

Stratosphere to troposphere ozone event characterisation and distribution over Melbourne, Macquarie Island, and Davis with ozonesondes.

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Abstract

We develop a quantitative method to determine Stratosphere to Troposphere Transport events (STTs) from ozone sonde profiles. Using this method we estimate the quantity of ozone transported across the tropopause over Melbourne (38°S), Macquarie Island (54°S), and Davis (69°S). STT seasonality is determined from a 7–9 year long time series of ozone profiles from each site. STT events primarily occur during summer above Melbourne and Macquarie Island, while there is little seasonal cycle in STT events above Davis. The majority of tropospheric ozone due to STT events occur within 3 km below the tropopause at Melbourne and Macquarie Island, and within 2 km below the tropopause at Davis. Overall, the fraction of total tropospheric ozone attributed to STT events is 2 – 4% at each site, however, during individual events, an STT event can contribute more than 10% of the total tropospheric ozone at that time. TODO: We also need to note favourable or unfavourable comparisons between our ozone flux results and previous model studies; also a sentence about GEOS-Chem results.

1 Introduction

Tropospheric ozone, which constitutes only 10% of the total ozone column, is an important oxidant, greenhouse gas and is toxic to biological life. Over the

industrial period, increasing tropospheric ozone, has been estimated to exert a radiative forcing equivalent to a quarter of the CO₂ forcing [Forster et al., 2007]. Tropospheric ozone increases above pre-industrial levels are estimated to result in global losses up to \$USD₂₀₀₀ 35 billion per annum until 2030 due to food crop impacts [Nawahda, 2013] and up to \$USD₂₀₀₀ 580 billion by 2050 (11.8 billion per year) due to health impacts [Selin et al., 2009]. Tropospheric ozone is produced anthropogenically by fossil fuel and biomass combustion emissions (NO_x and volatile organic compounds) that subsequently undergo photochemistry. Natural sources of tropospheric ozone include the downward transport from the ozone-rich stratosphere, wildfires and lightning photochemical production (?Jacobson and references therein).

Stratosphere to troposphere transport (STT) of ozone can increase regional surface ozone levels above air quality standard thresholds [?Lefohn et al., 2011, Langford et al., 2012, Zhang et al., 2014]. A review of photochemical models by Stohl et al. [2003] (STACCATO) concluded that between 25-50% of tropospheric ozone can be attributed to SST events. A lower estimate was derived from the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP), Stevenson et al. [2006] with 5100 ± 600 Tg/yr (90%) and 550 ± 170 Tg/yr (10%) of tropospheric ozone is due to chemical production and STT, respectively. Models are challenged to correctly represent STT, and process studies seem to be key in determining the relative role of SST in the tropospheric ozone budget.

STT events have been observed due to deep overshooting convection [Frey et al., 2015], tropical cyclones [Das et al., 2016] and mid-latitude synoptic scale disturbances (e.g. Stohl et al. [2003], Mihalikova et al. [2012]). STT events observed over the Mediterranean region estimate a 10% contribution to tropospheric ozone budget between 2000 and 2003 [Galani, 2003], with other observational studies noting significant occurrences and strong seasonal dependence (i.e. Lefohn et al. [2011]), contributing up to 30% of the surface ozone over the Western US in spring [Lin et al., 2012]. To date, while this topic has received significant attention in the tropics and northern hemisphere, observational estimates from the southern hemisphere extra-tropics is noticeably absent in the literature.

In the extra-tropics, ozone has a longer photochemical lifetime and STT events most commonly occur during synoptic-scale tropopause folds [Sprenger et al., 2003, Tang and Prather, 2012] which are characterised by tongues of high Potential Vorticity (PV) air descending to low altitudes. These tongues become elongated and filaments disperse away from the tongue and mix irreversibly into the troposphere. STT events have been observed in tropopause folds around both the polar-front jet [Vaughan et al., 1993, Beekmann et al., 1997], and the subtropical jet [Baray et al., 2000] and cut-off lows [Price and Vaughan, 1993, Wirth, 1995]. A high correlation is found between lower stratospheric and tropospheric ozone [Terao et al., 2008] with the highest STT associated with jet-streams over the oceans in winter.

In section 2 we use several years of ozonesonde flights from three locations spanning the 38°S - 69°S latitudes to characterise the seasonal cycle of STT

events and determine their contribution to the total amount of tropospheric ozone. We examine the depth and frequency of the intrusions and use case studies to relate these STTs to meteorological events. Lastly, we calculate the fraction of total tropospheric column ozone attributed to STT events.

2 Data and Methods

2.1 Ozonesonde record in the Southern Ocean

Ozonesondes are weather balloons with an attached instrument which measures ozone concentrations roughly every 100m from the surface to around 30km. These ozonesondes provide a high-vertical resolution profile of ozone. Ozonesondes are estimated to provide around 2% precision in the stratosphere, with increasing precision at lower altitudes [NOAA].

Ozonesondes are launched approximately weekly from Melbourne (38°S, 145°E), Macquarie Island (55°S, 159°E) and Davis (69°S, 78°E). For this study, we use the data collected from 2004-2013 for Melbourne and Macquarie, and 2006-2013 for Davis. Around twice as many ozonesonde launches occur at Davis prior to and during the ozone hole season (June-October) than at other times of the year [Alexander et al., 2013].

2.2 Characterisation of STT events and associated fluxes

Stratospheric ozone typically mixes irreversibly down (vertically) into the troposphere in kilometres-scale tongues of air. The altitude of the tropospheric ozone peak due to an STT event and the amplitude of the event above the background tropospheric ozone profile vary in space and time. While ozone sondes are released every week or so, ozone events may only be detectable for a matter of hours [Tang and Prather, 2012]. The vertical ozone profile recorded by the ozonesonde is highly dependent on the time of launch [Sprenger et al., 2003], and we cannot guarantee that detected ozone enhancements are fully separated from the stratosphere.

