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

Jesse Greenslade[1], Robyn Schofield[2], and Simon Alexander[3]

[1]*Center for Atmospheric Chemistry, University of Wollongong*
[2]*School of Earth Sciences, University of Melbourne*
[2]*ARC Centre of Excellence for Climate System Science, University of New South Wales*
[3]*Australian Antarctic Division*

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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, which is the third most potent greenhouse gas, has been estimated to exert a radiative forcing equivalent to a quarter of the CO2 forcing. 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, estimated using a model ensemble to be 5100 ± 600 Tg/yr and 550 ± 170 Tg/yr, respectively [Stevenson et al., 2006]. The primary sinks are chemical destruction and dry deposition, estimated to be 4700 ± 700 Tg/yr and 1000 ± 200 Tg/yr, respectively [Stevenson et al., 2006].

need some refs in here

Ozone-rich air mixes irreversibly down from the stratosphere during meteorologically conducive conditions [Sprenger et al., 2003, Mihalikova et al., 2012] X, ~~these are~~ referred to as Statosphere - Troposphere Transport events (STTs). In anything more recent?

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 irreversibly 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].

This belongs in your thesis chapter but is too basic for the audience of your paper.

While the amount of tropospheric ozone is small compared with that found in the stratosphere, it is an important constituent. 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 is important for explaining tropospheric ozone variability [Tang and Prather, 2011].

some of this would fit better in previous paragraph

refs?

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 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^{-1} relative to 1965 in the southern (northern) hemisphere due to an acceleration in the Brewer Dobson circulation.

needs context! % change? or 1965 levels?

Using several years of ozonesonde flights from three locations spanning ~~the latitudes of the Southern Ocean~~ XX-yy°S, 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~~ use case studies, to relate these STT to meteorological events.

Would be clearer as a Roadmap; e.g. In Section 2...

Paper introductions don't typically have sub-parts; make this Section 2.1?

~~1.~~2 ~~Instruments and Data~~ "Ozonesonde Record in the Southern Ocean" (or something)

Ozonesondes are ~~weather balloons with an attached~~ balloon-borne instruments ~~which~~ that measure ozone concentrations roughly every 100m ~~up~~ 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. While ozonesondes were launched prior to 2004 at the two northern sites, they were less frequent than after 2004, the analysis focuses on results from these more recent 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].

Doesn't add anything, irrelevant

dates?

This sentence is irrelevant (and misleading!) if you aren't using O3 from the reanalysis. I suggest you wait until you use ERA to introduce it.

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 in itself useful to diagnose the evolution of STT exchange over time-scales associated with normal synoptic scale weather patterns present in the extra-tropics. The ozonesonde data are supplemented with the ERA-Interim reanalysis [Dee et al., 2011] to enable construction of an STT exchange climatology.

2 Methods

2.1 Characterisation of STT~~s~~ events and associated flux

We characterise (try to use active voice throughout)

STT events ~~are characterised~~ in the ozonesonde~~s~~ vertical profiles ~~of ozone~~ as altitudes in the troposphere where the ozone mixing ratio exceeds a specified threshold. ~~Usually~~ Stratospheric ozone typically mixes irreversibly ~~down~~ into the troposphere in a synoptic-scale tongue of air; The vertical ozone profile recorded ~~observed~~ by the ozonesonde depends ~~up~~on the time in this cycle that ~~it is observed~~ [Sprenger et al., 2003] the ozonesonde is launched (right?). 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.

Need a transition. Why are you talking about tropopause height now?

~~Two definitions of the~~ Tropopause height ~~are calculated:~~ can be defined based on the standard lapse rate tropopause [WMO, 1957]~~,~~ or ~~and~~ the ozone tropopause [Bethan et al., 1996]. → Quick 1-sentence description of each? At Davis, the ozone tropopause defintion is modified for polar sites, following Tomikawa et al. [2009], Alexander et al. [2013]. → explain, not common knowledge. 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 two tropopause altitudes ~~definitions is referred to as the tropopause~~ we use for this study. This choice ~~of the lowest altitude of the tropopause~~ avoids occasional unrealistically high tropopause heights due to perturbed ozone or temperature measurements. → in ozonesonde records?

new paragraph

Figure 1 shows...

