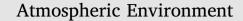
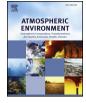
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Impact of chemical lateral boundary conditions in a regional air quality forecast model on surface ozone predictions during stratospheric intrusions



Diane Pendlebury^{a,*}, Sylvie Gravel^b, Michael D. Moran^a, Alexandru Lupu^a

Environment and Climate Change Canada, 4905 Dufferin St., Toronto, M3H 5T4, Ontario, Canada

^b Environment and Climate Change Canada, 2121 Trans Canada Highway, Dorval, H9P 1J3, Quebec, Canada

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ABSTRACT

A regional air quality forecast model, GEM-MACH, is used to examine the conditions under which a limited-area air quality model can accurately forecast near-surface ozone concentrations during stratospheric intrusions. Periods in 2010 and 2014 with known stratospheric intrusions over North America were modelled using four different ozone lateral boundary conditions obtained from a seasonal climatology, a dynamically-interpolated monthly climatology, global air quality forecasts, and global air quality reanalyses. It is shown that the mean bias and correlation in surface ozone over the course of a season can be improved by using time-varying ozone lateral boundary conditions, particularly through the correct assignment of stratospheric vs. tropospheric ozone along the western lateral boundary (for North America). Part of the improvement in surface ozone forecasts results from improvements in the characterization of near-surface ozone along the lateral boundaries that then directly impact surface locations near the boundaries. However, there is an additional benefit from the correct characterization of the location of the tropopause along the western lateral boundary such that the model can correctly simulate stratospheric intrusions and their associated exchange of ozone from stratosphere to troposphere. Over a three-month period in spring 2010, the mean bias was seen to improve by as much as 5 ppbv and the correlation by 0.1 depending on location, and on the form of the chemical lateral boundary condition.

1. Introduction

The primary source of tropospheric ozone is in situ photochemical oxidation of tropospheric precursors such as carbon monoxide (CO) and hydrocarbons in the presence of nitrogen oxides (NO and NO₂) during daylight (Crutzen and Gidel, 1983). However, stratospheric ozone may also be transported into the troposphere during tropopause folding events, also known as stratospheric intrusions, which are the primary mechanism for stratosphere-troposphere exchange (STE) at mid-latitudes. Deep stratospheric intrusions are capable of directly influencing surface ozone concentrations by transporting stratospheric air quickly to the surface (e.g., Lin, et al., 2012). Shallow intrusions (i.e., those for which the stratospheric air does not reach the surface within 48 h) can enhance tropospheric ozone by producing potential vorticity streamers in the mid-troposphere (e.g., Appenzeller and Davies, 1992; Sprenger et al., 2007; Kunz et al., 2015) that have the chemical characteristics of stratospheric air; high ozone and low carbon monoxide concentrations, and low relative humidity. At these altitudes ozone has a reasonably long lifetime (~2 weeks), and subsequent down-welling can bring high ozone concentrations to the surface (Stohl et al., 2000).

Stratospheric intrusions are episodic in nature, with deep exchanges occurring most frequently in the winter (James et al., 2003) due to the strong jet stream in that season and the resulting propensity for baroclinic wave activity. They are largely confined to mid latitudes in the vicinity of the storm tracks (Wernli and Bourqui, 2002; Skerlak et al., 2014). Transport across the tropopause is the result of non-conservative processes (e.g., radiative cooling, turbulence, gravity waves), and is followed by almost adiabatic descent along downward sloping isentropes and subsequent quasi-horizontal dispersion in the lower troposphere (Bourqui and Trépanier, 2010). The downward slope of the isentropes is due to a negative potential temperature anomaly at the tropopause combined with a baroclinic zone downstream of the intrusion at the surface.

