

**Transport analysis
during BAQS-Met**

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This discussion paper is/has been under review for the journal Atmospheric Chemistry and Physics (ACP). Please refer to the corresponding final paper in ACP if available.

Transport analysis of ozone enhancement in Southern Ontario during BAQS-Met

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Received: 5 May 2010 – Accepted: 31 May 2010 – Published: 24 June 2010

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Published by Copernicus Publications on behalf of the European Geosciences Union.

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Abstract

Twice-daily ozonesondes were launched from Harrow, Ontario (east of Detroit) during the BAQS-Met (Border Air Quality and Meteorology Study) campaign in the summer of 2007. A co-located radar windprofiler measured tropopause height continuously.

Simulation results from the dispersion model FLEXPART, using the output of the Environment Canada Global Environmental Multiscale (GEM) weather forecast model, indicate the occurrence of stratospheric ozone intrusion events during the BAQS-Met campaign. This interpretation is supported by the ozonesonde observations, one-minute average surface ozone data measured by the chemistry supersite at Harrow, and geostatistical interpolation results of satellite ozone data that were observed from TES (Tropospheric Emission Spectrometer) onboard NASA's Aura satellite and AIRS (Atmospheric Infrared Sounder) onboard NASA's Aqua satellite. Source-receptor analysis using the GEM-FLEXPART model shows the stratosphere over the Northwest Territories region close to the Beaufort Sea to be the main source of the enhanced ozone at Harrow on 1 July 2007.

1 Introduction

Stratosphere-troposphere exchange (STE) of mass and chemical species across the tropopause is an ongoing process at a variety of scales: mesoscale, synoptic-scale and global-scale (Holton et al., 1995). Although the global flux of ozone from the stratosphere is controlled non-locally, by the global circulation, it is episodic, being associated with tropospheric weather systems, and varies both spatially and with season. STE processes play an important role in the distribution of ozone in both the stratosphere and the troposphere. Most stratosphere-troposphere exchange occurs at midlatitudes, mainly in association with tropopause folds and cutoff lows (Singh et al., 1980; Price and Vaughan, 1993; Ancellet et al., 1994; Beekmann et al., 1997; Sprenger et al., 2007). Other processes include convectively breaking gravity waves,

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deep convection, quasi-isentropic transport, radiative heating, and turbulent mixing. Because tropopause folds and cut-off lows are the principal mechanisms of STE, abrupt variations of tropopause height are usually observed with stratospheric ozone intrusions (Hocking et al., 2007).

5 Although tropopause folds rarely reach lower than 5 km a.s.l. at mid-latitudes, deep stratospheric ozone intrusions down into the boundary layer have been observed in many studies (Davies and Schuepbach, 1994; Elbern et al., 1998; Wakamatsu et al., 1989; Zanis et al., 2003a; Cooper et al., 2005; Bourqui and Trepanier, 2010).

10 The stratosphere is believed to be a significant source of ozone in the troposphere, e.g. Stevenson et al. (2006). Recent interest in STE events has focussed on their importance to the tropospheric ozone budget, which depends on their distribution with geography and season, and on the fate of the exchanged air parcels, particularly their vertical penetration and residence time (Collins et al., 2003; Stohl et al., 2003; Cooper et al., 2004; Thompson et al., 2007; Tarasick and Slater, 2008).

15 Balloon-born ozonesondes with fine vertical resolution (~ 100 m) and with moderate temporal resolution (e.g. twice a day) during campaigns have been used to study STE at specific locations and times (Oltmans et al., 1989; Wakamatsu et al., 1989; Cooper et al., 1998). The primary challenge with using ozonesondes for studying stratospheric ozone intrusions is the limited regional coverage, i.e., a relatively small number of mea-
20 surements at different locations during specific campaigns.

Satellite observations, in contrast, have limited vertical resolution but offer the potential of long-term and global coverage. While nadir-viewing satellite observations have limited vertical resolution, they offer wide geographic coverage that can complement detailed profile information from ozonesondes. Over the last few years, observations
25 from TES (Tropospheric Emission Spectrometer) onboard NASA's Aura satellite, AIRS (Atmospheric Infrared Sounder) onboard NASA's Aqua satellite, GOME (Global Ozone Monitoring Experiment) onboard the European Remote Sensing (ERS-2) satellite, OMI (Ozone Monitoring Instrument) onboard the Aura satellite, and HIRDLS (High Resolution Dynamics Limb Sounder) onboard the Aura satellite, have provided a global

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continuous data set for atmospheric ozone. In recent years, satellite observations have been used in an attempt to provide a more complete three-dimensional view of stratosphere-troposphere exchange events (Pan et al., 2006, 2007a, b; Kollonige et al., 2007; Parrington et al., 2008).

5 The Lagrangian particle dispersion model FLEXPART (Bonasoni et al., 1999, 2000; Stohl et al., 2002, 2003a, b, c, 2005; Cristofanelli et al., 2003; James et al., 2003a, b; Zanis et al., 2003b; Meloen et al., 2003; Cooper et al., 2004, 2005; Hocking et al., 2007; Carey-Smith et al., 2010) has been used extensively to model stratospheric intrusions.

10 STE events were studied using radar windprofilers, balloon-borne ozonesondes, supplemented by simulation results from FLEXPART, in several ozone campaigns in 2005, 2006 and 2007 in southeastern Canada (Hocking et al., 2007; Carey-Smith et al., 2010). Excellent qualitative agreement between experimental measurements and model results were observed, although quantitative agreement was not ideal.

15 This study presents observations of stratospheric ozone intrusions during a summer campaign, the Border Air Quality and Meteorology Study (BAQS-Met) in 2007, in Southern Ontario, Canada. Section 2 introduces the BAQS-Met campaign and describes measurements using a radar windprofiler, ozonesondes, and in-situ ozone monitors at Harrow, and presents satellite ozone measurements by TES and AIRS. Section 3 describes the Kriging interpolation of the satellite data and the FLEXPART
20 simulation. Section 4 presents the analysis of ozone profile data, radar tropopause heights, surface ozone data, and satellite ozone observations. This section also shows the Kriging interpolation results using TES and AIRS, and compares the observations with simulation results from FLEXPART and from the AURAMS (A Unified Regional Air-quality Modeling System) chemistry transport model, both driven by the Canadian
25 operational weather forecast Global Environmental Multiscale (GEM) model. Conclusions and discussion are presented in Sect. 5.

