728, Line 23: I am not following this scaling to 2003 emissions at all and suggest further explanations. Or, if this is standard practice in GEOS-CHEM, then it would help to, at least, include a reference.

0.0.9

The scaling from 2000 to 2003 uses the standard practice for GEOS-Chem. It is mentioned explicitly here because the model version used does not have scaling factors for these years. The text has been updated to mention that this is a standard method of the model, including the citation to the paper describing the scaling method.

0.0.10

Page 8729, Line 3: Is the trend calculated for entire Asia, or separately for China and East Asia? Can you comment if and how changes in other sources (biomass burning) might impact such a trend and the derived emissions?

0.0.11

The trends calculated from the SCIAMACHY data are performed at the model resolution (2\(^\circ\) × 2.5\(^\circ\)). The numbers given for China and East Asia are simply averaged over these respective regions. The text has been updated to clarify.

0.0.12

Trends in other emission sources to which SCIAMACHY NO\(_2\) columns are sensitive could impact the derived emissions. For instance, an increase in biomass burning emissions over the four years would cause an overestimate of the trend, which would result in estimates of anthropogenic emissions that are too high since we attribute the trend solely to changes in anthropogenic emissions. We expect the trend in anthropogenic emissions to be the dominant influence in
NO_2 columns over this period. Furthermore, we chose to use a trend over the four years rather than only the two endpoint years to reduce the sensitivity of the estimate to outliers. The text has been updated.

Page 8731, Figure 5: Can you comment on the fact the model does not represent the low end of observed values?

In Figure 5, the observed propane mixing ratio goes as low as about 6 pptv, while the model has no values below about 20 pptv. Most of the low-end values in the plot come from the same flight, and were all sampled in the lower atmosphere (pressure > 550hPa) and had smaller than average carbon monoxide mixing ratios as well. It is hypothesized that the aircraft encountered a plume of clean marine air, but the coarse model is not able to represent these lower-than-background values. Text has been added on the issue.

Page 8731, Line 20 ff: Can you explain the different biases between the data sets, e.g. a large positive bias for C-130 and a large negative bias for Cessna data? Overall, I would not second what is described as a “good agreement”. O3 compared to C-130 has a rather larger bias at all altitudes and PNs and NOx for DC-8 are significantly overestimated in the model. It is also disconcerting to me that, for C-130, the model agrees well with PNs and NOx but has a large bias in O3, while the model compared to DC-8 has a large bias in PNs and NOx, but agrees rather well with O3.

As the intercomparison flights indicate, the C-130 ozone measurements are several ppbv higher than coincident measurements by the Cessna. Beyond that, differences between the Cessna and C-130 profiles reflect processes occurring over North America that are beyond the scope of this paper.

The simulation of the DC-8 measurements is high with respect to PANs and NOx, but agrees with ozone. The simulation of the C-130 measurements agrees with PAN and NOx, but is low with respect to ozone. In both cases, the simulation is producing too little ozone per molecule of reactive nitrogen for both the nitrogen species and ozone to match the measurements. Two possible reasons for this are already mentioned in the text (the non-steady state of NOx, and the large overestimate of nitric acid).

Page 8733, Figure 9: I am surprised by the high model bias compared to OMI. Compared to the aircraft and sonde data, the model seemed biased rather low and on page 8732, Line 18 you mention that other studies found that GEOS-Chem underestimates ozone of stratospheric origin. Can you comment on this?

On further review, we located a bug in the calculation of the tropospheric columns. The figure has been remedied and the mean model bias is now 3.7
DU lower than OMI, which is more consistent with the bias with respect to ozonesonde and aircraft observations.

Page 8734, Line 15: Because of the non-linear chemistry, it is not the “actual contribution” that is calculated by differencing model results with and without Asian emissions, but rather an estimate of how the ozone production is changed if there were no Asian emissions. Can you comment on how the derived estimate would differ from the “actual contribution”?

The absence of the Asian source will artificially raise the ozone production efficiency in the simulation without the Asian source, so the derived estimate will be an underestimate of the net production due to the Asian source. This distinction is made in the preceding section, so was not reiterated here.

Page 8734, Figure 10: Do I understand that correctly that, at least in an average sense, net ozone production from Asian sources only occurs over Asia, meaning that ozone transported into North America is ozone that was produced in Asia and then transported across the Pacific? Or is this only true for 800 hPa? How representative is the 800 hPa level for other altitudes?