The monthly mean tropopause altitudes at each location ~~are shown in Figure 1~~, 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 poleward. awkward phrasing

Figure 2 shows that...

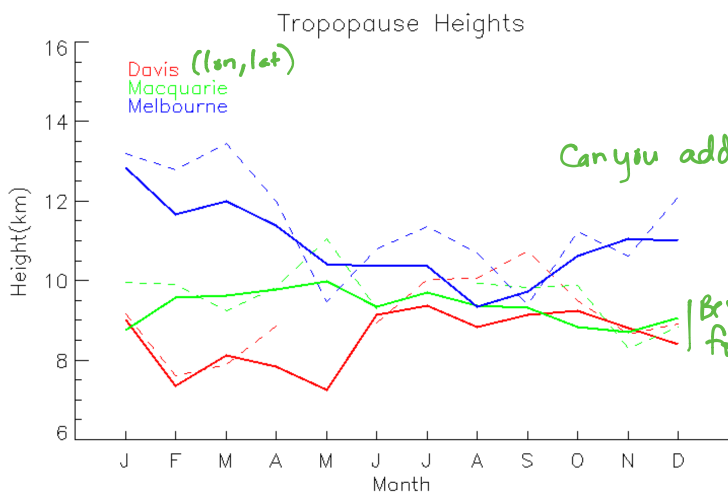
Seasonally averaged ozone as recorded over the three stations ~~(Figure 2)~~ shows increased ozone extending down through the stratosphere during the peak STT months over Melbourne. ? be explicit ~~It is worth noting that t~~ Tropopause altitudes at Davis may exceed 11 km altitude under certain synoptic conditions [Alexander et al., 2013]: the relation of tropopause altitude with individual STT events will be investigated in detail below. → this should come w/ Fig 1 not after Fig 2

Isn't there more to say from Fig 2? Other sites?

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 ~~so as~~ to retain perturbations ~~which have~~ with vertical scales between 0.5 km and 5 km. The choice of band limits ~~is~~ was set empirically; ~~but we note that to define an~~ for an event to qualify as STT ~~event~~, a clear increase above the background ozone level is needed, and a vertical limit of ~ 5 km removes seasonal-scale effects. We exclude from our analysis perturbations ~~The ozone perturbation profile is analysed at altitudes from~~ below 4 km ~~above the surface~~ (to avoid surface pollution events) and those occurring within 1 km ~~below~~ of the tropopause (to avoid the sharp transition to stratospheric air producing spurious false positives). Perturbations above the 99th percentile (locally?) of all ozone levels (anomalies?) 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 ppbv km^{-1} and simultaneously require that the perturbation profile does not drop below zero between the event peak

What does unclear mean here?



Can you add +/- 1 std dev?

Best not to use red + green for colour blind readers

Figure 1: Monthly mean tropopause altitudes (minimum of lapse-rate and ozone defined tropopause~~s~~) @ 3 sites. Dashed lines show 'event only' seasonal tropopause altitudes.

↳ define/explain in figure caption

~what %?

and the tropopause. The addition of these filters removes several events~~, each~~ with an ozone peak ~~which~~ that could not be definitively ~~said to be~~ seperated from the stratosphere.

We conservatively estimate ~~To provide a conservative estimate of~~ the ozone flux into the troposphere ~~for~~ associated with each event, The ozone concentration is integrated vertically over the ~~interval~~ altitude range for which an STT event is identified. ~~An example of an ozone profile is illustrated in~~ Figure 3 shows for an example ozone profile ~~and indicates~~ how the algorithm detects an STT event, defines the event boundaries, and calculates the ozone flux.

2.2 Removing ~~STT events related to smoke plumes~~ of biomass burning influence

Need a transition here. Why are we talking about fire?