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2 Data

2.1 BAQS-Met campaign

The Border Air Quality and Meteorology Study (BAQS-Met) during the summer of 2007 was a measurement-intensive field campaign, which examined the impact of lake-breeze meteorological conditions on air pollution in the region between Lake Erie and St. Clair, to address the need for improved understanding of the air quality and meteorology along the US-Canada border region.

During the BAQS-Met campaign, from 20 June to 7 July 2007 (except 1 and 2 July), ozonesondes were launched about every 8–12 h at Harrow (42.03° N, 82.92° W), Ontario (20 launches in total). The balloon-borne instruments used in the study were GPS-equipped EN-SCI model 2Z-ECC ozonesondes and Vaisala RS80 radiosondes. They provide vertical profiles of ozone concentration, temperature, humidity, wind speed and wind direction from the surface to the stratosphere, with vertical resolution of ~100 m.

The WindTracker (Wind-Turbulence tracker) windprofiler radar, which provided real-time wind information, as well as backscattered power, turbulence strength and tropopause heights, was located close to the ozonesonde site. The radar-derived tropopause height is defined as the height in the altitude region between 6 and 14 km where the secondary maximum value of backscattered power occurs in the troposphere. It shows good agreement with the thermal tropopause (Gage et al., 1986; Hocking, 1997; Hooper and Arvelius, 2000; Vaughan et al., 1995). One-minute averages of surface ozone at the nearby chemistry “supersite” are also available through the BAQS-Met campaign.

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2.2 Satellite data

2.2.1 TES data

The Tropospheric Emission Spectrometer (TES), on the NASA's Earth Observing System Aura satellite, launched in late September 2004 (Beer et al., 2001; Beer, 2006; Schoeberl et al., 2006), is an infrared, high-resolution Fourier transform spectrometer which provides vertical profiles of tropospheric ozone, and other atmospheric constituents such as carbon monoxide. TES constituent's vertical profiles are retrieved by an optimal method following Rodgers (2000); Osterman (2004); Worden (2004); and Kulawik et al. (2006). This study uses Version 3 (V003) of the TES Level 2 data products, whose observation footprint is 5 km by 8 km. Version 3 of TES data represents significant improvements in nadir ozone data quality over version 2. Initial validation studies have shown that TES ozone profiles show a 5–15% high bias in the troposphere (for TES Version 2) compared to ozonesondes (Nassar et al., 2008), and more recent work (Boxe et al., 2009) shows similar biases for TES Version 3. The data used here were obtained from 16-orbit global surveys from 20 June to 07 July 2007 and are available from the NASA Langley Atmospheric Science Data Center (ASDC): <http://eosweb.larc.nasa.gov/HPDOCS/datapool>.

2.2.2 AIRS data

The Atmospheric Infrared Sounder (AIRS), on the NASA Aqua satellite, provides profiles of high-resolution atmospheric temperature, water vapor, and a number of trace gas species including ozone, using a nadir cross-track scanning infrared instrument with a 15 km field of view (Fetzer et al., 2006). This study will use Version 5 (V005) of the AIRS standard level 2 ozone products (Olsen et al., 2007), given in 45 km pixels and 28 vertical layers. The data used here are available from the Goddard Earth Sciences (GES) Data Information and Services Center. Previous validation studies have shown that these data have a consistent high positive bias in the upper troposphere

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and a low bias in the lower stratosphere, compared to ozonesondes (Bian et al., 2007; Monahan et al., 2007; Divakarla et al., 2007). The AIRS ozone profiles of version 5 have ozonesonde climatology as the first guess, so improved tropospheric ozone directly from first guess, even the instrument does not have much sensitivity (McPeters et al., 2007; Olsen et al., 2007a). AIRS version 5 products overestimate upper tropospheric (UT) ozone and underestimate lower stratosphere (LS) ozone, at mid and high latitudes (Olsen et al., 2007b; Pan et al., 2007b). Compared with the ozonesonde profiles, AIRS ozone profiles have a tendency of larger positive bias than TES ozone profiles in the troposphere. However, AIRS ozone profiles have a finer latitude-longitude resolution than TES ones, which make the AIRS ozone and water vapor profiles good data sets to diagnose stratospheric ozone intrusions.

3 Methods

3.1 Kriging interpolation of satellite ozone data

Satellite observations are of particular interest to this study when no ozonesonde data are available. Since satellite data can not provide observations at specific positions on consecutive days, a geostatistical interpolation method is used to obtain approximate observations at that location.

Kriging is a popular linear least square interpolation method widely used in mining, mapping, hydrogeology, and environmental science, e.g., Bayraktar and Sezer (2005), as well as remote sensing (Stein et al., 2002; Tranchant and Vincent, 2000). Kriging considers the observations as a realization of a random spatial process. Ordinary Kriging (OK) allows one to construct an unbiased estimator that does not require the quantity of interest have a time-invariant probability distribution. The ordinary Kriging interpolation, $\hat{y}(x_0)$, at a unobserved point x_0 , is a weighted linear combination of all observations $y(x_n)$, $n = 1, \dots, N$, i.e.,

$$\hat{y}(x_0) = \sum_{n=1}^M \lambda_n(x_0) y(x_n), \quad (1)$$

with $\sum_{n=1}^N \lambda_n(x_0) = 1$. The weighting coefficients $\lambda_n(x_0)$ for each interpolated location x_0 is determined from the semi-variogram equations,

$$\sum_{n=1}^N \lambda_n(x_0) \gamma(x_n, x_m) + \mu = \gamma(x_m, x_0), \quad m = 1, \dots, N, \quad (2)$$

5 based on the minimum estimated variance criterion. μ is the Lagrangian coefficient, and the semi-variance $\gamma(x_n, x_m)$ is a function of the separation distances between observations, e.g., $\gamma(x_n, x_m) = f(0.5 \|x_n - x_m\|^2)$ where $\|x_n - x_m\|^2$ is the square of the Euclidean distance between two locations x_n and x_m . The experimental semi-variogram is estimated from scattered locations and observations, and the model $f(\cdot)$ semi-variogram used in Eq. (2) is a mathematical model, usually a Gaussian, spherical, or
 10 exponential function, which fits the trend in the experimental variogram. In this paper, an exponential function $f(h) = a + b \cdot (1 - \exp(-c \cdot h))$ is used, considering ozone's spatial variation.

