

Our responses are interspersed with the reviewers' comments (in bold font) and are given in normal black text (changes to the manuscript are shown in red). The line numbers where those changes appear in the revised paper are also given at that point.

Reviewer 1

The paper presented by Ilann Bourgeois and a large number of co-authors is not bringing a large number of new knowledge, but it presents a large number of observations of ozone over the Atlantic and the Pacific oceans during the 4 seasons. In this regard, the paper is very important because it helps getting a global view of the ozone distribution in regions that are not too close to sources of precursors. The paper makes use of important field campaigns including ATom and HIPPO. What is really nice is that the data from these airborne campaigns are compared with ozone sonde measurements (when possible) and data from IAGOS in the North Atlantic (civil aircraft measurements). I have no major comments to make except to congratulate the teams involved for the wonderful airborne missions conducted under the scientific leadership of Steve Wofsy. The paper is very clearly written with appropriate references and justifications of the statements that are made. The methodology is clearly stated. The results are well presented and the discussion is clear.

I would have wished to see a bit more background material in the introduction, and maybe this can be added. What did we know about ozone above the two oceans from previous experimental studies including space observations and ozonesondes? What did these studies show regarding ozone in the tropics and in the extra-tropics as a function of season? What did we know about the influence above the oceans of African and American biomass burning and lightning? What about the plumes from industrialized countries? And at the end of the paper, does the study confirm what was known or is there any new findings that would change our understanding of the processes involved? Perhaps a few sentences on these issues in the introduction and the conclusions would make the paper more attractive

Response #1: We would like to thank the reviewer for the comment. We agree that the manuscript would benefit from more background context in the introduction and conclusions. We added a whole new paragraph in the introduction that provides contextual knowledge on the current understanding of O₃ distribution and climatology over the Pacific and Atlantic Oceans, L.56-82:

“Airborne campaigns have targeted both the Pacific and Atlantic Oceans, providing novel characterization of O₃ sources, distribution, and photochemistry in the marine troposphere (Browell et al., 1996a; Davis et al., 1996; Jacob et al., 1996; Pan et al., 2015; Schultz et al., 1999; Singh et al., 1996c) and the low-O₃ tropical Pacific pool (Kley et al., 1996; Singh et al., 1996b), the pervasive role of continental outflow on O₃ production (Bey et al., 2001; Crawford et al., 1997; Heald et al., 2003; Kondo et al., 2004; Martin et al., 2002; Zhang et al., 2008), and the marked influence of African and South American biomass burning on O₃ production in the Southern Hemisphere (Browell et al., 1996b; Fenn et al., 1999; Mauzerall et al., 1998; Singh et al., 1996a; Thompson et al., 1996). Ozonesondes have been launched from remote sites for more than three decades in some places, and have provided additional constraints on the sources and photochemical balance of tropospheric O₃ including a deep understanding of vertically-resolved tropospheric O₃ climatology in select locations (Derwent et al., 2016; Diab et al., 2004; Jensen et al., 2012; Liu et al., 2013; Logan, 1985; Logan and Kirchhoff, 1986; Newton et al., 2017; Oltmans et al., 2001; Parrish et al., 2016; Sauvage et al., 2006; Thompson et al., 2012). Spatially-resolved O₃ climatology has been provided by

routine sampling by commercial aircraft, but has mostly been limited to the upper troposphere or over continental regions (Clark et al., 2015; Cohen et al., 2018; Logan et al., 2012; Petetin et al., 2016; Sauvage et al., 2006; Thouret et al., 1998; Zbinden et al., 2013), and by satellite observations (Edwards et al., 2003; Fishman et al., 1990, 1991; Hu et al., 2017; Thompson et al., 2017; Wespes et al., 2017; Ziemke et al., 2005, 2006, 2017), somewhat tempered by large uncertainties (Tarasick et al., 2019). Recent overview analyses depict the current understanding of global tropospheric O₃ sources, distribution, and photochemical balance and underscore the insufficiency of observations in the remote free troposphere (Cooper et al., 2014; Gaudel et al., 2018; Tarasick et al., 2019) necessary to improve the current representation of tropospheric O₃ in global chemical models (Young et al., 2018). Spatial and temporal representativeness of O₃ observations is currently the biggest source of uncertainty when inferring O₃ climatology in the free troposphere, even in regions where observation are abundant but not ideally distributed (Lin et al., 2015; Tarasick et al., 2019).”

