

Simon's comments.  
01/02/16

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

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## Abstract

We develop a quantitative method for determining Stratosphere to Troposphere Transport events (STTs) and a minimum bound for this transported ozone quantity using ozonesondes over Melbourne, Macquarie Island, and Davis.

## 1 Introduction

### 1.1 Background

Tropospheric ozone is important for both air quality and climate change. Over the industrial period, tropospheric ozone, the third most potent greenhouse gas, has been estimated to exert a radiative forcing equivalent to a quarter of the CO<sub>2</sub> forcing. The primary sources of tropospheric ozone are chemical creation and stratospheric input, estimated using a model ensemble to be  $5100 \pm 600$  Tg/yr and  $550 \pm 170$  Tg/yr, respectively. The primary sinks are chemical destruction and dry deposition, estimated to be  $4700 \pm 700$  Tg/yr and  $1000 \pm 200$  Tg/yr, respectively Stevenson et al. [2006].

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. Ozone-rich air mixes irreversibly down from the stratosphere during meteorologically conducive conditions Sprenger et al. [2003], Mihalikova et al. [2012]; these are referred to as Statosphere -

LaTeX:

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\citet

Use \citep here

& \citet when quoting  
reference  
in a  
sentence.

⊗ Thus, stratospheric ozone accounts<sup>1</sup> for around 10% of  
tropospheric ~~ozone~~ sources [correct?]

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citation?  
Stevenson 2006?

Troposphere Transport events (STTs). In the extra-tropics, STTs most commonly occur during synoptic-scale tropopause folds Sprenger et al. [2003] and are characterised by tongues of high Potential Vorticity (PV) air descending to low altitudes. These tongues become elongated and filaments separate from the tongue which mix into tropospheric air. 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].

irreversibly

While the amount of tropospheric ozone is small compared with that found in the stratosphere, it is an important constituent. The relative contributions to the tropospheric ozone budget of photochemistry and STT (dynamical transport) is still uncertain Zanis et al. [2003] (TODO: more recent source). A high correlation is found between lower stratospheric and tropospheric ozone Terao et al. [2008] with the highest STT associated with the jet-streams over the oceans in winter. Irreversible STT of ozone have been shown to be important for explaining tropospheric ozone variability Tang and Prather [2011].

but you quote that model in 1st paragraph?

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

Using several years of ozonesonde flights from three mid-latitude locations in the Southern Hemisphere, we will characterise the seasonal cycle of STT events and determine their contribution to the total amount of tropospheric ozone. We will examine the depth of the intrusions and using case studies, relate these STT to meteorological events.

spanning the latitudes of the Southern Ocean,

## 1.2 Instruments and Data

Ozonesondes are weather balloons with an attached instrument which measures ozone concentrations roughly every 100m up to around 30km. These ozonesondes provide a vertical profile of ozone. (TODO: precision of ozone recordings?)

Ozonesondes are launched approximately weekly from Melbourne (145E, 38S), Macquarie Island (159E, 55S) and Davis (78E, 69S). For this study, we use the data collected from 2004-2013 for Melbourne and Macquarie, and 2006-2013 for Davis. A larger number of ozonesonde flights occurred in these more recent years compared with earlier times. More frequent ozonesonde launches occur at Davis during the spring ozone hole season than at other times of the year Alexander et al. [2013].

Ozonesondes provide much higher vertical resolution profiles of ozone than that available from reanalyses products. However, one data point per week from an ozonesonde flight is too low to be useful to diagnose the evolution of STT exchange over time-scales associated with normal synoptic scale weather patterns present in the extra-tropics. Instead, the ozonesonde data are supplemented with the ERA-Interim reanalysis Dee et al. [2011] to enable construction of an STT exchange climatology.

high-vertical resolution

\$145^{\circ}\$ circ \$^{\circ}\$ etc.

in itself

① While ozonesondes were launched prior to 2004 at the two northern sites, these were less frequent than launches after 2004 so the analysis focuses on results from these more recent times

## 2 Methods

### 2.1 Characterisation of STTs

STT events are characterised in the vertical profiles of ozone as altitudes in the troposphere where the ozone mixing ratio exceeds a specified threshold. Usually stratospheric ozone mixes irreversibly down into the troposphere in a synoptic-scale tongue of air: the vertical ozone profile observed by the ozonesonde depends upon the time in this cycle that it is observed Sprenger et al. [2003]. As such, 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.

