Dear Editor,

Thank you very much for your attention to our paper.
We thank the two anonymous reviewers for their effort to our paper.
All their comments are taken into account in the revised manuscript.

Please find below two response letters and the revised manuscript.

Text with blue color in the revised manuscript denote revision or addition, and text with gray color denote deletion.

We found some minor mistakes in the submitted version, so that they have corrected as listed below:
1. In Supplement, the pressure level for isobaric trajectories was 83 hPa (not 80 hPa).
2. Livesey et al. 2015 was incorrect; This was not the ACP paper, but the MLS quality document. I find the latest quality document (version Rev.C, 2017), so I put this reference.
3. The terms of Bry and Cly were not explained in Section 2.3, so that we put "total inorganic bromine" and "total inorganic chlorine" in that section.

As a corresponding author, I confirm that all co-authors concur with the submission in its revised form.

Yours sincerely,

Takafumi Sugita, Dr.
National Institute for Environmental Studies
Center for Global Environmental Research
16-2 Onogawa, Tsukuba, Ibaraki 305-8506 JAPAN
Tel: +81-298-50-2460
mailto:tsugita@nies.go.jp
http://www.cger.nies.go.jp/en/

Anonymous Referee #1

Received and published: 6 September 2017

General Summary:

"This study evaluates the agreement between DIAL profiles, MLS, and CTM in austral spring in Rio Gallegos, Argentina. The material is appropriate for AMT and I have provided some comments below to improve the overall manuscript. Additional references should be added. A location map would improve the understanding of the manuscript in the context of the polar vortex."
Reply: We thank the referee for the effort to carefully reading the manuscript and providing us useful comments. All of the comments are considered properly as listed below. Several references are included properly. A map showing the DIAL observation site in Río Gallegos is also included.

A list below shows differences in Figures after and before this revision.

New	Old
Fig.1: map of Rio Gallegos	
Fig.2: vertical profiles (example)	Fig.1: vertical profiles (example)
Fig.3: time series of DIAL profiles	
Fig.4: sPV maps	
Fig.5: time series (18hPa)	Fig.2: time series (18hPa)
Fig.6: time series (56hPa)	Fig.3: time series (56hPa)
Fig.7: abs diff. vs. sPV diff.	Fig.4: abs diff. vs. sPV diff.
Fig.8: abs diff. vs. MERRA-2 O3 diff.	
Fig.9: abs diff. vs. distance	Fig.5: abs diff. vs. distance
Fig.10: abs/rel diff. vs. prs	Fig.6: abs/rel diff. vs. prs
Fig.11: abs diff. (w/filtered) vs. prs	Fig.7: abs diff. (w/filtered) vs. prs
Fig.12: time series of abs diff. (8hPa)	

Technical Comments:

1. *"A site lat/lon map would improve the discussion in the introduction surrounding the location of the site and vicinity to the polar vortex. Including a map with model/satellite overlay during the case studies would also improve the understanding of the horizontal scale of the variability within the latitude bands of interest."*
Reply: The new Figure 1 shows location of Río Gallegos. We have added a sentence in Section 1 Introduction: "A map showing the OAPA site is shown in Figure 1." A map with model/satellite overlay for one case study on Oct. 3, 2009 is shown in Figure S2. We would like to leave this figure as it is in Supplement.

2. *"P2L27 -It seems you have already evaluated the DIAL, this is more of an evaluation of the MLS/CTM."*
Reply: This DIAL system in Río Gallegos is not extensively evaluated so far (Wolfram et al, 2008; Wolfram et al, 2012). The MLS ozone data have shown the long-term stability and the small bias relative to ozonesonde and ozone lidar (Hubert et al., 2016). Thus, this DIAL ozone data will not be dedicated for validating the MLS ozone data, but the comparison is a good opportunity to evaluate the inter-consistency. On the other hand, the DIAL/MIROC-CTM comparison will be dedicated for evaluating performance of the model in the southern polar vortex season where we have no such a model comparison so far.

3. *"P3L6 -The reference Hubert et al., 2016 is used quite extensively. As DIAL has a very* *robust heritage, consider referring to additional investigators in locations such as this."*

Reply: We have revised these sentences: "DIAL is a laser-based active remote sensing system operated from the ground, aircraft, and ship, and has a robust heritage (e.g., Megie et al., 1977; Browell et al., 1983; Steinbrecht et al., 1989). O3 measurements from DIAL have a high vertical resolution and measurements have shown long-term stability (Nair et al., 2012; Hubert et al., 2016), owing to the stratospheric ozone lidar sites of NDACC (e.g., Leblanc and McDermid, 2000; Brinksma et al., 2002; Godin-Beekmann et al., 2003, Steinbrecht et al., 2009)."

4. *"P3L18-24 -This should be more concisely written as it is very qualitative. Backscatter is from the entire atmosphere, including aerosols. Consider adding in further references."*

Reply: We have revised a corresponding sentence: "The O3 number density profile is computed using the DIAL equation from the difference between the signal slopes originating from Rayleigh scattering of the emitted laser beams (n_{O3}). Since the returned signals include scattering and attenuation by atmospheric molecules, aerosols, and other atmospheric components, this complementary term could be minimized with laser wavelength chosen in the DIAL instrument. The laser wavelength chosen in the DIAL instrument minimizes the complementary term in the stratosphere to less than 10% of n_{O3} measured, in the presence of low aerosol loading (Pelon et al., 1986)."

5. *"P3L28 -'horizontal spatial resolution' – what altitude are you assuming this wind field at? Later on the manuscript interprets differences due to spatial locations, this seems counter to that."*

Reply: This is referred to some typical conditions in the lower stratosphere, so that we have evaluated horizontal distance using an air-parcel trajectory analysis at 83 hPa and summarized in new Table S1. The measurement duration for each date is also summarized in new Table S1. This sentence was revised as: "Most measurements were performed for 3-5 h to obtain a good signal-to-noise ratio (see Table S1 for detailed numbers). If we assume some typical wind speed of 30 m/s in the lower stratosphere, a horizontal spatial resolution becomes 300-500 km. In actual, we have evaluated horizontal distances using air-parcel trajectory analysis at 83 hPa (Tomikawa and Sato, 2005) and the results are summarized in Table S1."

6. *"P3L30 – Total measurement uncertainty – can you describe this more? Does this involve the uncertainty from ozone absorption cross-section, Pulse-pile up, background subtraction? Are you using the retrieved MLS temperature and number density for these comparisons (i.e. ruling out metadata as a source of difference)? Is for a 3 or 5 hour measurement?"*

Reply: We have added sentences in Section 3 Method for comparisons between DIAL and MLS/CTM: "For the total measurement uncertainty (Wolfram et al., 2008), we evaluated the effect of ozone absorption cross section, which is temperature dependent, and found the error is not larger than 2%. The other source is from correction of aerosol contamination. The methodology uses a Fernald inversion algorithm to evaluate the aerosol backscatter signal at 355 nm and extrapolated to 308 nm. In order to increase the signal to noise ratio, the signal registered is averaged over the full acquisition time of the measurement. The acquisition time is typically three to four hours, according to weather conditions. Before processing the signal using the DIAL equation, we make two corrections: 1) subtraction of the background signal using a linear regression within the range of altitudes where the lidar signal is considered negligible, typically between 80 and 150 km; 2) dead time correction of the detector, in order to correct the saturation of the photocounting signals (pile-up effect) in the lower altitude ranges (Godin et al., 1999)."

We do not use MLS temperature and pressure for converting O3 number density to mixing ratio. We used temperature, pressure, and geopotential height of the NCEP reanalysis data (Kalnay et al., 1996) for conversion. Thus, this is another source of uncertainty for the pressure/mixing ratio coordinate comparison. We have added a sentence in Section 3 Method for comparisons between DIAL and MLS/CTM: "For converting the original DIAL geometric altitude and O3 number density to pressure and O3 mixing ratio, the NCEP reanalysis data

(Kalnay et al., 1996) are used. These data are registered in the NDACC database. Possible deviations could be expected if we use other meteorological data for the conversion process in DIAL. However, in this study, we used the DIAL data that registered in the NDACC database."

The actual measurement duration is listed in new Table S1, ranging from 02:24 to 05:45.

7. *"If there is large uncertainty in the DIAL measurements below a certain altitude range, consider removing them from the manuscript."*
Reply: As in new Figure 3, the total error is not so large, but the O3 mixing ratio itself becomes small values in the lowermost stratosphere, providing large relative difference. Thus, we will leave the results of 83 hPa and 100 hPa levels.

8. *"P5L14 – 'In this study step one was not necessary' - then remove this discussion, confusing for reader."*
Reply: We have deleted the corresponding sentences and simply mention as below: "For comparison between DIAL and MLS, the DIAL profile is convolved using the following equation (Livesey et al., 2017): ..."

9. *"P5L16 – specify great circle lat/lon"*
Reply: It becomes between 47.1S and 56.1S for 69.3W. These values were added in the sentence.

10. *"P5L30 – Was eq.1 used or not? If so, revisit this section. If not, drop Eq. 1 entirely from manuscript."*
Reply: Eq. 1 was used. We have revised sentences: "Figure 2a shows vertical profiles of O3 measured with DIAL compared with those of MLS on the same day (November 14, 2009) as an example. The plus-crosses and dotted-line show the converted DIAL profile using Equation (1) and the original high vertical resolution DIAL profile, respectively."

11. *"Fig 1. – I understand the need for the MLS profiles, but are all of the CTM profiles necessary? They seem to cause more confusion and are not even compared in the difference plot. Also, the CTM profiles are significantly different above 10 hPa, this should be discussed."*
Reply: Figure 1 has revised as new Figure 2. CTM profile nearest the OAPA site is shown. We have also revised a corresponding sentence: "We have extracted data from six locations between 48.8S and 54.4S in latitude at 67.5W and 70.3W in longitude, but the nearest grid data was plotted in Figure 2a (see Figures 5 and 6 for the variability in six model grids)."

Above 10 hPa, the deviation of the CTM profile found on November 23, 2009 was actually discussed in Section 4.4 Comparison at other levels. So that, we have added a sentence in the last of Section 4.1 Example of vertical profile comparison: "This is discussed in Section 4.4."

12. *"P6L5 – add in plots of MLS potential temperature in Fig 1 if that is the case"*
Reply: Figure 1 has revised as new Figure 2. The potential temperatures (PT) for MLS computed from the MERRA-2 data are shown as text, instead of plots, for both new Figures 2a and 2b. Corresponding to PT levels of Wolfram et al. (2012), we show 475, 550, 650, and 850 K levels. We have also added a sentence in Section 4.1 Example of vertical profile comparison: "Several PT levels corresponding to pressure are also shown as text in Figure 2a."

13. *"P6L25 – Consider adding in a sentence for the reader to better understand PV and the relationship to the polar vortex. A map of the vortex using scaled PV would be helpful to understand why/where the boundaries were drawn on certain days."*
Reply: We have added a sentence: "The degree of PV values at each measurement or model grid is a robust indicator of the location relative to the polar vortex." sPV maps for selected days are now shown in new Figure 4. Sentences are added in Section 4.2 Time series comparison: "Figure 4 shows sPV maps from MERRA-2 for selected days on September 26, October 3, November 14, and November 23, 2009. At 20 hPa, the polar

vortex significantly diminishes on November 23 compared to that on September 26. Whereas at 50 hPa, the polar vortex still exists on November 23 with smaller area than that on September 26."

14. *"Does MIROC-CTM provide a PV product? Is MERRA-2 meteorological variables driving the CTM? If not, couldn't differences in the modeled PV be driving large differences? MERRA-2 provides ozone as well – if this is used, why not compare it as well?"*
Reply: We can compute the PV value from the output of MIROC-CTM, but we used PV values from MERRA-2 for location and time of all DIAL, MLS, and MIROC-CTM in this study to unify the data source. We have performed a model run with MERRA-2 but not achieved a detailed comparison yet (Akiyoshi et al., a presentation in Meteorological Society of Japan, 2017). Since a possible deviation could be expected if we use MERRA-2 for the nudging process in MIROC-CTM, some discussion has added in Section 3 Method for comparisons between DIAL and MLS/CTM: "... Another possible deviations could also be expected if we use other meteorological data for the nudging process in MIROC-CTM. The different reanalysis data may cause different vertical and horizontal motions of air in the model, providing different tracer correlations, hence ozone field. However, in this study, we analyze owing to the model of Akiyoshi et al. (2016) to examine the performance." (See also in Point 6 of Referee#2.)

We have analyzed the O3 value from MERRA-2, and added new Figure 8 and some discussion in Section 4.3 Dependency in distance and sPV difference: "Since the MERRA-2 data set also provide the O3 value (Wargan et al., 2017), we examined those data instead of the sPV value. Figure 8 shows the O3 difference versus MERRA-2 O3 difference between DIAL and MLS (MLS - DIAL). The mean difference is computed from the horizontal axis, resulting in -0.12 ppmv at 18 hPa and -0.02 ppmv at 56 hPa. The measured O3 difference is well reproduced by the MERRA-2 O3 that assimilates Aura MLS as well. At 56 hPa, a compact correlation is found between the two differences with a slope of one-by-one. A similar positive correlation is also found at 18 hPa."