3.2 GEM-FLEXPART model study

15 Stratospheric ozone intrusions can be modeled using a Lagrangian particle dispersion model FLEXPART, which does not include any chemistry (Stohl et al., 2005). FLEXPART has been extensively validated (Stohl et al., 1998, 2002; Forster et al., 2004). FLEXPART parameterizes convections using the scheme presented in (Emanuel and Zivkovic-Rothman, 1999; Seibert et al., 2001) and parameterizes turbulence in the free
 20 troposphere and boundary layer by solving Langevin equations (Stohl and Thomson, 1999). FLEXPART uses a domain filling procedure where a large number of particles are released within the stratosphere of the model domain at the beginning of a model run. The dynamic tropopause, defined using isentropic potential vorticity (PV), is used

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in FLEXPART to locate the surface that separates the stratospheric and tropospheric air masses. Initially particles that are in the stratosphere, with PV value greater than 2 pvu ($1 \text{ pvu} = 10^{-6} \text{ km}^2 \text{ kg}^{-1} \text{ s}^{-1}$), e.g., Fischer et al. (2000) and Zahn and Brennike-meijer (2003), are given an ozone concentration calculated using potential vorticity and the relation $\text{O}_3 = S \times \text{PV}$, where S is a constant which varies seasonally (between 51 and 60 ppb between June and July) (Stohl et al., 2000). New particles are created and initialized in the same way on inflow boundaries. Values used for S were 60 and 51 ppb pvu⁻¹ for June and July, respectively, following Stohl et al. (2000).

Wind field data from the Canadian operational weather forecast Global Environment Multiscale (GEM) model, (Côté et al., 1998), are used as the input to FLEXPART to advect parcels. The standard regional version 3.2.2 of GEM (58 vertical levels to 10 hPa) was run at 0.1375 by 0.1375 degree resolution on a domain covering North America, and produced hourly output fields which were interpolated onto a 0.5×0.5 degree resolution latitude-longitude grid.

4 Observations

Figure 1 shows ozone volume mixing ratios in parts per billion (ppb) as a function of height and time, from all the ozonesonde flights (upper panel), and the one-minute averages of surface ozone volume mixing ratios from the chemistry supersite (bottom panel), at Harrow (42.03° N, 82.92° W), Ontario, during the BAQS-Met campaign. All observations and model simulation results are displayed in Greenwich Meridian Time (GMT). Each ozonesonde launch is represented by a vertical column of coloured boxes, with the time rounded to the nearest half day. Radar tropopauses (at about 9.5–13.5 km) are shown by the red solid line in the upper plot. A rapid ascent in tropopause height on 20 June suggests the possibility of an ozone intrusion from the stratosphere (Hocking et al., 2007). The enhancement of ozone (typically 80 ppb) appears down to an altitude of 4–5 km from 21 June to 23 June. Low relative humidity in these layers confirms the stratospheric origin. However, the irreversible character of the

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intrusion can not be confirmed by these observations. Numerical modeling with GEM-FLEXPART is used below to support this identification.

Figure 2 shows the occurrence of all ozone intrusions during the BAQS-Met campaign, using the illustration method of Hocking et al. (2007). Although a level 3 (see definition in the caption of Fig. 2) radar tropopause excursion rate on 20 June is clearly associated with the occurrence of an ozone intrusion which was detected from 21 June to 23 June, consistent with the conclusions of Hocking et al. (2007), two ozone intrusions (on 26–27 and 30 June) are not associated with level 2 or 3 radar tropopause excursion rates. Unfortunately, ozonesonde data are not available on 1–2 July, although high surface ozone concentration is observed on 1 July. High surface ozone is also observed approximately coincident with the ozone enhancement observed in ozonesonde measurements on 7 July. High relative humidity (about 80%) suggests that this last ozone enhancement was not the result of a stratospheric intrusion. However, as will be shown, GEM-FLEXPART simulation results support a stratospheric origin for this enhancement.

The upper panel of Fig. 3 shows vertical profiles of TES ozone mixing ratio (ppb) along a global survey track (shown in the lower panel), on 1 July of 2007. Close to the Harrow station (shown by a black cross), ozone enhancement in the lower troposphere is observed. The TES profile also appears to show high ozone in the boundary layer, consistent with the surface observations in Fig. 1. The upper plot also shows what appears to be a large stratospheric intrusion in the upper troposphere, down to about 5 km, to the north and east of Harrow. On 2 July, there is no TES global survey data close to the Harrow station.

The upper plots in Fig. 4 display the AIRS ozone measured at 500 hPa, which is approximately 5 km a.s.l., on 1–2 July. An air mass rich in ozone (~100 ppb) was located initially to the northeast of Harrow (as seen in the TES profiles of Fig. 3), moved eastward. Low water vapor values coincident with the high ozone concentrations are evidence of their stratospheric origin.

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Figure 5 shows the average values of Kriging interpolations of TES and AIRS ozone data at Harrow. No correction has been made for the known high bias of the satellite instruments, because the comparison between Kriging interpolations and ozonesonde measurements is only in a qualitative way. Since ozone observations in the troposphere have little correlation if their horizontal distance is greater than 600–1000 km (Liu et al., 2009), only TES and AIRS ozone profiles with horizontal distance with the Harrow station less than 1000 km are selected for the interpolation. At an altitude of about 5 km on 20 and 21 June, high ozone concentration values (about 80 ppb) can be observed in the satellite data. Another significant enhancement of ozone (typically 80 ppb) reaches down to an altitude of about 6 km on 7 July 2007, in agreement with the ozonesonde observations in Fig. 1. High ozone concentrations (approximately 80–100 ppb) are also observed down to the ground on 1 and 6 July, when the surface record (Fig. 1) also shows high ozone.