Two definitions of the tropopause height are calculated: the standard lapse rate tropopause WMO [1957], and the ozone tropopause Bethan et al. [1996]. At Davis, the ozone tropopause definition is modified for polar sites, following Tomikawa et al. [2009], Alexander et al. [2013]. 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]. The lower of these tropopause altitudes is referred to as the tropopause for this study. This definition avoids occasional overly high tropopause height recordings due to perturbed ozone or temperature measurements. The monthly mean tropopause altitudes at each location are shown in Figure 8 along with the 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 poleward. It is worth noting that tropopause altitudes at Davis may exceed 11 km altitude under certain synoptic conditions Alexander et al. [2013]: the relation of tropopause altitude with STT events will be investigated in detail below.

The vertical profiles of ozone volume mixing ratio are linearly interpolated to a regular grid with 20m resolution up to 14km altitude and are then bandpass filtered so as to retain perturbations on altitude scales between 0.5km - 5km. The choice of band limits is set empirically, but we note that to define an STT event, a clear increase above the background ozone level is needed, and a vertical limit of  $\sim 5$  km removes seasonal-scale effects. The ozone perturbation profile is analysed at altitudes from 2 km above the surface (to avoid surface pollution events) and 1 km below the tropopause (to avoid the sharp transition to stratospheric air producing spurious false positives). Perturbations above the 99th percentile (locally) of all ozone levels are initially classified as STT events.

In order to remove unclear 'near tropopause' anomalies we remove events where the gradient between the maximum ozone peak and the ozone at 1 km below the tropopause is greater than  $-20 \text{ ppbv km}^{-1}$  and simultaneously require that the perturbation profile does not drop below zero between the event peak and the tropopause.

To provide a conservative estimate of ozone flux into the troposphere for each event, the ozone concentration is integrated vertically over the interval for which an STT event is identified. An example of an ozone profile is illustrated

ozonesondes'

two definitions

subset of

which have vertical

unrealistically

Make this Figure 1

individual

(A) choice of the lowest altitude of the tropopause

(B) [We need to comment that our algorithm is not overly sensitive to the specified band limits nor the  $-20 \text{ ppbv km}^{-1}$  limits  $\Rightarrow$  well I hope it's not anyway!]

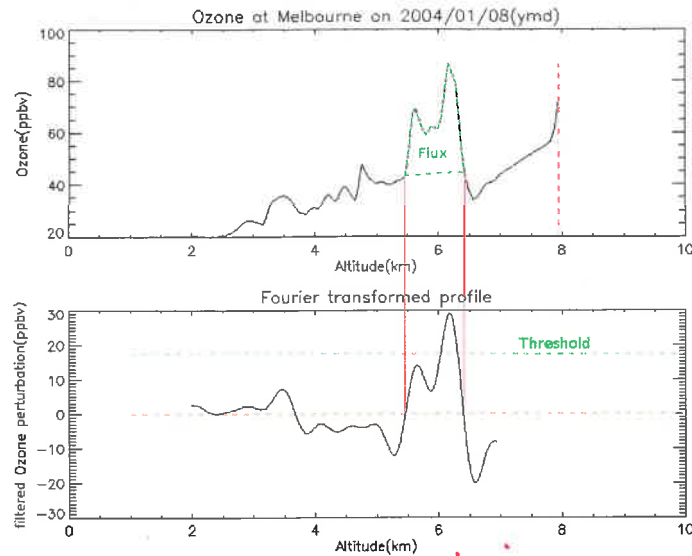


Figure 1: (a) An ozone profile between 2km altitude and the tropopause (indicated by the dashed vertical line). The 'flux' area shows the estimate of stratospheric impact on ozone. (b) The 99th percentile of filtered ozone perturbations (green dashed line) and the technique for determining the vertical extent of the 'event' (red dashed and solid lines).

in Figure 1 and indicates how the algorithm detects an STT event, defines the event boundaries, and calculates the ozone flux.

