The impact of chemical lateral boundary conditions on regional forecasting of surface ozone during stratospheric intrusions

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Introduction

Surface ozone can have negative impacts on human health and on plant and animal life since it can oxidize biological tissue. Episodic high ozone concentrations are associated with acute respiratory health effects and are known to contribute to crop damage. Ozone also influences the oxidizing capacity of the atmosphere since it is the primary precursor of OH, and it is an important green-house gas (infrared absorber), especially in the upper troposphere.

The primary source of tropospheric ozone is the photochemical oxidation of surface pollutants during daylight. However, stratospheric ozone may also be transported into the troposphere during tropopause folding events, also known as stratospheric intrusions. Deep stratospheric intrusions are capable of directly influencing surface ozone by transporting stratospheric air quickly to the surface. More common shallow intrusions on the other hand can enhance mid-tropospheric ozone by producing streamers that have the chemical characteristics of stratospheric air (high ozone, low carbon monoxide, low relative humidity). In the mid-troposphere, ozone has a reasonably long lifetime (~ 2 weeks), and subsequent downward transport can also bring high ozone concentrations to the surface (e.g., Stohl et al., 2000).

Regional air quality forecasting models require that chemical concentrations be stipulated along the lateral boundaries, and that their definition be computationally simple. For the ECCC operational forecast model, GEM-MACH, lateral boundary conditions for chemical species are taken from a seasonal average. Here we discuss the problems with this approach and explore the possibility of improving surface ozone forecasts by testing ozone lateral boundary conditions from three other sources that allow for the evolution of the ozone across the tropopause along the lateral boundaries.

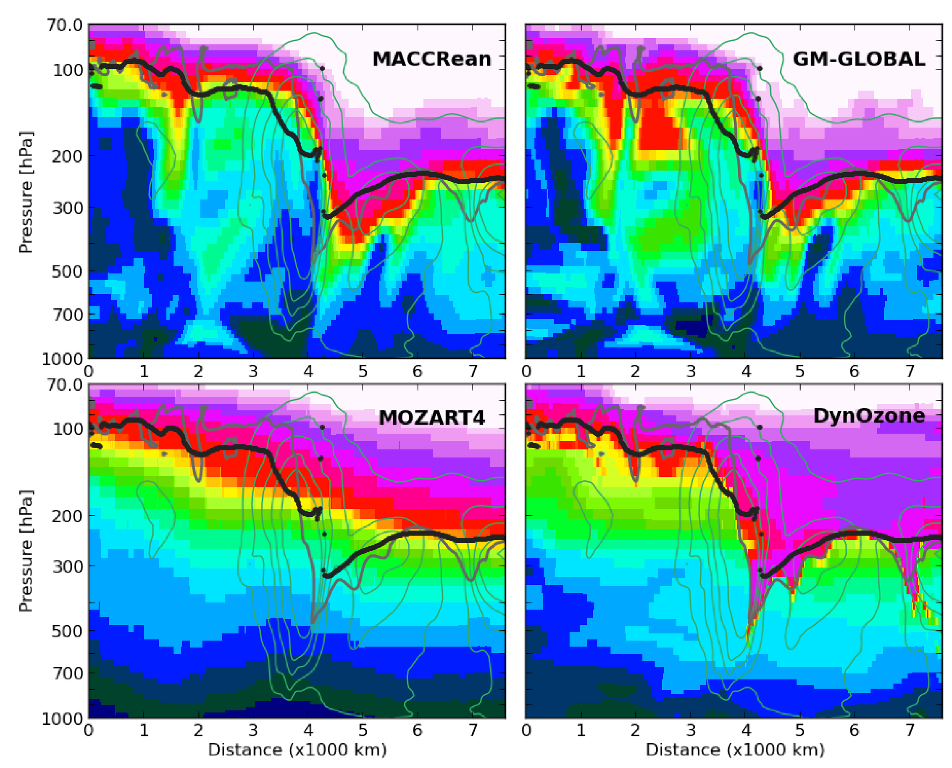

Figure 1: The ozone concentrations along the western boundary from GEM-MACH v2 on 2010-June-12 00:00GMT for MACC Reanalysis (top left), GEM-MACH-GLOBAL (top right), MOZART4 (bottom left) and DynOzone (bottom right). Also shown are the thermal tropopause (black dots) and the dynamical tropopause (gray; potential vorticity 2.0 PVU contour) from GEM-MACHv2, overlaid on all plots. The kinetic energy from GEM-MACH v2 for is shown as teal contours, indicating the location of the jet stream through the lateral boundaries. The western boundary runs from south (0 km) to north (7500 km).