Figure 6 shows ozone mixing ratios from a GEM-FLEXPART simulation at Harrow, Ontario, from 20 June to 7 July 2007. These values were extracted from the FLEXPART output grid at Harrow. It should be noted that the color scale is different from that of the ozonesonde observations in Fig. 1, since the FLEXPART model has been set up to simulate the transport and dispersion of a stratospheric ozone tracer and has no tropospheric background ozone. The high ozone in the upper troposphere on 20 June in the ozonesonde profiles is reflected in the GEM-FLEXPART simulation, confirming that it is of stratospheric origin. Figure 7, showing the 80 ppb ozone mixing ratio iso-surface at 09:00 UT on 20 June (left-hand plot), clearly displays the large scale of this intrusion. Another significant stratospheric ozone intrusion on 1 July, penetrating below 5 km altitude is indicated in Fig. 6. Unlike the case on 20 June, high ozone concentration is not observed in the upper troposphere on 1 July. The right-hand plot of Fig. 7 clearly indicates a deep influx of ozone from the stratosphere, resulting in a tongue of ozone penetrating to 5 km altitude over Harrow. Unfortunately there are no ozonesonde data for 1 July or 2 July, but this intrusion is clearly seen in the satellite data (Figs. 3, 4 and 5). Figure 6 also shows frequent ozone enhancements in the upper

troposphere down to ~8 km between 28 June and 7 July, that agree qualitatively well with the TES and AIRS composite plot in Fig. 5.

Figure 8 shows ozone mixing ratio (ppb) from a GEM-FLEXPART simulation at 5.5 km altitude over North America on 1–2 July 2007. An ozone intrusion moves south-east from the west of Hudson Bay, passing over Southern Ontario before continuing east to the east shore of the United States. This movement is similar to that observed from AIRS in Fig. 4.

Despite the size of the stratospheric intrusion on 1 July, no large change in radar tropopause height appears to have preceded it (Fig. 2), in contrast to the findings of Hocking (2007) and Carey-Smith (2010). Figure 8 also suggests that the intrusion may have originated to the northwest of Harrow. In order to analyze the origin and pathways of the air masses leading to the observed ozone enhancement in the mid-troposphere (3–8 km above the ground) on 1 and 7 July 2007, the FLEXPART model was run in backward mode. The FLEXPART backward model takes into account both resolved motions and parameterized subgrid scale convection and turbulence, using the same parameterizations as the forward mode (described in Sect. 3.2). The backward FLEXPART simulations released 20 000 particles between 3 and 8 km above Harrow (42.03° N, 82.92° W) on 1 and 7 July, and followed them for 10 d backward in time. The upper plots in Fig. 9 show the total column residence time in 10^3 s for the particles. In both cases it appears that the air over Harrow between 3 and 8 km originated primarily from the northwest.

The complex output of FLEXPART calculations can be condensed by clustering particles into several clustered positions at every output time (Stohl et al., 2002). The lower plots in Fig. 9 show the cluster (with the cluster parameter equal to 4) back trajectories for the first 48 h proceeding the release. The lower left-hand plot indicates that the high ozone concentration in the mid-troposphere on 1 July 2007 at Harrow originates in part from the stratosphere over Nunavut and the Northwest Territories. Horizontal transport from the direction of Hudson Bay also contributes to the observed ozone enhancement on 1 July. That the source of the high ozone was apparently the stratosphere over the

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Arctic could explain why the radar did not show rapid tropopause height changes right before the enhancement.

The lower right-hand plot indicates that, unlike the case on 1 July, the high ozone concentration observed at altitudes of 3–8 km on 7 July was the result of horizontal ozone transport from the the direction of the Northwest Territories and did not have a significant recent stratospheric contribution. The high relative humidity observed there is consistent with this conclusion.

That several significant stratospheric intrusions were observed in a short campaign in high summer is surprising, given the prevailing view that STE is more active in spring, e.g., Appenzeller et al. (1996). In the summer, it is generally expected that direct transport from the stratosphere is a minor source of ground level ozone. Since FLEXPART simulates the trajectories of ozone particles originating from the stratosphere, one can calculate the stratospheric ozone tracers and compare them with ground observations of ozone. From the comparison of GEM-FLEXPART simulations with ozonesonde profiles at Harrow, it can be found that the model underestimated ozone in the lower stratosphere. The ozone mixing ratios of the GEM-FLEXPART simulations are corrected, according to ozonesonde measurements in the stratosphere and the mean value of modeled/measured ratios. From the surface ozone measurements and bias-corrected GEM-FLEXPART ozone mixing ratios at the lowest level (190 m a.s.l., which at Harrow is about on the ground level), the fraction of surface ozone of stratospheric origin can be estimated.

AURAMS (A Unified Regional Air-quality Modeling System) is an episodic regional chemical transport modeling system that is driven by GEM model meteorological fields. More detail comparisons between AURAMS output and observations may be found in Makar et al. (2010a), and the impact of ozone boundary conditions in AURAMS may be found in Makar et al. (2010b). Figure 10 displays the hourly averages of surface ozone measurements, bias-corrected GEM-FLEXPART ozone mixing ratios at an altitude of 190 m, and GEM-AURAMS ozone mixing ratios at the altitude of 190 m. Estimates from AURAMS simulations agree with the surface ozone observations very well.

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Stratospheric ozone intrusions contributed most to surface ozone on 27, 28 and 30 June. On these days, ozone of stratospheric origin makes up to 25–50% of surface ozone. The fraction of stratospheric origin is calculated as the ratio of FLEXPART ozone to ozonesonde measurements at ground level. Averaged over the entire campaign period, however, FLEXPART calculates the stratospheric contribution to boundary layer ozone (defined as the 0–1 km average ozone measured by ozonesondes) to be only 1.3 ppb, or 3% (see Table 1). This is much less than the stratospheric contribution of 4.7 ppb estimated for 1–3 km (mean value of FLEXPART calculations at an altitude of 1–3 km), or 5.5 ppb for 3–8 km (mean value of FLEXPART calculations at an altitude of 3–8 km). In either case, it represents that 9% of measured ozone by sondes originated from the stratosphere. This may indicate issues with modeling of transport across the boundary layer, since it is not obvious why ozone above the boundary layer should not eventually be mixed downward. We note that these estimates of the stratospheric contribution to mid-tropospheric ozone are smaller than those of Cooper et al. (2006) and Thompson et al. (2007) for the 2004 IONS summer campaign, possibly because of the limited regional domain of our model, which limits long-range transport. Nevertheless our results suggest that despite a significant incidence of STE during the BAQS-Met campaign, stratospheric ozone made at most a modest contribution to the tropospheric budget. Generally it appears that active photochemistry plays the most important role in controlling ozone amounts in the boundary layer in Southern Ontario during the summer of 2007.