Net ozone production due to the Asian anthropogenic NO_x source seems to occur primarily over the continent. The gross production extends much farther downwind, but is more or less balanced by the gross loss shown in the bottom left panel of Figure 10 (now Figure 11). At least in part, the gross production downwind is due to PAN that is transported out of the boundary layer and later subsides to produce NO_x over the ocean, as shown in the top right panel of this figure. This same pattern holds true throughout the lower troposphere (up to 400 hPa, at least), although the production over the continent is obviously larger as one moves toward the surface. Text has been added.

Page 8737, Line 12: How do total O3 and PANs look along the flight track? Do they resemble the measurements?

The model reproduces the general shape of the measurements along these flight tracks, but has insufficient spatial resolution to capture the peaks in concentration encountered as the aircraft traverses the sub-grid scale plumes. A note of this has been added to the text.
Walker et al. investigates transport of ozone and reactive nitrogen from Asian to Canada using an ensemble of aircraft, satellite, and ozonesonde observation interpreted with the GEOS-Chem chemical transport model. Four sensitivity simulations are conducted to examine contributions from various NOx sources and the chemical mechanism. A transport event on May 4-5, 2006 is examined in detail to infer ozone production from PAN decomposition. The study is well designed, makes a comprehensive use of INTEX-B observations, and contributes to our knowledge of trans-pacific transport of ozone pollution. However, I have some specific comments that would suggest the authors to address to improve the manuscript before publication.

We thank the reviewer for their comments. Specific reviewer comments are addressed individually below.

Specific comments:

Page 8722, Line 9-15: I suggest at the beginning of this paragraph adding 1-2 sentences that introduce the employ of chemical transport models for studying long-range transport.

Two sentences were added to this paragraph to re-introduce the use of CTM’s in the modelling of long-range transport. CTM’s are also discussed in this context in the third paragraph of the introduction.


Added a citation to the intercomparison paper.

Page 8725, Line 24: WMO thermal tropopause is described here, but this information is not used in the following analyses. Please clarify.

A sentence was added at the end of the paragraph indicating how the WMO tropopause was used. Each sonde profile was truncated at the WMO thermal tropopause calculated for that sounding before any averaging or other analysis.
was performed.

Page 8728, Line 18-19: I think another challenge is the systematic bias in the satellite measurements of NO2 columns that can cause a bias in estimating the emission for a specific time. If so, please clarify.

Clarified in the text.

Page 8729: The method used to derive the emission trend needs a better explanation. As I understand, the values of beta in Equ. (1) can be affected by the contributions of lightning NOx and other NOx sources outside the region to the NO2 columns. What is the typical value of $\beta$? How differ from 1 ($\beta = 1$ if following Equ. (2))? In equation 1, $\beta$ is calculated from the baseline simulation and a perturbation simulation. The only difference between the two is a 30% increase to Asian anthropogenic emissions in the perturbation (although 10% and 20% increases were also tested and yielded similar values). Other sources of NOx in the simulation do affect the value of $\beta$ in that they provide the background state against which the perturbation is measured. Values of $\beta$ are within a few percent of 1 everywhere except over the East Asian cities where the large perturbation to NOx emissions caused a disproportionately smaller increase in the NO2 column (peak value is 8.8).

Page 8729, Line 14-15: Suggest changing the sentence to: “The factor $\beta$ is then used to infer the emission trend between 2003 and 2006 from SCIAMACHY observed NO2 columns.”

Updated the text.

Page 8730, Fig 4: Can you comment on the 30 percent difference between the SCIAMACHY and model NO2 column?

This magnitude of bias is typical of comparisons between GEOS-Chem and remotely sensed NO2 columns. A sentence and a number of references have been added to the text to clarify.
Page 8731, Line 24: Suggest using negative values to represent model underestimates and positive values for model overestimates. This also applies for describing comparisons of NOx and PAN on Page 8732, Line 5-7, and for ozone on Page 8732, Line 17.

The sign of the bias has been reversed, and a clarification added to the text.

Page 8733, Line 3: Do you use the same tropopause pressure for computing the tropospheric ozone columns from OMI and GEOS-Chem? Please mention.

Yes, the same tropopause pressure is used for both calculations. The text has been updated to indicate this.

Page 8736, Fig. 12: Can you comment on the negative values over the US in the top left panel of Fig. 12? Also from Fig.12 and Fig.13, over the eastern Pacific the regions where PAN transport is important (left panels) tend to be regions where Asian ozone contributions are small (right panels), can you comment on that?