Ozone production ~~due to fire smoke plumes~~ from biomass burning is complex and affected by photochemistry, fuel nitrogen load, and atmospheric plume interactions? (chemistry? dilution?) both during transport (time since emission?) and at the plume's destination (point of measurement?). [Ozone precursors include nitrogen oxides ($NO_x = NO + NO_2$) and non methane volatile organic compounds (NMVOCs).] (too basic here) Large biomass burning~~(BB)~~ 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].

This paragraph is a bit rambling. Need to be clear that some BB (but not all) leads to O_3 prod which could affect your stats.

refs? | Biomass burning influence in the southern hemisphere comes mostly from ~~South~~ southern Africa and South America, however Australian and Indonesian fires can also influence ~~our~~ the 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

Australian fires could have equiv. influence if from midlats – rephrase

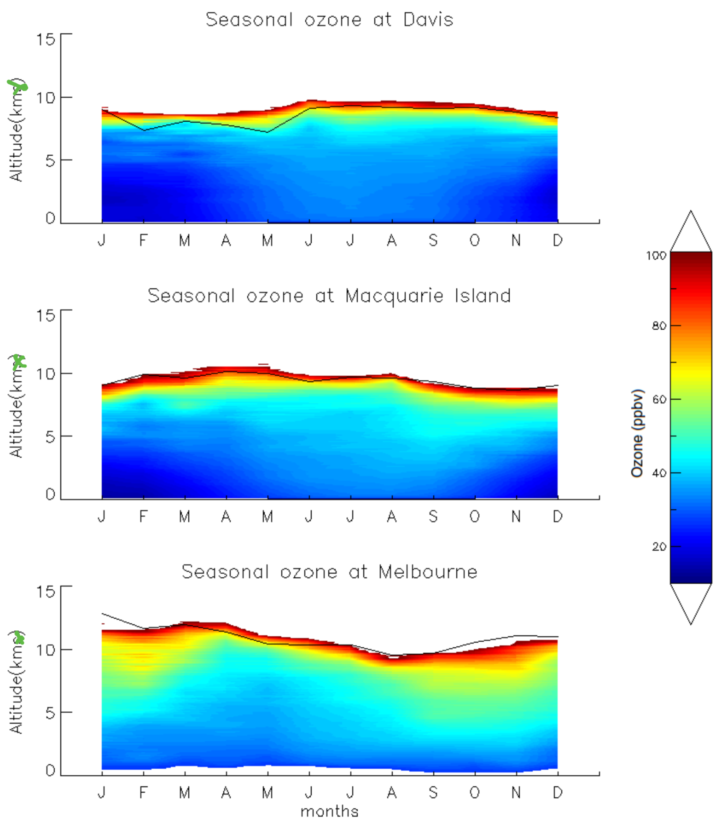
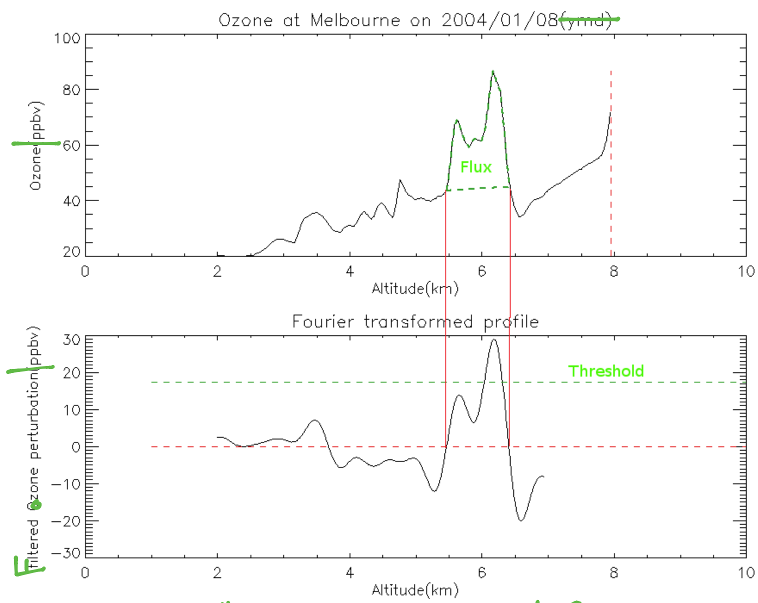