15. *"There is significant vertical motion occurring during the polar vortex breakup, is it worth looking at more vertical levels than just two to evaluate MLS/CTM? Lidar is powerful at analyzing the entire vertical profile. It would be useful to isolate a series of lidar profiles that demonstrate the variability in the polar vortex sPV regimes. Fig 5 highlights the differences that may be associated with horizontal differences, but there is no mention of how the vertical gradients may affect the overall differences between measurements."*
Reply: We have extended discussion on the low bias in MIRCO-CTM above 10 hPa (relate to Point 11). To examine the effect of the vortex breakup on the O3 difference, we have added the O3 difference time series at 8 hPa in new Figure 12. A new Figure 3 shows a series of 23 DIAL profiles. Data are color-scaled based on sPV values. With these figures, we have revised sentences in Section 4.4 Comparison at other levels: summary: "To examine the low bias in MIROC-CTM, the time-series in O3 difference between DIAL and MIROC-CTM at 8 hPa is shown in Figure 12. Larger negative deviations in MIROC-CTM are found in October and November, especially for data with sPV values between -1.0 and -1.5 x 10^{-4} s^{-1}. Similar results are also found from 6 hPa and 7 hPa levels. The peak altitude of ozone in MIROC-CTM is lower than that of DIAL, as shown in Figure 2. Both the vertical and horizontal motions of air in the model are responsible for this different feature, but the cause is not known. As was shown in Figure 3, the vertical gradient of O3 from DIAL above 15-20 hPa shows rather week inside the polar vortex, but occasionally strong outside or edge of the polar vortex. Thus, the vertical gradient of O3 may affect the result for such occasions with the steeper gradient."

Also, we have added a sentence for the new Figure 3 in Section 4.2 Time series comparison: "Figure 3 shows all the 23 profiles of O3 obtained by DIAL. Data are color-scaled based on sPV values. The difference in the O3 value is found depending on the sPV value especially above 30-40 hPa."

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Anonymous Referee #2

Received and published: 26 September 2017

1. *"This article describes stratospheric ozone comparisons between the differential absorption lidar (DIAL) measurements of Rio Gallegos, Argentina, the Aura Microwave Limb Sounder (MLS) satellite observations, and the MIROC Chemistry-Transport Model (MIROC-CTM) outputs. The manuscript is well-written, and contains results from rare observations from a southern hemisphere ground-based station, making this contribution worth-publishing, after the few minor comments listed below can be addressed adequately."*

Reply: We thank the referee for the effort to carefully reading the manuscript and providing us useful comments. All of the comments are considered properly as listed below.

A list below shows differences in Figures after and before this revision.

New	Old
Fig.1: map of Rio Gallegos	
Fig.2: vertical profiles (example)	Fig.1: vertical profiles (example)
Fig.3: time series of DIAL profiles	
Fig.4: sPV maps	
Fig.5: time series (18hPa)	Fig.2: time series (18hPa)
Fig.6: time series (56hPa)	Fig.3: time series (56hPa)
Fig.7: abs diff. vs. sPV diff.	Fig.4: abs diff. vs. sPV diff.
Fig.8: abs diff. vs. MERRA-2 O3 diff.	
Fig.9: abs diff. vs. distance	Fig.5: abs diff. vs. distance
Fig.10: abs/rel diff. vs. prs	Fig.6: abs/rel diff. vs. prs
Fig.11: abs diff. (w/filtered) vs. prs	Fig.7: abs diff. (w/filtered) vs. prs
Fig.12: time series of abs diff. (8hPa)	

2. *"Page 5, lines 8-15: What altitude variable is being used for conversion to pressure? (is it the MLS-provided geopotential height?) Is geopotential height converted to geometric altitude? Please provide more details here"*

Reply: For conversion of the DIAL altitude/O3 number density into pressure/O3 mixing ratio, we used the NCEP reanalysis data, and the data are registered in NDACC. The geopotential height of the NCEP data is converted to geometric altitude. We have added a sentence in Section 3 Method for comparisons between DIAL and MLS/CTM: "For converting the original DIAL geometric altitude and O3 number density to pressure and O3 mixing ratio, the NCEP reanalysis data (Kalnay et al., 1996) are used. These data are registered in the NDACC database."

3. *"Figure 1, right panel: Can the combined (MLS and DIAL) uncertainty be added to the plot. This would show the differences in the context of their uncertainty estimates"*

Reply: We missed to mention the bars in MLS O3 profiles in the submitted version. This is precision reported for individual profiles. The combined uncertainty is added in new Figure 2 (right panel). The total uncertaintiy for DIAL is also added in new Figure 2 (left panel). We have added sentences in Section 4.1 Example of vertical profile comparison: "The bar in MLS O3 profiles shows the precision reported for individual profiles. The bar in DIAL O3 profile shows the total uncertainty. The combined uncertainty (root sum square) is shown in the right panel."

4. *"Figures 2 and figures 3: Please add approximate geometric altitude for convenience Also, I would recommend showing differences in percent as well."*

Reply: Approximate geometric altitude of DIAL are now added in new Figure 5 and Figure 6, and relative differences as 100*(X - DIAL) / DIAL are also added as another panels in new Figure 5 and Figure 6. We have added sentences in Section 4.2 Time series comparison: "For reference, Figures 5e and 5f show the relative differences for DIAL/MLS and DIAL/MIROC-CTM comparisons, respectively." and "Figures 6e and 6f show the relative differences for DIAL/MLS and DIAL/MIROC-CTM comparisons, respectively."

5. *"Page 7, lines 26-29: The explanation of model high bias is not convincing. Could the bias be related to inaccurate/incomplete chemistry causing in-vortex ozone loss to be underestimated? Please provide additional details supporting this statement."*
Reply: For this high bias in the MIROC-CTM, one possible explanation was as follow: A higher N2O value at 18 hPa than that of MLS was seen. A higher N2O value corresponds a smaller value of Cly (ClOx), providing a higher O3 value owing to a weaker O3 destruction. We have added some sentences in Section 4.2 Time series comparison: "Another possible explanation could be due to a weaker vertical motion of air in MIROC-CTM. Although not shown, a vertical profile of nitrous oxide, N2O, from MIROC-CTM on November 14, 2009 is different from that from MLS. A tight correlation between N2O and Cly is found in the stratosphere (e.g., Schauffler et al., 2003), and used to infer the Cly value from a measured N2O value (e.g., Wetzel et al., 2010; Strahan et al., 2014). At 18 hPa, the MIROC-CTM N2O value is higher than that of MLS, resulting in a smaller value of Cly in MIROC-CTM. Thus, a smaller active chlorine (ClOx) induces a higher O3 amount in MIROC-CTM."

6. *"On the use of meteorological fields: MIROC-CTM apparently uses ERA-based meteorological fields. However meteorological fields from GEOS-5/MERRA-2 are used for the other work described here (PV calculation, pressure/altitude conversion etc.). Would it be possible to use the same dynamical fields for improved consistency? If not, some discussion on the implications of using different met fields should be added, for example in section 3."*
Reply: We did not use MERRA-2 for the conversion of the DIAL altitude/O3 number density to pressure/O3 mixing ratio. The difference between NCEP and MERRA-2 will affect the DIAL pressure/O3 mixing ratio. This will be done in a future work when we register such data to NDACC.
Regarding a nudging other meteorological data, we performed CTM runs using MERRA-2 and NCEP reanalysis data. However, we have not achieved a detailed comparison among those meteorological data (Akiyoshi et al., a presentation in Meteorological Society of Japan, 2017). This will also be done in a future work. We have added some sentences in Section 3 Method for comparisons between DIAL and MLS/CTM: "Possible deviations could be expected if we use other meteorological data for the conversion process in DIAL. However, in this study, we used the DIAL data that registered in the NDACC database. Another possible deviations could also be expected if we use other meteorological data for the nudging process in MIROC-CTM. The different reanalysis data may cause different vertical and horizontal motions of air in the model, providing different tracer correlations, hence ozone field. However, in this study, we analyze owing to the model of Akiyoshi et al. (2016) to examine the performance."

7. *"Figure 6b, (X-DIAL)/(X+DIAL)*200: I think plotting differences between instruments should not be done with respect to the mean of the 2 instruments. Biases between instruments are better identified when one instrument is used as the reference (typically, the instrument believed to have a best accuracy). I would recommend to modify figure 6b by taking DIAL as the reference, i.e., plot (X-DIAL)/DIAL*100 instead."*
Reply: The right panel of Figure 6 has been revised in new Figure 10. According to this revision, the numbers shown in Section 4.4 Comparison at other levels: summary and in Section 5 Conclusions have revised to 116% for DIAL/MLS and 292% for DIAL/MIROC-CTM.

8. *"Page 9, lines 26-34: Below 70 hPa, large percent differences between observations are typically expected due to the lower ozone mixing ratio values at the bottom of the stratosphere, and occasionally also due to the proximity of the tropopause. The lidar signal saturation is a possible reason for the low bias, but the large percent differences are likely associated with the loss of sensitivity in this region of low ozone concentration"*
Reply: Thank you for pointing out this. We have added a sentence: "Since the O3 mixing ratio from DIAL is very small below about 70 hPa, the sensitivity might be degraded along with the saturation effect."

9. *"Conclusion: There is little discussion on the CTM outputs, especially the low ozone bias inside the vortex at 18 hPa. This finding deserves some digging to my opinion, including references to published works on the subject. Finally the conclusion should emphasize the crucial importance of the DIAL station location and the dearly-needed continuation for long-term measurements there for NDACC."*

Reply: From a view of the mean difference between DIAL and CTM, there is a low bias in CTM above 18 hPa, as shown in Figure 6 (new Figure 10). However, looking at data inside the vortex, there are high biases in CTM, as shown in Figure 2d (new Figure 5d). As I mentioned above, the high biases may be associated with a weaker vertical motion of air in CTM. This partly cancelled the underestimate of O3 value, providing a mean difference of nearly zero (0.04 ppmv in Figure 2d, i.e., new Figure 5d). We have added a sentence in Section 5 Conclusions: "An insufficient model vertical motion may also be partly responsible for the O3 differences, especially inside the polar vortex."

We have also added a sentence to emphasize the continuation of DIAL observations: "Because of very sparse observations from S.H. ground-based stations, continuation for long-term measurements there for NDACC is highly recommended."

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Comparison of ozone profiles from DIAL, MLS, and chemical transport model simulations over Río Gallegos, Argentina during the spring Antarctic vortex breakup, 2009

Takafumi Sugita[1], Hideharu Akiyoshi[1], Elián Wolfram[2], Jacobo Salvador[2,4,5], Hirofumi Ohyama[3,1], and Akira Mizuno[3]

[1]National Institute for Environmental Studies, Tsukuba, Ibaraki, Japan
[2]Laser Application Research Center (CEILAP)-UNIDEF (MINDEF-CONICET), UMI-IFAECI-CNRS-3351, Villa Martelli, Argentina
[3]Institute for Space-Earth Environmental Research (ISEE), Nagoya University, Nagoya, Aichi, Japan
[4]Universidad Tecnológica Nacional, Facultad Regional Bs. As. (UTN-FRBA) Medrano 951 CABA, Argentina
[5]Universidad Nacional de la Patagonia Austral Unidad Académica Río Gallegos and CIT Santa Cruz, Argentina

Correspondence to: T. Sugita (tsugita@nies.go.jp)

Abstract. This study evaluates the agreement between ozone profiles derived from the ground-based DIfferential Absorption Lidar (DIAL), satellite-borne Aura Microwave Limb Sounder (MLS), and 3-D chemical transport model simulations (MIROC-CTM) over the South Patagonian Atmospheric Observatory (OAPA, 51.6°S, 69.3°W) in Río Gallegos, Argentina from September to November 2009. In this austral spring, measurements were performed in the vicinity of the polar vortex, and inside it on some occasions; they revealed the variability in potential vorticity (PV) of measured air masses. Comparisons between DIAL and MLS were performed between 6 hPa and 100 hPa with 500 km and 24 h coincidence criteria. The results show a good agreement between DIAL and MLS with mean differences of ±0.1 ppmv (MLS – DIAL, n = 180) between 6 hPa and 56 hPa. MIROC-CTM also agrees to DIAL, with mean differences of ±0.3 ppmv (MIROC-CTM – DIAL, n = 23) between 10 hPa and 56 hPa. Both comparisons provide mean differences of 0.5 ppmv (MLS) to 0.8-0.9 ppmv (MIROC-CTM) at the 83-100 hPa levels. DIAL tends to underestimate ozone values at this lower altitude region. Between 6 hPa and 8 hPa, the MIROC-CTM ozone value is 0.4-0.6 ppmv (5-8%) smaller than those from DIAL. Applying the scaled PV criterion for matching pairs in the DIAL/MLS comparison, the variability in the difference decreases 21-47% between 10 hPa and 56 hPa. However, the mean differences are slight for all pressure levels, except 6 hPa. Because ground measurement sites in the Southern Hemisphere are very sparse at mid- to high-latitudes, i.e., 35-60°S, the OAPA site is unique for evaluating the bias and long-term stability of satellite instruments. The good performance of this DIAL system will be useful for such purposes in the future.