## 2.2 Filtering Smoke Plumes

Ozone production due to fire smoke plumes is complex and affected by photochemistry, fuel nitrogen load, and atmospheric plume interactions both during transport and at the plume's destination. Ozone precursors include nitrogen oxides ( $NO_x = NO + NO_2$ ) and non methane volatile organic compounds (NMVOCs). 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 South Africa and South America, however Australian and Indonesian fires can also influence our ozonesonde release sites. Transported BB plumes influence the southern mid latitudes generally between July and December Pak et al. [2003]. Ozone production due to fire smoke plumes is complicated and dependent on many chemical and meteorological factors. Due to this complication and the possible influence of smoke plumes on ozone concentrations, this work excludes

③ During the frontal passage, stratospheric air descends and streamers of high-~~er~~ ozone-rich air likely break off and mix into the troposphere, [cite {Sprnger 2003}].

from analysis any dates where this influence is likely.

CO has a long enough lifetime to ~~observe transport~~, and 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 in order to determine where fire smoke plumes exist, ~~has been done~~ in several studies (eg: Edwards [2003], Sinha et al. [2004], Edwards et al. [2006], Mari et al. [2008]) and is the method used here. The AIRS (Atmospheric Infrared Sounder) instrument on board the AQUA satellite records column CO. In this work a visual inspection of AIRS' vertical columns of CO (provided by NASA AIR [2013]) over the southern hemisphere is used to exclude possible foreign smoke plume influence on the ozone profile at our three sites. Whenever high CO concentrations coincide with sonde-detected ozone events it's possible that the tropospheric ozone spike could be due to transported ozone or ozone precursors. All occasions where these coincidences occur are removed. ~~Using a scale of  $1 \times 10^{18}$  up to  $3.5 \times 10^{18}$  molecules/cm<sup>2</sup> shows possible burning influence, as exemplified in figure 2.~~ All days where an ozone event is detected are checked except for one event which occurs within AIRS' 15 missing days within January 2010. This results in 15 of the 72 events over Melbourne being discarded, and 8 of the 48 events over Macquarie island being discarded.

be an effective transport tracer.

is a well-established method.

[Define "high" → I guess this is a certain threshold?]

intrusion

at all three ozonesonde launch sites

### 3 Results

#### 3.1 Case Studies

We examine two STT case studies in detail to ~~investigate~~ <sup>illustrate</sup> the synoptic scale conditions in which they can occur above Melbourne.

A cut-off low pressure system passed over Melbourne on 3 February 2005 (Figure 3b). The ozonesonde profile indicated low lapse-rate and ozonesonde tropopauses (both > 450 hPa, see Figure 3a). An ozone intrusion into the troposphere is identified by our detection algorithm at ~ 520 hPa.

STT events also occur during frontal passages, an example of which is illustrated in Figure 3d over south-eastern Australia. The tropopauses are much higher at this time and an ozone intrusion is identified centred around 200 hPa. ~~The lowered stratospheric air can be seen along the low pressure front in figure 4.~~

The relative humidity profiles are anticorrelated with ozone in the upper troposphere for these events, indicating again the stratospheric origin of the ozone-rich air mass. ~~[same paragraph]~~

Some of this stratospheric air gets mixed into the troposphere, with one ozonesonde column showing an intrusion at around 200 hPa, with dry ozone rich air peaking below the tropopause (see figure 3 panel c).

Note the separation between this intrusion and the ozone tropopause (marked by the green dashed line), indicating above Melbourne its separation from the stratosphere.

① A day where possible biomass burning possibly affects tropospheric ozone is shown in Figure 2a. Elevated CO levels can be seen above Australia, likely due to long-range transport from African or South American biomass burning. As such, this day is discarded from the ozonesonde database at Melbourne.