We also added in several places in the conclusion a few sentences indicating how ATom and HIPPO findings confirmed and extended the scientific community’s understanding of global O₃ distribution, sources and relevant processes.

L.551-552:

“The data highlight several regional- and large-scale features of O₃ distributions, and provide **new** insight into **current** O₃ distributions in remote regions.”

L.567-568:

“Higher O₃ loading in the NH compared to the SH is consistent with the heterogeneous distribution of O₃ precursor emissions around the globe, mostly concentrated in the NH, **a result consistent with previous modeling studies and satellite observations.**”

L.570-575:

“**Similar O₃ distributions across latitude bands have been suggested in the past, but these studies were limited to the northern midlatitudes. Conversely, other observation-based studies indicated significant O₃ longitudinal gradients. Here, our findings are consistent with zonal transport smoothing the baseline O₃ distribution longitudinally from the Pacific to the Atlantic.**”

L.578-581:

“A comparison of seasonal O₃ vertical profiles does not reveal a marked seasonality in the tropics, but instead highlights the influence of specific events, most notably BB emissions from Africa and South America, **which have been extensively documented in the literature.**”

L.586-590:

“**ATom and HIPPO provide the first airborne in situ vertically-resolved O₃ climatology covering both the Atlantic and Pacific Oceans in the NH and in the SH. They confirm and extend the current understanding of O₃ variability in the remote troposphere, built over several decades by airborne campaigns, monitoring networks, and satellite observations.**”

L.594-596:

“In addition, ATom and HIPPO in situ measurements are invaluable to establish the quantitative legacy of pollution transport and chemistry globally by looking at the covariation of key species – in this case O₃ and CO.”

Reviewer 2

This paper reports tropospheric ozone distributions measured in ATom and HIPPO, compares them to ozonesondes and IAGOS to determine the consistency between the air sampled by ATom/HIPPO and long-term climatology, and comments on different features of the ozone distributions including vertical and seasonal distributions in the tropics and extratropics, and differences between the Pacific and Atlantic. Ozone curtain data from ATom and HIPPO are worth reporting in the literature, and it is also of interest to place ATom and HIPPO in a broader climatological context, as can be done with ozone data but has broader implications for the ensemble of ATom/HIPPO measurements for which such long-term records don't exist. This is a useful though limited contribution.

However, a big problem with the current paper is that it claims to discover what are very well-known features of the global ozone distribution, and it fails to credit previous work (notably from Jennifer Logan, Ed Browell's group, and many model papers) that made exactly the same points 1-3 decades ago. GTE campaign papers from Ed Browell's group using DIAL (PEM series, INTEX-B, SONEX, TRACEA, TRACE-P...) report very similar ozone curtains as ATom and HIPPO. All the findings reported here in the Abstract and Conclusions can be found in the previous literature. Once this is corrected and previous literature is properly credited, then it is not clear what is actually new in this paper in terms of scientific findings. It represents a limited contribution, and whether this is worth publishing in ACP is an Editor's decision.

Response #2: We would like to thank the reviewer for the time taken evaluating our manuscript. We understand the issue raised by the reviewer regarding acknowledgment of prior work dealing with O₃ distribution over the Pacific and Atlantic Oceans. We have added a whole paragraph in the introduction that provides contextual knowledge on the current understanding of O₃ distribution and climatology over the Pacific and Atlantic Oceans, as suggested by Reviewer 1 (L.56-82, or see Response #1). References in this paragraph are in part related to the GTE campaigns, as suggested by the reviewer 2.