1 Introduction

Ozone depleting substances (ODS) have been decreasing since the Montreal Protocol and its subsequent adjustments and amendments. As a result, stratospheric ozone (O_3) is expected to increase in the future. The last WMO/UNEP ozone assessment concluded that increasing O_3 has been observed in the upper stratosphere around 42 km, or 2 hPa, in altitude (WMO, 2014). Positive trends have been evaluated for both the tropics and 35-60° latitude bands of both hemispheres above 5 hPa levels from 2000 to 2016 (Steinbrecht et al., 2017). However, the trend is still not statistically significant below 10 hPa levels. Steinbrecht et al. (2017) found 0.7 $\pm$ 0.9 and –0.2 $\pm$ 1.4% per decade changes at 10 hPa and 70 hPa, respectively, for 35-60°S. The satellite measurement has an advantage for estimating long-term trends because of its global coverage on a daily basis. However, its drift, i.e., the long-term measurement stability, should be quantitatively assessed with independent instruments. Ground-based ozone lidar is a potential candidate for such purposes, and can be used to estimate drift (e.g., Nair et al., 2011, 2012; Eckert et al., 2014; Hubert et al., 2016).

Hubert et al. (2016) comprehensively evaluated the bias and drift of 14 limb-viewing satellite sensors using ozonesonde and ozone lidar measurements. They concluded that biases in the satellite sensors were within $\pm$5% between 20 and 40 km and drifts were at most $\pm$5% per decade. They suggested that several instruments have significant drifts; caution is needed for multi-instrument comparisons to derive drift. Hubert et al. (2016) also showed a comparison spread, which is a measure of the short-term variability, with values of $<$5-12% for the same altitude range.

The ozone DIfferential Absorption Lidar (DIAL) system was installed at the South Patagonian Atmospheric Observatory (Observatorio Atmosférico de la Patagonia Austral, OAPA, 51.6°S, 69.3°W) in Río Gallegos, Argentina in 2005 (Wolfram et al., 2008). A map showing the OAPA site is shown in Figure 1. This site has been a stratospheric ozone lidar site within the Network for the Detection of Atmospheric Compositions Change, NDACC (www.ndsc.ncep.noaa.gov) since December 2008. NDACC sites in the Southern Hemisphere (S.H.) are very sparse at mid- to high-latitudes (e.g., 35-60°S). In springtime, this site is occasionally inside the southern polar vortex, which has shifted off the pole or elongated. A long persistent coverage ($\sim$20 days) of the polar vortex over the southern tip of South America occurred in 2009 for the first time since 1979 (de Laat et al., 2010; Wolfram et al., 2012). In the 2009 austral spring between September and November, measurements at OAPA were performed in the vicinity or, on some occasions, inside of the polar vortex, revealing a variability in potential vorticity (PV) of measured air masses inside and outside the vortex. Accordingly, the largest variability in O_3 values would be expected in such a latitude band (35-60°S). Therefore, this event was a good opportunity to assess the impact of O_3 variability on biasing behavior. To evaluate the performance of the DIAL system under such variability, O_3 data from Aura Microwave Limb Sounder (MLS) satellite measurements (Waters et al., 2006) were used for comparison. In addition to the DIAL/MLS comparison, we also used O_3 values from a 3-D chemical transport model simulation, which is based on version 3.2 of the Model for Interdisciplinary Research on Climate (MIROC) (Akiyoshi et al., 2016). Therefore, a secondary objective of this study was to examine the performance of the model simulation. Measurement and model simulation data used here are described in Section 2. The methodology used for the comparison is provided in Section 3. Vertical profiles of O_3 and their time series at the two selected pressure levels are shown in Section 4. The results of differences that depend on coincidence criteria are also shown in Section 4 and summarized for all pressure levels (from 6 hPa to 100 hPa). The conclusions of this study are described in Section 5.

2 Measurements and model simulations

2.1 Stratospheric ozone lidar

DIAL is a laser-based active remote sensing system operated from the ground, aircraft, and ship, and has a robust heritage (e.g., Mégie et al., 1977; Browell et al., 1983; Steinbrecht et al., 1989). O_3 measurements from DIAL have a high vertical resolution and measurements have shown long-term stability (Nair et al., 2012; Hubert et al., 2016), owing to the stratospheric ozone lidar sites of NDACC (e.g., Leblanc and McDermid, 2000; Brinksma et al., 2002; Godin-Beekmann et al., 2003; Steinbrecht et al., 2009). To target the stratosphere, O_3 number density is usually retrieved between 15 km and 45 km in geometric altitude. The DIAL system installed at the OAPA site in Río Gallegos, Argentina, began operating in August 2005 (Wolfram et al., 2008). The DIAL system operated at this site is fully described in Wolfram et al. (2008) and included in a review (Antuña-Marrero et al., 2017). The instrument is briefly described here. The DIAL technique requires two emitted wavelengths generated by two emitter lasers. An excimer (XeCl) laser emitting at 308 nm with 30 Hz repetition rate and maximum energy per pulse of 300 mJ is used for ozone absorption. The reference wavelength corresponds to the third harmonic of the Nd-YAG laser emission at 355 nm with 30 Hz repetition rate and maximum energy per pulse of 130 mJ. The optical receiver that collects the backscattered photons is composed of four Newtonian (*f*/2) telescopes defining an array of telescopes. Each has a 50-cm diameter with parabolic aluminized surfaces, 48-cm in diameter. This produces a total reception area of 7238 cm^2. An optical fiber, 0.27 db km^{-1} with attenuation at 308 nm, is placed at the focus of each telescope. The other end of the fiber is positioned at the focus of a quartz lens placed inside a spectrometer used to separate the received wavelengths. A mechanical chopper is positioned at the entrance of the spectrometer. It has a rotational frequency of 300 Hz (18,000 rpm), and its role is to block the strong lidar signals originating from the lower part of the atmosphere, typically below 10 km.

The O_3 number density profile is computed using the DIAL equation from the difference between the signal slopes originating from Rayleigh scattering of the emitted laser beams (n_{O3}). Since the returned signals include scattering and attenuation by atmospheric molecules, aerosols, and other atmospheric components, this complementary term could be minimized with laser wavelength chosen in the DIAL instrument. The laser wavelength chosen in the DIAL instrument minimizes the complementary term in the stratosphere to less than 10% of n_{O3} measured, in the presence of low aerosol loading (Pelon et al., 1986). Because lidar signals cover a large dynamic range, they have to be attenuated for measurements in the lower stratosphere. Therefore, the final O_3 profile corresponds to a composite profile computed from the “low" and “high" energy Rayleigh signals which are detected simultaneously (e.g., Godin et al., 1989).

In the 2009 spring, measurements began on September 6 (UTC, Coordinated Universal Time) during clear-sky local nighttime. Because the latitude of OAPA is 51.6°S, the short night lengths with increased seasonal cloud cover made it challenging to perform measurements after December (Wolfram et al., 2012). In total, 23 vertical profiles of ozone were obtained between September and November 2009, which were used for this study. Most measurements were performed for 3-5 h to obtain a good signal-to-noise ratio (see Table S1 for detailed numbers). If we assume some typical wind speed of 30 m/s wind speed in the lower stratosphere, a horizontal spatial resolution becomes 300-500 km. In actual, we have evaluated horizontal distances using air-parcel trajectory analysis at 83 hPa (Tomikawa and Sato, 2005) and the results are summarized in Table S1. The actual vertical resolution ranged from 0.7 km to 4 km at 14 km and 35 km in altitude, respectively. The total measurement uncertainty also ranged from 3% to 15% at the same altitudes.

For the total measurement uncertainty (Wolfram et al., 2008), we evaluated the effect of ozone absorption cross section, which is temperature dependent, and found the error is not larger than 2%. The other source is from correction of aerosol contamination. The methodology uses a Fernald inversion algorithm to evaluate the aerosol backscatter signal at 355 nm and extrapolated to 308 nm. In order to increase the signal to noise ratio, the signal registered is averaged over the full acquisition time of the measurement. The acquisition time is typically three to four hours, according to weather conditions. Before processing the signal using the DIAL equation, we make two corrections: 1) subtraction of the background signal using a linear regression within the range of altitudes where the lidar signal is considered negligible, typically between 80 and 150 km; 2) dead time correction of the detector, in order to correct the saturation of the photocounting signals (pile-up effect) in the lower altitude ranges (Godin et al., 1999).

2.2 Aura MLS

The MLS measurement covers latitudes between 82°N and 82°S since August 2004 (Waters et al., 2006). It is onboard the National Aeronautics and Space Administration (NASA) Earth Observing System (EOS) Aura satellite. MLS measures millimeter- and submillimeter-wavelength thermal emission from the limb of the Earth's atmosphere every 25 seconds, from which vertical profiles of more than 15 chemical species are retrieved. We used the standard O_3 data product (240 GHz radiances) retrieved with the version 4.2 data processing algorithm, which is publicly available from mls.jpl.nasa.gov/. The quality of the O_3 data is as follows from Livesey et al. (2017). The vertical × horizontal resolutions are 3 km × 300 km at 100 hPa and 3 km × 500 km at 10 hPa. The precision is 0.03 ppmv at 100 hPa and 0.1 ppmv at 10 hPa. The accuracy estimated from systematic uncertainty characterization tests are 0.05 ppmv at 100 hPa and 0.3 ppmv at 10 hPa. Data screening was accomplished according to Livesey et al. (2017). The former versions of the MLS O_3 values are evaluated from comparisons with DIAL (Jiang et al., 2007). They showed a good agreement of $\sim$5% from 5 hPa to 100 hPa.

2.3 Nudged chemistry-climate model based on MIROC3.2 GCM

As described in Akiyoshi et al. (2016), NIES developed nudged chemistry-climate models (CCMs) using the MIROC model. The CCM was nudged toward European Center for Medium-Range Weather Forecasts (ERA)-Interim data below 1 hPa (Dee et al., 2011). In these nudged-CCMs, a set of model variables for zonal-wind (u), meridional wind velocities (v), and temperature (T) were nudged. Above 1 hPa, where no ERA-Interim pressure level data exist, the zonal-means of zonal wind and temperature are nudged toward the COSPAR international Reference Atmosphere 1986 data (CIRA, 1990). The time scale for nudging the meteorological data (u, v, and T) was set to one day.

The model used in this study is a spectral model with T42 horizontal resolution (2.8° × 2.8°) and 34 vertical atmospheric layers above the surface. The top layer is located at approximately 80 km (0.01 hPa). Hybrid sigma-pressure coordinates are used for the vertical direction. The chemical constituents included in this model are O_x, HO_x, NO_x, ClO_x, BrO_x, hydrocarbons for methane oxidation, heterogeneous reactions for sulfuric-acid aerosols, supercooled ternary solutions, nitric-acid trihydrate, and ice particles. The CCM contains 61 chemical constituents including 42 for prediction and 19 for photochemical equilibrium, 165 gas-phase reactions, 42 photolytic processes, and 13 heterogeneous reactions on multiple aerosol types. The reaction rates and absorption coefficients are based on JPL 15-10 (Burkholder et al., 2015). The bromine budget is increased for consistency with observations using additions of $CHBr_3$ and CH_2Br_2, which results in approximately 21 pptv total inorganic bromine, Br_y, in the stratosphere around the year 2000. The volume mixing ratio of total inorganic chlorine, Cl_y, is approximately 3.3 ppbv in the stratosphere over the same period. This nudged CCM is hereafter termed the MIROC-chemical transport model (MIROC-CTM).

3 Method for comparisons between DIAL and MLS/CTM

The O_3 profiles from DIAL are used to evaluate the bias and drift, i.e., long-term stability of satellite measurements (e.g., Nair et al., 2012; Eckert et al., 2014; Hubert et al., 2016). Therefore, it is important to show the quality of the respective ground-based DIAL performance. Although O_3 profiles at OAPA are included in Hubert et al. (2016), the result for OAPA alone is not shown. Wolfram et al. (2012) also did not show coincident O_3 profiles with any limb-viewing satellite instruments. Therefore, we revisit the quality of the DIAL O_3 profiles obtained in the 2009 austral spring.