PTO



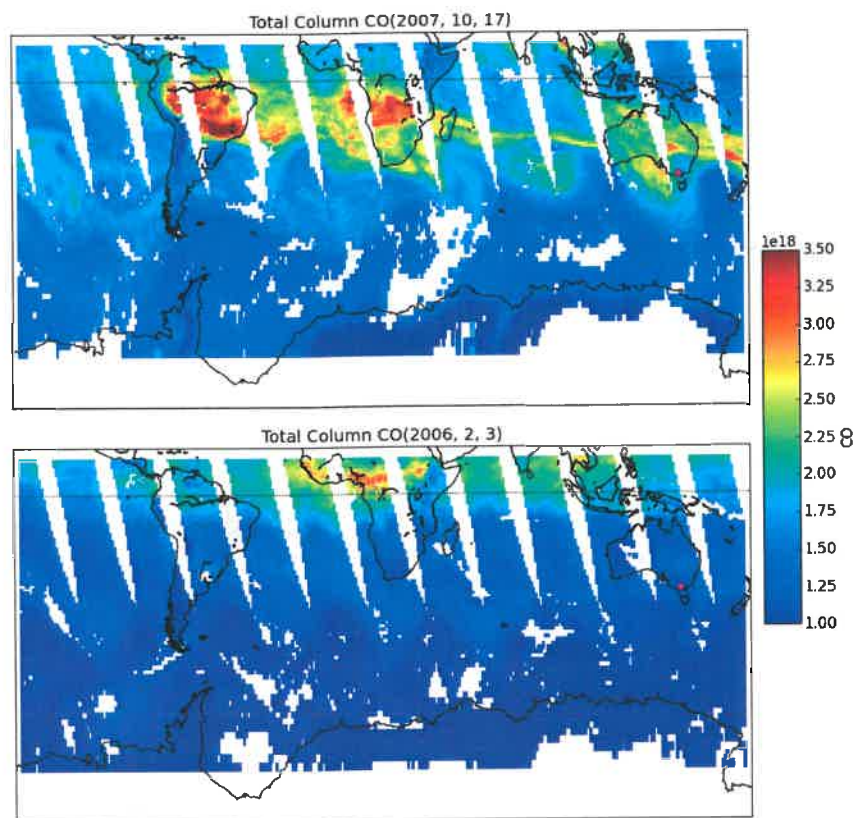


Figure 2: AIRS total column CO image showing two days separate days of swathes. The top panel shows an example of an excluded ozone event which could have been caused by a transported biomass burning plume on ~~October~~ 17th, 2007. The bottom panel shows an example of a non-excluded ozone event.

(17 October 2007)

above Melbourne  
(marked by the red dot)

(3 February 2006)

→ This day can be contrasted with the bottom panel in Figure 2b, where low CO levels are observed over the entire Southern Hemisphere, and in particular, over Australia. Our rejection criteria removes the ~~October~~ 17 October 2007 event but not the 3 February 2006 event from the Melbourne ozonesonde database.

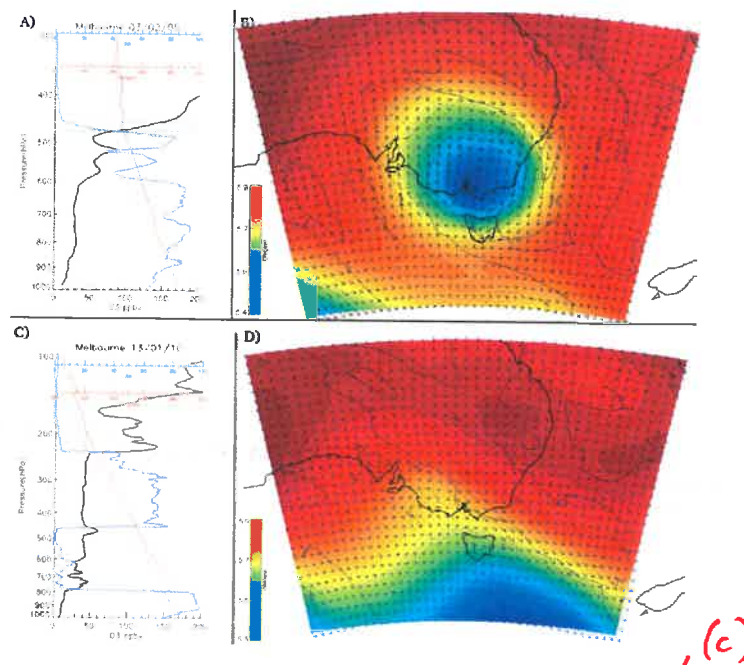


Figure 3: Vertical profiles show ozone ppbv (black line), relative humidity (blue line), and temperature (red line) for (a) 3 February 2015 and (b) 13 January 2010. Synoptic weather maps show the 500 hPa pressure level taken from the ERA-Interim reanalysis on (c) 3 February 2015 and (d) 13 January 2010. Vectors show wind direction and speed while the colour indicates the geopotential height. Also visible are the vertical wind contours, rising air represented by dashed lines, descending air represented by dotted lines, and solid lines where the vertical wind speed is zero.

- 1) Remove vertical wind contours  $\rightarrow$  not important here  
on Fig 3b & Fig 3d
- 2) Instead, overlay 500 hPa PV surfaces e.g.  
1, 2, 3 PVU  
7  
[2 PVU commonly used to indicate the dynamical tropopause]
- 3) Remove Figure 4

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Combine with  
Fig 3.