In order to characterise tropospheric ozone events a clear definition of where the stratosphere begins is necessary. Tropopause height can be defined based on the standard lapse rate tropopause [WMO, 1957] or the ozone tropopause [Bethan et al., 1996]. The lapse rate is the negative altitudinal temperature gradient, while the lapse rate tropopause is defined as the lowest altitude where the lapse rate is below $2^{\circ} \text{ C km}^{-1}$, provided the lapse rate between this altitude and all subsequent altitudes within 2 km is also below $2^{\circ} \text{ C km}^{-1}$. The ozone tropopause is defined as the lowest altitude satisfying these three conditions [Bethan et al., 1996]:

1. Vertical gradient of ozone mixing ratio (OMR) is greater than 60 ppbv km^{-1}
2. OMR is greater than 80 ppbv

3. OMR between 500 m and 2000 m (1500 m in the Antarctic) above exceeds 110 ppbv.

At Davis, the ozone tropopause definition is modified since the site is Antarctic. While the ozone tropopause can be less robust during stratosphere-troposphere exchange, it performs better than the lapse rate tropopause at polar latitudes in winter and near jet streams in the lower stratosphere [Bethan et al., 1996, Tomikawa et al., 2009, Alexander et al., 2013]. We use the lower of these two tropopause altitudes for this study. This choice avoids occasional unrealistically high tropopause heights due to perturbed ozone or temperature measurements in ozonesonde records.

Figure 1 shows the monthly mean tropopause altitudes at each location, along with the subset of altitudes from profiles for which an STT event was determined. The seasonal cycle in tropopause altitude at Melbourne is clearly apparent, as is the decreasing tropopause altitude which occurs at higher southern latitudes. The tropopause recorded by sonde is generally higher during an STT event at all three sites. Tropopause altitudes at Davis may exceed 11 km altitude under certain synoptic conditions [Alexander et al., 2013]; the relation of tropopause altitude with individual STT events is investigated in detail below.

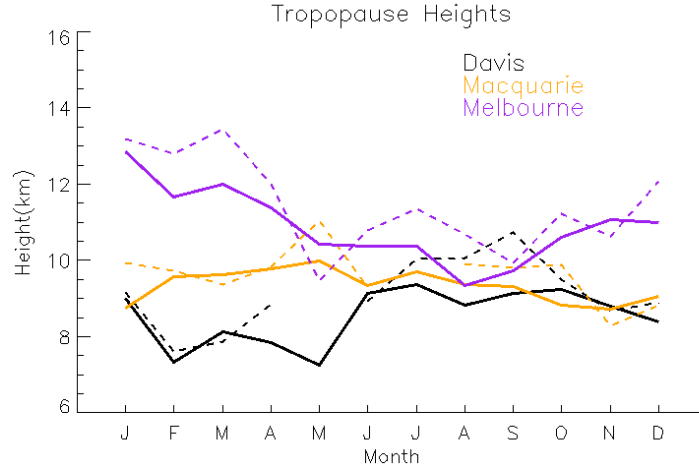


Figure 1: Monthly mean tropopause altitudes (minimum of lapse-rate and ozone defined tropopause at 3 sites) determined from ozone sondes. Dashed lines show the average monthly altitude when only considering dates when STTs occurred.)

Figure 2 shows seasonally averaged ozone as recorded over the three stations. Increased ozone extending down through the stratosphere is apparent during December to March and September to November over Melbourne. These increased tropospheric ozone months are due to STTs (in summer), and possible

fire smoke plume influence (in winter). Over Davis and Macquarie Island the tropospheric ozone is higher between March and October, although the effect is subtle compared to Melbourne.

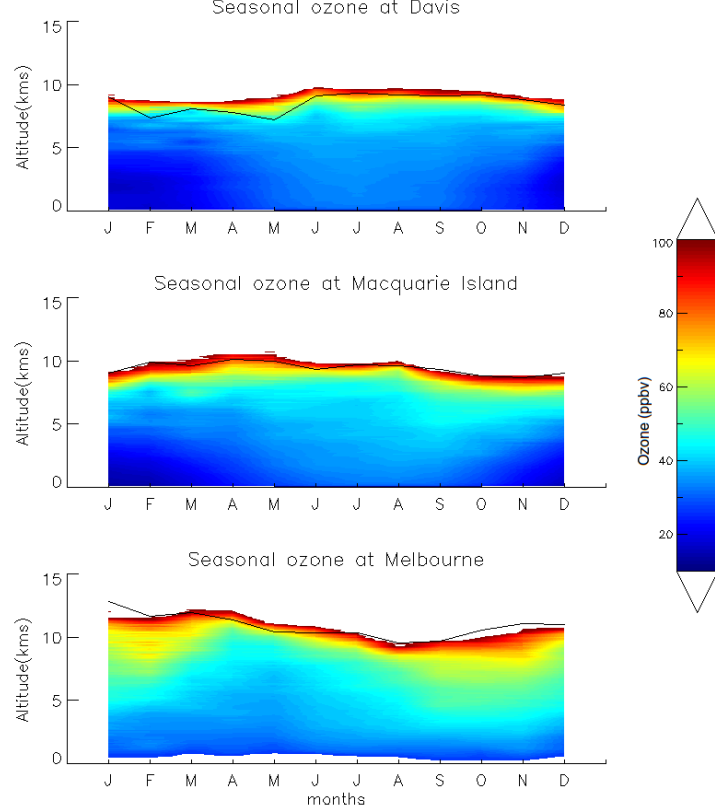


Figure 2: Seasonal cycle of ozone over Davis, Macquarie, and Melbourne measured by ozonesondes, where measurements are binned monthly. Black solid lines show seasonal tropopause heights, defined as described in the text.