The top left panel shows the difference in ozone concentrations between the baseline simulation and the “no PAN” simulation. Negative values indicate regions where the reaction of NOx into PAN has reduced the ozone concentration, because the local ozone production is lowered when NOx is partitioned into a reservoir species and transported away (see Figure 11, top right panel).

It should be noted that the contributions from PAN transport and from Asian sources are not mutually exclusive. Some of the PAN contribution comes from radicals produced over North America or elsewhere, and not all of the Asian contribution uses the PAN pathway. In Figure 12 (now Figure 14), the westward branch of the flight still has high amounts of PAN, so the influence on ozone there is small. In Figure 13 (now Figure 15), the contribution from PAN transport at the aircraft location becomes more coincident with the Asian contribution. The text in Section 4.2 has been rewritten to better emphasize this. Also, Figures 12 and 13 now show the contributions at 4 km, which is nearer to the altitude where the aircraft sample the plume.

Technical comments:

This reference has been updated.

Page 8725, Line 4: Suggest replacing “consider” with “use”

Replaced.

Page 8725, Line 7: Suggest deleting “aboard”

Removed.

Page 8734, Line 10: Delete “including”

Modified surrounding text to clarify. Changed to “... regions with net photochemical production including downward flux from the stratosphere, and in situ...”

All the references are ended with numbers of “87xx”. Please correct.

Page 8751, Fig 1: please use a larger font size for longitude and latitude labels.

Label size has been increased on Figure 1.
This paper aims to describe the chemical and transport processes that allow Asian emissions to affect the Canadian troposphere during the INTEX-B springtime intensive. The analysis covers topics previously addressed by several studies examining the impact of Asian emissions on the USA. While the paper lacks originality in many aspects, it's still important to see how Asian emissions impact the large area covered by western Canada. This paper could be appropriate for publication in ACP, but before it can be considered further it requires a major revision to address the points described below.

We thank the reviewer for their comments and suggestions. Specific issues are dealt with individually below.

Major comments:

The introduction is not clear as to what the main purpose of the paper is. Instead it just lists several topics that the paper covers with no apparent common link. The order in which they appear in the Introduction is: 1) role of PAN 2) role of LNOx 3) using SCIAMACHY for a top-down emission inventory 4) need for understanding pollution transport to Canada 5) hydrocarbon ratios 6) ozone source attribution. Judging by the title of the paper, the main purpose of the paper is to address #4, with all the other topics being covered so as to support this goal. If this is the case then the Introduction needs to be re-written to clearly convey this point.

The introduction has been rewritten to make the point more plain.
Page 8723 line 19 In the description of the aircraft intercomparison it says that measurements within 200 hPa of each other were compared. This is a very large vertical distance of 2-3 km and air parcels that are spaced so far apart in the vertical often have very different chemical characteristics and transport histories. I really don't see how this vertical grouping can provide a valid intercomparison. Please justify.

The wording of the criteria for data inclusion was poor. Both intercomparison flights included near-simultaneous profiles at close horizontal range; however, on the May 3 flight the Cessna performed its ascent about 15 minutes ahead of the C-130. The wide altitude range was used to accommodate this separation in time (by virtue of profiling first, the Cessna was about 200hPa above the C-130 at the same time). The wording has been changed to indicate that the aircraft both sampled the same altitude within a short time period of one another. This definition alters the included data only slightly.

Page 8725 line It seems that the ozonesondes were only used to verify the model. Why were ozonesondes from Bratt's Lake used instead of sondes from Trinidad Head which is closer to Whistler and more representative of the air from the North Pacific Ocean?

The ozonesondes from Trinidad Head have already been used to validate the GEOS-Chem model and are shown in Figure 5 of the Zhang et al., 2008 manuscript (“Transpacific transport of ozone pollution and the effect of recent Asian emission increases on air quality in North America: an integrated analysis using satellite, aircraft, ozonesonde, and surface observations”). One emphasis in this work is transport into western Canada, which makes the model performance at Bratt’s Lake relevant.