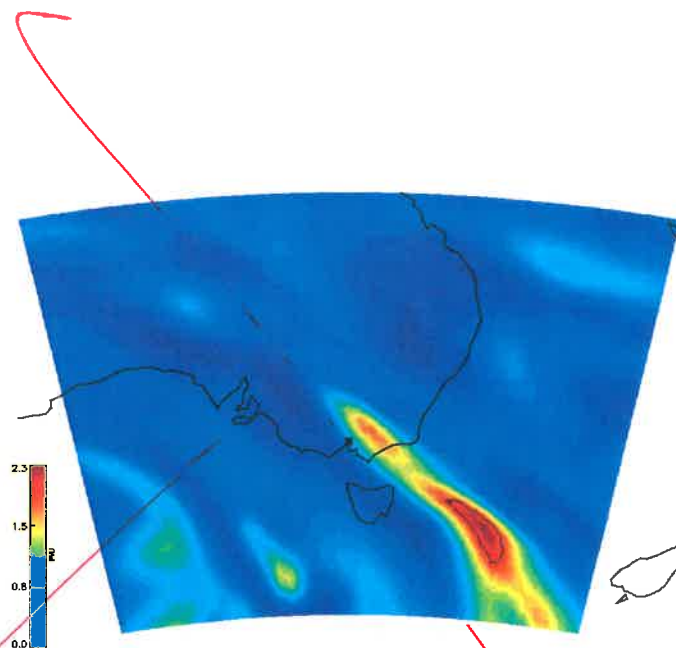


Figure 4: Melbourne synoptic PVU at 500hPa on the 13th of January, 2010, taken from the ERA-Interim dataset.

### 3.2 Climatologies

*seasonal cycles and event climatologies of the STT events*  
The event summaries for each of the three locations are presented in Figure 5 to Figure 7. There is an annual cycle in the occurrence frequency of STT events with a summertime peak above Melbourne and Macquarie Island. However, the occurrence frequency of STT events above Davis is relatively constant throughout the year.

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. STT event altitudes most commonly occur at 6 – 10 km above Melbourne and below 8 km at Davis but are distributed more evenly in altitude at Macquarie Island.

Using 458 ozonesonde profiles over Melbourne, 72 ozone events are detected, of which 15 are discarded as possibly caused by transported fire smoke plumes. Over Macquarie island 48 events are detected from 380 ozonesondes of which 8 are discarded due to possible smoke influence. For both of these sites, events occur mostly in Summer and mostly during storms which can increase convection and upper tropospheric turbulence. For Davis, 36 events are detected from 240 ozonesonde profiles. There is no clear seasonal cycle over Davis. These events contribute at least an average of 2.8 – 3.8% of the tropospheric ozone column.

*the STT which are unlikely fire-related*

### 3.3 Seasonal influences

Seasonally averaged ozone as recorded over the three stations (figure 9) shows increased ozone extending down through the stratosphere during the peak STT months over Melbourne.

*I think you can remove this & move Fig 9 to be the new Fig 2 because it provides the background O<sub>3</sub> climatology*

*(A) We also include on Fig 5 → Fig 7 the events which have a possible fire influence. These events are concentrated in spring at Melbourne and Macquarie Island.*



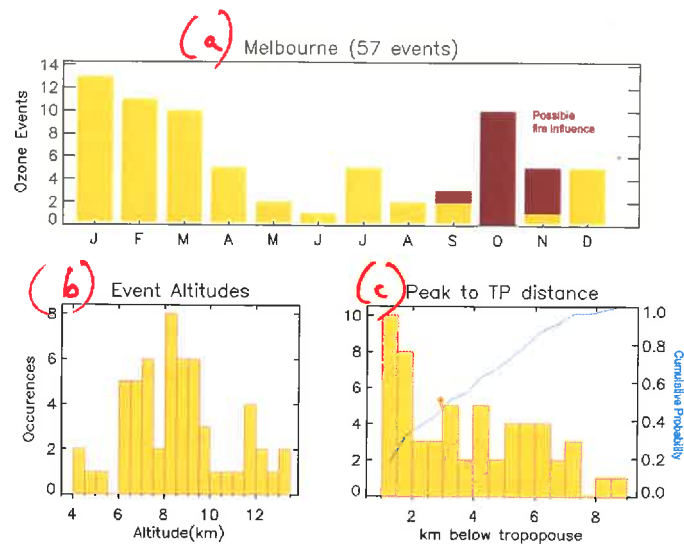


Figure 5: The climatology of STT events at Melbourne: (a) Events sorted by month from the entire Melbourne ozonesonde dataset. Additionally shown in red are the events filtered out as possibly smoke plume influenced. (b) The occurrence distribution of the ozone peak altitude. (c) The distance between the ozone peak and the tropopause (bars) and the cumulative probability function of these distances (blue line).