While the mass exchange between the stratosphere and troposphere peaks in the winter, ozone levels in the lower stratosphere relative to the tropopause are greatest in early spring to summer (Wang et al., 2006), and so the peak effect of stratospheric ozone on the surface occurs in spring. There are numerous case studies focused on the western United States since the morphology of the jet stream combined with the high surface elevation and downstream subsidence makes this

Corresponding author. E-mail addresses: diane.pendlebury@canada.ca (D. Pendlebury), sylvie.gravel@canada.ca (S. Gravel), mike.moran@canada.ca (M.D. Moran), alexandru.lupu@canada.ca (A. Lupu).

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a likely place for deep stratospheric intrusions (Skerlak et al., 2014). Langford et al. (2012) found that 13% of the variance of daily maximum 8-h average ozone over Los Angeles during the CalNex field study in spring 2010 was due to the passage of upper-level troughs that were coincident with stratospheric intrusions. For the same period, Lin et al. (2012) found that at high-elevation sites in the western U.S. the stratospheric contribution to surface ozone was between 15 and 25 ppbv, indicating that stratospheric intrusions can play a major role in contributing to high surface ozone mixing ratios. Langford et al. (2012) showed that this impact can be higher than 30 ppbv, with long-range transport from Asia contributing < 10 ppbv in their results. Lefohn et al. (2011) noted that the high-elevation site at Yellowstone National Park in Wyoming exhibited the most stratospheric enhancements during the spring and summer, although lower elevation sites across the western and northern U.S. also showed stratospheric enhancement of surface ozone, especially during spring. Mountainous regions are more susceptible to stratospheric influence on ozone for two main reasons: 1) they are higher in elevation so that stratospheric air can reach the surface more directly and is therefore less diluted or mixed with tropospheric air, and 2) the mountainous topography itself is responsible for the dynamical conditions that contribute to stratospheric intrusions (Stohl et al., 2000).

Although there has been much attention focused on the western U.S. during the spring, it should be noted that stratospheric intrusions can occur over other regions of North America, for example over southern Ontario in eastern Canada during the winter (Dempsey, 2014) and summer (Bourqui and Trépanier, 2010; He et al., 2011; Bourqui et al., 2012). Lefohn et al. (2011) showed that there is substantial year-to-year variability in the impact of stratospheric intrusions, and although there is greater impact at high-elevation sites during spring, STE events are coincident with enhanced surface ozone concentrations for all months (lowest in autumn) and for all regions and elevations. In addition, Langford et al. (2015) have suggested that there is increased fire danger in California as a result of strengthened Santa Ana winds with the passage of upper level troughs and the associated strong northerly flow on the western flank of very deep troughs. Their study focused on a La Niña year with a polar jet that was located southward of the climatological mean, leading to more frequent deep events (see Lin et al., 2015). Hence, deep stratospheric intrusions may contribute to enhanced lower-tropospheric ozone directly by downward transport of ozone from the stratosphere, and indirectly by in situ ozone production in the troposphere due to enhanced occurrence of wildfires.

For operational air quality forecasting, Environment and Climate Change Canada (ECCC) uses the GEM-MACH chemical weather model, which consists of GEM (Global Environmental Multiscale model), ECCC's multiscale operational weather forecast model, and MACH (Modelling Air quality and CHemistry), an on-line chemical transport module (CTM) that is embedded within GEM (Gong et al., 2015; Pavlovic et al., 2016). The GEM model was used by Bourqui and Trépanier (2010) to investigate some of the dynamical aspects of stratospheric intrusions, namely dispersion and mixing with the surrounding tropospheric air, and necessary conditions for stratospheric air reaching the lower troposphere. They evaluated the accuracy of the GEM simulations first by comparing the evolution of potential vorticity and specific humidity on the 330K isentrope with the corresponding analyses, and second by comparing model vertical profiles of ozone with balloon soundings. Their study suggested that GEM captures well the dynamics of these events. He et al. (2011) examined stratospheric intrusions over southern Ontario during the BAQS-Met field campaign in June and July of 2007 and found that GEM, using FLEXPART (FLEXible PARTicle dispersion model) for trajectory analysis and AURAMS (A Unified Regional Air-quality Modelling System), an off-line CTM, well represented the meteorology of these sporadic events, although they noted that too much ozone was brought down during the intrusions and attributed the excess transport to poor vertical resolution. Barré et al. (2012) used assimilation of MLS (Microwave Limb Sounder) data into the MOCAGE (Modélisation de la Chimie Atmosphérique Grande Echelle) CTM using two horizontal resolutions: 2° and 0.2° . They found that at the higher resolution, filamentary structures associated with the STE events were well resolved and realistic despite a low aspect ratio between horizontal and vertical grid spacing. The regional GEM-MACH uses a horizontal grid-spacing of 0.09° and should therefore be capable of simulating such events.

