

Interactive comment on “Tropospheric ozone climatology at two southern subtropical sites, (Reunion Island and Irene, South Africa) from ozone sondes, LIDAR, aircraft and in situ measurements” by et al.

O. Cooper (Editor)

Owen.R.Cooper@noaa.gov

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Review by Owen Cooper, co-editor of ACPD/ACP and editor of this manuscript.

This paper provides a first look at free tropospheric ozone trends in the region of South Africa and the Indian Ocean using 15-17 year ozonesonde records from two locations. The trend on the ozone values above Irene is very interesting, with the topic being appropriate for ACP. These data and results will be very useful for the scientific community in its effort to fully quantify the tropospheric ozone budget, especially in data-poor re-

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gions.

I have not yet read the comments of the two anonymous referees that are reviewing this paper and their opinion will have a strong influence on my decision as to whether or not the paper will be published with ACP. My current opinion is that the paper could be published but not before it first undergoes a major revision to 1) correct errors and inaccuracies, 2) clearly delineate between items that can be supported by previous research and those items that are purely conjecture, and 3) to improve the writing style and grammar.

Major concerns:

1) The paper was updated to include the Irene data from the early 1990s, which I think has improved the analysis. As we discussed earlier the gap in the Irene data in the mid-1990s may cause some readers to question the trend analysis. Therefore please also include the original trend analysis using just the most recent 8 years of data, as these data also show a significant trend. Please carefully check all of your trend numbers in Tables 1-2 and figures 7-10 and in the text as I have found many errors.

Table 1 lists Irene as having 17 years of data but it actually only has about 13-14.

In Table 1 define IRN, RUN and JOB

Table 2, the number 0.54 ± 0.09 should be 0.054 ± 0.009

Two of the numbers in %/Decade for Reunion should have negative signs. I'm not sure how you get -3.6 ± 0.3 for A-4km at Reunion. I get -0.4 to -0.44 using a column of 5-5.5 DU.

In Figure 8 make sure Reunion's lower troposphere is listed as 1-4 km, while Irene is listed as 2-4 km.

Page 11080 lines 5-6: shouldn't 4.2 DU/decade be 0.87, and shouldn't -0.3 DU/decade be 0.22?

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Page 11080 lines 16-17: You report that the lower free troposphere has almost no trend. The trend is -0.053 ± 0.09 which includes zero, so the trend could be zero or positive as well as negative. So shouldn't you just state that there is no significant trend instead of almost no trend?

2) From the evidence provided in the manuscript it seems clear that there is a significant ozone increase in the lower troposphere of Irene and in the upper troposphere above Reunion. The causes of these trends are not clearly shown in the paper, and to their credit the authors do call for further research into these issues at the end of the paper, which I agree is necessary.

However the paper is written in a very confusing manner that spreads observations, trends, literature review and speculation throughout the paper, which leaves the reader uncertain regarding the robustness of the findings of the previous studies, and uncertain regarding the likelihood of the speculation in the paper. The paper needs to be reorganized in the following manner:

1) Provide in the Introduction a thorough literature review summarizing all that is known about the vertical and horizontal distribution of biomass burning, anthropogenic and lightning emissions across the study region. Also review the seasonal variation of STE.

2) Then review all that is known about trends in emissions and STE.

3) present the measurements and trends in the Results section, without speculation or literature review

4) Then in the Discussion speculate on the causes of the ozone distribution and trends, while being very clear about what is supported by previous studies and what is purely conjecture.

For example, in this paper there are several instances where high ozone in the mid and upper troposphere is linked to biomass burning. Seeing as this paper provides

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no actual in situ measurements of high ozone and smoke in the mid- and upper troposphere, much of this relationship is assumed based on previous works. I don't have the time to go through all of the literature, but I have looked at some of the more highly cited papers dealing with the topic and haven't found any that provide clear in situ measurements of ozone and smoke in the upper troposphere (although the 1992 TRACE-A data show an enhancement up to ~8km). If papers exist (measurement or modeling) that describe smoke in the upper troposphere above Africa and the Indian Ocean then I would like the authors to reference them and comment on them. If there are no such measurements, how can we be sure that smoke is having an impact on ozone in the upper troposphere?