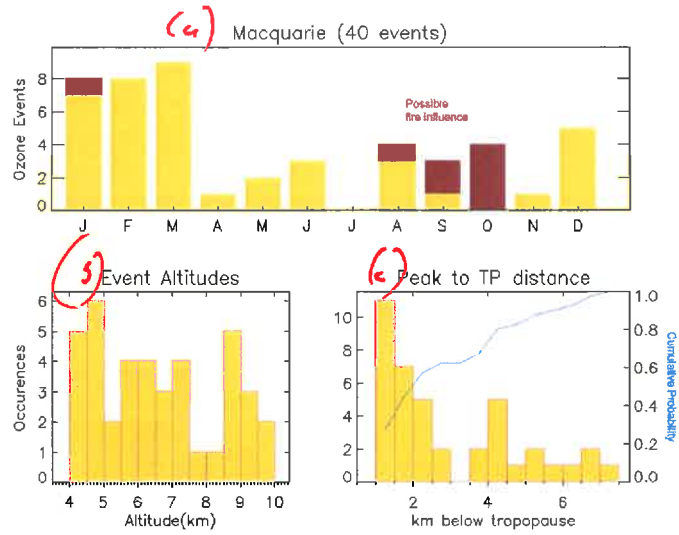


Figure 6: As for Figure 5 except showing the Macquarie Island STT events.

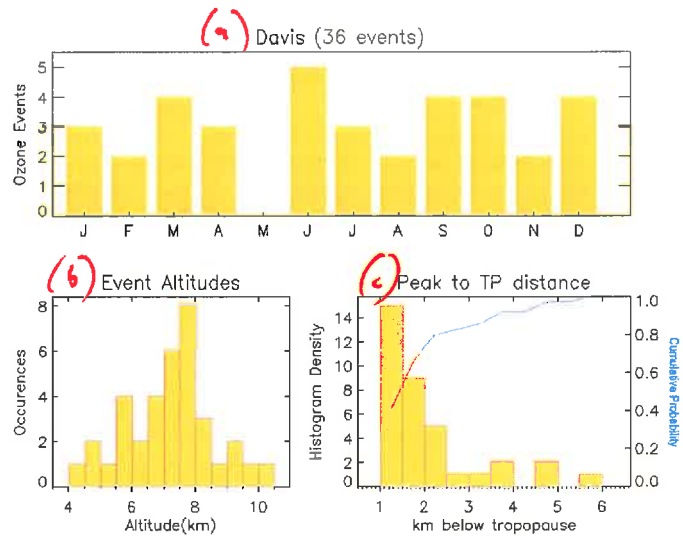


Figure 7: As for Figure 5 except showing the Davis STT events.

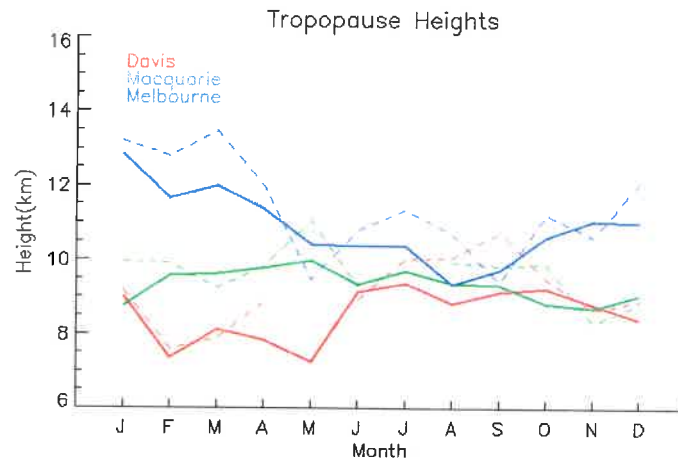


Figure 8: Monthly mean tropopause altitudes (minimum of lapse-rate and ozone defined tropopauses). Dashed lines show 'event only' seasonal tropopause altitudes.