5 Conclusions

We have presented a study of stratospheric ozone intrusion using both observations and models. Three significant ozone enhancements in the troposphere were observed at Harrow, Ontario, during the BAQS-Met campaign in 2007.

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- Ozone concentration observations from ozonesondes and satellite ozone interpolation results in the middle and upper tropopause, coincident low relative humidity in those layers, and a rapid ascent in tropopause height from the radar observations, all suggest that a stratospheric ozone intrusion occurred on 20 June.
- 5 – Kriging interpolation of TES and AIRS ozone profiles shows high ozone concentration in the upper and lower troposphere on 1st July. GEM-FLEXPART simulation results show that this was because stratospheric ozone intruded down below 5 km above the ground. The satellite data showed the intrusion clearly, its horizontal and vertical extent, and its eastward motion. These features were reproduced by the FLEXPART simulation. However, the radar at Harrow did not detect a rapid tropopause height change at this time. Source-receptor analysis using the GEM-FLEXPART model shows the stratosphere over the Northwest Territories region close to the Beaufort Sea to be the main source of the enhanced ozone near Harrow on 1 July.
- 10 – Both ozonesonde measurements and Kriging interpolations of satellite ozone showed ozone enhancement on 7 July. However, neither the radar tropopause height data nor GEM-FLEXPART simulation (forward and backward) suggest a recent ozone intrusion. Long-range transport from the upper troposphere over the Northwest Territories is the main source of this enhancement. Despite this high level of activity, transport contributed modestly on average to background ozone, and did not contribute measurably to the high variability observed in the surface measurements.

Acknowledgements. This work was supported by funding from the Canadian Foundation for Climate and Atmospheric Sciences (CFCAS). The Harrow radar was installed with support from the Canada Innovation Foundation and Ontario Innovation Trust. We thank Jonathan Davies, Rad Sakura and Ildiko Beres for ozone-launch support. We thank members of the TES team and AIRS teams for their measurements and data products.

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**Table 1.** Means of ozone mixing ratio.

ppb/km	0–1	1–3	3–8
FLEXPART	1.34	4.71	5.52
Sonde	43.53	55.31	62.07

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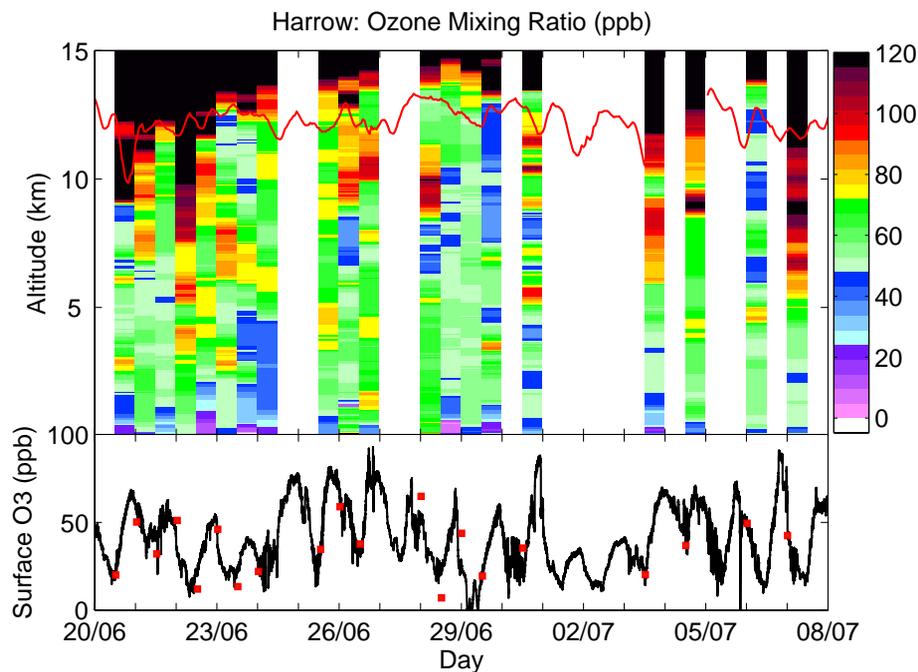


Fig. 1. Upper panel: vertical profiles of ozone mixing ratio (ppb) for the BAQS-Met campaign at Harrow, Ontario, in the summer of 2007. The red solid line shows the tropopause height derived from the radar data. Lower panel: one-minute averages of surface ozone mixing ratio (ppb) from 20 June to 7 July 2007, measured by the chemistry supersite at Harrow (42.03° N, 82.92° W), Ontario. The red squares show the ozonesonde measurements at the release time at ground.

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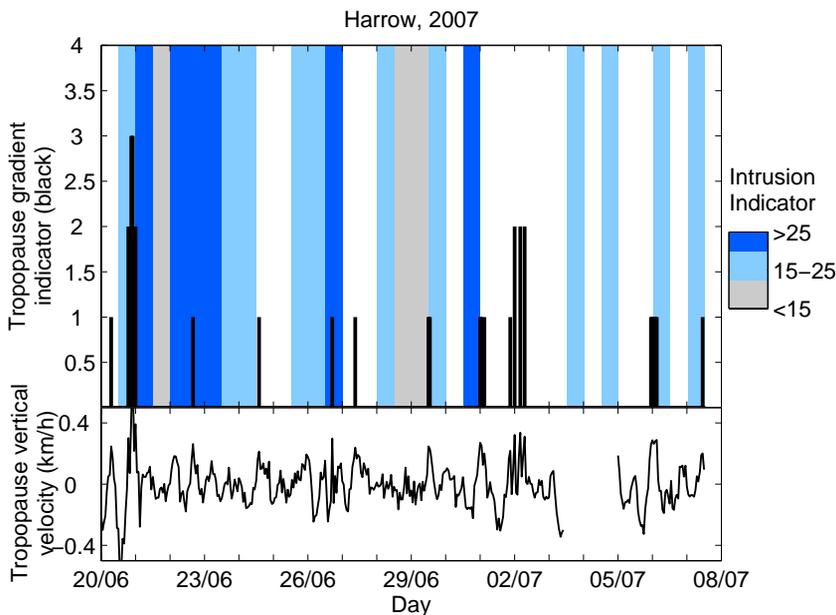


