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

Anonymous Referee #3

Received and published: 7 June 2010

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.

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

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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.

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.

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?

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.

page 8733 lines 27-29 Here you state that the greatest Asian contribution occurs be-

tween 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.

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

page 8721 line 7 Here you call for additional work to disentangle the various contri-

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butions 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.

Zhang L, Jacob DJ, Kopacz M, et al., Intercontinental source attribution of ozone pollution at western US sites using an adjoint method , GEOPHYSICAL RESEARCH LET-TERS Volume: 36 Article Number: L11810 Published: JUN 6 2009

Zhang L, Jacob DJ, Boersma KF, et al., 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, AT-MOSPHERIC CHEMISTRY AND PHYSICS Volume: 8 Issue: 20 Pages: 6117-6136 Published: 2008

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.

Minor comments:

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.

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?

page 8719 line 22 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. page 8719 line 24 "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"

page 8720 line 1 Two recent papers that make the case for Asian pollution being a concern for North America are:

Parrish et al., Increasing ozone in marine boundary layer inflow at the west coasts of North America and Europe, ATMOSPHERIC CHEMISTRY AND PHYSICS Volume: 9 Issue: 4 Pages: 1303-1323 Published: 2009

Cooper et al., Increasing springtime ozone mixing ratios in the free troposphere over western North America, Nature, 463, 344-348, 2010

page 8720 line 6 Here and throughout the paper the geographically correct term for the ocean of interest is the North Pacific Ocean

page 8720 line 15 drop the word "present"

page 8720 line 25 Heald et al. 2003 is a TRACE-P paper, not an ITCT paper.

page 8720 line 27 A WCB is an airstream within an extra-tropical cyclone and is not a circulation pattern.

page 8722 line 14 over several e-folding lifetimes.

page 8728 line 7 I assume this is a bottom-up inventory that is being discussed?

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

page 8731 line 20 24 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

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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

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

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

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?

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

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?

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.

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

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.

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.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 8717, 2010.

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