page 8732 lines 15-19 At Kelowna and Bratt’s Lake the model is biased low, possibly because it underestimates transport of ozone from the stratosphere. But Whistler is close to Kelowna and experiences the same stratospheric intrusions, but here the model overestimates ozone when compared to the Cessna flights. This is a major discrepancy and needs to be explored further. Did the Cessna only fly at times when stratospheric intrusions were unlikely to be present? This may be the case seeing as the Cessna had many more ozone values close to 30 ppbv in the free troposphere, which would likely have a southerly origin with less stratospheric influence. You need to compare relative humidity between Kelowna and the Cessna. If the air sampled by the Cessna is much more humid it was probably biased towards non-stratospheric influenced air masses. Back trajectories would also indicate if one site had more transport from the stratosphere.
The backtrajectories for these two locations are quite similar, as are the humidity profiles. Differences between the Cessna and ozonesonde profiles reflect processes occurring over North America that are beyond the scope of this paper.

page 8733 lines 27-29 Here you state that the greatest Asian contribution occurs between 600-700 hPa. But looking at the plots I just don’t see a maximum influence occurring in this altitude range. The altitude of greatest Asian impact would be an important and useful piece of information and you need to show clearly where this occurs. Please create a plot that shows the difference between the model runs (with and without Asian emissions) in units of ppbv (and percent) vs altitude and include all three aircraft datasets and both ozonesonde datasets. This plot will clearly show the altitude of maximum Asian influence.

This plot has been created (Figure 10) and is now described at the appropriate point in the text.

Section 4.2 This section requires a thorough revision. It begins by stating that there are two branches of Asian outflow that affect N. America, it then goes on to say that the section will “explore how the two branches affect Canadian air quality.” Surprisingly, the section does not describe the two branches, and only discusses measurements and model simulations from one of the branches. It also fails to say which branch of the outflow was sampled by the aircraft. When revising the description of the transport pathways please also reference Holzer and Hall, 2007, a very good paper that describes trans-Pacific transport pathways. There are also problems regarding the interpretation that the aircraft sampled the same polluted air mass on consecutive days. The authors suggest that the sampling on consecutive days illustrates the chemical aging of the air mass. But I don’t see how the aircraft sampled the same air mass. The air mass on May 4 was heading towards the northeast while the air mass sampled on May 5 was southeast of the air mass sampled on May 4. These can’t be the same air masses. Please provide forward trajectories from the May 4 air mass and backward trajectories from both the May 4 and May 5 air masses to see if they have similar transport histories. Also, the discussion of Figure 12 states that the contribution from PAN transport is > 2 ppbv, but the figure clearly shows that in the region of interest, the contributions is only about 1 ppbv. Holzer M, Hall TM, Low-level transpacific transport, JOURNAL OF GEOPHYSICAL RESEARCH-ATMOSPHERES Volume: 112 Issue: D9 Article Number : D09103 Published: MAY 2 2007

The description of the transport pathways has been clarified as to which branch of the Asian transported plume is being examined here. Also, a reference to Holzer and Hall, 2007 has been included.

48-hour ensembles of backtrajectories from each of the six marked points in the two figures all originate from higher altitudes just south of the Aleutian islands. The forward trajectories from the May 4 flight do coincide intersect the flight
track on the following day, and the backtrajectories from the May 5 flight intersect the previous day’s track. Forward trajectories from the May 5 flight show the air circulating to the southeast around the high pressure system off the California coast. It seems that the aircraft here sample the southern, descending branch identified in Zhang et al 2008. The text interpreting these figures has been updated to highlight this. Also, Figures 12 and 13 (now 14 and 15) have been updated to show the ozone contributions at a higher altitude that is nearer to where the aircraft is sampling (4 km instead of 2 km).

0.0.85

page 8721 line 7 Here you call for additional work to disentangle the various contributions to ozone production over Asia, the North Pacific and North America. But this type of work has recently been reported by the following papers which deserve greater credit. These papers also need to be mentioned in the conclusions as they support your findings.

0.0.86

Both these references are now included in the introduction as well as the conclusions.

0.0.87

The conclusions end by stating that the transport pathway studied in this paper is a potential transport pathway into the Arctic. This topic appears nowhere else in the paper and its mention in the Conclusions comes as a surprise and seems very out of place. This final paragraph is unnecessary and needs to be removed.

0.0.88

While this is an issue of importance, this paragraph has been removed.

0.0.89

Minor comments:

0.0.90

Abstract: page 8719 lines 4 and 5 would sound better as: to evaluate sensitivities of the free troposphere above the North Pacific Ocean and North America to Asian anthropogenic emissions.

0.0.91

The text has been updated.