However, we want to point out that

- i) the aim of this paper is not to provide a review of all the work that has been done regarding tropospheric O₃ distribution. This would be a daunting, yet certainly useful, exercise that is not the goal here. We had rather opted for the option to provide adequate examples of literature that have previously shown similar, or different, results than found in our study (and acknowledged by the reviewer 1). In any case, our intent was certainly not to claim that most of our results are new findings. Therefore, we have changed the phrasing in the manuscript wherever it might have seemed ambiguous (e.g., L.19, 22, 30, 96-97, 395-397, 552, 567-568, 570-575, 580-581), and hope that this new version of the manuscript will meet the reviewer's transparency standards. As stated above, we also added more literature references in the introduction to acknowledge prior work and provide a more expanded contextual background to the study (see Response #1).
- ii) we believe that this paper will be an important contribution to the literature, for several reasons.
First, evaluating the representativeness of in situ observations from airborne campaigns by comparing with longer-term observational records is a critical exercise, never achieved before to this extent (i.e., global-scale coverage). We think our paper will be a milestone in proving the usefulness of airborne observations to establish an O₃ - and other similarly long-lived species -

climatology, that have thus far been inferred from spatially-limited ozonesondes (Derwent et al., 2016; Diab et al., 2004; Liu et al., 2013; Logan, 1985; Logan and Kirchhoff, 1986; Oltmans et al., 2001; Parrish et al., 2013, 2016; Thompson et al., 2012) and commercial aircraft (Clark et al., 2015; Cohen et al., 2018; Kumar et al., 2013; Logan et al., 2012; Petetin et al., 2016; Sauvage et al., 2006; Zbinden et al., 2013), or from satellite and modeling work that has higher uncertainties (Edwards et al., 2003; Fishman et al., 1990, 1991; Hu et al., 2017; Thompson et al., 2017; Wespes et al., 2017; Ziemke et al., 2005, 2006, 2017). In addition, ATom and HIPPO in situ measurements help to establish the quantitative legacy of global pollution transport and chemistry by through the evaluation of key, covarying species – in this case O₃ and CO, as added L.594-596.

Second, Reviewer #2 suggests no new findings result from this analysis. An important distinction to make here is that most studies reporting global O₃ distribution result from satellite observations (e.g., Edwards et al., 2003; Fishman et al., 1990, 1991; Thompson et al., 2017; Wespes et al., 2017; Ziemke et al., 2005, 2006, 2017), modelling studies (e.g., Hu et al., 2017; Young et al., 2018) or from observations spatially expanded using back trajectory calculations (e.g., Liu et al., 2013; Tarasick et al., 2010). While useful, these studies come with somewhat large uncertainties, as recently noted by reports from the Tropospheric Ozone Assessment Report (TOAR), and thus require *in situ observations* to be used as a validation bench-mark (Tarasick et al., 2019; Young et al., 2018). ATom and HIPPO datasets provide exactly that, and therefore confirm and extend our understanding of global tropospheric O₃ distribution, sources, and processes, as noted L.586-590.

Third, there are several results highlighted in this paper that significantly extend tropospheric O₃ state of knowledge. For instance, our finding of a longitudinally similar O₃ distribution in the mid- and high-latitudes of both hemispheres has been disputed previously in the literature. The similarity of the distribution has been suggested for the northern mid-latitudes (Logan, 1985; Parrish et al., 2020) but it is not always evident in satellite-, modelling-, or ozonesonde-derived maps (e.g., Gaudel et al., 2018; Hu et al., 2017; Liu et al., 2013; Tilmes et al., 2012; Ziemke et al., 2017). Our observations show that this result mostly holds true for each season in the remote free troposphere (see section 4.2 of the manuscript). Another example is the O₃ to CO scatterplots shown in Figure 13, that highlight the wide-spread, year-round influence of continental outflow on O₃ in the remote troposphere in both oceans, and at almost all latitudes. Other studies have shown this for more spatially and temporally limited environments (e.g., Bey et al., 2001; Heald et al., 2003; Jaffe et al., 2004; Lin et al., 2017; Liu et al., 2002; Martin et al., 2006; Trickl et al., 2003), but our results show in situ evidence of frequent, large-scale occurrence of such events.

