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Stratosphere to troposphere ozone event characterisation and distribution over Melbourne,

Macquarie Island, and Davis.

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May 26, 2016

Abstract

We develop a quantitative method for determining Stratosphere to Troposphere Transport events (STTs) from ozone sonde profiles over southern high latitudes. Using this method we estimate transported ozone quantity using over Melbourne, Macquarie Island, and Davis. STT seasonality is determined from a 7-9 year long time series of ozone profiles. An examination of causes of tropospheric ozone enhancement follows.

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1 Introduction

Tropospheric ozone is important for both air quality and climate change. Over the industrial period, tropospheric ozone, which is the third most potent greenhouse gas, has been estimated to exert a radiative forcing equivalent to a quarter of the $\rm CO_2$ forcing (TODO: citation?). Ozone is present in the troposphere due to a variety of dynamical and photochemical processes, including downward transport from the ozone-rich stratosphere and anthropogenic pollution. The primary sources of tropospheric ozone are chemical creation and stratospheric input. Using 17 models from the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP), Stevenson et al. [2006] estimates that 5100 ± 600 Tg/yr and 550 ± 170 Tg/yr of ozone in the troposphere is due to chemical creation and stratospheric transport respectively. Primary sinks are

5TT events have been observed is topopour around 50th the polar-front jet and the subtropical (Vaughan et al 1994, jet (Baray et al 2000) and cut-off laws (Price Luanghan 1993,

> chemical destruction and dry deposition, estimated to be $4700 \pm 700 \,\mathrm{Tg/yr}$ and $1000 \pm 200 \text{ Tg/yr}$, respectively. The total radiative forcing caused by tropospheric ozone is estimated at 377±65 mWm⁻². — reference.

> Ozone-rich air mixes irreversibly down from the stratosphere during meteorologically conducive conditions [Sprenger et al., 2003, Mihalikova et al., 2012], referred to as Statosphere - Troposphere Transport events (STTs). In the extra-tropics, STTs most commonly occur during synoptic-scale tropopause folds [Sprenger et al., 2003, Tang and Prather, 2012] and are characterised by tongues of high Potential Vorticity (PV) air descending to low altitudes. These tongues (also called ozone folds) become elongated and filaments disperse away from the tongue and mix irreversibly into the troposphere. Stratospheric ozone brought deeper (lower) into the troposphere is more likely to affect the surface ozone budget and tropospheric chemistry [Zanis et al., 2003, Zhang et al., 2014]. 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. Irreversible STT of ozone is important for explaining tropospheric ozone variability [Tang and Prather, 2012].

> In a future climate, a warmer, wetter troposphere will change the chemical processing of ozone. Dynamical processes such as STT, boundary layer ventilation and convection will change and alter tropospheric ozone distributions [Hegglin and Shepherd, 2009]. Hegglin and Shepherd [2009] estimate that climate change will lead to increased STT of the order of 30 (121) Tg yr⁻¹ relative to 1965 in the southern (northern) hemisphere due to an acceleration in the Brewer Dobson circulation.

In section 2 we use several years of ozonesonde flights from three locations spanning the 69°S - 37°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. Additionally a new method to deterministically define a tropospheric ozone fold or STT events is tested and some basic analysis is performed. Lastly, we calculate the

2 Data and Methods

2.1Ozonesonde 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. (TODO: precision of ozone recordings?)

Ozonesondes are launched approximately weekly from Melbourne (145°E, 38°S), Macquarie Island (159°E, 55°S) and Davis (78°E, 69°S). For this study, we use the data collected from 2004-2013 for Melbourne and Macquarie, and 2006-2013 for Davis. More frequent ozonesonde launches occur at Davis during the spring ozone hole season (September-December) than at other times of the

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year. Over the time span we observed, around twice as many ozone launchesoccurring between June and October than during the other months at Davis station

2.2 Characterisation of STT events and associated fluxes

Stratospheric ozone typically mixes irreversibly down into the troposphere in a kilometres-scale tongue 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 $2*\circ Ckm^{-1}$, provided the lapse rate between this altitude and all subsequent altitudes within 2 km is also below $2*\circ Ckm^{-1}$. 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 $\rm km^{-1}$
- 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 defintion 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 souther latitudes. 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.

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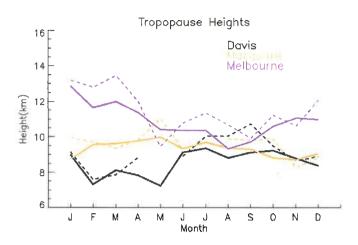


Figure 1: Monthly mean tropopause altitudes (minimum of lapse-rate and ozone defined tropopause at 3 sites) determined from ozone sondes. Dashed lines show the average monthly altitude when only considering dates when STTs occured.)