Tang and Prather [2010] define one method of detecting these stratospheric tongues (or tropospheric ozone folds) as follows: From 5 km altitude, if the ozone level exceeds 80 ppb and then within 3 km decreases by 20 ppb or more to a value less than 120 ppb, then a tropopause fold has occurred. Their definition is based on subjective analysis of sondes released from 20 stations in the latitudinal range from 35°S to 40°N. We also characterise STT events using the ozonesondes vertical profiles, looking for tropospheric ozone enhancement above a local background (in moles per billion moles of air, or ppb). In this paper we define tropospheric ozone events based on a subjective analysis of ozonesonde profiles at three sites at 38°S, 55°S, and 69°S.

TODO: More of simon’s changes here, clarify analysis and sensitivity To identify STT events, the vertical profiles of ozone volume mixing ratio are linearly interpolated to a regular grid with 20 m resolution up to 14 km altitude and are then bandpass filtered to retain perturbations with vertical scales between 0.5 km and 5 km. From here onwards the filtered vertical profile is referred to as the perturbation profile. The choice of band limits was set empirically; for an event to qualify as STT, a clear increase above the background ozone level is needed, and a vertical limit of ~ 5 km removes seasonal-scale effects. We exclude from analysis perturbations at altitudes below 4 km above the surface to avoid surface pollution events and those occurring within 0.5 km of the tropopause to avoid the sharp transition to stratospheric air producing spurious false positives. Then using ozone perturbations from 2 km above the surface up to 1 km below the tropopause, we create a threshold for each launch site at the 99th percentile. Profiles with perturbations exceeding this threshold are initially classified as STT events.

The ozone peak is defined at the altitude where the OMR is greatest within the lowest range of altitudes where the perturbation profile exceeds the percentile based threshold. If the OMR between this ozone peak and the tropopause drop below 80 ppb and are more than 20 ppb lower than the peak ozone then the initially classified event is confirmed, otherwise the profile is rejected as a non-event. This confirmation is only required if the perturbation profile does not drop below zero between the event peak and the tropopause. This happens in order to remove ‘near tropopause’ anomalies for which there is no evidence of detachment from the stratosphere.

We conservatively estimate the ozone flux into the troposphere associated with each event. The estimate is conservative since it does not take into any secondary ozone enhancements which may have been caused by the STT, as well as ignoring any heightened ozone background levels which may be due to synoptic-scale stratospheric mixing into the troposphere. The ozone concentration is integrated vertically over the altitude range for which an STT event is identified. Figure 3 shows an example ozone profile, and how the algorithm detects an STT event, defines the event boundaries, and calculates the ozone flux.

2.3 Sensitivities and limitations

There are several observationally defined thresholds and limits which have an effect on how many events are detected, what altitude within which they can be detected, and how strongly the events are separated from the stratosphere.

The cutoff threshold (defined locally to each site) is determined from the 99th percentile of the filtered ozone profile between 2 km and the tropopause height minus 1 kilometer. If an ozonesonde’s filtered profile (between 4 km and the tropopause minus 500 m) goes above this threshold then the profile is flagged as an event. Changing either of these altitude ranges, or the cut-off threshold, changes how many events are detected. For example, using the 98.5th percentile increased detected events by 26 and Melbourne, 18 at Mac-

Ozone at Melbourne 2004/01/08

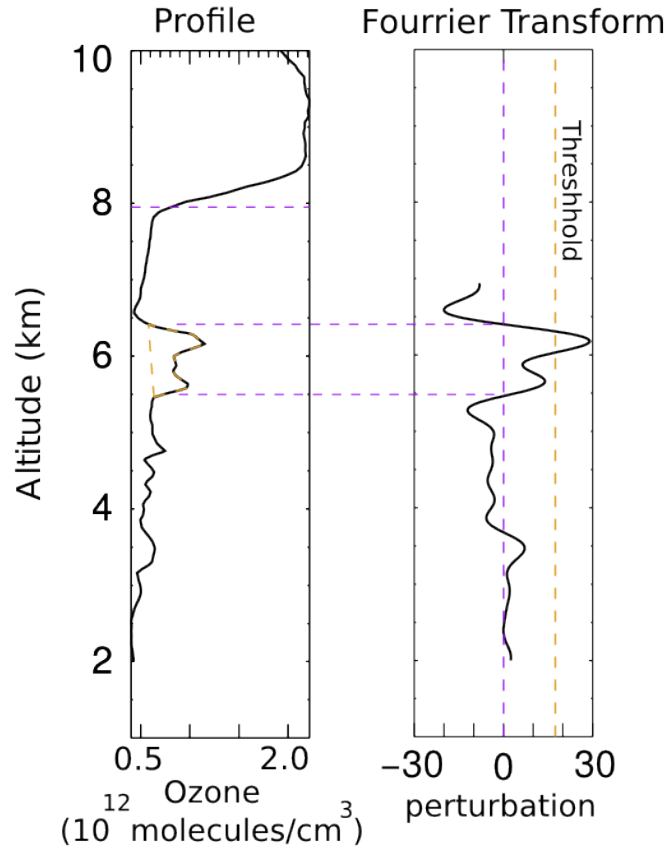


Figure 3: Left: an example illustrating methods used for STT identification and flux estimation using an ozone profile from 2km to the tropopause (dashed vertical line). At Melbourne on the 8th of January 2004 the flux area shows the estimate of stratospheric impact on tropospheric ozone. Right: band pass filtered O₃ ppb perturbation profile. Coloured lines show the 99th percentile of filtered ozone perturbations (purple dashed) and the technique for determining the vertical extent of the event (orange dashed) is outlined in the text. See text for details.

quarie Island, and 9 at Davis. We use the 99th percentile because at this point the filter locates clear events with no obvious false positives.