Model Description and Experiments

GEM-MACH is a chemical transport model that is used as a limited-area model for operational AQ forecasting. It has a horizontal grid spacing of 10 km with 80 vertical levels. Chemical processes represented in the operational GEM-MACH include gas-phase, aqueous-phase and heterogeneous chemistry, and aerosol processes. Here, we perform a set of four experiments over the spring of 2010 (April, May, June) using four different ozone lateral boundary conditions (LBCs) :

a. Seasonally averaged ozone from MOZART4 (current operational)
b. MACC Reanalysis from ECMWF - a reanalysis product that assimilates temperatures along with ozone, CO and several other species)
c. GEM-MACH-GLOBAL - a global configuration of the GEM-MACH
d. DynOzone - a method that uses separate tropospheric and stratospheric ozone climatologies derived from ozonesonde data (Liu et al. 2013a; 2013b). The ozone values along the lateral boundary are chosen from each climatology based on the location of the dynamical tropopause.

The run using MOZART4 LBCs will be referred to as the control run; other runs will be referred to as the modified runs. For all other fields, the LBCs are unchanged between experiments. An example of the difference in the ozone along the western LB is shown in Figure 1. In all three modified run, ozone follows the dynamical tropopause better than in the control run.

References

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Liu, G., et al., (2013a). A global tropospheric ozone climatology from trajectory-mapped ozone soundings. *Atmos. Chem. Phys.,* **13**, 10659-10675, doi:10.5194/acp-13-10659-2013.
Liu, J. T., et al., (2013b). A global ozone climatology from ozone soundings via trajectory mapping: A stratospheric perspective. *ACP,* **13**, 11441-11464, doi:10.5194/acp-13-11441-2013.
Stohl, A., et al., (2000). The influence of stratospheric intrusions on alpine ozone concentrations. *Atmos. Env.,* **34**, 1323-1354.