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Figure 2 shows seasonally averaged ozone as recorded over the three stations. Apparent is the increased ozone extending down through the stratosphere 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.

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 20ppb 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.

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 \sim 5 km removes seasonal-scale

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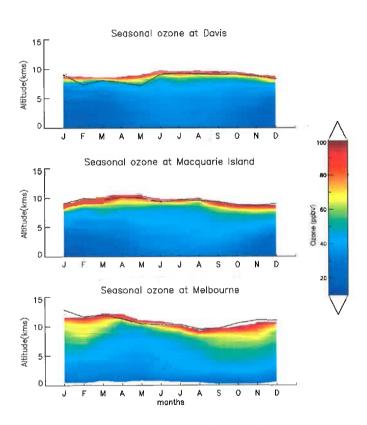


Figure 2: Seasonal cycle of ozone over Davis, Macquarie, and Melbourne measured by ozonesondes, where measurements are binned monthly. Black solid lines show seasonal tropopause heights, defined as described in the text.

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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 threshhold for each launch site at the 99th percentile. Profiles with perturbations exceeding this threshhold 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 threshhold. If the OMR between this ozone peak and the tropopause drop below 80ppb and are more than 20ppb 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 stratospheric mixing. 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 threshholds 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 cutoff threshold, changes how many events are detected. For example, using the 98.5th percentile increased detected events by 26 and Melbourne, 18 at Macquarie 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 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.

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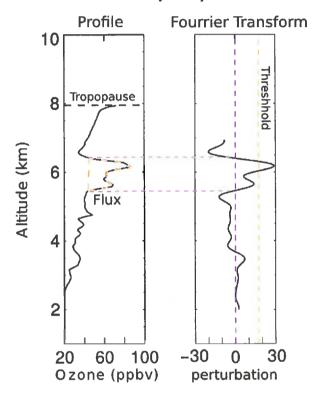


Figure 3: Left: an example illustrating methods used for STT identification and flux estimation using an ozone profile from 2km to the tropopause (dashed vertical line). At Melbourne on the 8th of January 2004 the flux area shows the estimate of stratospheric impact on tropospheric ozone. Right: band pass filtered ${\rm O}_3$ profile from the left. 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). NB: The area denoting the flux is actually calculated using the ozone density profile (molecules/cm³) rather than the ozone ppb profile shown here.

See text for details.

oke so why not show 7 og dusity profile? 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 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 ($\approx 2*10^{18}$ molecules cm⁻² or higher) CO columns appear and when these occur near our sites during a sond edetected ozone event we remove the event from STT analysis.

we remove the event from STT analysis.

Figure 4(top) shows a day where smoke plumes are near the sonde launch site on the day of a detected event. This detected event 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 BB 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

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with missing AIRS data (January 2010). We discarded 15 of 72 events over Melbourne, 8 of 48 events over Macquarie island, and none over Davis. Nearly all of the discarded events occur within the burning season of the southern hemisphere.

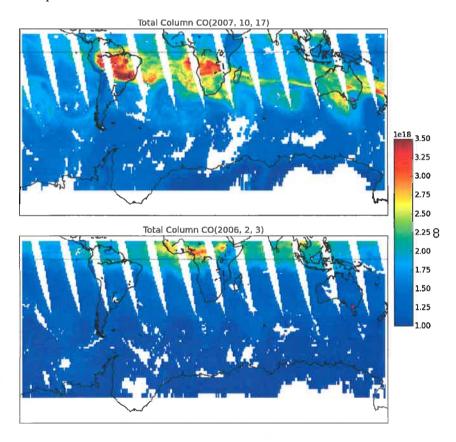


Figure 4: AIRS total column CO. The top panel (17 October 2007) is a day when ozone above Melbourne (purple dot) could have been caused by a transported biomass burning plume, and so was excluded from analysis. The bottom panel (3 February 2006) shows an example of a day when Melbourne ozone was likely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

Using 458 ozonesonde profiles over Melbourne, 72 ozone events are detected, of which 14 are discarded as possibly caused by transported fire smoke plumes. Over Macquarie island 47 events are detected from 380 ozonesondes of which 8 are discarded due to possible smoke influence. 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. For Davis, 45

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events are detected from 240 ozonesonde profiles, none of which are discarded due to smoke influence.