Fig. 2. Upper panel: Stratospheric ozone intrusion indicator. Lower panel: radar tropopause vertical velocity (km/h). Tropopause vertical velocities (in km/h) have been classified into four categories of tropopause gradient indicator: level 3 if the tropopause vertical velocity >0.4 , level 2 if in $(0.3, 0.4]$, level 1 if in $(0.2, 0.3]$, and level 0 if ≤ 0.2 . The tropopause gradient indicators are shown in the upper panel by the black line. The intrusion indicators (in ppb) are computed as the difference between the maximum ozone mixing ratios between 3 km altitude and the radar tropopause, and the background ozone mixing ratio calculated as average of values before, after, above and below the maximum. If the intrusion indicator is larger than 25 ppb and the tropospheric ozone maximum is coincident with a local minimum in relative humidity, a dark-blue vertical band is shown. Light-blue bands indicate tropospheric ozone maxima exceeding the background ozone by 15–25 ppb, grey differences less than 15 ppb, and white bands where no ozonesonde data are available.

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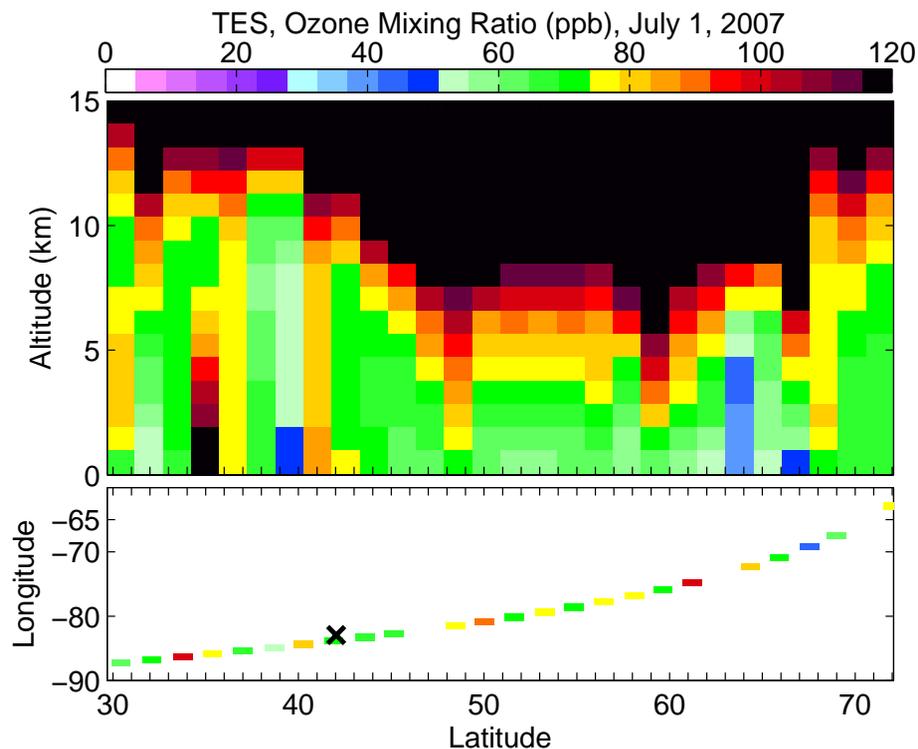


Fig. 3. Upper panel: vertical profiles of TES ozone mixing ratio (ppb) along a global survey track (shown in the lower plot), on 1 July 2007. Lower panel: the TES ozone mixing ratio (ppb) at 6 km a.s.l.. The black cross shows the location of Harrow.

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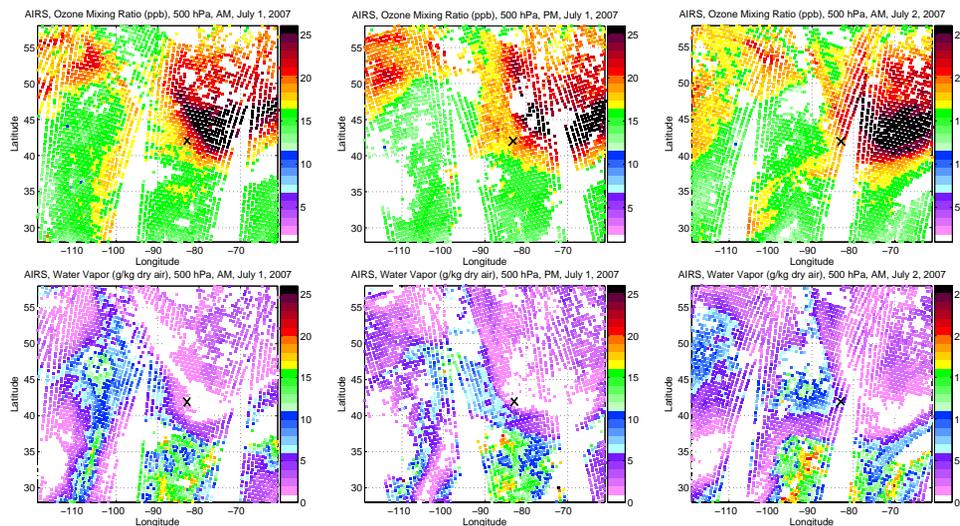


Fig. 4. AIRS observations at 500 hPa (~ 5.5 km above the mean sea level), on 1–2 July 2007. Upper plots: ozone mixing ratios (ppb); lower plots: water vapor content (g/kg dry air). The black cross shows the location of Harrow.