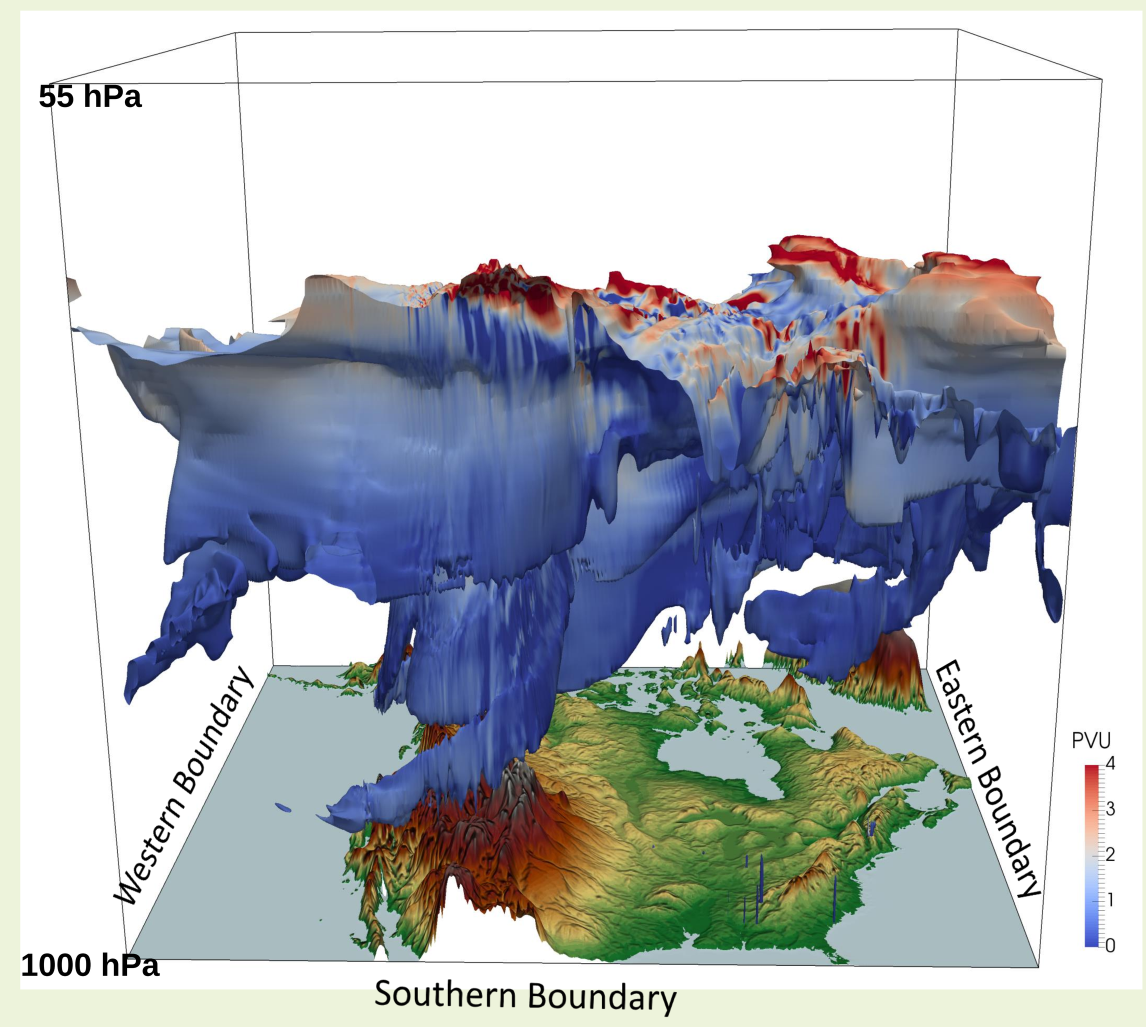

Figure 2: A 3-d rendering of the 100 ppbv isosurface from the run using ozone LBCs from the MACC Reanalysis for 13-June-2010 21:00 GMT, at the peak of the stratospheric intrusion. The isosurface is coloured by potential vorticity.

Stratospheric Intrusion Event

In spring 2010, 13 stratospheric intrusion events occurred over the period April 1 and June 30. The 7-15 June event was a deep intrusion that increased surface ozone by ~30 ppbv (Lin et al., 2012). Figure 2 shows a 100 ppbv isosurface for ozone that impacts the lower troposphere in the Rocky Mountains near the peak of the event on 13-June-2010 21:00GMT.

Figures 3 shows the impact on surface ozone concentrations at the height of the intrusion event for the three modified runs compared against the control run. In all three runs there is more ozone entering through the western lateral boundary at the surface, which affects the surface values over Alaska and western/northern Canada. The impact of the intrusion is seen over the western US, with the largest impacts seen from the DynOzone LBCs and the smallest impact from the GEM-MACH-GLOBAL LBCs.

Figure 4 shows a cross section of ozone along the dark green line indicated in Figure 3 for each run. Also plotted is the 2.0 PVU contour for potential vorticity, indicating the location of the dynamical tropopause. Between 116°W and 107°W the tropopause dips very low due to the passage of an upper-level trough. While all of the modified runs have the large gradient in ozone following the tropopause, the control run does not. This deficiency in ozone for the control can be traced back directly to the deficiency in ozone in the lower stratosphere along the western lateral boundary ~2 days prior.