Usually, comparisons between DIAL and limb-viewing satellite instruments are conducted considering the differences in their vertical resolution and retrieval strategies (Hubert et al., 2016). MLS has covered the location of OAPA (51.6°S) on a daily basis since measurements began in 2005. The long-term stability of the MLS ozone dataset has been shown to be very good (e.g., Nair et al., 2012; Hubert et al., 2016). For comparison between DIAL and MLS, the DIAL profile is convolved using the following equation (Livesey et al., 2017): DEL: As recommended in Livesey et al. (2017), comparisons of MLS with other higher vertical resolution data are achieved by two-step procedure. First, high vertical resolution data, such as those from ozonesondes, should be interpolated with a least squares fit to the profile for each MLS retrieval pressure level, avoiding any layered structures or jagged features, if they exist. The resultant profile is then convolved using the following equation:

$$X_{comv} = X_a + A[X_{DIAL} - X_a] \quad (1)$$

where X_a is the a priori profile for each retrieval and A is the averaging kernel functions (matrix) of MLS. X_{DIAL} is the DIAL ozone profile, and X_{comv} is the convolved DIAL ozone profile, which is converted to each MLS grid for comparison. DEL: However, in this study, each DIAL profile was already very smooth, and step one was not necessary. We used A for the polar winter condition from two A that have been provided in the MLS dataset, the other is for the tropical condition.

We used 500 km in distance, the great circle (between 47.1°S and 56.1°S for 69.3°W) , and ±24 hours for coincidence criteria between DIAL and MLS measurements. Because the mid-point for the DIAL measurement duration was usually 2-3

UTC, the time differences (MLS – DIAL) were 0-4 hours or 13-17 hours on the same day that correspond to night or day paths of the EOS-Aura orbit. When no MLS measurements were available on the same day (9 cases), measurements one day before were used. In those cases, the time differences were –6 to –10 h or –20 to –24 h. For the DIAL measurement on October 27, an MLS measurement on October 28 was used, resulting in a 26 h difference. Both DIAL measurements on October 7 and 8 used ten MLS measurements on October 7 for matching pairs. In total, 180 matching pairs were used in this study.

For comparisons between DIAL and MIROC-CTM, we also unified the vertical grids for comparison. The DIAL profiles were linearly interpolated onto the pressure grids for the MLS data; the vertical increments of the DIAL profile are as small as 150 m. The MIROC-CTM profiles on the day of each DIAL measurement were interpolated onto the pressure grids for the MLS data using a cubic-spline. Both interpolated values were used to compute differences (MIROC-CTM – DIAL) (see Figures 2, 5, and 6).

For converting the original DIAL geometric altitude and O_3 number density to pressure and O_3 mixing ratio, the NCEP reanalysis data (Kalnay et al., 1996) are used. These data are registered in the NDACC database. Possible deviations could be expected if we use other meteorological data for the conversion process in DIAL. However, in this study, we used the DIAL data that registered in the NDACC database. Another possible deviations could also be expected if we use other meteorological data for the nudging process in MIROC-CTM. The different reanalysis data may cause different vertical and horizontal motions of air in the model, providing different tracer correlations, hence ozone field. However, in this study, we analyze owing to the model of Akiyoshi et al. (2016) to examine the performance.

4 Results and discussion

4.1 Example of vertical profile comparison

Figure 2a shows vertical profiles of O_3 measured with DIAL compared with those of MLS on the same day (November 14, 2009) as an example. The plus-crosses and dotted-line show the converted DIAL profile using Equation (1) and the original high vertical resolution DIAL profile, respectively. Each MLS profile was color-scaled with its measurement latitude to observe the latitudinal difference between DIAL and MLS. The bar in MLS O_3 profiles shows the precision reported for individual profiles. The bar in the DIAL O_3 profile shows the total uncertainty. The combined uncertainty (root sum square) is shown in the right panel. In addition to the DIAL and MLS profiles, we also compared the 24 h average O_3 profiles from MIROC-CTM at 12 UTC. We have extracted data from six locations between 48.8°S and 54.4°S in latitude at 67.5°W and 70.3°W in longitude, but the nearest grid data was plotted in Figure 2a (see Figures 5 and 6 for the variability in six model grids).

On this day, the DIAL profile above 50 hPa, i.e., pressures smaller than that level, revealed lower O_3 values, which was suggested in Wolfram et al. (2012) due to the edge of the southern polar vortex located near OAPA on November 14. Wolfram et al. (2012) also suggested that an altitude region around a potential temperature (PT) of 650 K was just inside the vortex. Several PT levels corresponding to pressure are also shown as text in Figure 2a. This DIAL profile agrees well with MLS profiles observed at similar latitudes, 51.7°S with green lines. The MLS profiles revealed a larger latitudinal difference of ∼2.5 ppmv over ∼8° especially at ∼50 hPa level. For reference, the MIROC-CTM profiles also revealed latitudinal differences of

$\sim$1 ppmv over 5.6° at the same pressure level (not shown), suggesting a weaker latitudinal gradient in the model simulation for these conditions. In addition, the MIROC-CTM O_3 value is higher than from DIAL around 20 hPa levels. We will discuss this feature in the MIROC-CTM in Section 4.2.

In the right panel of Figure 2a, the differences between MLS O_3 and DIAL (MLS – DIAL) are shown. In addition, the difference between DIAL and the nearest MIROC-CTM is shown. In general, the MLS profiles of similar latitudes (51.7°S) with OAPA are in good agreement with the DIAL profile within ±0.5 ppmv between 100 hPa and 6 hPa. The largest negative value is found at 46 hPa, with 2.0 ppmv for a profile of the highest latitude measured (54.7°S). In contrast, the largest positive value is found at 22 hPa, with 1.2 ppmv for a profile of the lowest latitude measured (48.8°S). This indicates that lower O_3 values still exist inside the vortex, i.e., depleted ozone in the spring time has not yet recovered, and larger O_3 values are found outside the vortex at the lower latitudes in the middle stratosphere.

Another example is shown in Figure 2b. On this day, November 23, there were less latitudinal differences in ozone field compared to the result on November 14 as observed by MLS. Consequently, the latitudinal difference in MIROC-CTM is also smaller on November 23 than on November 14 (not shown). Similar to the former result, the MLS profile at a similar latitude with OAPA is in good agreement with the DIAL profile within ±0.5 ppmv between 83 hPa and 6 hPa. Whereas, the MIROC-CTM O_3 is lower than DIAL by $\sim$–2 ppmv between 10 hPa and 6 hPa. This is discussed in Section 4.4.

4.2 Time series comparison

All 23 DIAL profiles obtained in September-November 2009 were evaluated for their variability with time. The PV values at the location and time of all O_3 profiles from DIAL, MLS, and MIROC-CTM were investigated to place the measurements inside or outside the polar vortex. The degree of PV values at each measurement or model grid is a robust indicator of the location relative to the polar vortex. Here, we used meteorological data from the NASA Global Modeling and Assimilation Office (GMAO) Modern-Era Retrospective Analysis for Research and Applications-2 (MERRA-2) reanalysis (Molod et al., 2015; Gelaro et al., 2017) (gmao.gsfc.nasa.gov/reanalysis/MERRA-2/). We calculated the scaled PV (sPV) for pressures between 100 hPa and 6 hPa from the PV values from MERRA-2 and PV/sPV ratios as a function of PT. The PV and sPV values are provided through the MLS website as the derived meteorological products (DMPs) (Manney et al., 2007). We used version 2 of DMP (GEOS5MERRA2 for the version 4 MLS data). sPV values (s^{-1}) are nearly constant at levels throughout the stratosphere (e.g., Dunkerton and Delisi, 1986; Manney et al., 1994, 2007). Figure 3 shows all the 23 profiles of O_3 obtained by DIAL. Data are color-scaled based on sPV values. The difference in the O_3 value is found depending on the sPV value especially above 30-40 hPa. Figure 4 shows sPV maps from MERRA-2 for selected days on September 26, October 3, November 14, and November 23, 2009. At 20 hPa, the polar vortex significantly diminishes on November 23 compared to that on September 26. Whereas at 50 hPa, the polar vortex still exists on November 23 with smaller area than that on September 26.

An sPV value of $\sim 1.4 \times 10^{-4}\ s^{-1}$ has been used to define the Northern Hemisphere (N.H.) polar vortex edge center (e.g., Ryan et al., 2016, and references therein). In addition, values of $\sim$1.6 and $\sim 1.2 \times 10^{-4}\ s^{-1}$ have been used to define the inner and outer edges, respectively. Those vortex edge definitions, i.e., center, inner, and outer, are according to Nash et al. (1996). We examined these values using the DMPs for the MLS measurements for the period studied here (i.e., September to November

2009). The results were somewhat different from those from the N.H. depending on time and altitude. For example, center, inner, and outer boundaries are defined by the absolute sPV values of 1.6, 1.9, and $1.3 \times 10^{-4}\ s^{-1}$ at 68 hPa in November. The sPV values shown in the following figures are useful guides for showing positions relative to the vortex.

As representatives for the middle and the lower stratospheres, results at 18 hPa and 56 hPa are shown in Figure 5 and Figure 6, respectively. Figure 5a shows the time variation of O_3 values obtained from DIAL and MLS at 18 hPa. Both O_3 values are color-scaled using sPV values. On several occasions, O_3 values below 4 ppmv were measured by DIAL in air masses with larger sPV values, i.e., larger negative values indicated with blue and purple colors, in conjunction with the polar vortex dynamics.

For both September 26 and October 5, the polar vortex shifted toward the South American side, covering the OAPA site. On November 13-14, the O_3 values were low again. Correspondingly, the MLS O_3 values also show lower values with higher sPV values. In general, the DIAL O_3 values are within the variations of MLS O_3 values for each coincident date during all comparison periods. To quantitatively evaluate the degree of agreement, the differences between the two (MLS – DIAL) are shown in Figure 5c. These values are color-scaled using the sPV value from each MLS measurement. We computed mean and root-mean-square (rms) differences of O_3 from all 180 data points. At 18 hPa, the mean difference is –0.03 ppmv and the rms difference is 0.78 ppmv. Although the mean value shows a good agreement, the variance is large especially in September. We will discuss this large variance in Section 4.3.

Figure 5b for 18 hPa also shows time variations in O_3 values obtained from DIAL and those simulated with MIROC-CTM. Figure 5d shows the O_3 differences between DIAL and MIROC-CTM (MIROC-CTM – DIAL). In this plot, mean and rms differences in O_3 are calculated from all data points of the nearest model grid, 51.6°S/70.3°W, to the OAPA site (the number is 23). As a result, the mean difference is 0.04 ppmv and rms difference is 0.72 ppmv. For reference, Figures 5e and 5f show the relative differences for DIAL/MLS and DIAL/MIROC-CTM comparisons, respectively.

Similar to the DIAL-MLS comparison, both the DIAL and MIROC-CTM O_3 values show low values with larger sPV values, which indicate that the locations are inside the polar vortex, or that the air masses originate from the polar vortex. However, MIROC-CTM overestimates O_3 values with the larger sPV values compared to DIAL. When those higher deviations in MIROC-CTM are found, the DIAL O_3 values show smaller amounts below $\sim$4 ppmv (Figure 5b). This is also observed in the vertical profile in Figure 2a. The overestimate of MIROC-CTM may be partly due to the relatively coarse horizontal resolution of the model with regard to a complicated spatial structure near the boundary of the polar vortex in the breakup season. The polar vortex begins to breakup at higher altitudes, and then propagates downward. Another possible explanation could be due to a weaker vertical motion of air in MIROC-CTM. Although not shown, a vertical profile of nitrous oxide, N_2O, from MIROC-CTM on November 14, 2009 is different from that from MLS. A tight correlation between N_2O and Cl_y is found in the stratosphere (e.g., Schauffler et al., 2003), and used to infer the Cl_y value from a measured N_2O value (e.g., Wetzel et al., 2010; Strahan et al., 2014). At 18 hPa, the MIROC-CTM N_2O value is higher than that of MLS, resulting in a smaller value of Cl_y in MIROC-CTM. Thus, a smaller active chlorine (ClO_x) induces a higher O_3 amount in MIROC-CTM.

Figures 6a and 6c show time variations in O_3 values from DIAL and MLS, and the difference between the two at 56 hPa, similar to Figures 5a and 5c. Figures 6b and 6d also show time variations in O_3 values at 56 hPa from DIAL and MIROC-

CTM, and the difference between the two, similar to Figures 5b and 5d. Figures 6e and 6f show the relative differences for DIAL/MLS and DIAL/MIROC-CTM comparisons, respectively. Unlike the characteristics of the 18 hPa result, significant lower ozone values relative to the other dates were not found inside the polar vortex on September 26 and October 5. Whereas, on November 13-14 and 23-24, lower O_3 values inside the polar vortex were found from both of DIAL and MLS. This is in agreement with the long-lasting polar vortex dynamics in the 2009 spring (de Laat et al., 2010; Wolfram et al., 2012). The mean differences between DIAL and MLS/MIROC-CTM are as small as 0.06 ppmv and 0.16 ppmv, respectively. The rms differences are 0.46 ppmv and 0.36 ppmv for DIAL/MLS and DIAL/MIROC-CTM comparisons, respectively, which are smaller values than those at 18 hPa. The overestimate of MIROC-CTM with larger sPV values, as seen at 18 hPa is not evident at 56 hPa. One explanation may be that the polar vortex is more stable at 56 hPa than at 18 hPa, even on November 23-24.

