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Discussions

Interactive comment on "Observations of ozone-poor air in the Tropical Tropopause Layer" *by* Richard Newton et al.

Anonymous Referee #2

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General comments:

This paper addresses core elements of the hypothesis, originally proposed by Kley et al. (1996) and subsequently addressed by Folkins et al. (2002) and others, that low values of ozone in the TTL over the Pacific Warm Pool are a consequence of the convective transport of ozone-depleted air from the boundary layer over the region. To address this hypothesis, the paper employs data the coordinated airborne measurements from the CAST, CONTRAST and ATTREX airborne campaigns staged from Guam in early 2014, together with a concurrent CAST equatorial ozonesonde campaign at Manus Island. The analysis presents three significant findings: (a) that very low ozone (<15 ppbv) occurs at least episodically in the Warm Pool boundary layer, (b) that similarly low ozone values occur in the TTL south of the equator over the tropical Western Pacific but only rarely north of the equator, and (c) an association of of dimethyl sulfide, methyl iodide and other very short-lived substances (VSLS) of marine origin with lower ozone in the upper troposphere. While findings (a) and (c) are reasonably robust, (b) suffers from an unknown level of uncertainty due to the high degree of noise in the ATTREX ozone data which are the basis of the finding. This is a major shortcoming of the paper.

Scientific significance:

Taken together the main findings are not inconsistent with the original hypothesis. However, due to the operational constraints on the three aircraft, based as they were at 13°N, relatively few measurements were obtained in the Southern Hemisphere, where deep convection was presumably providing the hypothesized direct path of low-ozone air in the boundary layer to the TTL. The NASA Global Hawk used in ATTREX did have the range and duration to sample effectively south of the equator from Guam, however it only did so on one occasion, and this was after the CAST and CONTRAST campaigns had concluded.

Despite the limitations of the sampling, the finding of a hemispheric contrast in TTL ozone is significant. As the authors state in their conclusions, this contrast may be a result of differences in the ozone content of the air entering convection in the two hemispheres or a difference in convection. They favor the latter as it is consistent with seasonal boundary layer measurements from earlier campaigns, including from GTE, BIBLE and HIPPPO.

There is one important shortcoming in the analysis however which renders the finding of hemispheric contrast as presented less than robust; this is the high level of noise in the Global Hawk UCATS ozone data. As I discuss below, this noisiness and its impact on the uncertainty in the analysis needs to be addressed directly in the paper.

The second finding is the association of trace constituents of marine origin with lowered ozone in the upper troposphere. I suspect that if more observations had been obtained from the active Hadley branch south of the Equator, the association would have stood out much more clearly. Nevertheless, it is an important finding as it stands. Maybe not a ‘smoking gun’, but tantalizing nonetheless.

Scientific quality:

The paper provides a somewhat limited review of the scientific literature of the TTL low-ozone problem. In its simplest form, the hypothesis is straightforward. However, testing the hypothesis is made difficult for a number of reasons, not the least of which is the challenge of sampling a phenomenon so vast in scale. A discussion of this and the challenges that it presented to the utilization of the disparate datasets in the paper would have been as important as the review of the basic elements of the phenomenon. Thus, the paper would benefit from a more focussed detailing of what they are looking for in the various analyses they lay out and what the train of the argument will be. While this becomes clearer as you go along in the text, it would have been better if the reasoning had been presented at the outset.

There are four sections of analysis focusing respectively on the CAST/Manus ozonesonde profiles (Section 3), the TTL ozone measurements on the Global Hawk (Section 4), the CAST and CONTRAST ozone measurements (Section 5) and the very short-lived substances measured by the Whole Air Samplers on all three aircraft (Section 6). In Section 3, a time-height section of ozonesonde profiles is presented that clearly shows the presence of sub-15 ppbv ozone in both the TTL and the boundary at this south equatorial location, though the low ozone in the TTL was not linked in any way with the local boundary layer values. Indeed, the HYSPLIT trajectory results, which place the origin of the latter to the east of Manus, illuminate the challenge of making inferences from a single location’s data. While the authors don’t say this in so many words, they clearly wanted to set the stage here for their subsequent analysis of the ATTREX Global Hawk TTL measurements.

Indeed, Section 4, which focuses on those measurements, comprises the bulk of the

paper, and of this, most is devoted to the results from the single ATTREX flight (RF05) that sampled the Southern Hemisphere. The key figure here is Fig. 7 which plots the average TTL ozone along extended sections of the RF05 track. This averaged presentation is an accommodation to the fact that the Global Hawk ozone data were extremely noisy. As is stated in the paper, ozone data were plotted as averages in each slow ascent of the aircraft from 150 hPa to the tropopause; left unexplained is why the ascent segments appear in the figure to be coterminous. (Are the descents nearly instantaneous?) In any event, if the ozone data had been of higher quality, then it would have been helpful to augment the flight track figure with a standard line plot.

Given that the UCATS ozone data are so central to the analysis in this section, indeed the whole paper, the rather severe shortcomings of UCATS ozone measurement mentioned in passing in Section 4 ought to have been discussed fully in Section 2 on instrumentation. Particularly concerning is the possible negative bias of up to 5 ppbv at low ozone concentrations. Given the possibility of such a substantial bias, the reader might fairly ask how much confidence can be placed in the hemispheric difference suggested in Fig. 7. I would suggest that the authors show at least an extended section of the ozone data in time series format to give the reader a better sense of the uncertainty in the averaged values.

In contrast to Section 4, the subsequent section is compelling, showing striking similarities between the Manus ozonesonde data and the ozone data on board the CONTRAST Gulfstream V and the CAST BAe 146, the rather different regions of sampling notwithstanding. Notably absent in this section are the ATTREX ozone data, which but for their extreme noisiness would have filled in the region above the ceiling of the Gulfstream V. So the picture is unfortunately incomplete.

Section 6 shows in Figure 17 some separation between profiles of Very Short-lived Substances originating in the marine boundary layer on the basis of ozone. Here again, though, the noisiness of the UCATS ozone data cloud the picture in the critical TTL altitudes. By lumping all the WAS data together, the differences in the quality of the ozone data between aircraft are essentially blurred. Would, for example, we see the same strong difference in methyl iodide at 300 hPa if each aircraft were plotted separately? Indeed, where do the Global Hawk data leave off? Are Global Hawk descent profiles at Guam used at all in Figure 17? Here again then, as in Section 5, questions about the quality of the UCATS ozone data limit the confidence in the authors' conclusions.

Presentation quality:

This paper is very well written, and is almost devoid of grammatical errors. The figures, with the exception of Figure 7 noted above, are well conceived and competently drafted.

Recommendation:

I recommend that the paper be accepted with major revisions. The most important of these would be a thorough analysis of the UCATS ozone data quality and the uncertainties in the averaging they base their central argument upon. I would also suggest that the WAS data from all three aircraft not be combined as they are in Figure 17 unless they can present evidence that there aren't biases between the ozone measurements on board the three aircraft.

With regard to the flow of the text itself, I would recommend a more detailed background section in the introduction. In particular, I think that the authors should provide the reader with some sense of what questions can be robustly addressed by the datasets acquired in this unprecedented joint effort - and what questions are likely to be left unanswered. With that greater clarity, this paper may well stand as an important contribution.

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