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LETTERS

Detection of stratospheric ozone intrusions by windprofiler radars

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Stratospheric ozone attenuates harmful ultraviolet radiation and protects the Earth's biosphere[1]. Ozone is also of fundamental importance for the chemistry of the lowermost part of the atmosphere, the troposphere[1–8]. At ground level, ozone is an important by-product of anthropogenic pollution[7], damaging forests and crops[5,6], and negatively affecting human health[9]. Ozone is critical to the chemical and thermal balance of the troposphere[10] because, via the formation of hydroxyl radicals, it controls the capacity of tropospheric air to oxidize and remove other pollutants[1]. Moreover, ozone is an important greenhouse gas, particularly in the upper troposphere[1]. Although photochemistry in the lower troposphere is the major source of tropospheric ozone[2,7,11], the stratosphere–troposphere transport of ozone[12–19] is important to the overall climatology, budget and long-term trends of tropospheric ozone[3,4,8,12]. Stratospheric intrusion events, however, are still poorly understood. Here we introduce the use of modern windprofiler radars[20–22] to assist in such transport investigations. By hourly monitoring the radar-derived tropopause height[23–25] in combination with a series of frequent ozonesonde balloon launches, we find numerous intrusions of ozone from the stratosphere into the troposphere in southeastern Canada. On some occasions, ozone is dispersed at altitudes of two to four kilometres, but on other occasions it reaches the ground, where it can dominate the ozone density variability. We observe rapid changes in radar tropopause height immediately preceding these intrusion events. Such changes therefore serve as a valuable diagnostic for the occurrence of ozone intrusion events. Our studies emphasize the impact that stratospheric ozone can have on tropospheric ozone, and show that windprofiler data can be used to infer the possibility of ozone intrusions, as well as better represent tropopause motions in association with stratosphere–troposphere transport.

Ozone enters the troposphere from the stratosphere as part of the Brewer–Dobson circulation[13], through episodic events, but details of the process are not well known. In this work, we observed a number of such events, using a unique combination of windprofiler radars, frequent ozonesonde launches and computer modelling. Windprofiler radars at two locations were used to provide tropopause heights and vector winds as a function of time, at a resolution of typically one hour, as described in the Methods Summary and illustrated in Fig. 1a.

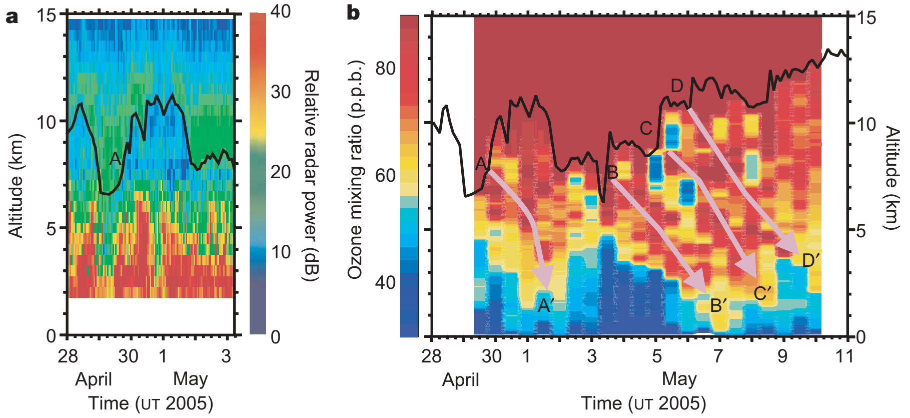

Figure 1 | Simultaneous radar and ozonesonde measurements from the Montreal Campaign of April–May 2005. a, Altitude–time intensity plot of backscattered radar power observed with the McGill windprofiler radar, expressed as relative power in decibels. Absolute maximum values of backscattered powers occurred in the lower atmosphere, but a secondary maximum appeared in the altitude region between 6 and 14 km. The lower edge (region of largest local power gradient as a function of height) of this secondary maximum (green in the figure) is shown as a black line. This represents the height of the tropopause, as has been shown in a variety of studies[23–25]. Radar data were recorded from 28 April to 11 May, but only a subset of the radar data are shown. **b**, Ozone densities (measured in parts per billion) are plotted as a function of height and time for the period 29 April to 10 May. Each vertical column of coloured boxes represents a different launch. The tropopause height as determined by the radar is marked as the solid black line at 6–14 km altitude. Regions of rapid tropopause ascent are labelled as A, B, C and D. Stratospheric ozone intrusion trajectories are highlighted approximately by the hand-drawn pink arrows (A–A′, B–B′, C–C′, D–D′), although these should only be taken as a guide, because the detailed trajectories are complicated by dispersive processes, by the presence of pre-existing tropospheric ozone, and, in cases C and D, by the close proximity of the two jumps (making their effects hard to separate).