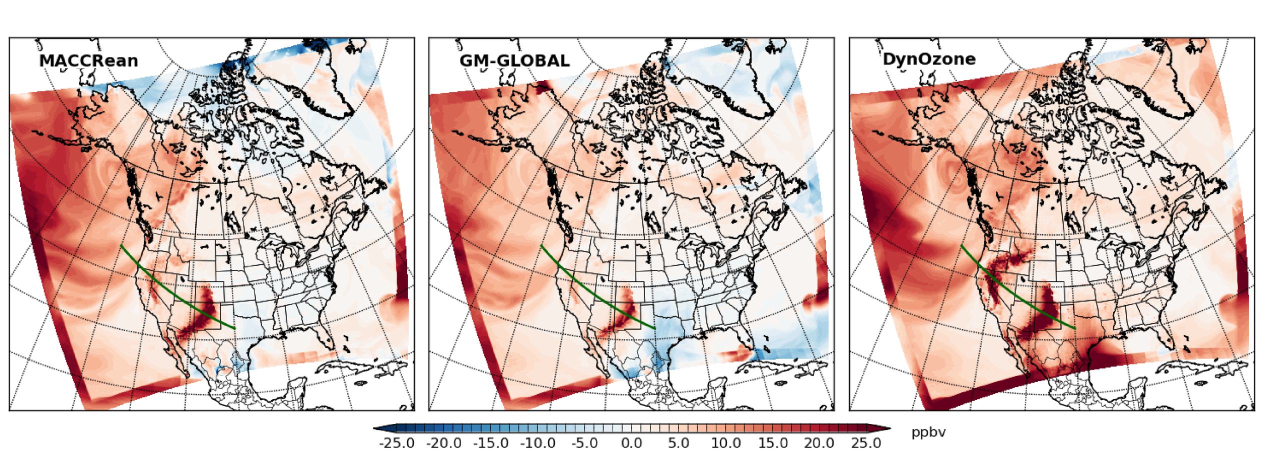

Figure 3: Differences in surface ozone for 13-June-2010 21:00GMT between the control run (MOZART4) and the three modified ozone LBC runs, at approximately the peak impact of the stratospheric intrusion. The dark green line denotes the location of the cross section shown in Figure 4.

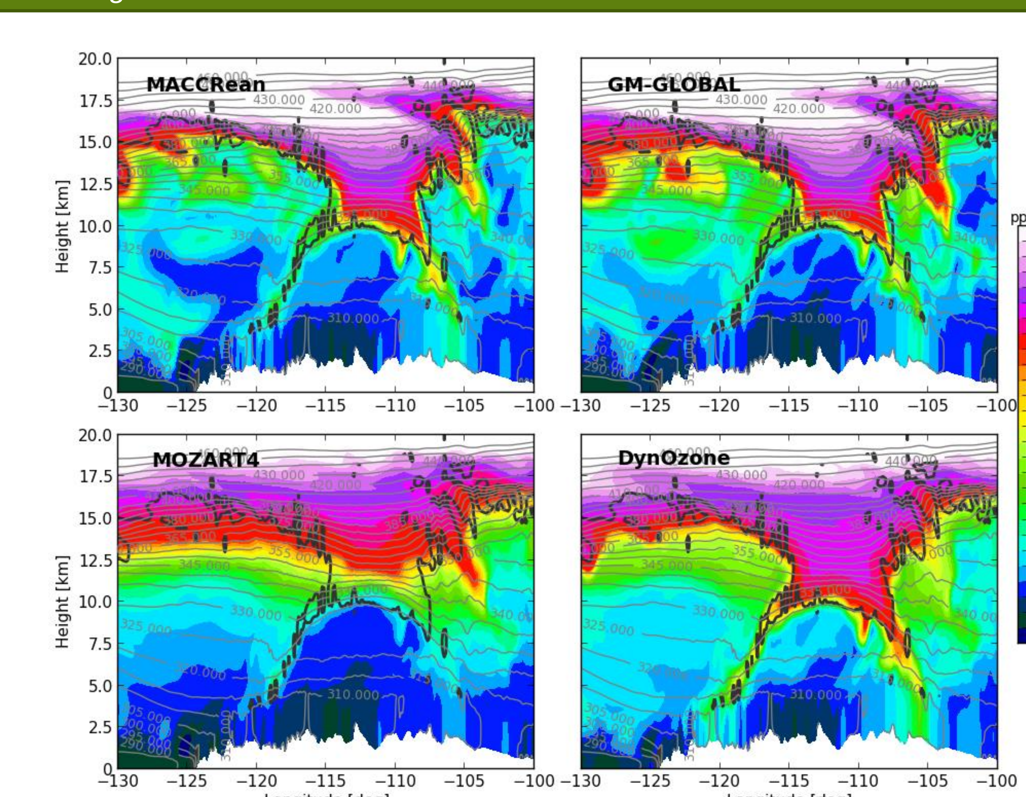

Figure 4: Cross section of ozone concentration (colour contours), and the 2.0 PVU surface (gray) for 13-June-2010 21:00GMT. The location of the cross section is shown in Figure 3.