Figure 2: Seasonal~~ly~~ ~~averaged~~ cycle of ozone over Davis, Macquarie, and Melbourne measured by ozonesondes and interplolated between weekly measurements (right?). Black solid lines show seasonal tropopause heights, defined as described in the text



Rotate to have alt as y-axis

change Red/green

An example illustrating methods used for STT identification & flux estimation.

Figure 3: (a) ~~An~~ ozone profile ~~between~~ from 2km ~~altitude and~~ to the tropopause (~~indicated by the~~ dashed vertical line). The "flux" area shows the estimate of stratospheric impact on tropospheric 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).

at Melbourne on 8 Jan 2004

Band pass filtered O3 profile from (a). Coloured lines show repetitive; combine these two paragraphs

on many chemical and meteorological factors. Due to this complication and the possible influence of smoke plumes on ozone concentrations, we exclude from analysis any dates where this influence is likely. ↳ more accurately, influence on identification of STT events.

Transition! e.g. "Here we identify transported smoke plumes using carbon monoxide (CO)." We use data from

CO has a long enough lifetime to be an effective ~~transport~~ tracer. 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~~ for smoke plumes ~~exist~~ is a well established method (eg: Edwards [2003], Sinha et al. [2004], Edwards et al. [2006], Mari et al. [2008]) ~~and is used here~~. The AIRS (Atmospheric Infrared Sounder) instrument on board the ~~AQUA~~ Aqua satellite ~~records column CO. In this work a~~ We visual inspection ~~of~~ AIRS' vertical columns of CO ~~(provided by NASA [AIR, 2013])~~ over the southern hemisphere ~~is used~~ to exclude possible events with ~~foreign~~ smoke ~~plume~~ influence ~~on the ozone profile~~ at our three sites. Whenever high (~~approximately 2e18~~ molecules cm^{-2}) CO ~~concentrations~~ columns 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 are detected (through visual inspection) are removed. I would say something like "We diagnose smoke plumes when ... and remove these days from our analysis." Also specify you are only looking @ high CO at your sites.

only primary in SH! need refs.

give web address & product / version e.g. V6, Level 3 data

compared to a background of XX molec. cm^{-2}

Fig. 4a shows AIRS CO on a day where BB possibly affects tropospheric ozone ~~is shown in figure 4a~~. Elevated CO levels can be seen ~~above~~ over Australia, likely due to long-range transport from African and/or South American BB. This day can be contrasted with figure 4b where low CO levels are observed over the entire Southern Hemisphere. ~~Our rejection criteria removes the 17 October 2007 event but not the 3 February 2006 event from the Melbourne ozonesonde database.~~ All days at all three sites where an ~~ozone intrusion~~ STT event is detected ~~are checked~~ were screened except for one event ~~which~~ that coincided with missing AIRS data ~~15 missing days within~~ (January 2010). ~~This results in~~ We discarded 15 of ~~the~~ 72 events over Melbourne ~~being discarded, and~~ 8 of ~~the~~ 48 events over Macquarie Island ~~being discarded~~, and none over Davis.

the example in just work into previous sentence.

Comment on timing of these relative to expected SH BB.