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. 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} 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 highly anticorrelated relative humidity provides evidence that the air is descended from the stratosphere. Note the separation between this intrusion and the ozone tropopause (marked by the black dashed line), which suggests that the sonde passes through regular tropospheric air after hitting a stratospheric intrusion but before reaching the tropopause. Figure 6(right) shows a frontal low 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 the frontal passage, stratospheric air descends and streamers of ozone-rich air beely break off and mix into the troposphere [Sprenger et al., 2003].

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 is as one would expect since Summer weather is more storm prone, with low pressure systems bringing storms and turbulence along with a lowered tropopause level [Reutter et al., 2015]. TODO: weather and summer storms citation.

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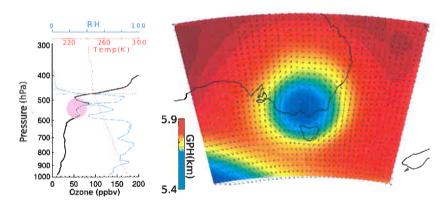


Figure 5: (Left) Vertical profile of ozone (black line), relative humidity (blue line), and temperature (red line) for 3 February 2005. The STT ozone event is highlighted in pink. The tropopause heights using both the ozone definition (black dashed line) and lapse rate definition (red dashed line) are 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.

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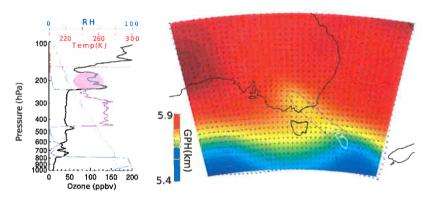


Figure 6: As figure 5, for 13 January 2010.

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.

Davis Macquarie Melbourne 12 8 4 0 J F M A M J J A S O N D

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.

Event Altitudes Davis (45 events) Altitude(km) Macquarie (39 events) Occurences Altitude(km) Melbourne (58 events) 4 2 0 Altitude(km)

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.

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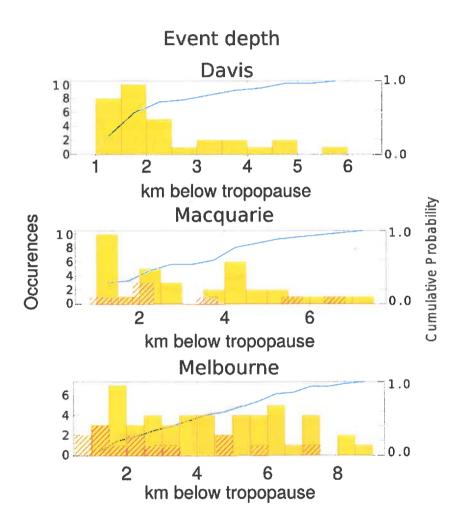


Figure 9: The distance between the ozone peak and the tropopause, and the cumulative probability fraction of these distances (blue line) for Davis, Macquarie Island, and Melbourne. This shows the depth of the event into the troposphene, starting from the tropopause. Red diagonal lines show the removed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

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Stratosphere to troposphere ozone flux from 5 STT events

Stratosphere to troposphere ozone transport can potentially increase regional surface ozone levels above safe levels [Zhang et al., 2014]. Based on the integrated ozone amount associated with each STT event (see section 2.2), we find a lower bount 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. Shown in figure 11 shows the data is the same figure using absolute terms, showing the mean STT event impact is around 2 * 10¹¹ 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).

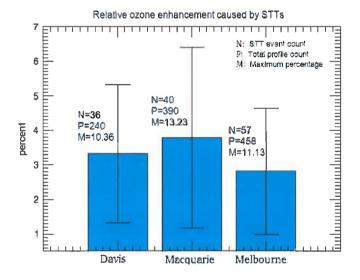


Figure 10: Fraction of total tropospheric column ozone attributed to stratospheric air intrusions during STT events. Error bars indicate one standard deviation.

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 over the southern ocean. This can be done by multi-

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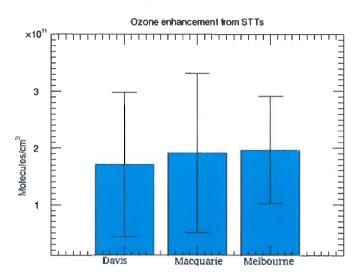


Figure 11: Tropospheric ozone attributed to stratospheric air intrusions during STT events. Error bars indicate one standard deviation.

plying 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 m^2

6 Conclusion

Using ozonesonde data in the southern hemisphere can allow an overview of STT ozone transport which is independent of satellite data. 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 anti-note summer peak is seen for STT occurences at both 38°S and 55°S, although not so-clearly at 69°S.

Running a Fourier filter allows deterministic and quantitative analysis of 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, OMR 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

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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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See 3 can you make some comment shot a typical STT meteorlogy is like?

about Davis?

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