4.3 Dependency in distance and sPV difference

The good correlation between sPV and O_3 values near the vortex boundary in austral spring has been previously shown in satellite measurements (e.g., Manney et al., 1999, 2001, 2005). Therefore, a horizontal gradient in O_3 should have been present at the vortex boundary in the 2009 spring. A previous study suggested that a better agreement is found when the comparison is performed with matching meteorological conditions using parameters such as sPV and equivalent latitude (Manney et al., 2001). Therefore, we further examined the larger variability between DIAL and MLS at 18 hPa, from the perspective of different sPV values. Figure 7a shows the O_3 difference (MLS – DIAL) versus sPV difference between DIAL and MLS (MLS – DIAL). Similar to Figure 5b, the data points are color-scaled based on the sPV values of the MLS measurements. A positive correlation between O_3 and sPV differences is found, suggesting lower O_3 values in MLS (negative in the y-axis) with a more poleward MLS profile, i.e., negative in the x-axis. Conversely, higher O_3 values in MLS, i.e., positive in the y-axis, with the lower latitude side profile in MLS, i.e., positive in the x-axis, is also seen, although the correlation is weaker than in the negative value area. After filtering out matching pairs over a certain sPV difference, e.g., below or above $\pm 0.3 \times 10^{-4}$ s^{-1}, the rms difference between DIAL and MLS at this pressure level decreases significantly. Such an sPV criterion is useful for suppressing the large rms difference found in O_3 measurements affected by the motion of polar vortex. Whereas, the mean difference less changes applying such the sPV criterion. This is consistent with the result from Holl et al. (2016) who showed differences in CH_4 values observed in the northern high latitude and sPV criterion with a value of 0.2×10^{-4} s^{-1} has little effect below 25 km in altitude.

We also examined results from 56 hPa in Figure 7b. Similar to the results from 18 hPa, larger O_3 differences are found with larger sPV differences. Applying certain sPV criterion to these data, the mean difference changes only slightly, but the rms difference decreases, similar to the results from 18 hPa. The results for other pressure levels are summarized in Section 4.4.

Since the MERRA-2 data set also provide the O_3 value (Wargan et al., 2017), we examined those data instead of the sPV value. Figure 8 shows the O_3 difference versus MERRA-2 O_3 difference between DIAL and MLS (MLS – DIAL). The mean difference is computed from the horizontal axis, resulting in –0.12 ppmv at 18 hPa and –0.02 ppmv at 56 hPa. The measured O_3 difference is well reproduced by the MERRA-2 O_3 that assimilates Aura MLS as well. At 56 hPa, a compact correlation is found between the two differences with a slope of one-by-one. A similar positive correlation is also found at 18 hPa.

In addition to the sPV differences examined, we evaluated the correlation between the O_3 difference and distance in the DIAL/MLS measurements (Figure 9). In these figures (Figure 9a for 18 hPa and Figure 9b for 56 hPa), data points are color-scaled based on the sPV difference between DIAL and MLS (MLS – DIAL). Clearly, larger O_3 differences, especially those with negative values in Figure 9a, have large sPV differences, i.e., below -0.5×10^{-4} s^{-1}). As shown in the figures, the O_3 difference does not depend critically on the distance between the two measurements.

In summary, the O_3 differences between DIAL and MLS can be partly attributed to differences in the measurement points. Furthermore, the O_3 difference is more correlated with sPV differences between than the difference in distance. Therefore, it is important to analyze O_3 values with sPV (or PV) values near the polar vortex boundary, which has been suggested previously (e.g., Manney et al., 2001).

4.4 Comparison at other levels: summary

The mean and rms differences computed from the time-series comparisons in Section 4.2 were extended for other pressure levels to summarize the degree of agreement between DIAL and MLS or MIROC-CTM. These results are plotted versus pressure in Figure 10. Absolute differences are shown in the left panel. Relative differences, the absolute differences divided by their mean values of O_3, are shown in the right panel. In the left panel, mean differences (open circle and cross) for both DIAL/MLS and DIAL/MIROC-CTM comparisons, along with rms differences (dotted lines) are shown. The mean differences of the DIAL/MLS comparison are almost within ±0.1 ppmv between 6 hPa and 56 hPa with 180 data points for each level. This corresponds to the relative values, in the right panel, of ±3%. Figure 11 shows differences between DIAL and MLS using the sPV criterion. The mean and rms differences shown in this figure as blue lines are identical to Figure 10. The mean and rms differences after filtering with the sPV criteria ($\pm 0.3 \times 10^{-4}$ s^{-1}) are shown as green lines. Clearly, the rms differences decrease 21-47% between 10 hPa and 56 hPa; the number of data points was reduced from 146-180 to 107-144. However, the mean differences only change slightly for all pressure levels, except for the 6 hPa level.

For the DIAL/MIROC-CTM comparison, the mean differences are almost within ±0.3 ppmv between 10 hPa and 56 hPa, with 23 data points for each level. This corresponds to relative values of ±8%. Above 8 hPa, the absolute differences increase to –0.6 ppmv which corresponds to relative values of –8%. To examine the low bias in MIROC-CTM, the time-series in O_3 difference between DIAL and MIROC-CTM at 8 hPa is shown in Figure 12. Larger negative deviations in MIROC-CTM are found in October and November, especially for data with sPV values between –1.0 and -1.5×10^{-4} s^{-1}. Similar results are also found from 6 hPa and 7 hPa levels. The peak altitude of ozone in MIROC-CTM is lower than that of DIAL, as shown in Figure 2. Both the vertical and horizontal motions of air in the model are responsible for this different feature, but the cause is not known. As was shown in Figure 3, the vertical gradient of O_3 from DIAL above 15-20 hPa shows rather week inside the polar vortex, but occasionally strong outside or edge of the polar vortex. Thus, the vertical gradient of O_3 may affect the result for such occasions with the steeper gradient. DEL: Although not shown in the figure, a time-series for 6-8 hPa shows that larger negative deviations in MIROC-CTM are observed in October and November. The peak altitude of ozone in MIROC-CTM is lower than that of DIAL, as shown in Figure 2. Both the vertical and horizontal motions of air in the model are responsible for this different feature, but the cause is not known. The feature presented here suggests a difficulty in the reproduced ozone field for those pressure levels (6-8 hPa) in these latitudes and season using this version of MIROC-CTM. As discussed in Section 4.2, the polar vortex breakup process may cause a highly variable spatial structure. This may be partly responsible for the difference because of the insufficient spatial resolution of the model to distinguish this process.

Both the DIAL/MLS and DIAL/MIROC-CTM comparisons show increasing rms differences with increasing altitudes above the 20-30 hPa levels, reaching more than 1 ppmv. This is partly due to the O_3 value increasing with increasing altitudes. Thus, relative values of the rms difference (Figure 10b) do not show strong vertical gradients compared to the absolute values (Figure 10a).

Both comparisons also show larger absolute differences below 68 hPa, reaching 0.5 ppmv (116%) for DIAL/MLS and 0.9 ppmv (292%) for DIAL/MIROC-CTM. This suggests a lower bias in the DIAL measurement at these lower altitudes (∼80-100 hPa) of some magnitude. As discussed in Wolfram et al. (2008), this DIAL system has some difficulty in measuring around 100 hPa and below due to saturation from backscattered photons in the low-energy channels. Since the O_3 mixing ratio from DIAL is very small below about 70 hPa, the sensitivity might be degraded along with the saturation effect. Therefore, DIAL data at this altitude range should be used with caution.

Another possible reason is the difference in measured ozone associated with the difference in original vertical resolution, ∼1 km for DIAL versus 3 km for MLS. In this period, lamina structures in O_3 profiles are often observed from ozonesonde measurements, especially below 20 km. DIAL may capture lower values of O_3 in these lamina structures while collecting measurements over 3-5 h, compared to MLS that measures instantaneously along the orbit, nearly the north-south direction (see Supplement). This may facilitate O_3 differences, to a certain extent, even while both measurements are accurate. In the other geophysical regions of the Asian monsoon anticyclone, difficulties in MLS retrievals within the strong vertical gradient of O_3 have been discussed (Yan et al., 2016). The largest O_3 difference between DIAL and MLS at 83 hPa was found on October 3, 2009; this case was studied using air mass trajectory analysis (Tomikawa and Sato, 2005) and the O_3 field from MIROC-CTM (see Supplement).

5 Conclusions

Ground-based DIAL measurements were performed at the OAPA in Río Gallegos (51.6°S, 69.3°W), Argentina, from September to November 2009, when a long-lasting southern polar vortex, and accompanying ozone depletion, occurred over the area for the first time since 1979 (de Laat et al., 2010; Wolfram et al., 2012). This site is one of the few NDACC DIAL sites in the S.H. Focusing on this period of large dynamical variability in measured air masses during the movement of the polar vortex, it is possible to analyze the effects of the polar vortex on O_3 variability. Twenty-three O_3 profiles were obtained by DIAL during the period. These profiles were compared with coincident MLS O_3 profiles with 180 matching pairs, based on time and space criteria.

The mean differences between DIAL and MLS are within ±0.1 ppmv (±3%) from 6 hPa to 56 hPa, showing good agreement regardless of the large sPV variability between each matching pair. The DIAL data are also compared with outputs from the MIROC-CTM model simulation. The mean differences between DIAL and MIROC-CTM are within ±0.3 ppmv (±8%) from

10 hPa to 56 hPa. Above 8 hPa, the mean differences increases to –0.6 ppmv (–8%). To measure variability in the comparison, rms differences between DIAL and MLS or MIROC-CTM are also evaluated. For both DIAL/MLS and DIAL/MIROC-CTM comparisons, the rms differences are nearly 0.5 ppmv for pressure levels between 30 hPa and 100 hPa, and increase with increasing altitudes up to 6 hPa, reaching 1.1-1.2 ppmv. From the DIAL/MLS comparison, the O_3 differences depend on sPV differences at 18 hPa. Therefore, another criterion for comparison is proposed: pairs with absolute sPV differences that exceed $0.3 \times 10^{-4}\ s^{-1}$ are discarded. As a result, the rms differences decreased significantly between 10 hPa and 56 hPa, but the mean differences only slightly change for all pressure levels, except for 6 hPa.

The comparison between DIAL and MLS indicates that the O_3 difference is partly due to sPV differences between measurement locations; however as yet unknown factors create additional differences. The comparison between DIAL and MIROC-CTM indicates that an insufficient model spatial resolution may be partly responsible for the O_3 differences above 18 hPa during polar vortex breakup. An insufficient model vertical motion may also be partly responsible for the O_3 differences, especially inside the polar vortex. Both the DIAL/MLS and DIAL/MIROC-CTM comparisons also show larger mean differences below 68 hPa, reaching 0.5 ppmv (116%) and 0.9 ppmv (292%) at 100 hPa, respectively. One possible cause may be a low bias in the DIAL O_3 measurement, but this hypothesis was not confirmed in this study. Nevertheless, finding good agreement between DIAL and MLS O_3 measurements between 6 hPa and 56 hPa is a necessary step for studies in evaluating bias and long-term stability of satellite sensors in the future. Because of very sparse observations from S.H. ground-based stations, continuation for long-term measurements there for NDACC is highly recommended. This study provides an outlook for continuing measurements at the OAPA site. The DIAL measurements at the OAPA site are available for all years since 2005, except 2016 when no measurements were collected. The result of the DIAL/MLS comparison using these long-term data will be published elsewhere.