~~3 Results~~

Just make these each their own section

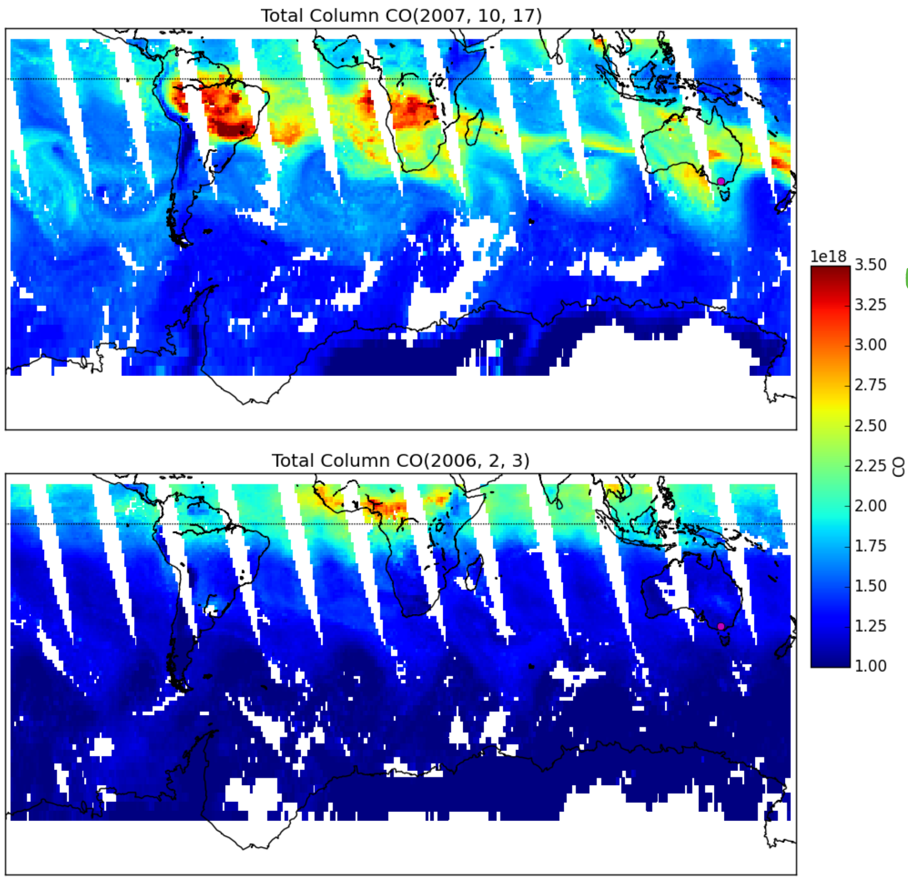
3.~~1~~ Case Studies of synoptic conditions during STT events

We examine two ~~STT~~ case studies in detail to illustrate the synoptic scale conditions in which ~~they can~~ STT events occur ~~above~~ over Melbourne. Fig. 5 shows ... for both events.

A cut-off low pressure system passed over Melbourne on 3 February 2005 (Figure 5b). The ozonesonde profile indicated low lapse-rate and ozonesonde tropopause heights (both > 450 hPa, see Figure 5a). An ozone intrusion into the troposphere is identified by our detection algorithm at ∼ 520 hPa.

Need more discussion of this event. What is going on meteorologically? How does this relate to what we know from STT literature?

STT events also occur during frontal passages, an example of which is illustrated in Figure 5d over south-eastern Australia. The tropopause heights are much higher at this time and an ozone intrusion is identified centred around 200 hPa. Note the separation between this intrusion and the ozone tropopause (marked by the green dashed line), indicating the start of the stratosphere above Melbourne. During the frontal passage, stratospheric air descends and streamers of ozone-rich air likely break off and mix into the troposphere [Sprenger et al.,



units?

Figure 4: AIRS total column CO ~~image showing two days seperate days of swathes.~~ The top panel (17 October 2007) ~~shows~~ is an example of a~~n excluded~~ day when ozone ~~event~~ above Melbourne (~~marked by the~~ red dot) ~~which~~ could have been caused by a transported biomass burning plume ~~on October 17th, 2007.~~ and so was excluded from our analysis. The bottom panel (3 February 2006) shows an example of a ~~non-excluded ozone event.~~ day when Melbourne was likely not influenced by transported and was retained for ozone our analysis

Need to fix resolution of these (unreadable)
I suggest post processing in inkscape
(save as postscript first, use !p.font=0 in IDL)

highlight your event on these plots. Also calculate flux associated with them.