The altitude range for flagging filtered profiles is set from 4 km above the surface to 500 m below the tropopause. This range removes possible ground pollution effects as well as local fire smoke plumes which are not likely to ascend above 4 km, as well as allowing event detection up to 500 m from the tropopause. Some events, including the storm-caused event examined in figure 5 are within one kilometer of the tropopause.

The altitude range used to determine the 99th percentile is set from 2 km up to 1 km below the tropopause. This range removes any anomalous edge effects of the Fourier filter, as well as discounting the highly variable ozone concentration which occurs near the tropopause.

2.4 Removal of biomass burning influence

Other sources of tropospheric ozone profile perturbation need to be analysed and excluded before drawing any conclusions about STTs based on recorded ozone profiles. The major possible ozone influence other than STTs in the troposphere above 4 km is smoke plumes from biomass burning.

Ozone production from biomass burning is complex and affected by photochemistry, fuel nitrogen load, time since emission, and atmospheric plume chemistry both during transport and at the point of measurement. Large biomass burning events emit substantial ozone precursors, some of which are capable of being transported far from their origins. Peroxyacetyl Nitrate (PAN) is a reservoir of NO_x which can lead to enhanced ozone far from the source of a fire [Jaffe and Wigder, 2012].

Biomass burning influence in the Southern Hemisphere comes mostly from Southern Africa and South America, however Australian fires from the mid-latitudes, and Indonesian fires can also influence the ozonesonde release sites. Transported biomass burning plumes influence the southern mid-latitudes generally between July and December [Pak et al., 2003]. Biomass burning smoke plumes can lead to enhanced ozone, however this is not always the case. Due to the possible influence of smoke plumes on identification of STT events we exclude from analysis any dates where they are seen near the ozonesonde launch sites.

Removal of any possible influence from biomass burning smoke plumes is performed by detection of smoke plumes through global CO measurements. Here we identify transported smoke plumes through enhanced carbon monoxide (CO) levels. CO has a long enough lifetime to be an effective tracer of transport. The primary source of atmospheric enhancement of CO is fires, making CO a good indicator of fire plumes. Using high CO levels as a proxy for smoke plumes is a well established method (eg: Edwards [2003], Sinha et al. [2004], Edwards et al. [2006], Mari et al. [2008]). We use data from the AIRS (Atmospheric Infrared Sounder) instrument on board the Aqua satellite [AIR, 2013]. We visually inspect AIRS' vertical columns of CO over the Southern Hemisphere to exclude events with possible smoke influence at our three sites. We diagnose

smoke plumes where high ($\approx 2 \times 10^{18}$ molecules cm^{-2} or higher) CO columns appear and when these occur near our sites during a sonde-detected ozone event we remove the event from STT analysis.

Figure 4(top) shows a day where smoke plumes are near the Melbourne sonde launch site on the day of a detected event. An event preliminarily detected on this day through the ozonesonde data is flagged as possibly due to fire. In the figure elevated CO levels can be seen over Australia, likely due to long-range transport from African and/or South American biomass burning. This day can be contrasted with the example in figure 4(bottom) where low CO levels are observed over the entire Southern Hemisphere. We screened all days at all three sites where an STT event is detected except for one event that coincided with missing AIRS data (January 2010). We flagged 15 of 72 events over Melbourne, 8 of 48 events over Macquarie island, and none from 45 over Davis. Nearly all of the discarded events occur within the burning season of the Southern Hemisphere.

We also include on Figure 7 to Figure 9 the events which have possible fire influence. These events are concentrated in spring at Melbourne and Macquarie Island.

3 Case Studies of synoptic conditions during STT events

We examine two case studies in detail to illustrate the synoptic-scale conditions in which STT events occur over Melbourne. Data from the European Center for Medium-range Weather Forecasts (ECMWF) Interim Reanalysis (ERA-I) [Dee et al., 2011] product is used for synoptic-scale examination of weather patterns over our three sites on dates matching detected STT events.

Figure 5(left) shows the ozonesonde profile recorded on the 3rd of February 2005 above Melbourne. Both tropopause definitions are between 400 and 500 hPa and the ozone spikes have clear anticorrelations with the relative humidity, suggesting dry stratospheric air is measured here. An ozone intrusion into the troposphere is identified by our detection algorithm at ~ 520 hPa. Figure 5(right) shows the synoptic weather system, a cut-off low pressure system which caused a large storm and lowered the local tropopause height for several days. The wind circles around the low pressure system in a clockwise direction, typical geostrophic flows which are caused by pressure gradients and coriolis forces. The flux of stratospheric ozone brought into the troposphere by this event is at least 3.1×10^{11} molecules cm^{-3} , or 8% of the tropospheric ozone column.

Figure 6(left) shows the vertical ozonesonde profile recorded on the 13th January 2010 over Melbourne. The tropopause heights are greater at this time and an ozone intrusion is identified centred around 200 hPa. Again, anticorrelated relative humidity provides evidence that the air is stratospheric in origin. Note the separation between this intrusion and the ozone tropopause (marked by the black dashed line), which indicates that the sonde passes through reg-