Discussion and Conclusions

The surface ozone forecasts for the entire 3-month period were compared to ozone observations from the surface networks NAPS, AQS and CASTNET. The changes in absolute mean bias and correlation with the observations between the control run and the modified runs for each station are shown in Figure 5. Note that there is significant improvement in mean bias of ozone in all runs with modified LBCs over the region most affected by the stratospheric intrusions (western North America). Improvement is also seen in the mean bias over of the southern US for the MACC Reanalysis and GEM-MACH-GLOBAL boundary conditions due to improvement in the surface ozone through the southern lateral boundary from these data sets. For DynOzone, the mean bias is worse than the control run due to the high ozone concentrations in the tropospheric ozonesonde climatology.

Table 1 shows the statistics aggregated by region. The regions were chosen to examine the separate impacts of the different lateral boundaries and the impact in the region most affected by the stratospheric intrusions. For all modified ozone LBCs, improvement is seen in all stats in almost all regions compared to the control run. The most notable exception is the *Southern US*, which is affected directly by the surface ozone entering through the southern lateral boundary. Only GEM-MACH-GLOBAL improves all the metrics in this region over the 3-month period.

Overall, the modified runs show improvement in the selected statistics for surface ozone over the Western US and Canada, and to a smaller extent over the Central US and Canada. This improvement comes from the proper characterization of the ozone gradient across the tropopause along the western lateral boundary, which allows the ozone field in the model domain to evolve with the meteorology. Use of the MACC Reanalysis and GEM-MACH-GLOBAL also allows the lateral boundaries to reflect daily to inter-annual changes in ozone (e.g. trans-continental transport). However, the surface ozone concentrations of the used data sets can adversely affect the results where the incoming surface ozone can directly impact the results.