Specific comments:

1. Abstract, lines 20-22: it is well known that there is more ozone in NH than SH.

Response #3: We do not claim here that this is fundamentally new knowledge. However, ATom and HIPPO datasets are the first in situ observations covering scales large enough to actually confirm the modeling and satellite works that have shown this hemispheric gradient. We added a precision to the abstract, L.21-23:

“We highlight a clear hemispheric gradient, with greater ozone in the northern hemisphere consistent with greater precursor emissions, **on par with previous modeling and satellite studies.**”

2. Abstract, lines 22-23: uniformity of free tropospheric ozone at northern mid-latitudes has been known since Logan JGR 1985 from ozonesonde data.

Response #4: We thank the reviewer for the reference. The reference has been added in the discussion where appropriate (L.487). However, our analyses expand beyond the northern midlatitudes, and provide the first in situ observation-based evidence of similar O₃ distribution longitudinally at the mid- and high latitudes of both hemispheres. Therefore, we would like to keep this sentence as is.

3. Abstract, line 24: the higher ozone over tropical Atlantic than Pacific is well known – there is extensive literature on this from SHADOZ, TRACE-A, satellite retrievals (Jerry Ziemke)

Response #5: Again, we do not claim to be the first to have found these results. That is why we chose to use the word “well-documented” (L.26). However, we have added substantial background context in the introduction regarding the high O₃ over tropical Atlantic citing appropriate references from GTE campaigns, ozonesonde networks, and satellite analyses (L.61-75):

“and the marked influence of African and South American biomass burning on O₃ production in the Southern Hemisphere (Browell et al., 1996b; Fenn et al., 1999; Mauzerall et al., 1998; Singh et al., 1996a; Thompson et al., 1996). Ozonesondes have been launched from remote sites for more than three decades in some places, and have provided additional constraint on the sources and photochemical balance of tropospheric O₃ including a deep understanding of vertically-resolved tropospheric O₃ climatology in select locations (Derwent et al., 2016; Diab et al., 2004; Jensen et al., 2012; Liu et al., 2013; Logan, 1985; Logan and Kirchhoff, 1986; Newton et al., 2017; Oltmans et al., 2001; Parrish et al., 2016; Sauvage et al., 2006; Thompson et al., 2012). Spatially-resolved O₃ climatology have been provided by commercial aircrafts routine sampling, but mostly limited to the upper troposphere or over continental regions (Clark et al., 2015; Cohen et al., 2018; Logan et al., 2012; Petetin et al., 2016; Sauvage et al., 2006; Thouret et al., 1998; Zbinden et al., 2013), and by satellite observations (Edwards et al., 2003; Fishman et al., 1990, 1991; Hu et al., 2017; Thompson et al., 2017; Wespes et al., 2017; Ziemke et al., 2005, 2006, 2017), somewhat tempered by large uncertainties (Tarasick et al., 2019).”

4. Abstract, lines 26-28: Continental influences over the NH oceans have been known for decades (PEM-Tropics B, INTEX-B over Pacific come to mind), vertical structure is a well-known feature from ozonesonde and GTE data and again this has been extensively discussed in literature including countless model papers.

Response #6: We do not claim to be the first to monitor continental influences on the ocean atmosphere. However, our study does provide the first in situ observation-based O₃ climatology over the Pacific and Atlantic Oceans in both hemispheres. We would also like to emphasize that East Asia’s economy is completely different today than it was back in the 1990s. So, results from the 1980s and 90s may or may not hold true today.

Modelling work has been useful to advance the scientific community's understanding of O₃ seasonal cycles in places where measurements were lacking or limited. Models can now be evaluated using ATom and HIPPO dataset, not only looking at O₃ distribution, but also at O₃ seasonality, a significant advancement. This is now noted L.98-100:

“In addition, ATom and HIPPO sampling strategies were designed to deliver an objective climatology of key species to enable modelling of air parcel reactivity of the remote troposphere (Prather et al., 2017).”