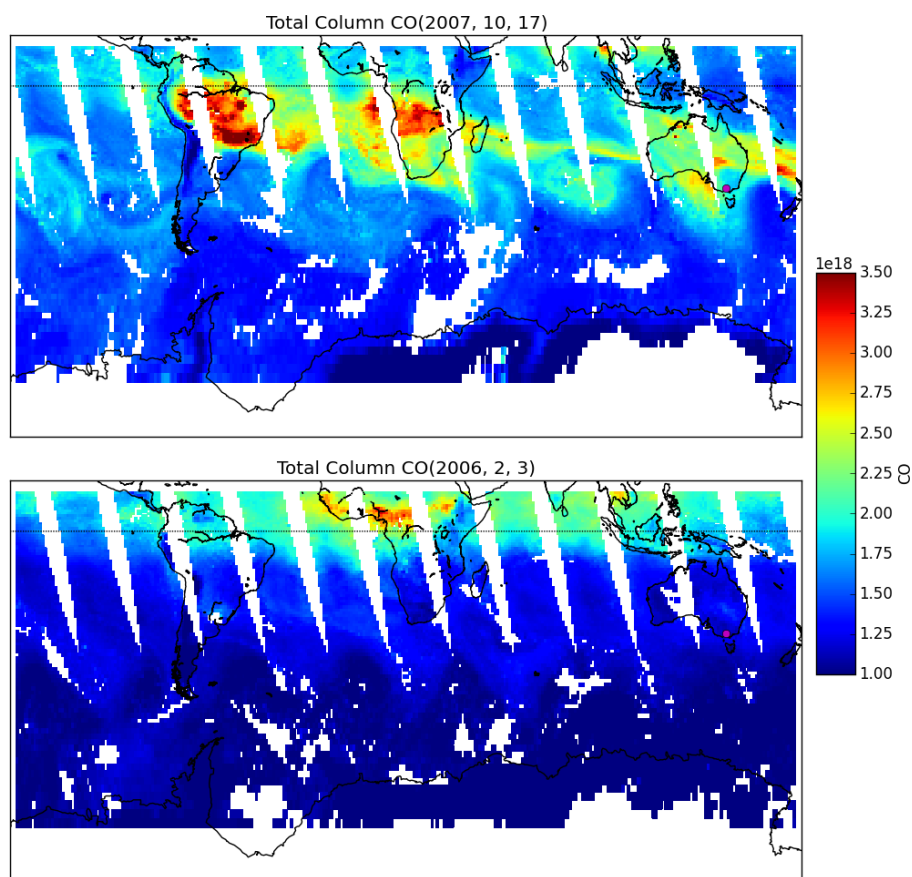


Figure 4: AIRS total column CO. The top panel (17 October 2007) is a day when ozone above Melbourne (purple dot) could have been caused by a transported biomass burning plume, and so was excluded from analysis. The bottom panel (3 February 2006) shows an example of a day when Melbourne ozone was likely not influenced by transported smoke plumes and was retained for analysis.

Melbourne 3/2/05

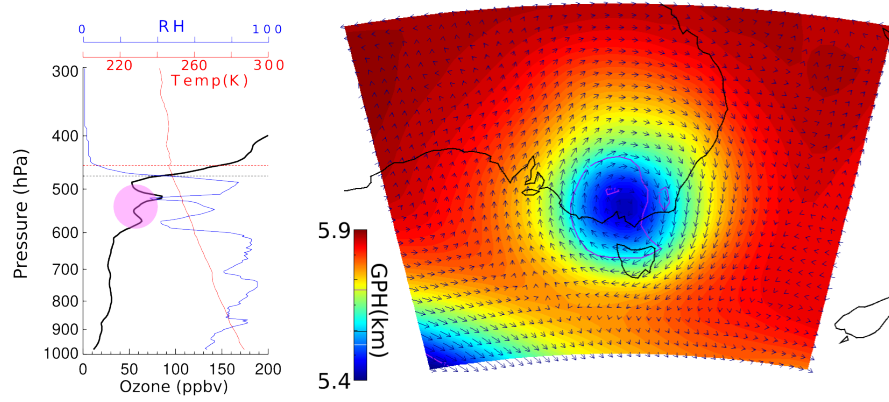


Figure 5: (Left) Vertical profile of ozone (black line), relative humidity (blue line), and temperature (red line) for 3 February 2005. The STT ozone event is highlighted in pink. The tropopause heights using both the ozone definition (black dashed line) and lapse rate definition (red dashed line) are shown. (Right) Synoptic weather map at 500 hPa from the ERA-Interim reanalysis. Vectors show wind direction and speed while colour indicates the geopotential height. Also visible are contours of potential vorticity units with 1 PVU in purple and 2 PVU (often used to determine dynamical tropopause height) in white.

ular tropospheric air after hitting a stratospheric intrusion but before reaching the tropopause. Figure 6(right) shows a trough of low pressure (front) passing over south-eastern Australia. This low pressure system crosses west to east and causes a wave of lowered tropopause height, which is often the cause of stratospheric mixing. During frontal passage, stratospheric air descends and streamers of ozone-rich air break off and mix into the troposphere [Sprenger et al., 2003].

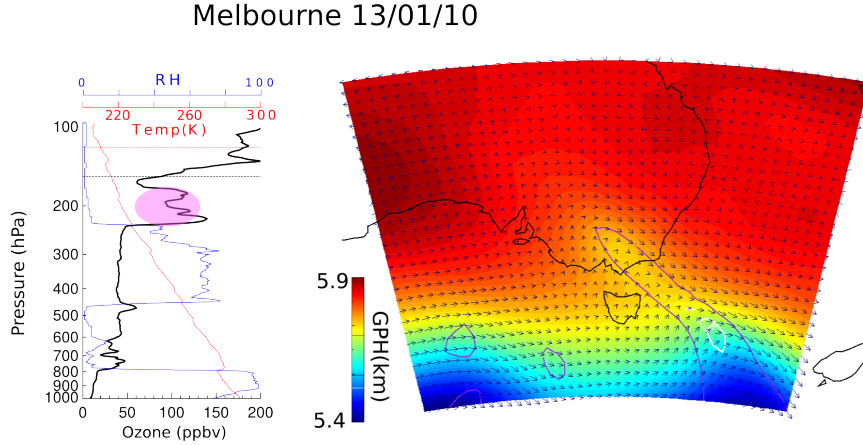


Figure 6: As figure 5, for 13 January 2010.