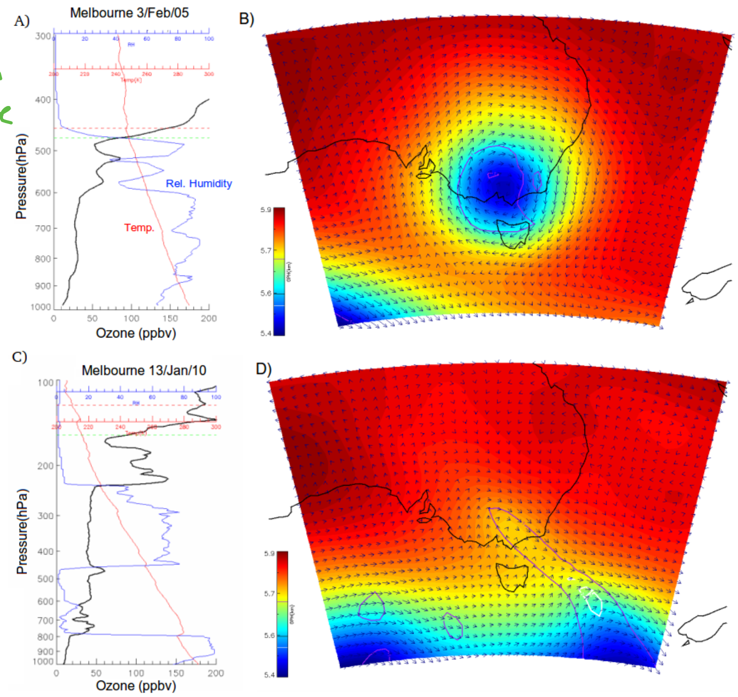


Figure 5: (a,c) Vertical profiles ~~show~~ of ozone ~~ppbv~~ (black line), relative humidity (blue line), and temperature (red line) for (a) 3 February 2005 and (c) 13 January 2010. (b,d) Synoptic weather maps ~~show the~~ at 500 hPa ~~pressure level taken~~ from the ERA-Interim reanalysis on (b) 3 February 2005 and (d) 13 January 2010. Vectors show wind direction and speed while ~~the~~ colour indicates the geopotential height. Also visible are ~~the line~~ contours of potential vorticity units, with 1 PVU in purple and 2 PVU (often used to determine dynamical tropopause height) in white.

2003]. More detail again. Relate to more than one paper!

The relative humidity profiles are anticorrelated with ozone in the upper troposphere for these events, indicating again ~~the~~ a stratospheric origin ~~of~~ for the ozone-rich air mass. 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(Figure 5c).

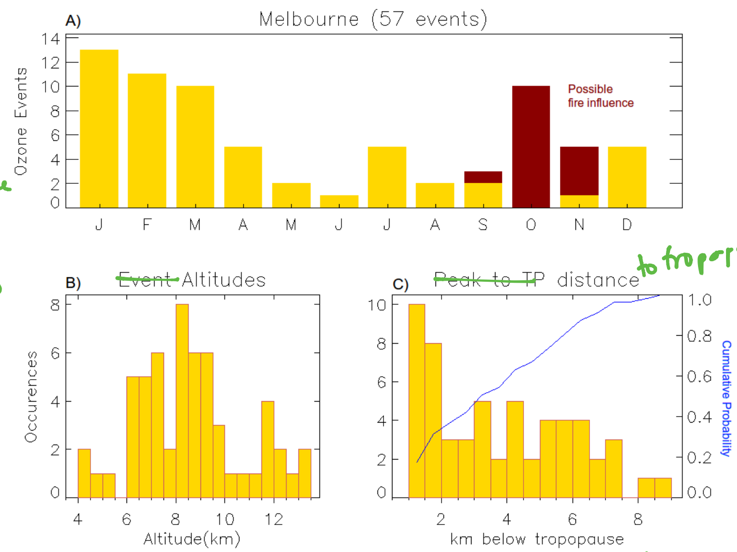
Does this say anything last paragraph didn't?