### 3.4 Flux

Stratosphere to Troposphere ozone transport can potentially increase regional surface ozone levels above safe levels Zhang et al. [2014]. Using our estimate of STT ozone flux we find a lower bound for the STT ozone flux over each of our three sites. Figure 10 shows the climatological mean fraction of total tropospheric column ozone attributed to stratospheric ozone intrusions at each site, on days when an STT event occurs. The mean fractions of stratospheric ozone are 2–4%, although the largest fractional ozone in the tropospheric column attributed to stratospheric air exceeds 10% at all locations.

⇒ Does this dot exclude fire days?

## 4 Conclusion

Using ozonesonde data in the southern hemisphere can allow an overview of STT ozone transport which is independent of satellite data. Using a simple Fourier filter allows deterministic and quantitative analysis of STT ozone transport events.

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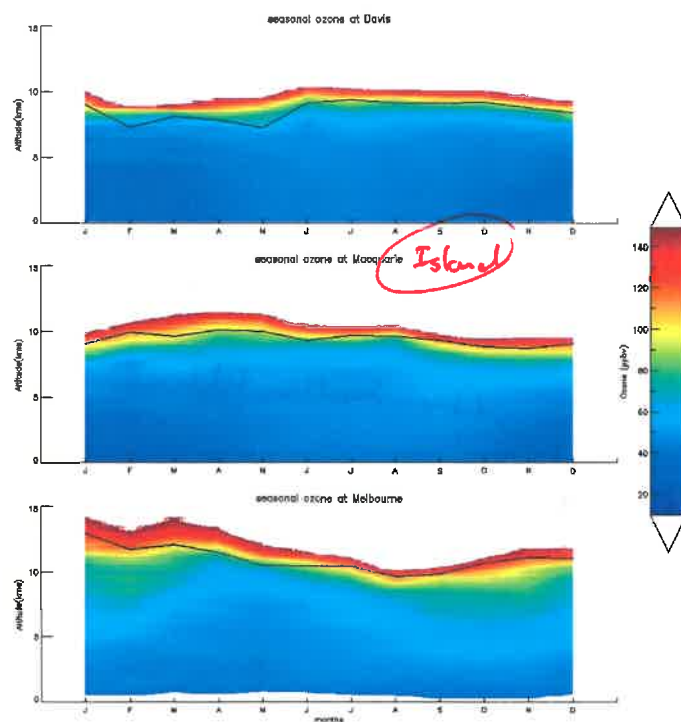


Figure 9: Seasonally averaged ozone over Davis, Macquarie, and Melbourne measured by ozonesondes. Black solid lines show seasonal tropopause heights.

- 1) *colours contour* Change/Scaling → cut at 100 ppbv
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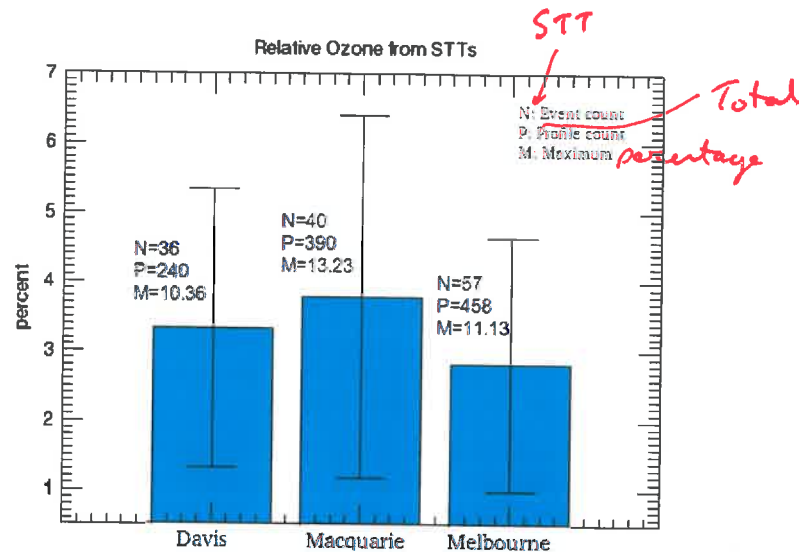


Figure 10: Fraction of total tropospheric column ozone attributed to stratospheric air intrusions during STT events. Error bars indicate one standard deviation.

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