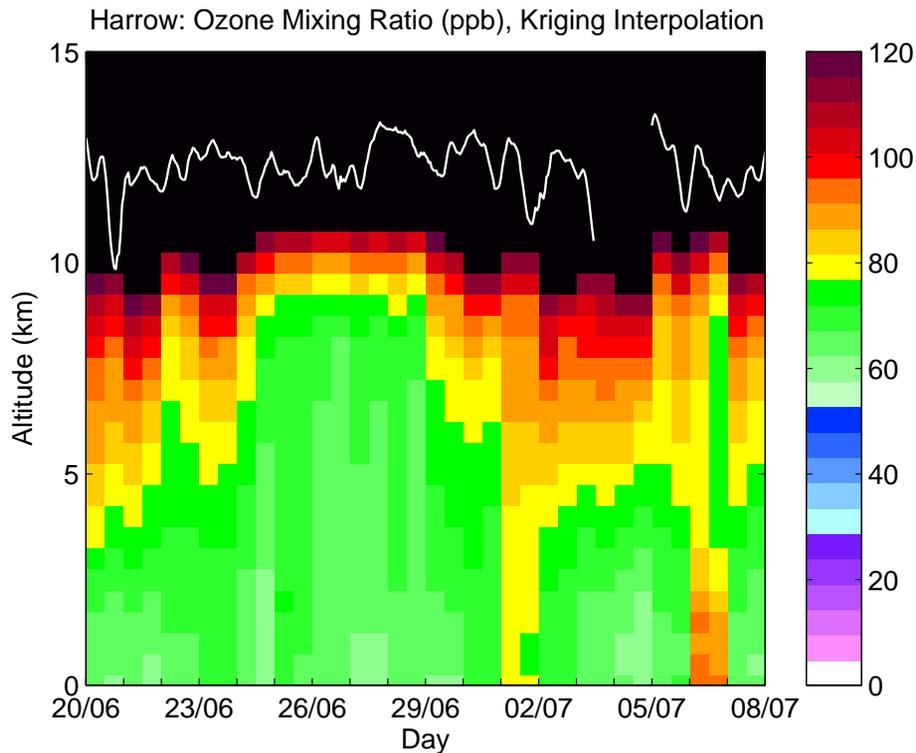


Fig. 5. Kriging interpolation results at Harrow (42.03° N, 82.92° W), Ontario, using TES and AIRS ozone profiles. The white solid line shows the tropopause height derived from the radar data.

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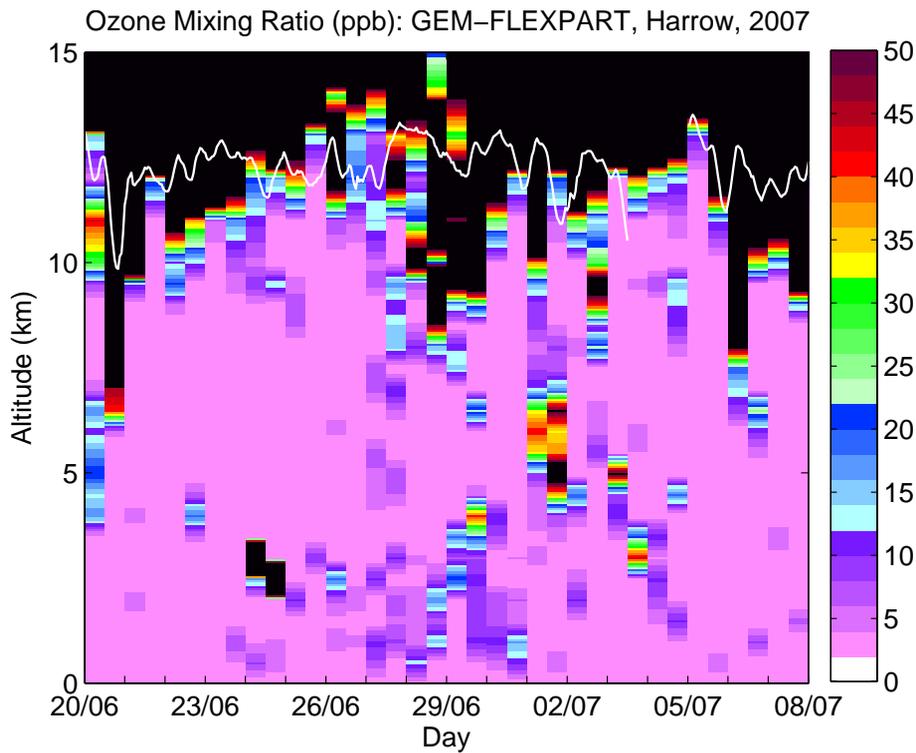


Fig. 6. Vertical profiles of ozone mixing ratios from a GEM-FLEXPART simulation, at the location of the Harrow site in the summer of 2007. The white solid line shows the tropopause height derived from the radar data.

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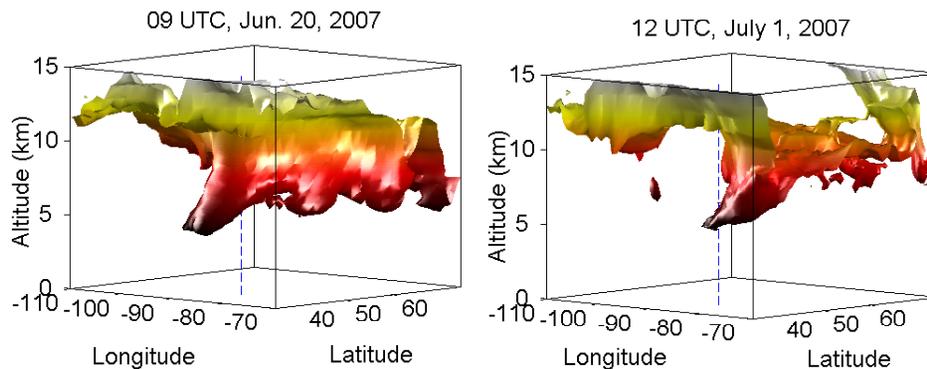


Fig. 7. FLEXPART output showing the 80 ppb ozone mixing ratio iso-surface (left: at 09:00 UTC on 20 June; right: at 12:00 UTC on 1 July 2007). The blue dashed line shows the location of Harrow. Lower the iso-surface, darker the color.