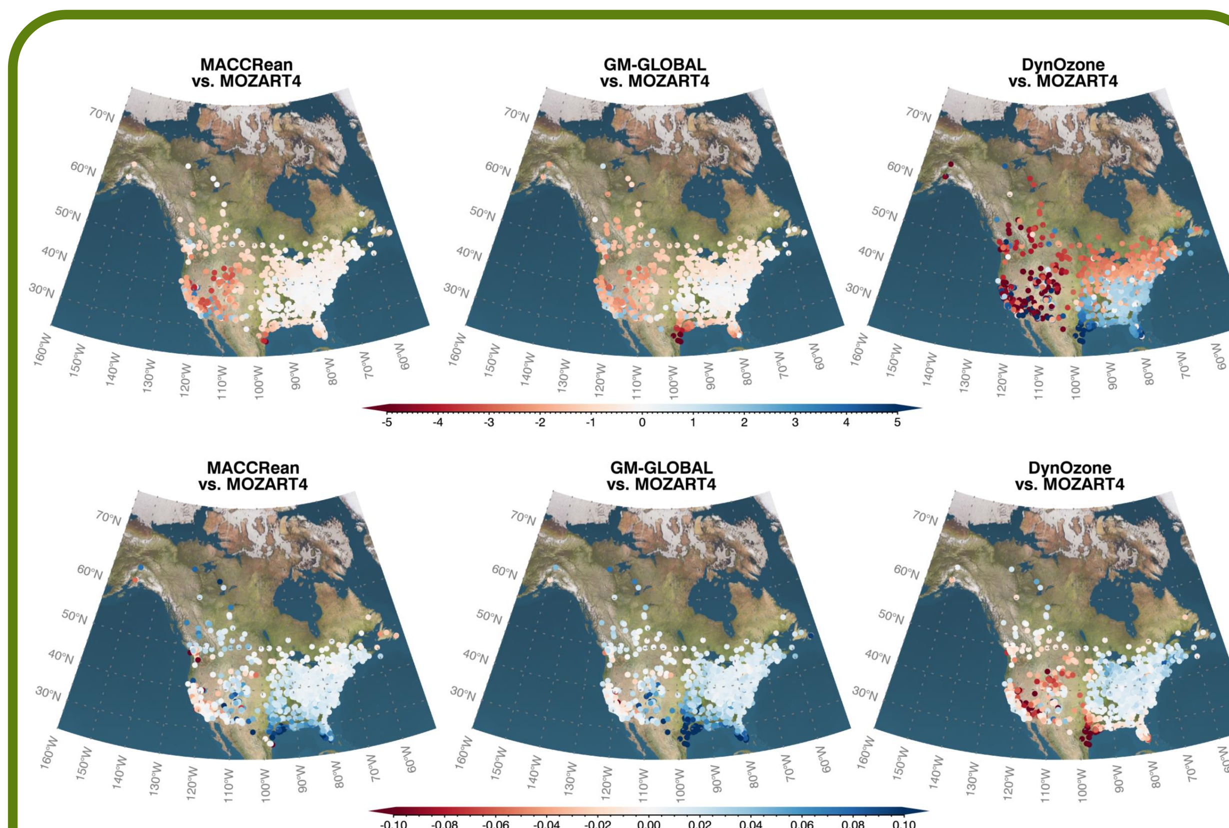

Figure 5: Spatial distribution of change in absolute mean bias (ppbv; top row) and correlation (bottom row) using MACC Reanalysis (left panels) vs MOZART4 boundary conditions, GEM-MACH-GLOBAL (centre panels) vs MOZART4, and DynOzone vs MOZART4 (right panels). Shown is the difference between the runs of the absolute value of the mean bias at each station over 3 months (April – June 2010). Blue (positive) means that the absolute value of the mean bias is smaller for MOZART4 boundary conditions, and red (negative) means that the modified run has a smaller mean bias. For the correlation changes, blue (positive) means that correlation is larger for the modified runs compared to MOZART4 lateral boundary conditions, and red (negative) means that the control run has larger correlation.

Table 1: Summary of statistics for April, May and June. Green shaded cells indicate the best run, lighter green cells indicate improvement over the control run (MOZART4) and red cells indicate that the modified runs is worse than the control run for that statistic and region.

	Region	MOZART4	MACC Reanalysis	GEM-MACH-GLOBAL	DynOzone
Mean bias:	Alaska & N. Canada	-1.82299	-0.87951	0.524220	4.45604
(ppbv)	Western NA	-6.51136	-4.26877	-5.03739	-1.02707
	E. Canada	-1.49661	-1.32525	-1.50278	0.596685
	Central US	-2.80774	-2.38825	-2.65021	-0.664292
	Southern US	4.66773	6.10807	4.49147	10.7723
	Northeast US	-4.76406	-4.35398	-4.53715	-3.10163
Correlation:	Alaska & N. Canada	0.633410	0.691490	0.694603	0.678929
(no units)	Western NA	0.615363	0.631296	0.622884	0.615734
	E. Canada	0.584721	0.582901	0.594382	0.600286
	Central US	0.589427	0.596501	0.599295	0.589863
	Southern US	0.660772	0.679729	0.714928	0.603204
	Northeast US	0.726970	0.729474	0.729764	0.739707
RMSE :	Alaska & N. Canada	7.34875	6.72061	6.62996	8.15134
(ppbv)	Western NA	14.4899	13.5403	13.8300	13.7474
	E. Canada	9.07909	9.03818	8.95502	8.89667
	Central US	13.5704	13.3995	13.3885	13.4170
	Southern US	12.6190	13.1099	12.1108	17.0732
	Northeast US	12.7574	12.5387	12.6052	11.8877
Slope of best	Alaska & N. Canada	0.427800	0.433324	0.488988	0.538360
fit line :	Western NA	0.572015	0.606088	0.585824	0.645207
(no units)	E. Canada	0.533387	0.526543	0.538509	0.561572
	Central US	0.593028	0.602210	0.603623	0.604794
	Southern US	0.688897	0.730730	0.800431	0.676738
	Northeast US	0.836579	0.837219	0.838318	0.842526