We investigate other STT events above Melbourne, Macquarie Island, and Davis, using reanalysis data. Similar characteristics to the case studies presented here occur over Macquarie Island: i.e. a prevalence of frontal and low pressure activity during STT events. Typically during STT occurrence, the upper troposphere is not calm, with low pressure fronts or cut-offs nearby at coincident time. Over Davis the weather systems are harder to distinguish, and the polar vortex may create ozone folds without other sources of upper tropospheric turbulence.

4 STT event climatologies

Figure 7 shows the seasonal cycles of the STT events for Melbourne, Macquarie Island, and Davis. There is an annual cycle with a summertime peak in the frequency of STT events above Melbourne and Macquarie Island. This summertime peak is due to a prevalence of summer storms, with low pressure systems bringing storms and turbulence along with a lowered tropopause level [Reutter et al., 2015]. TODO: weather and summer storms citation.

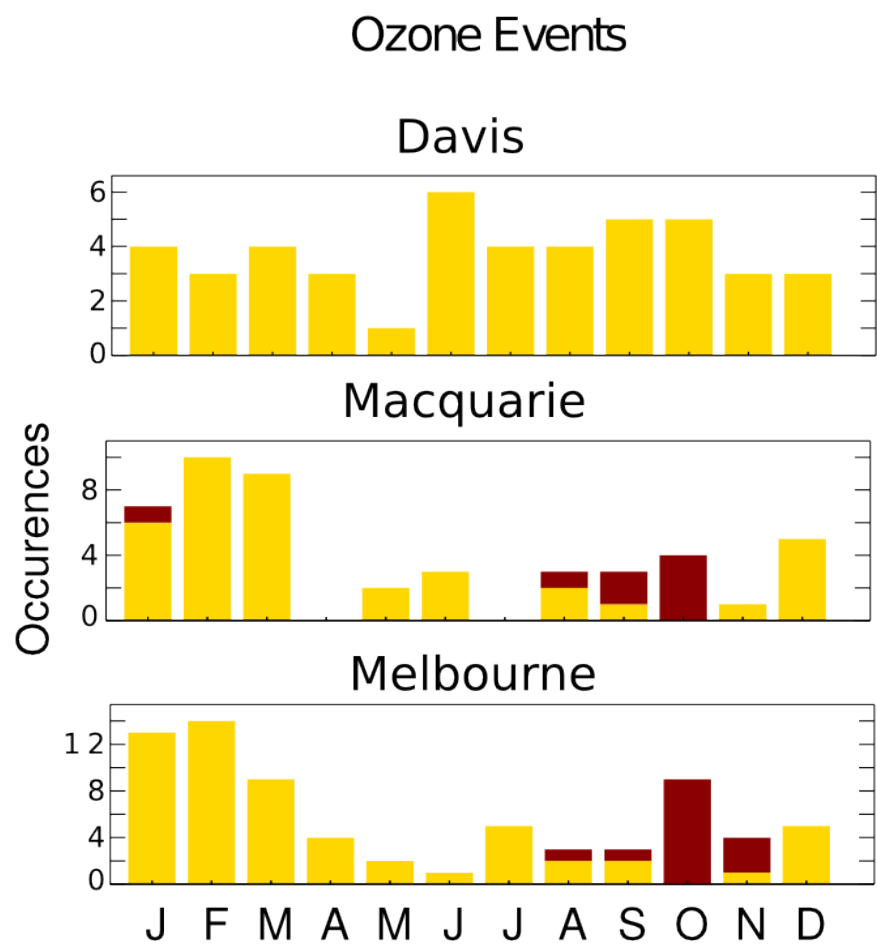


Figure 7: The seasonality of STT events at Davis, Macquarie Island, and Melbourne. The events filtered out as possibly smoke plume influenced are indicated in red.

The frequency of STT events above Davis is relatively constant throughout the year, with a slight increase in events during antarctic winter. The slightly increased winter time frequency can be attributed to the increased frequency of sonde releases during the June to October months over Davis. It could be that events are non seasonal at Davis, or else that the sample of 45 detected events over 10 years is too small or sparse to clearly show any cycle. It is possible that summer events caused by upper troposphere turbulence are balanced out by the events caused by the polar front jet stream, which is strongest during antarctic winter. The polar front jet stream is a band of wind extending from the mid troposphere up to the lower stratosphere, which is generally active from winter to spring. This vortex may be directly causing or impacting many of the STTs due to the lowered tropopause altitude which occurs south of the vortex edge (around 60° S).

Figure 8 shows the altitudes of detected events, based on the peak of tropospheric ozone. STT event altitudes most commonly occur at 6 – 10 km above Melbourne and below 8 km at Davis but are distributed more evenly at Macquarie Island. Figure 9 shows the depths of detected events, based on the ozone peak’s distance from the minimal determined tropopause. The majority of events occur within 3 km of the tropopause at both Melbourne and Macquarie Island, and within 2 km of the tropopause at Davis.

For both Melbourne and Macquarie Island, the STT events which are unlikely to be fire-related occur mostly in summer and mostly during low pressure synoptic systems which can increase convection and upper tropospheric turbulence.