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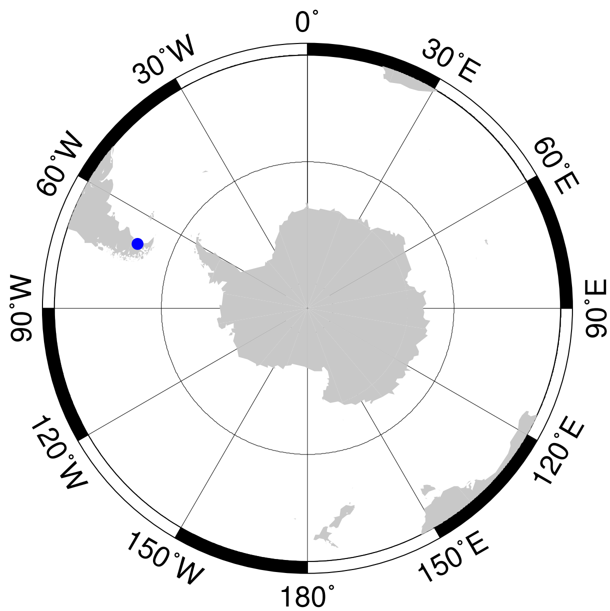
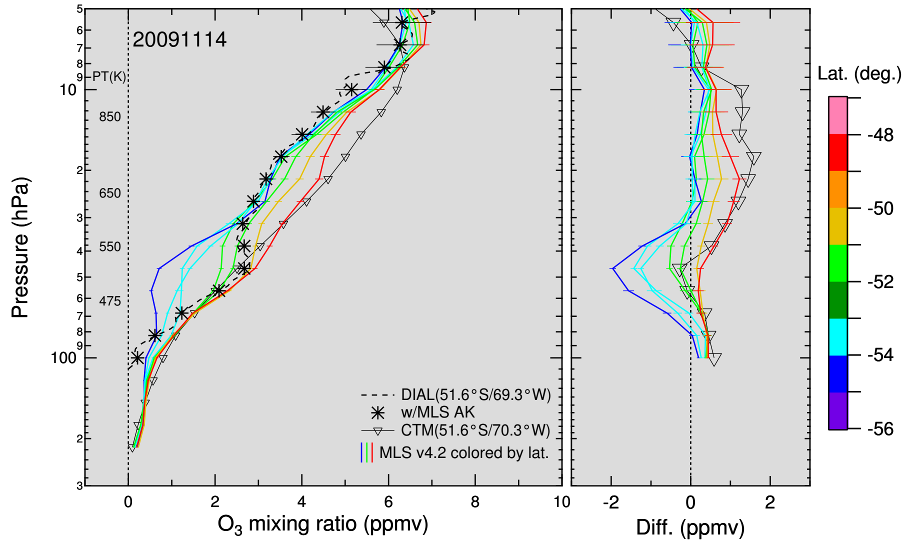
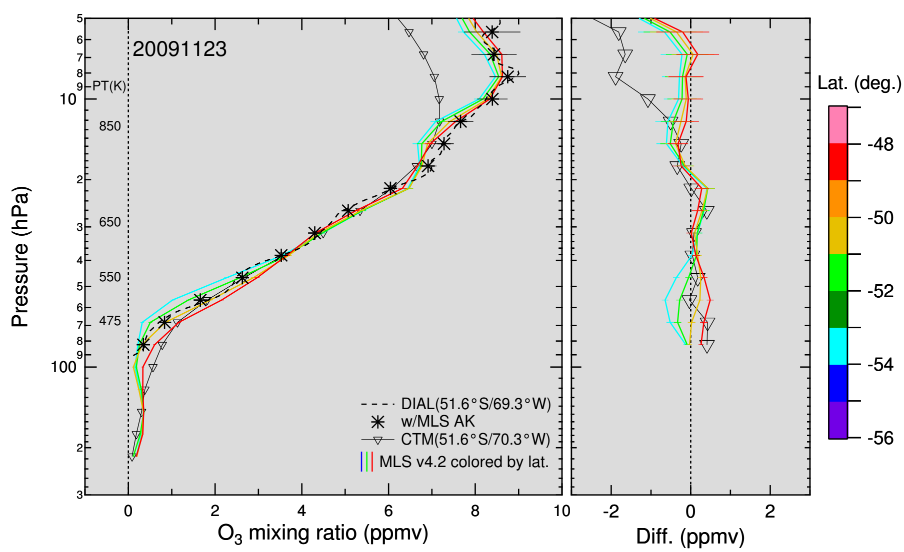


Figure 1. Location of the OAPA site in Río Gallegos, Argentina (51.6°S, 69.3°W), shown as a blue circle. Latitude ranges from 30°S to 90°S.



(a)



(b)

Figure 2. Vertical profiles of O_3 mixing ratios on November 14, 2009 (a) and November 23, 2009 (b) measured using DIAL (plus-crosses and dotted-line) and MLS (solid-lines with color) over the OAPA site (see text for additional description). A MIROC-CTM O_3 profile of nearest grid for the OAPA site is also shown. Corresponding potential temperatures for pressure are shown as text in the vertical axis. Differences between DIAL and X (MLS or MIROC-CTM) (X – DIAL) are shown in the right panel (see text). The MLS profiles are color-scaled based on their measurement latitudes.

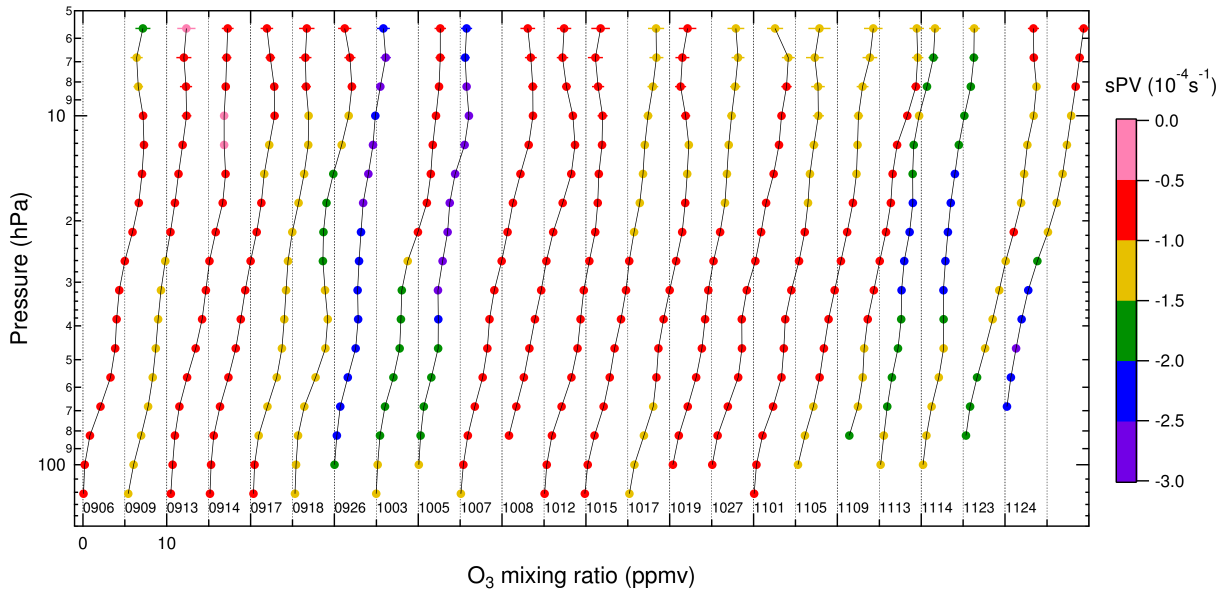


Figure 3. Time series of DIAL O_3 profiles at the OAPA site. Each profile is shifted 5 ppmv. Data are color-scaled based on sPV values. Observation dates in 2009 are shown as MMDD, e.g., 0906 is September 6, 2009.

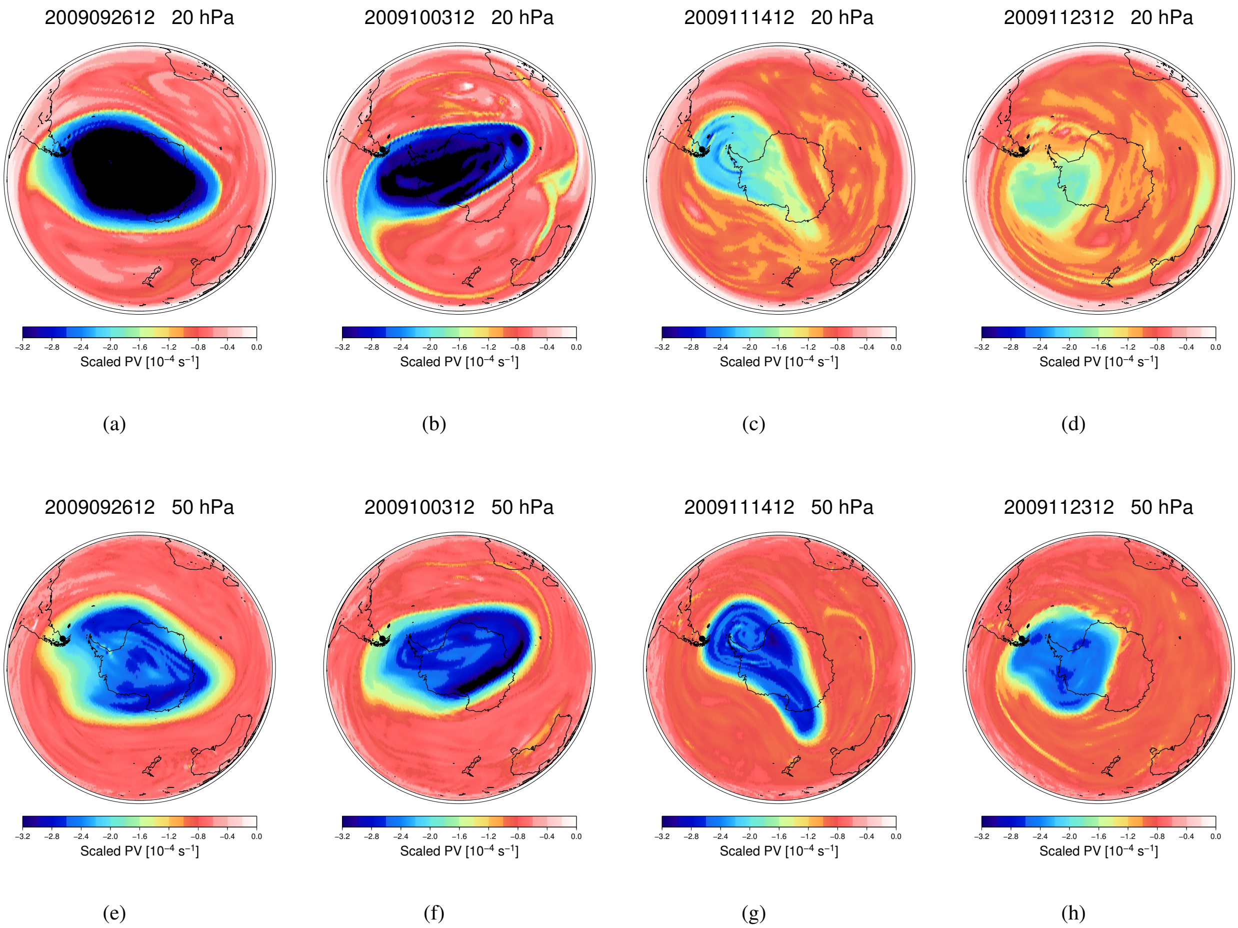


Figure 4. Scaled PV maps from MERRA-2 on September 26 (a, e), October 3 (b, f), November 14 (c, g), and November 23, 2009 (d, h). Top and bottom rows show pressure surfaces at 20 hPa and 50 hPa, respectively.