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The radars used were located at Montreal, Quebec (45.4°N, 73.9°W), and Walsingham, Ontario (42.6°N, 80.6°W). They had steerable beams with one-way beam half-power half-widths of 2.1°, which could be pointed vertically, or at 10.9° off-vertical in various azimuthal directions. The radio frequencies used were 52.00 MHz (Montreal) and 44.50 MHz (Walsingham). The vertical resolution was 500 m in each case.

Ozonesonde balloons were released close to the radars, and profiles of ozone concentration, temperature, humidity, pressure, wind speed and wind direction were obtained at intervals of typically 8–12 hours. In the Montreal case, the launches were made at the Canadian Space Agency Head Office in St Hubert, Quebec, while the radar was located on the MacDonald campus of McGill University, about 45 km away. At Walsingham, launches took place at the radar site. Studies were carried out in five campaigns, each about two weeks long. These simultaneous measurements of radar tropopause heights, together with dynamical parameters, ozone and water-vapour content, were supplemented by calculations using a three-dimensional dynamical lagrangian particle dispersion and tracking model of ozone transport, called FLEXPART[26–29], which used regional meteorological analyses of the Canadian operational weather forecast Global Environmental Multiscale (GEM) model[30] as input. More specifics can be found in the Methods Summary, and in the Supplementary Information. The results of our study demonstrate the capability of windprofilers to be used to understand ozone intrusions better.

The balloons used in the study carried EN-SCI model 2Z-ECC ozonesondes equipped with Global Positioning System (GPS) receivers and Vaisala RS80 radiosondes. The vertical resolution was about 100 m, and ozone measurement accuracy was about 5%. Over a hundred launches were carried out in five campaigns. Water vapour content was also measured by the ozonesondes, and low water vapour content in the middle troposphere, typically less than 0.2 mb, associated with ozone peaks, was taken as partial evidence that the ozone enhancement had a stratospheric origin. Every campaign showed evidence of stratospheric ozone intrusions into the troposphere, but in the first campaign, high levels of radio interference prevented useful radar measurements. Our discussions will therefore focus on the subsequent four campaigns.

Figure 1b shows a height–time ozone-density plot from Montreal for April–May 2005, as well as the tropopause height taken from Fig. 1a. The tropopause height closely follows the height at which ozone density increases markedly. Excursions of tropospheric ozone density above the background values (typically dark blue in Fig. 1b) are our main interest here. 'Background' ozone is generally defined as tropospheric ozone that is more than seven days old and therefore of uncertain origin, and typical values[2–4,11] are of the order of 20–40 p.p.b.

Three rapid ascents in tropopause height are labelled A, B and C, and a fourth, smaller one is labelled D. In each case there is evidence of ozone intrusion from the stratosphere. Case B is especially clear. In the other cases, the intrusion is less distinct, but increases are apparent in ground-level ozone density following the tropopause jumps. Jumps C and D occur close together, making separation of their effects harder, and the effect of event D seems delayed until after the campaign ended. Nevertheless, increases in low-altitude ozone densities are clearly associated with the tropopause jumps. In each case, decreased humidity served as an additional indicator that the air was stratospheric in origin. In case B–B′, a noticeable increase in surface ozone was also observed at stations from Montreal back to the Great Lakes, 1,000 km to the west. Back-trajectory calculations and meteorological analyses indicated that a significant part of this ozone enhancement was stratospheric in origin. Ozone had entered the troposphere both above Montreal and as much as 1,000 km upstream to the west and northwest on 3 May, and the whole ozone enhancement moved downward and downstream as a layer. An initially upstream component reached the ground in Montreal on 7 May. Ground-level values at these stations before and after the two events of 30 April–2 May and 6–9 May to were typically 15 p.p.b. at night and 30 p.p.b. during the day, but during the event of 6–9 May were typically 30 p.p.b. (night-time) to 50 p.p.b. (daytime). Stratospheric ozone had a large impact on the surface ozone densities, even in a large city in which photochemical effects might have been expected to dominate.