More specifically, on page 11075 the high springtime ozone measurements are attributed to biomass burning, but you are not clear as to how high this influence extends. You seem to imply that the high ozone values above 10 km are also linked to biomass burning, whereas to me it seems far more likely that STE is responsible. Please be more specific as to the likely source of the upper tropospheric ozone and provide supporting evidence.

On page 11075 you discuss the influence of STE. James et al. [JGR 2003, see reference below] give a very good climatology of global stratospheric intrusions and their results should be referenced when discussing the typical locations of STE events. Also on page 11080 you hypothesize that there may have been an increase in STE above Reunion, but is there any evidence, modeling or measurement to support this hypothesis? If not you must state that this hypothesis is purely speculative and requires further investigation. Then on page 11081 you seem to conclude that an increase in STE is the most likely explanation for the increase in free tropospheric ozone above Reunion, but again is there any evidence that STE has increased over this time period?

At the end of page you say that the two mechanisms can play a role above Irene. Which two mechanisms do you mean? Instead of "can play a role" say "may play a role"

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James, P., A. Stohl, C. Forster, S. Eckhardt, P. Seibert, and A. Frank, A 15-year climatology of stratosphere-troposphere exchange with a Lagrangian particle dispersion model, 2, Mean climate and seasonal variability, *J. Geophys. Res.*, 108(D12), 8522, doi:10.1029/2002JD002639, 2003.

Page 11082 lines 13-16 It's not clear if you are saying 1) that no research had been conducted on a biomass burning trend, or 2) that it has been shown that there is no trend. In which regions has the CO trend been explored? The full reference to Langerfelds et al. 2003, is missing from the Reference section.

On pages 11070 and 11076 the Pickering 1990 and 1996 papers are used to support claims that convection enhances ozone in the mid and upper troposphere above southern Africa. But the 1990 paper deals with the USA and the 1996 paper deal s with South America in springtime. So these papers are inappropriate for these claims and need to be removed. Please find other papers to support your claim.

3) All of the figures need to be improved such that the text and lines are clearly visible in the printed form. Please work with the copy editorial staff to ensure that the figures are clear and legible. Figures that are especially problematic are: Figure 3 (standard deviation lines are not clear), Figure 6 (I can barely see any of the profiles) and Figures 7-8 (the text if far too small to read).

Figure 1 is almost an exact copy of a figure in Diab, R. D., A. M. Thompson, K. Mari, L. Ramsay, and G. J. R. Coetzee (2004), Tropospheric ozone climatology over Irene, South Africa, from 1990 to 1994 and 1998 to 2002, *J. Geophys. Res.*, 109, D20301, doi:10.1029/2004JD004793.

You must receive permission from AGU (the copyright holder) to reproduce this figure. However it would be best to re-draft the figure yourself to remove items that are irrelevant to your paper, such as the site of Lusaka, and to add other features such as longitude lines (like the 25 E meridian that is important for delineating the biomass burning regions).

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Minor comments:

I have provided some extensive comments to fix several errors and to improve the grammar. However, I have not corrected all of the grammatical errors and a more thorough edit will be required after the paper is revised.

Throughout the paper ozone is reported as mixing ratios (ppbv) and therefore the measurements should be referred to as mixing ratios and not concentrations.

Throughout the paper please replace radiosonde with ozonesonde, and please replace "stratospheric-tropospheric exchanges" with "stratosphere-troposphere exchange"

Throughout the paper southern Africa is referred to as the African subcontinent. I have not been able to find any clear definition of what the African subcontinent includes. It does not appear to have any connection to a clear physical boundary such as a tectonic plate boundary or a mountain chain, and is not similar in structure to the Indian subcontinent. Instead of African subcontinent, please just describe this region as southern Africa.

Abstract: replace physico-chemical effects with photochemical and dry deposition processes

Introduction: Items discussed in the first two sentences need references.

page 11066 lines 7-8: Where is the ozone production occurring, in the mid and upper troposphere?

line 14: add a reference to the SAFARI experiment

line 23 The text gives the impression that the cut-off low formed in the tropics, which it did not. The mid-latitude cut-off low was advected into the tropics.

page 11067 lines 4-5, change to: Johannesburg (26.1 S; 28.0 E) and Irene (25.5 S; 28.1 E) are located about 3000 km southwest of Reunion.