0.0.92

page 8719 line 10 when talking about the contributions of Asian emissions and LNOx, specify the altitude you are talking about. And are you talking about lightning from just Asia or global lightning?
The text has been updated to mention the altitude range of the Cessna. All figures regarding lightning refer to global lightning, which is now also mentioned at this point in the abstract.

When stating that PAN is responsible for 2% of global ozone production, specify that this analysis is just for spring 2006 and is not a climatological value.

The value is derived using simulations for the spring 2006 period of interest. The text has been updated to reflect this.

"persistent winds" implies that the northeastward transport occurs all the time, when it surely does not due to the many cyclones that pass through this region, which modulate the transport. Maybe say something like “commonly occurring transport patterns”

Text has been switched to the latter wording.

Two recent papers that make the case for Asian pollution being a concern for North America are:

Inserted citations for the Parrish et al, 2009 and Cooper et al, 2010 papers.

Here and throughout the paper the geographically correct term for the ocean of interest is the North Pacific Ocean.

Fixed throughout the text.

Removed.
Heald et al. 2003 is a TRACE-P paper, not an ITCT paper.

Moved the Heald et al. 2003 citation to correct location.

A WCB is an airstream within an extra-tropical cyclone and is not a circulation pattern.

Changed wording to use “airstream” instead of “circulation pattern.”

page 8722 line 14 over several e-folding lifetimes.

Added to text.

I assume this is a bottom-up inventory that is being discussed?

Yes, it is the bottom-up inventory. Clarified this in the text.

page 8730 line 6 please make clear that global lightning NOx emissions are considered, not just Asian

Clarified that it is indeed the global lightning source.

Looking at the figure that compares model ozone to C-130 ozone, it’s clear that the model underestimates ozone and is biased low. But you report the bias as being positive, presumably because you calculate it by subtracting model ozone from measured ozone. Still this isn’t intuitive and it would be more consistent with the figure to report the model bias as -6.6 ppbv
The sign of the bias has been reversed, and a clarification added to the text.

page 8733 line 6 It would be helpful if you would also report the overestimate as a percentage.

Added to text.

page 8733 line 21 What do you mean by reasonable? Please provide some comparisons to other studies to show that this value is reasonable.

References to other studies of stratosphere-troposphere exchange have been added to the text.

Page 8735 line 22 As shown by several recent papers on ozone production by lightning NOx, the strongest NOx enhancements are found in the upper troposphere around 10-12 km at midlatitudes (also shown by your figure 7) and around 12-14 km in the tropics. These are also the altitudes where the strongest ozone enhancements due to LNOx are also found. So why do you use the 300 hPa layer (< 9 km) to show the influence of lightning on ozone?

The level at which ozone production due to lightning is shown in Figure 10 (now Figure 11) has been shifted to a higher altitude (225 hPa, about 11 km). The text has also been updated appropriately.

page 8736 line 11 Would read better as: Near the high, PAN leads to the production of > 4.7%.....

Altered text.

page 8739 line 7-9 Here you say it's important to understand the contribution of lightning if one is to understand the Asian impact on N. America. But couldn't you also say the same thing about needing to understand the contributions from Europe, N. America or the stratosphere?
This work concentrated on achieving good estimates of East Asian anthropogenic emissions and lightning emissions because of rapid growth in the former and the high level of uncertainty in the latter. It is true, however, that other sources also require consideration to accurately gauge the impact of any one particular source. The text has been expanded to address the other sources as well.

Figure 9 How many years of OMI data are used? Are the OMI retrievals just for spring and just for 2006? Also, please draw boxes around each plot.

The retrievals used in this analysis are for spring 2006 only, coincident with the other observations and modeling efforts. Boxes have been added to Figure 9.

Figure 10 Please enlarge the panels by 50% and use a different color scale as the shading in the North Pacific is very faint.

Fixed.

Figure 11 These plots are difficult to see and I can’t really tell which way the vectors are pointing. Please double the size of each panel, sharpen the drawing of the vectors and make the map outline white.

Fixed. Split Figure 11 into two parts (now Figures 12 and 13) to accommodate the larger panels.

Figure 12 and 13 Please make the longitude values for the aircraft data match the values in the map plots. Also please specify that the aircraft data are actual measurements and not model data extracted along the flight tracks.

The timeseries plots focus on a brief portion of the flight track, which is identified by the corresponding coloured markers on the maps. The broader scope of the map plots is used to give context to the large-scale transport features. The text and captions have been altered to specify that measurements are depicted.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/10/C6973/2010/acpd-10-C6973-2010-supplement.pdf