Figure 2 shows an example from the Walsingham campaign of November 2005, which ran from 17 to 25 November (23 launches). The period of greatest interest is the time frame covering 23–25 November. An enhancement of ozone appeared at an altitude of about 5–6 km, briefly descended, and then rose again to about 8 km in altitude (Fig. 2b). This layer had low water vapour content (<0.2 mbar), suggestive of stratospheric origin, but otherwise did not seem strongly associated with the stratosphere.

However, the radar data did suggest a stratospheric link. The scattered power as a function of height and time is shown in Fig. 2a, with the radar-determined tropopause indicated. At about the same time that the ozone enhancement appeared, the radar-determined tropopause showed a rapid descent (point P in Fig. 2a). The tropopause temperature profiles determined from the radiosondes showed a correlated behaviour, with strong temperature inversions tracking the radar-determined tropopause. The trajectory of this enhancement is shown by the broken line. When the radar tropopause descended, it left a region of only weak scatter at the 'normal' tropopause height (marked Q on the figure). The tropopause jet stream showed a very wavelike structure, indicating strong nonlinear planetary wave

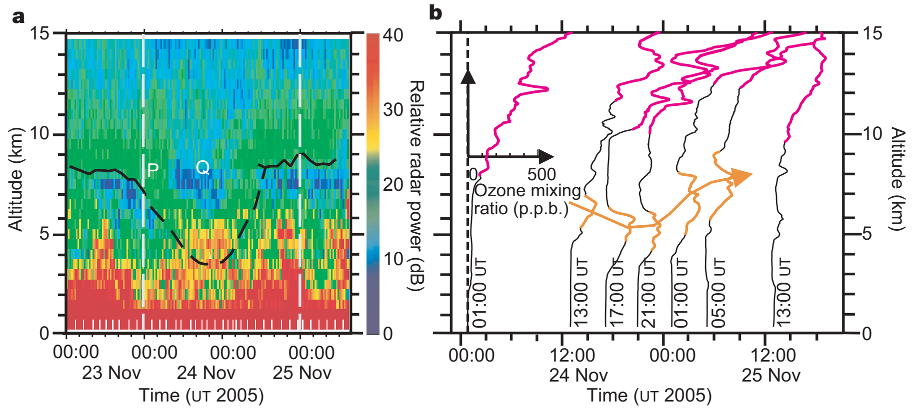

Figure 2 | Radar and ozone data recorded during the first campaign at Walsingham in November 2005. a, Altitude–time intensity plot of backscattered power during the campaign. The curved black broken line shows the radar-determined tropopause. The vertical grey broken lines show the start- and end-times of the period covering seven ozonesonde ascents of interest. **b**, Successive ozone profiles for the seven ozonesonde launches. The upper, red sections of the profiles indicate the stratosphere, based on the World Meteorological Organization (WMO) definition of the tropopause (which by definition generally occurs above 500 mb), and the centre, orange sections emphasize the region of ozone of interest.

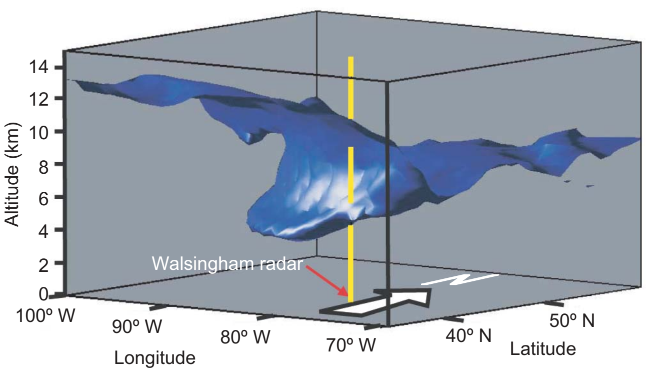

Figure 3 | Three-dimensional image of the 100 p.p.b. ozone surface during the event of 23–25 November 2005, as determined by the FLEXPART model. The time was specifically 24 November 2005 at 15:00 UT. The location of the Walsingham radar and radiosonde launch site is shown (vertical yellow line). The white arrow shows north.

activity in the lower stratosphere, as shown in maps provided by the Canadian Meteorological Centre (http://weatheroffice.gc.ca/analysis/index_e.html).