5 Stratosphere to troposphere ozone flux from STT events

Based on the integrated ozone amount associated with each STT event (see section 2.2), we find a lower bound for the STT ozone flux over each of our three sites (fire influence excluded). This is a conservative lower bound as the algorithm ignores secondary ozone peaks which may also be transported down from the stratosphere, as well as ignoring potential ozone dispersion from the ozone peak. Figure 10 shows the mean fraction of total tropospheric column ozone attributed to stratospheric ozone intrusions at each site, averaged over days when an STT event occurs. The mean fraction of tropospheric ozone attributed to STT events is 2–4%, on individual days this value can exceed 10% at all locations. Figure 11 shows the data in absolute terms, and indicates that the mean STT event impact is around 2×10^{11} molecules/cm³. Our flux estimates are relatively insensitive to our biomass burning filter; including smoke-influenced days changes the mean flux by less than 5% (relative).

Extrapolating out over the Southern Ocean using our estimate of 3% enhanced tropospheric ozone, we can create a rough estimate of the STT effect on tropospheric ozone in this region. This can be done by multiply-

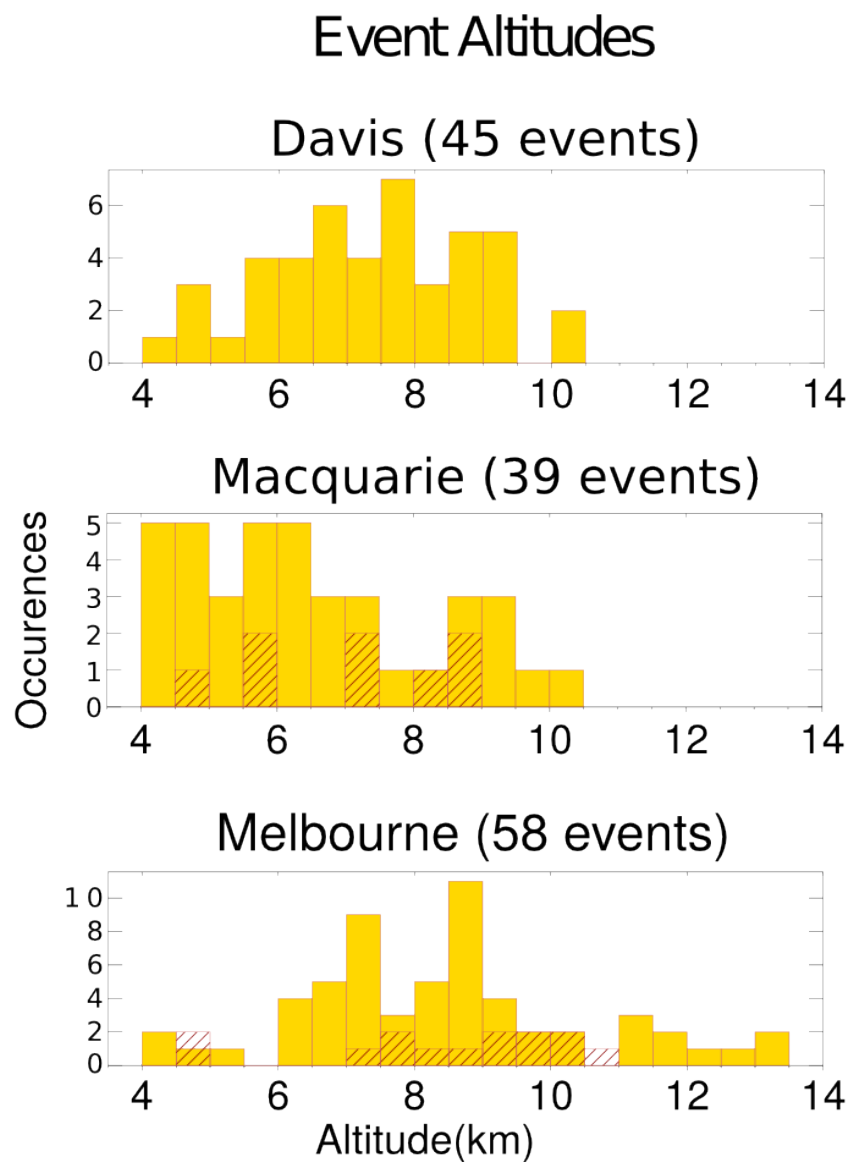


Figure 8: The distribution of the ozone peak altitude for Davis, Macquarie Island, and Melbourne. This shows the altitude of detected events, based on the tropospheric ozone enhancement peak. Red diagonal lines show the removed smoke influenced event altitudes.