For limited area CTMs, such as the GEM-MACH model, chemical lateral boundary conditions (CLBCs) have been shown to have a significant impact on predictions for longer-lived species such as ozone and carbon monoxide (Tang et al., 2007). Time-invariant CLBCs have been found to lead to biases in the predicted values. Using monthlyaveraged CLBCs instead of time-invariant CLBCs can improve the representation of the annual cycle but cannot capture synoptic or diurnal variations (Akritidis et al., 2013). The impact of CLBCs on surface ozone mixing ratio has a seasonal dependence; winters show more sensitivity with a low bias due to low ozone from the CLBCs (Solazzo et al., 2013). However, during summer, various near-surface processes (e.g. photochemistry, transport, emissions, and dry deposition) are more important in controlling ozone mixing ratios, thus dampening the impact from the CLBCs (see also Im et al., 2015). The same is true for both European and North American regional models, although actual results have been found to be highly dependent on the meteorological and chemical configurations, even within the same modelling system. Borge et al. (2010) found that ozone is very sensitive to the CLBCs over the Iberian Peninsula and that results improve if a larger domain is used to initialize CLBCs for the smaller domain, although the exact results depend on the season.

Simulations with regional CTMs are also improved if ozonesondes or satellite data are used to define the CLBCs. Pour-Biazar et al. (2011) found improvement in the simulation of ozone in the free troposphere with the use of OMI satellite data to define time-varying CLBCs, which helps in cases of transboundary transport of pollution and recirculation from the northeast to southeast U.S., compared to constant CLBCs. Tang et al. (2009) also found improvement in the prediction of ozone in the upper troposphere when using ozonesonde profile data sets, although the impact depended on the proximity of the sonde to the boundary, and most of the improvement was seen near the inflow boundaries. However, Andresson et al. (2015) found in their study that the use of time-varying CLBCs from a global model, as opposed to constant CLBCs based on observations, improved spatial correlations at 500 hPa and elsewhere in the free troposphere, but that the ground concentrations were only affected close to the model boundary even for relatively longlived species, leading them to the conclusion that the impact of CLBCs on ground concentrations may have been over-estimated in other studies.

Many studies have used time-varying CLBCs from other realistic sources such as the MACC (Monitoring Atmospheric Composition and Climate) Reanalysis or GEOS-Chem (e.g., Giordano et al., 2015; Im et al., 2015; Schere et al., 2012). However, even using the MACC Reanalysis with a 3-h temporal resolution, Wang et al. (2015) found that tropospheric ozone was over-predicted due to overestimates in the ozone profiles from the CLBCs. Kerkweg and Jöckel (2012) found that results could be improved when the model used to define the CLBCs and the regional AO model were consistent (that is, when all meteorological, physical and chemical processes had the same representation in both models). In addition, Pfister et al. (2011) noted that recirculation of local pollution produced within the regional model domain can impact the lateral boundaries, which emphasizes the need for consistency between the global model simulations used for the CLBCs and the regional model. The best results have been obtained using nested two-way coupled models. Yan et al. (2014, 2016) used a two-way coupled system for carbon monoxide and ozone such that the regional model took its CLBCs from the global model but also fed back information to the global model. This configuration allows the regional model to be consistent with the global model along the lateral

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