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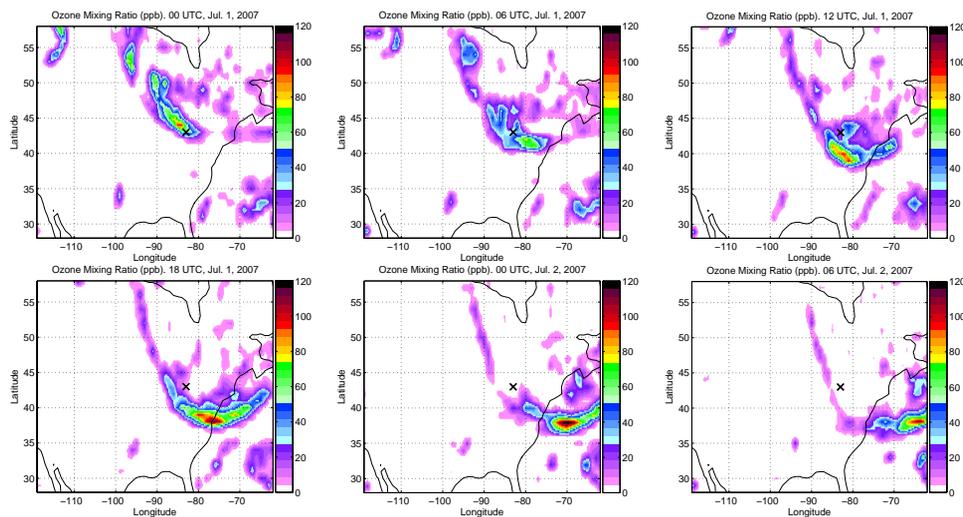


Fig. 8. Ozone mixing ratios (ppb) from a GEM-FLEXPART simulation, at 5.5 km altitude over the Harrow station, on 1 and 2 July 2007. The black cross shows the location of Harrow.

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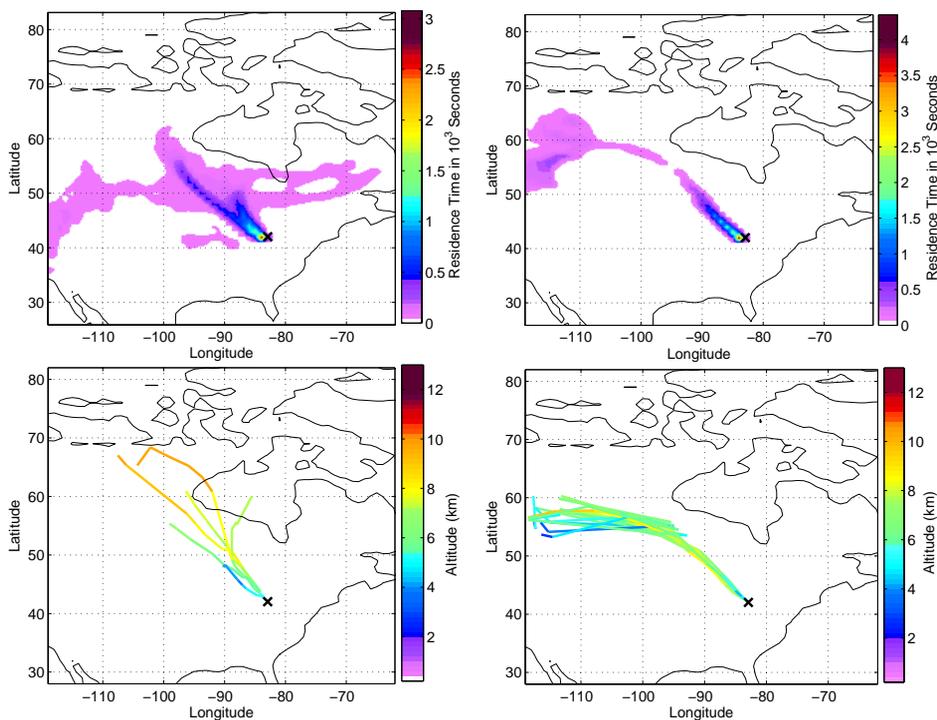


Fig. 9. GEM-FLEXPART 10-day backward simulation results, for air parcels arriving in the mid-troposphere (3–8 km a.s.l.) over Harrow, Ontario, on 1 and 7 July 2007. Upper: total column residence time given in 10^3 s; lower: cluster back trajectories for the previous 48 h. Left column: case study for 1 July; right column: case study for 7 July. The black cross denotes the location of Harrow.

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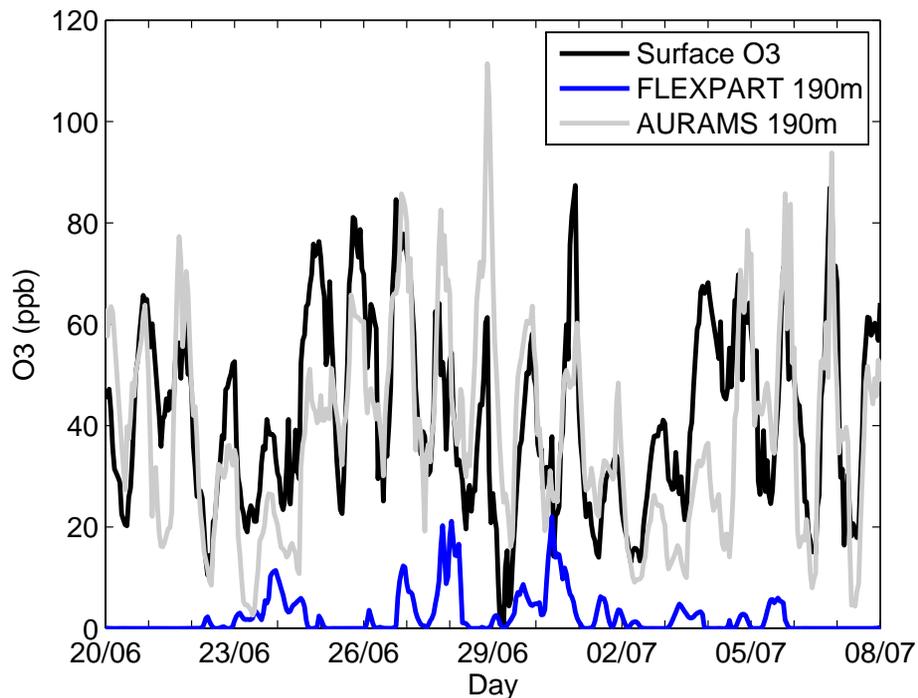


Fig. 10. Surface ozone measurements (black), bias-corrected GEM-FLEXPART ozone mixing ratio at 190 m altitude (blue), and GEM-AURAMS ozone mixing ratio at the altitude of 190 m (at the ground level of Harrow). Ozone mixing ratios are sampled once every hour.

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