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Page 11068 line 6: change to subsidence region

line 19: add a reference for the SAFARI experiment

Page 11069 line 12: I assume that when you talk about STE due to tropical cyclones you are talking about the Baldy et al 1999 paper "Tropical cyclone Marlene and STE", but this paper is not in the reference section.

lines 19-21, change to: "As a consequence a 5 km deep haze layer is formed over southern Africa, capped by a persistent subsidence inversion."

lines 24-27. This point is not clear to me. Does the export at 31 S occur continuously, regardless of the westerly waves, or does it only occur episodically when a westerly wave disrupts the anticyclone?

page 11070 lines 3-4 change to: In addition to stratospheric sources, ozone in the region surrounding Irene is produced photochemically from biomass burning, biogenic and anthropogenic emissions, as well as from lightning NO_x emissions.

line 7: it's not clear if the burning occurs east or west of the 25 E meridian.

line 8, change to: Although the strongest ozone enhancement is expected...

line 22, change to: "... can also be responsible for increasing tropospheric ozone via ozone production from lightning NO_x emissions [Schumann and Huntrieser, 2007]"
The Schumann and Huntrieser paper [ACP, 2007] is a good reference for the impact of lightning on tropospheric ozone and is recommended reading.

line 25: in which direction are the power plants from Irene?

Page 11071 line 1: a reference is needed to illustrate the seasonal variation in power-plant emissions in South Africa.

line 8: change to: ... a consistent database of tropospheric and stratospheric ozone ...

line 12: SHADOZ only covers a portion of the Southern Hemisphere, change to: ...good

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coverage across the tropical and subtropical Southern Hemisphere.

page 11072 line 10: change to: ...launched in 1994 by European scientists, Airbus and several commercial airlines....

line 12: MOZAIC does not measure nitrogen oxides (NO+NO₂), it measures NO_y. change to: ...ozone, water vapour, carbon monoxide and total odd-nitrogen (NO_y)....

lines 19-22: the paper still says it focuses on the 8-years of the SHADOZ data, but now it has been expanded beyond the 8 years, so please update the text accordingly

lines 11072: here the numbers don't add up because if there were at least 20 profiles per month at Irene for 8 years than Irene should have at least 240 profiles, but it only has 208. Please correct these monthly numbers.

page 11073 line 8: change to: ;...MOZAIC and LIDAR data are useful for describing ...

lines 9-10: delete this sentence as it isn't necessary and the point is made elsewhere

line 22: delete the word "set"

line 23-24: change to "Therefore the number of profiles used to build the climatological values are fewer in the upper troposphere , and profiles"

page 11074 lines 5-7 change to: In summer ozonesonde and lidar measurements are consistent in the lower troposphere below 5 km. Above 13 km, in summer (and all seasons) the LIDAR climatology yields smaller values than the ozonesonde profiles. In summer, between 5-13 km

lines 10-12 change to: ...profiles obtained in the presence and absence of tropical convection (Fig. 4 of Leclair de Bellevue et al., 2006).

lines 13-21 I understand the point you are trying to make but this paragraph is not very clear or well written and needs to be re-worked.

lines 23-25: delete first sentence and change second sentence to: Figure 4 depicts the

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mean monthly ozone profiles between 3 and 16 km.

page 11075 line 1: should read "mixing ratio is set to 130 ppbv"

page 11076 line 17, and page 11081 line 1: I think this should be accuracy rather than precision.

page 11078 lines 6-8: This sentence needs to acknowledge the influence of terrain on the ozone values. Change to: "Ground level measurements at a high altitude station on Reunion Island therefore allow documentation of the seasonal variations of regional lower free tropospheric ozone, although the data are still affected somewhat by up and down slope flow."

Page 11078 line 16 change to: "These two values are greater than the statistical error...."

Page 11080 line 23: "Mix then cook scheme" sounds like a very non-technical term. Don't you just mean to say that pollutants can be lofted to the free troposphere via convection and then over several days the aging air mass can undergo photochemical ozone production?

Page 11082 The last paragraph needs to be moved to the Conclusions.

line 29 You say that this work has highlighted the need to determine the trend with more precision. Precision means that the measurements can be reproduced with little variation. Do you mean to say precision or accuracy? What makes you think the current data suffer from problems with accuracy or precision? How can these deficiencies be improved? More frequent measurements?

Page 11083 lines 19-22 Delete this sentence

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 11063, 2008.

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