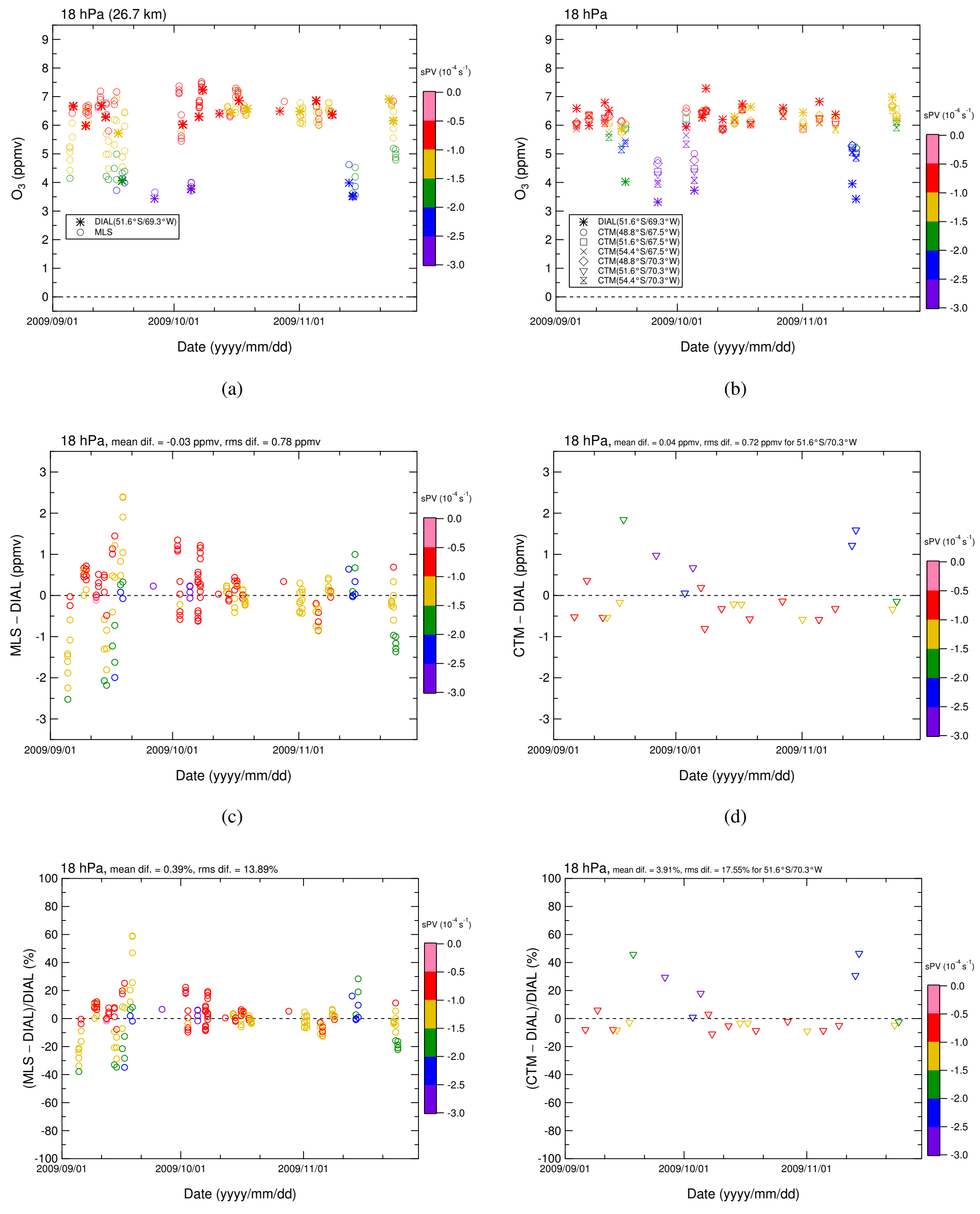


Figure 5. Time series of O_3 mixing ratios as measured by DIAL and MLS at 18 hPa (a) and absolute and relative differences between the two (c, e) from September to November 2009, over the OAPA site. (b) and (d, f) are same as (a) and (c, e), but for DIAL and MIROC-CTM. Data are color-scaled based on sPV values. For the absolute and relative differences, sPV values for MLS and MIROC-CTM are color-scaled. For MIROC-CTM, outputs from six grids are shown (see text for additional description).

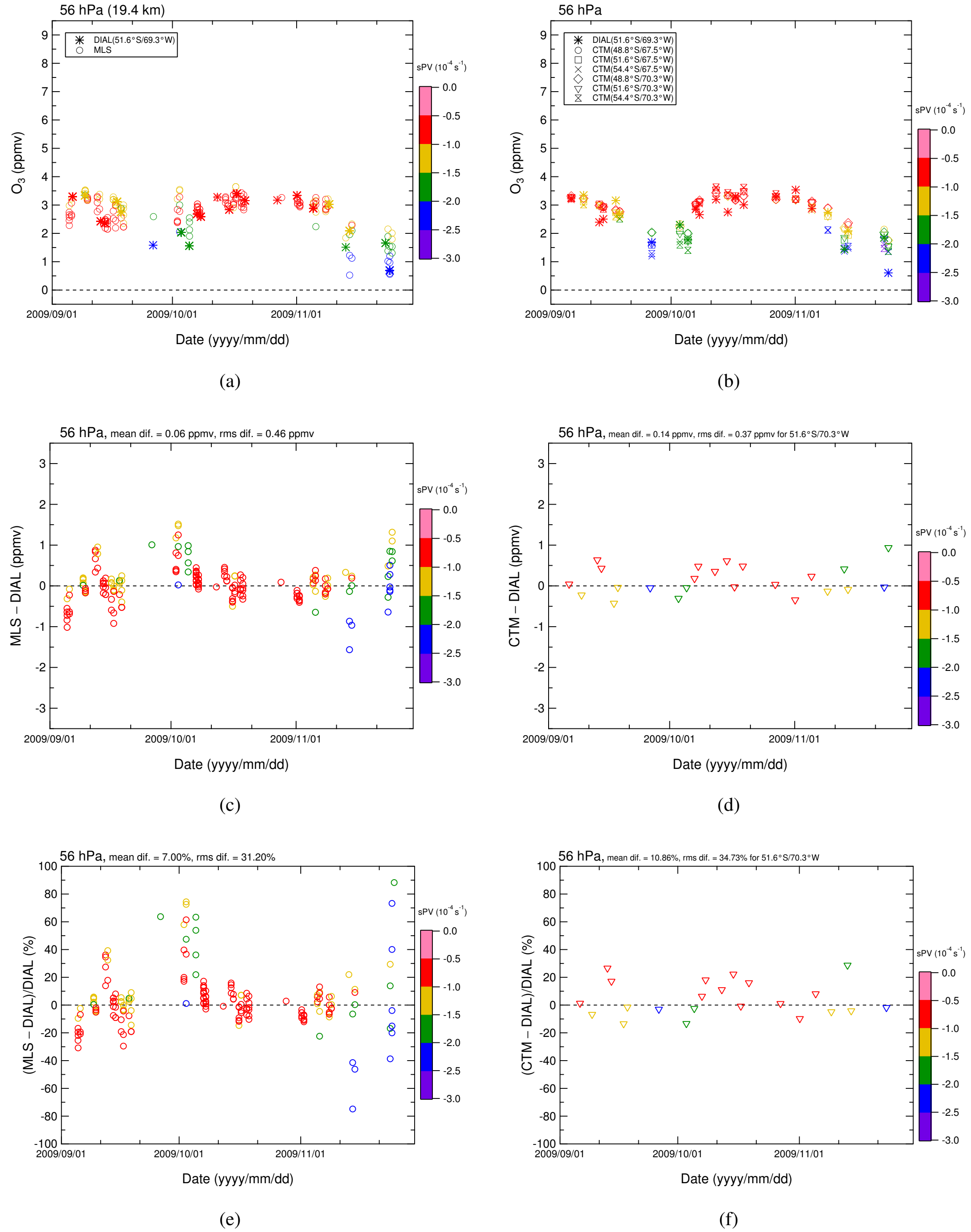
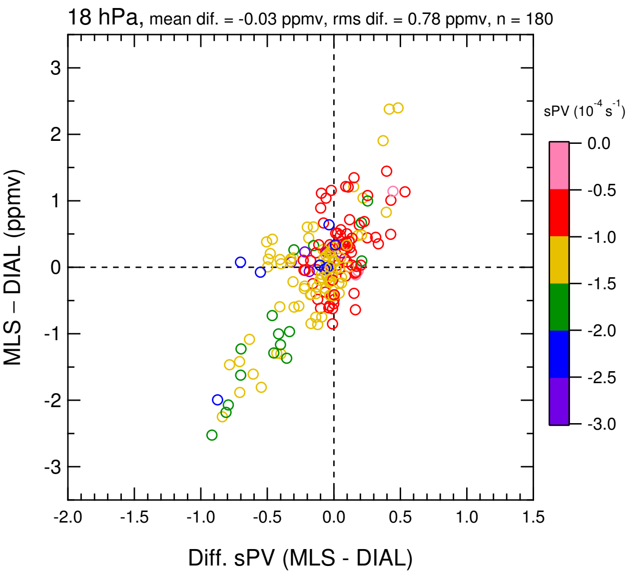
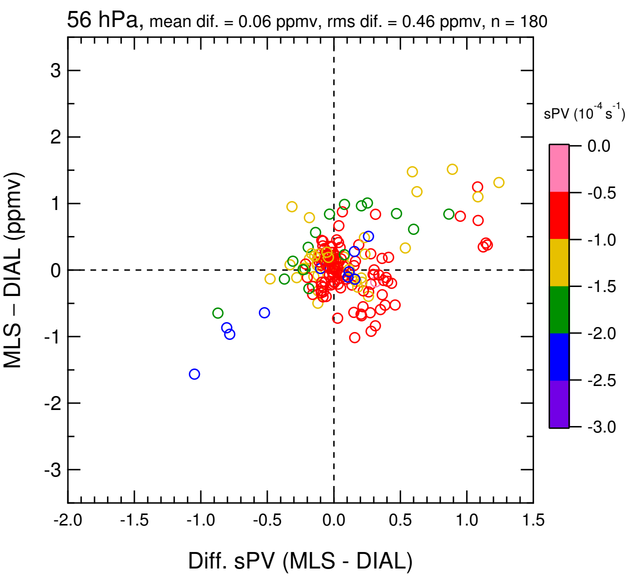


Figure 6. Time series of O_3 mixing ratios as measured by DIAL and MLS at 56 hPa (a) and absolute and relative differences between the two (c, e) from September to November 2009, over the OAPA site. (b) and (d, f) are same as (a) and (c, e), but for DIAL and MIROC-CTM. Data are color-scaled based on sPV values. For the absolute and relative differences, sPV values for MLS and MIROC-CTM are color-scaled. For MIROC-CTM, outputs from six grids are shown (see text for additional description).

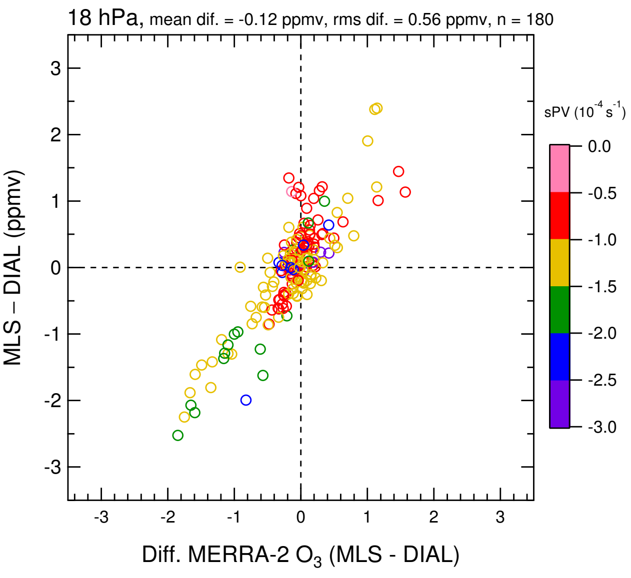


(a)

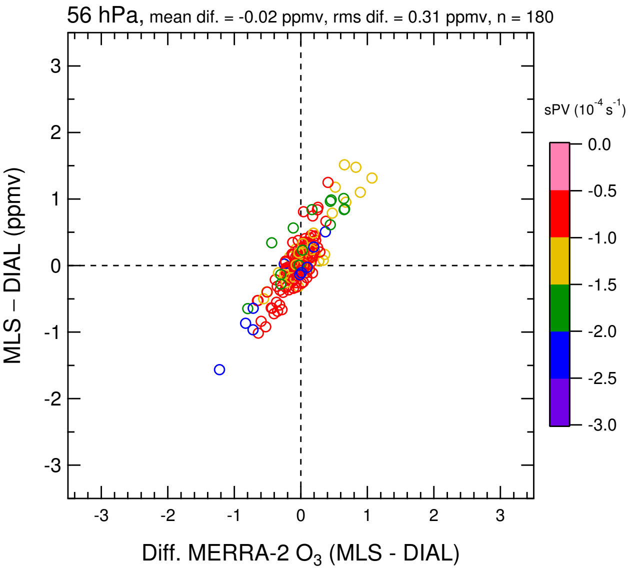


(b)

Figure 7. O_3 difference versus sPV difference for DIAL and MLS at 18 hPa (a) and 56 hPa (b). Data are color-scaled based on the sPV for the MLS measurements.

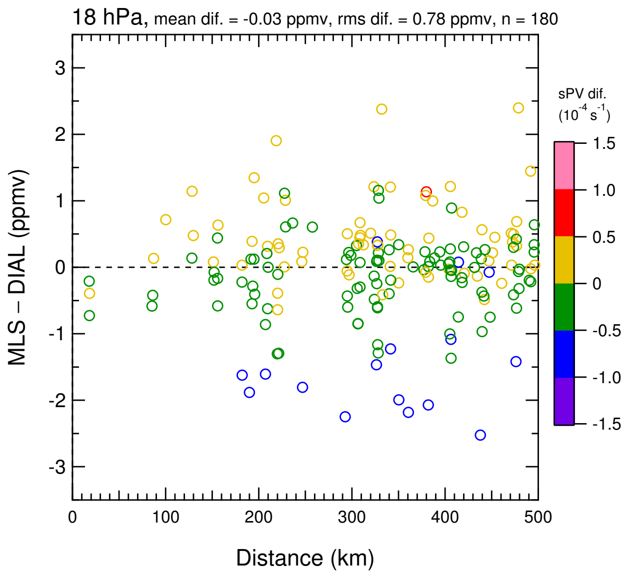


(a)

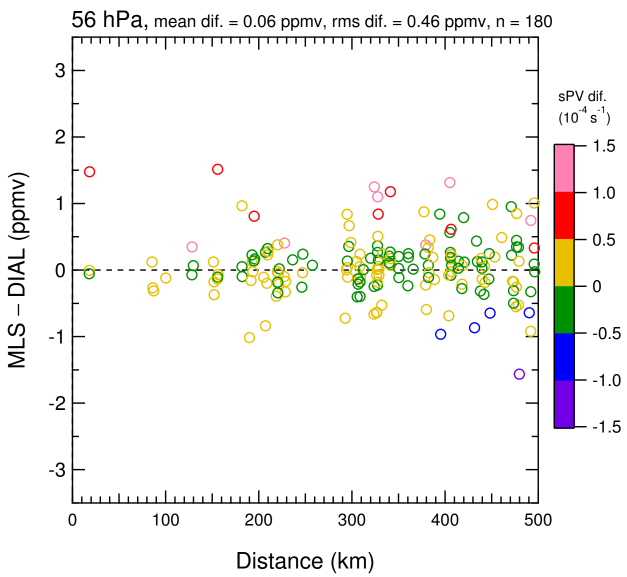


(b)

Figure 8. O_3 difference versus MERRA-2 O_3 difference for DIAL and MLS at 18 hPa (a) and 56 hPa (b). Data are color-scaled based on the sPV for the MLS measurements.