5. Abstract: that last sentence is gratuitous.

Response #7: We modified the last sentence (L.29-32):

“This new dataset provides additional constraints for global climate and chemistry models to improve our understanding of both ozone production and loss processes in remote regions, as well as the influence of anthropogenic emissions on baseline ozone.”

6. Introduction: there has to be some serious review of previous knowledge on the tropospheric ozone distribution from ozonesondes, GTE aircraft campaigns, satellites, models. The authors are addressing a very old problem, on which there is a lot of literature. Ignoring that literature is not right, particularly because it leads to claims in the Abstract and Conclusions that suggest that the authors are unaware of it.

Response #8: We have responded to this comment in Responses #1, #2, #3, and #4.

7. Figures: there are way too many figures, and too much information on the figures – the eyes glaze, there's a lot of repetitiveness. It's hard to care about Score.

Response #9: We think that 13 figures are well within the usual range of figures used in papers published in ACP, and that all are necessary to illustrate and support the discussion of our manuscript. If the reviewer could specifically recommend which figures are deemed gratuitous, we would consider changing or removing that figure. We would also like to point out that the S_{score} is a necessary metric to provide a quantitative assessment of ATom and HIPPO representativeness, an important component of this paper, and this evaluation has been commended by Reviewer #1.

8. Line 306: “airborne campaigns can capture global baseline O3 values”. Didn't we already know this from the GTE campaigns?

Response #10: The reviewer raises an interesting question. We argue that the comparison of airborne observations with longer-term observational records provides more statistically robust evidence that allows us to make this statement. In that sense, we think that no, we didn't already know that from the GTE campaigns, as an evaluation of GTE campaign representativeness has not been done.

9. Lines 364-366, also 399-400, 409-411: we do know that ozone can be >100 ppb in biomass burning plumes over the South Pacific in spring (several papers coming out of PEM-Tropics A in Sep-Oct1996), seems like ATom/HIPPO just didn't hit them.

Response #11: ATom and HIPPO did sample some BB plumes over the South Pacific in spring, as discussed L.512-514 and 539-545, and illustrated in Figure 13d.

10. Lines 452-453 – uniformity of free tropospheric ozone at northern mid-latitudes has been known since Logan 1985 and has been shown in countless models.

Response #12: Once again, our results are not limited to the northern mid-latitudes, but the mid- and high-latitudes of both hemispheres, a significant extension from the Logan et al. (1985) study. Nevertheless, we added this reference at L.487. If the reviewer could provide specific references for the models that have shown such results, we would certainly consider them. Regardless, ATom and HIPPO data provide the first in situ evidence of this pattern, and are therefore an important contribution to the O₃ literature.

11. Lines 496-497: continental influence on ozone over the North Pacific is hardly new.

Response #13: Agreed. That is why a full paragraph citing previous studies on the topic was included (L.500-507 in the original manuscript, now L.531-538). However, what is interesting and noteworthy from ATom and HIPPO datasets is the *year-round* influence of continental emissions on baseline ozone in the northern Pacific, indicated by elevated O₃ and CO in all seasons in Figure 13, whereas most of the existing literature focuses on spring and summer continental outflows (e.g., Heald et al., 2003; Jaffe et al., 2004; Lin et al., 2017).

12. Lines 516-, Conclusions: the conclusions list the same unwarranted claims of “valuable new insight into O₃ distributions in remote regions” (lines 520-521) as the Abstract. All of the features presented here about the global ozone distribution have been amply documented in the literature.

Response #14: We have modified the text L.551-552:

“The data highlight several regional- and large-scale features of O₃ distributions, and provide ~~valuable new~~ insight into O₃ ~~current~~ distributions in remote regions. The main findings are as follows:”

However, we disagree with the reviewer’s statement “All of the features presented here about the global ozone distribution have been amply documented in the literature”. Please refer to Response #2.

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