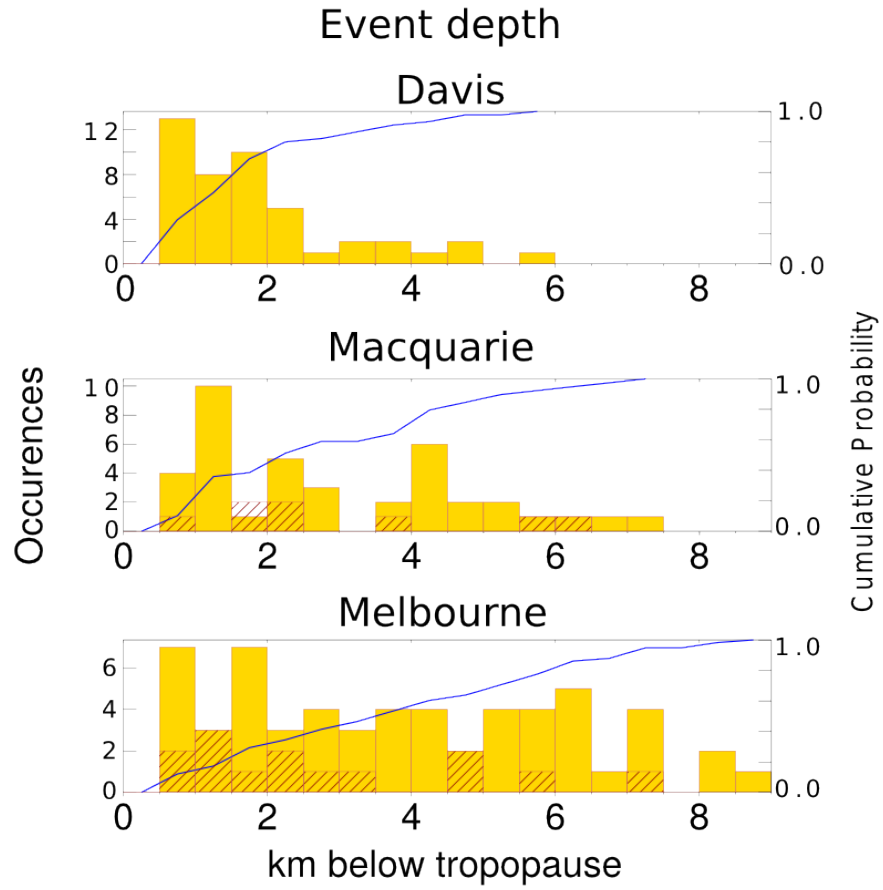


Figure 9: The distance between the ozone peak and the tropopause, and the cumulative probability of these distances (blue line) for Davis, Macquarie Island, and Melbourne. This shows the depth of the event into the troposphere, starting from the tropopause. Red diagonal lines show the removed smoke-influenced event depths

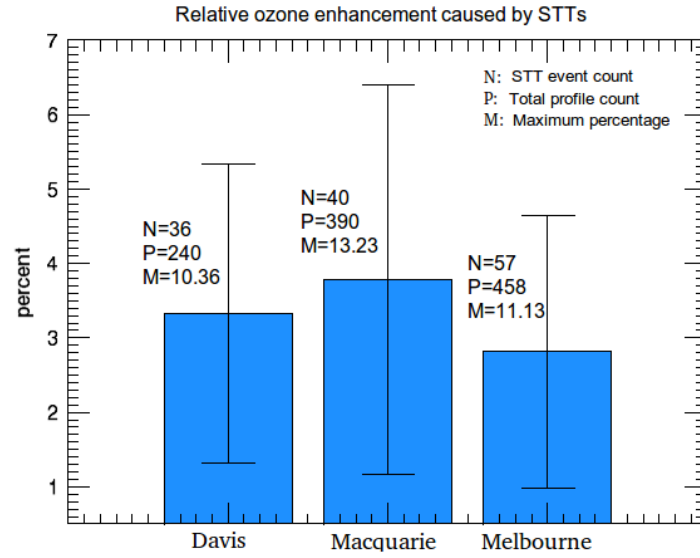


Figure 10: Fraction of total tropospheric column ozone attributed to stratospheric air intrusions during STT events. Error bars indicate one standard deviation.

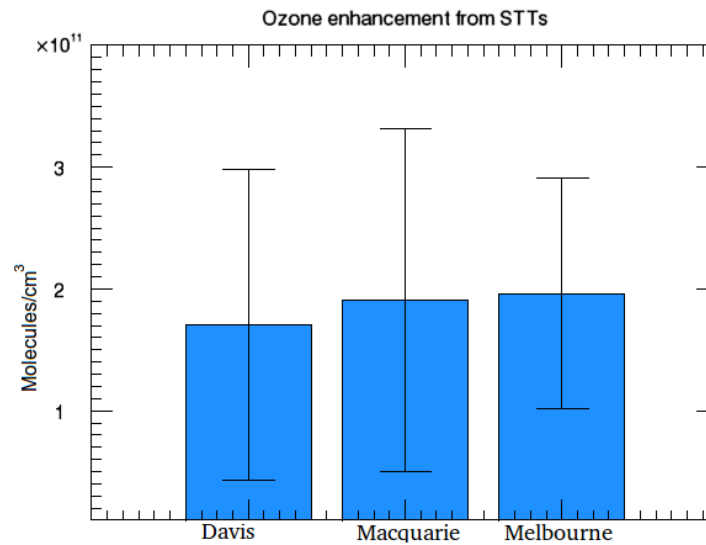


Figure 11: Tropospheric ozone attributed to stratospheric air intrusions during STT events. Error bars indicate one standard deviation.

ing the monthly likelihoods of STTs with the monthly tropospheric column ozone amounts multiplied by our mean flux fraction. Taking the monthly likelihood from our ozonesonde events count per sondes released during each month, and southern latitude tropospheric column ozone amount from GEOS-Chem, (TODO:Southern Oceanic Trop columns from GEOS-Chem.) The total amount of ozone from STT events over the southern ocean is at least (TODO:find x) X kg per m²

6 Conclusion

Ozonesonde data in the Southern Hemisphere provides a satellite-independant quantification of STT ozone transport. The frequency and amount of ozone descending from the stratosphere into the troposphere can be estimated from the long time series of tropospheric ozone profiles. Using almost ten years of ozonesonde profiles over the southern high latitudes, a clear summer peak is seen for STT occurences at both 38°S and 55°S, although not at 69°S.

We use a band-pass filter to determine STT ozone transport events. The filter removes seasonal tropospheric ozone influences and allows clear detection of ozone-enhanced tongues of air in the troposphere. By setting empirical checks, ozonesonde vertical profiles can clearly show tropospheric ozone enhancement which is separated from the stratosphere. The cause of these ozone enhancements is examined through the use of satellite and reanalysis datasets on case studies above Melbourne. The major causes of STT events found over Melbourne are turbulent weather in the upper troposphere due to low pressure fronts and cut-off low pressure systems.

Integration of the ozone enhancement along the altitude of the ozone profile allows a rough estimate of stratospheric transport for each event. Events typically cause a 3% enhancement of the tropospheric ozone column. This is around $2 * 10^{11}$ molecules/cm³ ozone enhancement over the southern high latitudes caused by STTs.

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