Figure 3 shows the results of a numerical simulation using FLEXPART. The model clearly indicates a deep influx of ozone from the stratosphere, resulting in a tongue of ozone penetrating to 4 km altitude, implying significant downward motion. The tongue of stratospheric air swept over the radar in such a way that its tip passed over the radar, producing the variation shown in Fig. 2b. Without the use of radar data, the stratospheric origin of this ozone tongue may well have been missed. In contrast to the Montreal case, in which the ozone reached the ground, the ozone in Fig. 3 appeared to mix with the surrounding air at 3–4 km.

Figure 4 shows the occurrence of all ozone intrusions during the four campaigns studied, shown placed end-to-end. Dark blue lines show occasions of strong intrusion, light blue represents weaker intrusions, and black lines represent strong upward tropopause motions. Large positive vertical velocities can arise due to true tropopause jumps (as in Fig. 1b) and can also be produced by split tropopauses, as in Fig. 2. In the latter case, the tropopause-determination software follows the descending tropopause from point P until it becomes so low that it would not normally be considered a tropopause, and then finds the higher-level, weaker, tropopause (closer to point Q), producing an apparent jump in tropopause height. Both types of jumps will be considered collectively.

Every occurrence of definite ozone intrusion is associated with a level 2 or level 3 radar-tropopause excursion rate at, or just before, the intrusion, with the exceptions of events A, B, C and D. For cases C and D, the tropopause was only intermittently visible with the radar (this happens on occasion). Of the remaining 13 intrusions, 11 (that is, all except A and B), were associated with a level 2 or level 3 tropopause excursion. Even more telling, every level 2 or level 3 tropopause excursion was associated with some form of ozone intrusion. Hence, a level 2 or level 3 tropopause jump is a very strong predictor for ozone intrusion. No matter whether the jumps are real, or a consequence of a split tropopause or discontinuity, they serve as a valuable diagnostic. The ability to detect ozone intrusions in this way is an important capability, and a major result of our study. This is particularly true because windprofilers generally operate 24 hours per day, 365 days per year, so even when ozonesonde data are not available, windprofiler data can be used as a proxy for the possibility of ozone intrusions. This will be useful for air-quality forecasts, stratosphere–troposphere transport research, and general understanding of the ozone circulation, transport, and budget.

METHODS SUMMARY

The simultaneous and co-located use of windprofiler radars combined with frequent ozonesonde launches represents the key experimental aspect of this project. Windprofilers[20–25] are radars that permit ground-based studies of the atmosphere from regions close to ground-level, to altitudes of 12 km and higher (depending on power output). A powerful transmitter initially emits repeated pulses of radio waves into the air, whereupon small portions of this signal are