4 ~~3.2~~ STT event Climatologies

are shown in Fig 6 for Melbourne, Fig 7 for ..., Fig 8 for ...

The seasonal cycles and ~~event~~ climatologies of the STT events ~~for each of the three locations are presented in Figure 6 to Figure~~ 8. There is an annual cycle in the ~~occurrence~~ frequency of STT events (with a summertime peak) above

Is there any difference in number of sondes launched per month @ each site? If so, needs to be commented on



Shouldn't there be red fire influence in b/c, or have those already been removed?

Figure 6: The climatology of STT events at Melbourne binned by (A) ~~Events sorted by~~ month, ~~from the entire Melbourne ozonesonde dataset~~. The events filtered out as possibly smoke plume influenced are indicated in red. (B) ~~The occurrence distribution of the ozone peak~~ altitude, and (C) ~~The~~ distance ~~between the ozone peak and~~ from the tropopause (bars) ~~and~~ along with the cumulative probability function of these distances (blue line).

In (b) and (c) altitude is at the event peak.

Melbourne and Macquarie Island. ~~However, the occurrence~~ The frequency of STT events above Davis is relatively constant throughout the year.

Again, you need more discussion. What does it mean? How can you relate to literature?

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. We also include on Figure 6 to Figure 8 the events which have possible fire influence. These events are concentrated in Spring at melbourne and Macquarie Island.] For both of these sites, the STT events which are unlikely to be fire-related 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. ← you said that already

Move to section on BB filtering.

expand this! →

Given the order of your discussion, I suggest you reshuffle subplots so Fig. 6 is seasonal cycle @ all 3 sites, Fig. 7 is altitudes, Fig 8. is distance to tropopause. Then discuss each in turn (as you sort of do now)

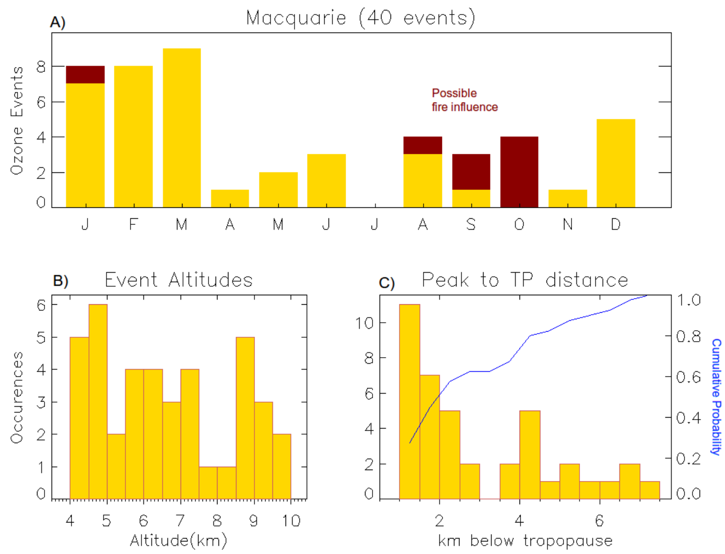


Figure 7: As for Figure 6 except showing the Macquarie Island STT events.

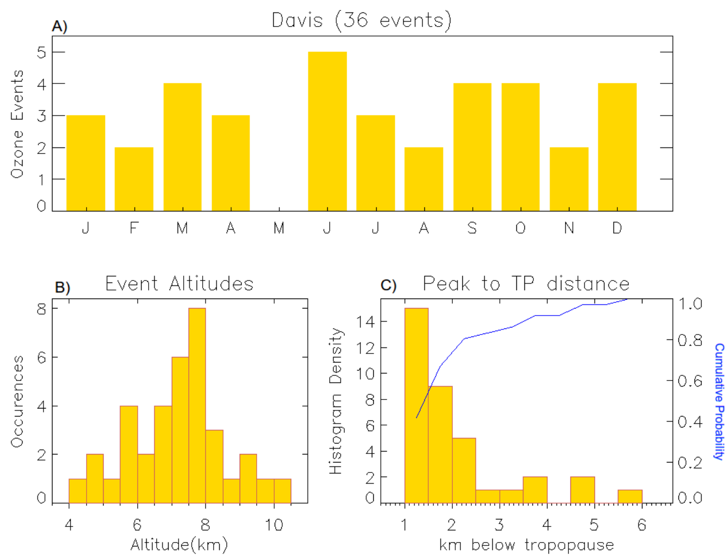


Figure 8: As for Figure 6 except showing the Davis STT events.

enhancement

Can you make another showing absolute enhancement?

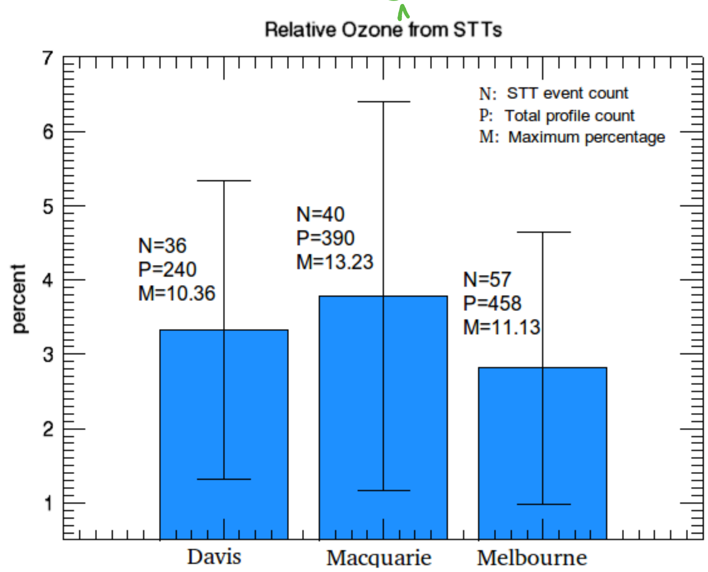


Figure 9: Fraction of total tropospheric column ozone attributed to stratospheric air intrusions during STT events. Error bars indicate one standard deviation.

Stratosphere to troposphere Ozone flux from STT events

4 ~~3.3~~ Flux

Based on the integrated ozone amount associated with each STT event,

How do you calculate the total column? Is that from the sonde? explain!

Stratosphere to ~~T~~troposphere ozone transport can potentially increase regional surface ozone levels above safe levels [Zhang et al., 2014]. ~~Using our estimate of STT ozone flux~~ (see section 2.1) we find a lower bound for the STT ozone flux over each of our three sites, (excluding possible fire influence) Figure 9 shows the ~~climatological~~ mean fraction of total tropospheric column ozone attributed to stratospheric ozone intrusions at each site, ~~on~~ averaged over days when an STT event occurs. ~~These flux amounts are calculated after removal of the biomass burning events, although leaving the burning events in changes the means by less than 5%.~~ The mean fractions of ~~strat~~ trop ospheric ozone ~~are~~ attributed to STT events 2–4%, ~~although the largest fractional ozone in the tropospheric column attributed to stratospheric air~~ exceeds 10% at all locations. Our flux estimates are relatively insensitive to our biomass burning filter; including smoke influenced days changes the mean flux by <5%.

why is it a lower bound?

On individual days, this value can

Would be nice to see a frequency distribution of flux associated w/ each event

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. TO BE CONTINUED! :)

To make this meaningful, I think you need at least a back of the envelope calculation of how much O3 we are talking about if we extrapolate from your sites. Should be able to find papers w/ trop. O3 columns, or we can grab some satellite data. If we combine w/ monthly likelihood + 3% should be able to estimate net O3 inputs to trop. over Southern ocean.

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