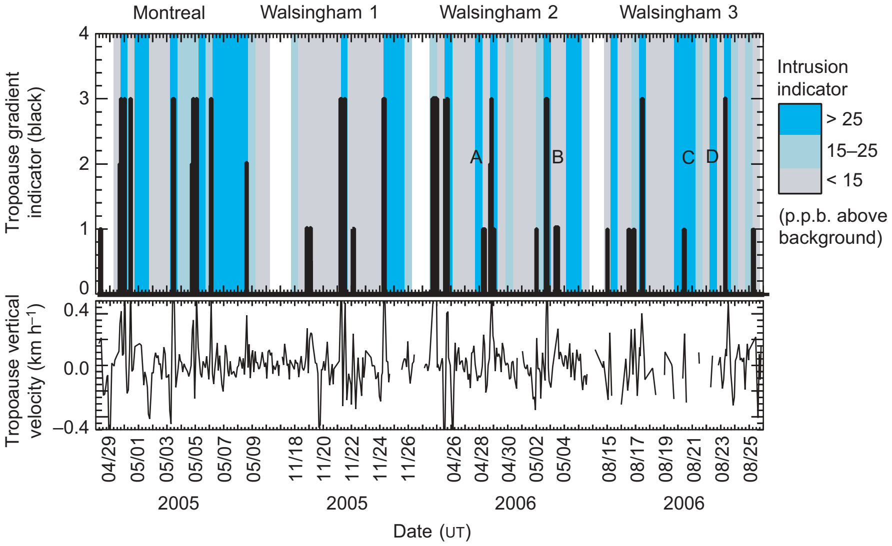

Figure 4 | Occurrence of ozone intrusions compared to radar-determined tropopause excursions. Dates for the campaigns are given as month/day. Dark-blue vertical bands represent occasions where a tropospheric ozone maximum occurred between 3 km altitude and the tropopause, and where this maximum exceeded the background value (that is, average of values before, after, above and below the maximum) by at least 25 p.p.b. A coincident local minimum in water vapour content was also required. This combination is taken as a strong indicator that a stratospheric intrusion had occurred, particularly if the layer showed evidence of descent. Light-blue shading indicates a weaker but nevertheless real intrusion, where in this case the excess of the local peak exceeded the background by 15–25 p.p.b. Grey bands represent occasions when there was no significant tropospheric maximum, and white bands indicate times when no ozone data were available. The date of intrusion is set according to the time at which the intrusion left the stratosphere, and not the time of arrival at the ground. The lower panel shows the vertical velocity of the tropopause, as determined from the radar data. Tropopause excursion velocities have been classified into four categories using the tropopause gradient indicator; specifically: type 3 means $>0.4\,\mathrm{km\,h^{-1}}$, type 2 means $>0.3\,\mathrm{km\,h^{-1}}$, type 1 means $>0.2\,\mathrm{km\,h^{-1}}$ and type 0 means $<0.2\,\mathrm{km\,h^{-1}}$ These categories are plotted as black vertical lines in the upper graph.

returned to the radar antennas and recorded for analysis. Proper interpretation of these returned signals allows wind and turbulence strengths in the atmosphere to be measured, and in our case the height of the tropopause can also be found. The radars run continuously, allowing unprecedented monitoring of tropopause height. Ozone measurements were made using EN-SCI ozonesondes and ancillary equipment, including a ground station. The ozonesondes were accompanied by Vaisala RS-80 radiosondes for pressure, humidity and temperature measurements. On-board GPS receivers were used to track the the sonde positions and allow wind velocity determination. Typically either 800 g or 1,200 g balloons were used, filled with sufficient helium that they achieved an ascent rate of $3–5\,\mathrm{m\,s^{-1}}$. Experimental studies were supported by application of the FLEXPART[26] computer model, which permitted modelling of ozone movement. FLEXPART required hourly wind fields produced by a regional analysis model called GEM[30], run at 0.1375° × 0.1375° resolution on a domain covering North America with 58 vertical levels to 10 hPa. Each FLEXPART run released 600,000 particles in the model domain, with those in the stratosphere initialized using an empirical relationship between potential vorticity and ozone concentration[15]. These were then advected using wind fields from GEM, and the resulting ozone field was output at 1° × 1° × 500 m resolution. Chemistry is not included in the model. FLEXPART has been extensively validated[19,27–29]. Please see the Supplementary Information for further details.

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Supplementary Information is linked to the online version of the paper at www.nature.com/nature.

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Author Contributions W.K.H. designed and built all the windprofiler radars used in the studies, and wrote all the on-line radar analysis software. He also originally proposed the concept of using the radars and ozone studies together to investigate stratosphere–troposphere transport, wrote the original proposal, and was the principal investigator on the grant used to obtain the data. T.C.-S. was a post-doctoral fellow on the project, and was responsible for all ozonesonde launches in regard to planning and implementation. He was also responsible for adaptation and implementation of the Flexpart model, and was responsible for data analysis after each flight. D.W.T. was the most experienced of the team in regard to ozone science, and was responsible for the direct supervision of T.C.-S. for significant parts of his tenure. He provided advice about ozonesonde launches and data interpretation, including initiating the use of FLEXPART and GEM to provide a four-dimensional view of the intrusion processes. P.S.A. is a research scientist who co-managed the ozonesonde programme, undertook much of the pre-campaign preparation work, and provided scientific direction during the experimental campaigns. K.S. operated the Toronto Atmospheric Observatory, which was used as a support facility to provide back-up data about the behaviour of various atmospheric chemical constituents. Y.R. supported T.C.-S. with advice. I.Z. and P.A.T., as principal investigators of various windprofiler projects, were responsible for the day-to-day running of the radars.

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