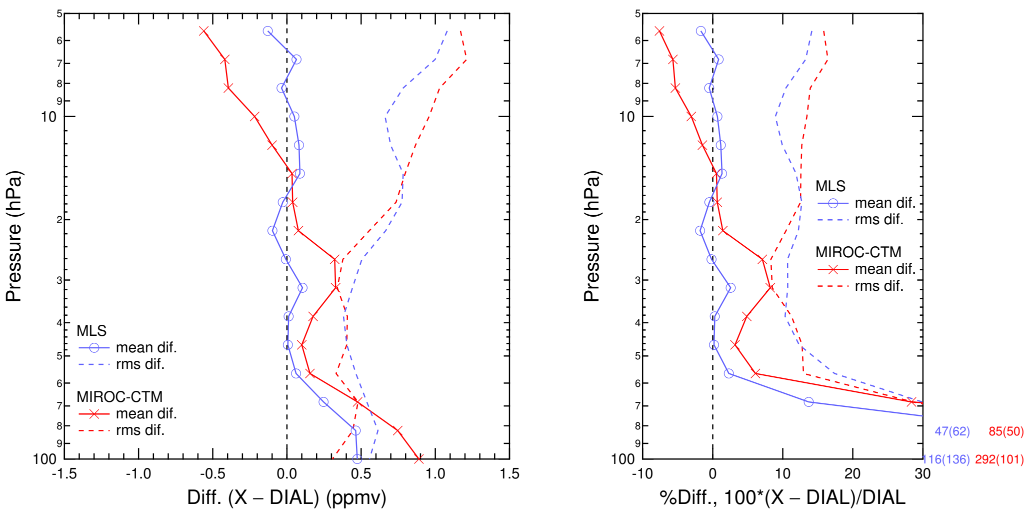


(a)



(b)

Figure 9. O_3 difference versus distance for DIAL and MLS at 18 hPa (a) and 56 hPa (b). Data are color-scaled based on sPV differences between DIAL and MLS measurements.



(a) (b)

Figure 10. Vertical profiles of mean and rms differences of O_3 values for DIAL and X (MLS or MIROC-CTM) (a) and those of relative values (b). Each value is computed from each pressure level in the time series as shown in Figures 5 and 6. The numbers outside the plot are values of the mean (rms in parentheses) difference at 83 hPa and 100 hPa.

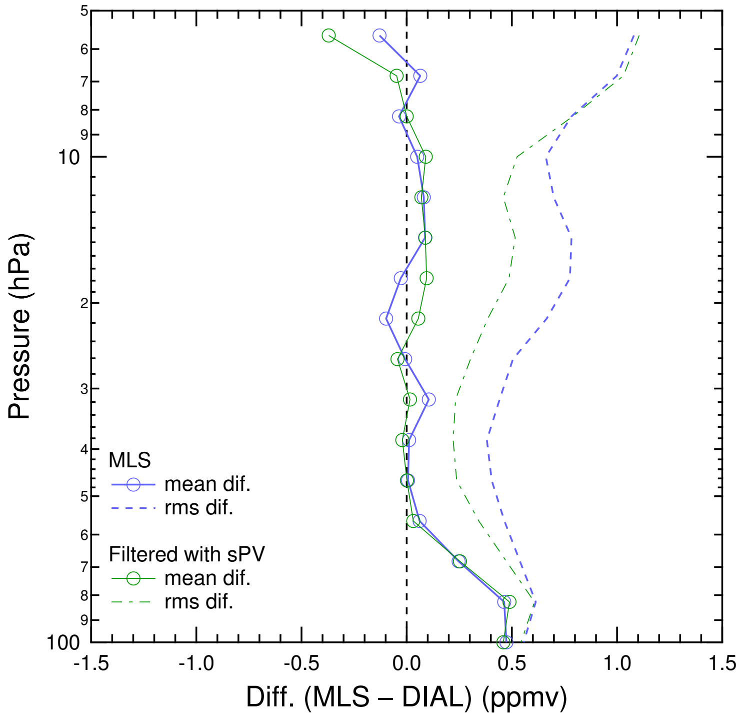


Figure 11. Vertical profiles of mean and rms differences with and without scaled PV criterion screening for the DIAL/MLS O_3 comparison.

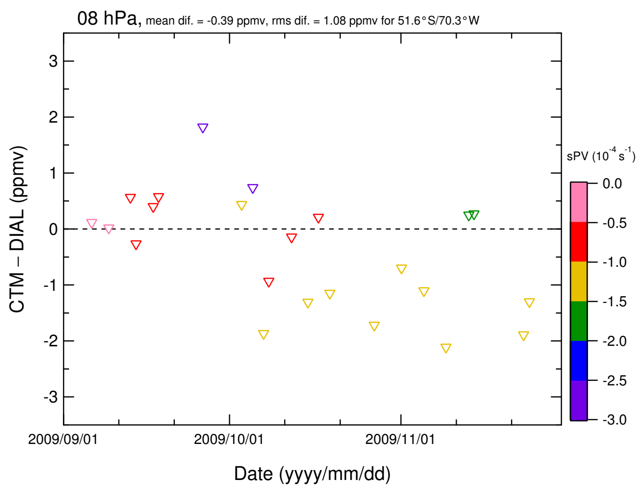
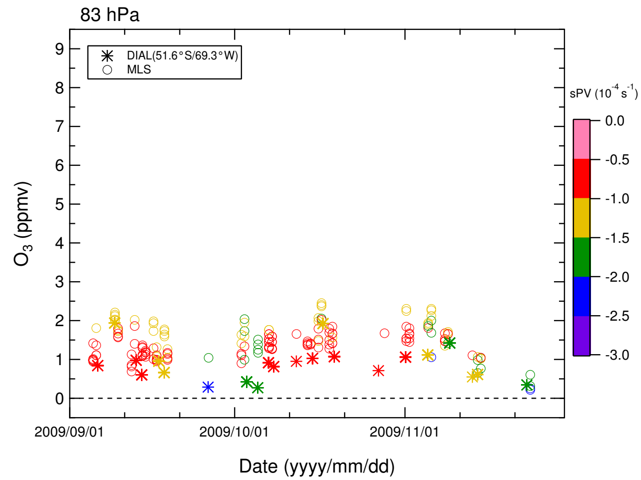


Figure 12. Time series of differences in O_3 mixing ratios as measured by DIAL and computed by MIROC-CTM at 8 hPa from September to November 2009, over the OAPA site. Data are color-scaled based on sPV values for MIROC-CTM.

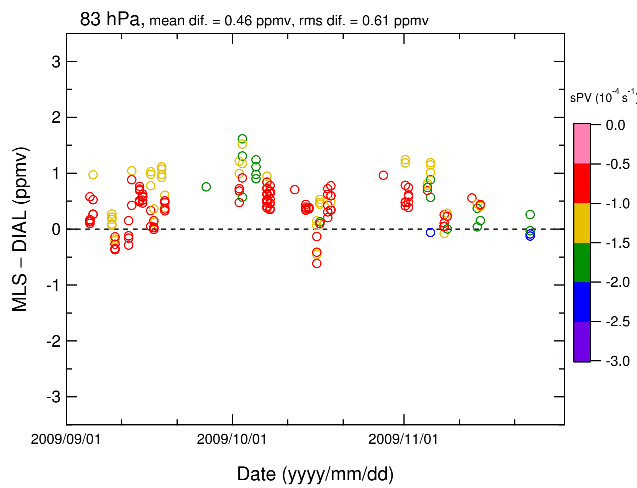
Supplement to the paper

In this Supplement, we examine the large difference between DIAL and MLS at 83-100 hPa. A case study is performed for October 3, 2009, when the largest difference was observed at 83 hPa (see Figure S1). The matching MLS pairs were extracted from one day before the DIAL measurement. The locations are plotted as pluses in Figure S2. The blue and red arrows in this figure indicate nighttime (∼0530 UTC) and daytime (∼1900 UTC) paths of the MLS measurement, respectively. The numbers for each MLS location show the O_3 mixing ratios of each measurement. The O_3 value of the DIAL measurement was 0.4 ppmv with an sPV value of -1.6×10^{-4} s^{-1}. The MLS line-of-sight is in the forward direction of the Aura spacecraft flight track, resulting in a nearly north-south direction (see Figure S2). The cross-track resolution is 6 km wide in the MLS 240 GHz field of view. The duration of DIAL measurements on this day was approximately from 0230 UTC to 0630 UTC. Therefore, we computed the backward air mass trajectories (Tomikawa and Sato, 2005) (www.firp-nitram.nipr.ac.jp/en/) at OAPA from 0430 UTC (the middle time) and 0630 UTC (the end time) to 0230 UTC (the start time), which are shown in Figure S2 as dotted and solid lines. The trajectories were calculated using the MERRA data on the isobaric surface at 83 hPa (see Table S1). This suggests that the DIAL-measured air mass was an average of an almost east-west direction, ∼800 km horizontally.

Next, we evaluated the MLS measurements at the region that the DIAL-measured air mass originated. There were several MLS measurements on October 3 at 83 hPa, as shown by crosses in Figure S2, although no measurements were found within 500 km of the OAPA site. The O_3 values were from 0.5 ppmv to 1.2 ppmv, which are smaller than those on October 2. Thus, the large O_3 differences between DIAL and MLS observed in Figure S1 could be partly explained by techniques measuring different air masses. Such a difference in O_3 field is also reproduced in the MIROC-CTM simulation, as shown in Figures S3 (October 2) and S4 (October 3). From the simulations, it is clear that there are larger O_3 values over the OAPA site on October 2 than on October 3. As shown in Figure S2, the sampling volume of the model is largest, i.e., 2.8° × 2.8° × 1 km, in the vertical at this pressure level based on the daily average. Therefore, the fine scale structure of the O_3 field can not be reproduced in this model.



(a)



(b)

Figure S1. Time series at 83 hPa of O_3 mixing ratios measured using DIAL and MLS (a) and differences between the two (b) from September to November 2009 over the OAPA site. Data are color-scaled based on sPV values.

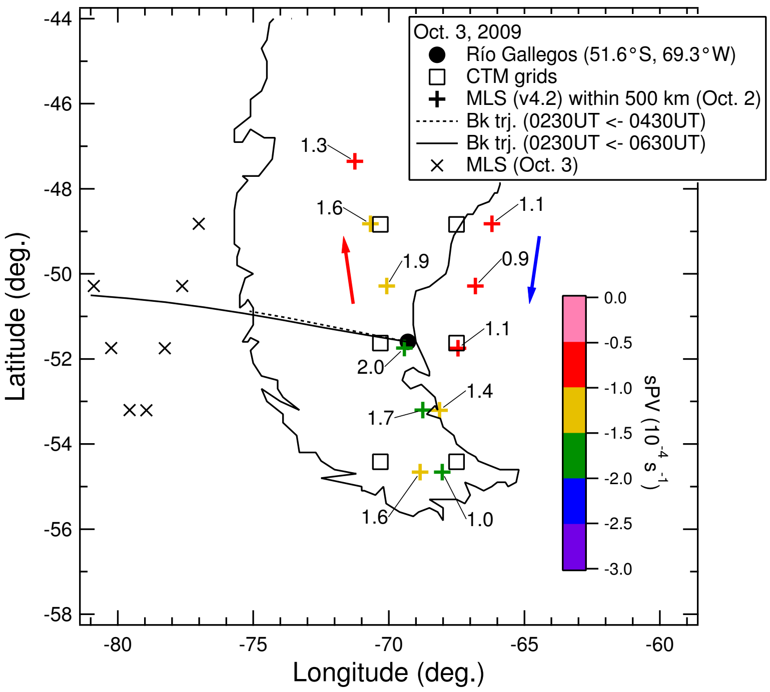


Figure S2. Locations of DIAL and MLS measurements on October 3, 2009. MLS data are color-scaled based on sPV values. Numbers are O_3 mixing ratios in ppmv. The six MIROC-CTM grids are also shown. The air mass trajectories from OAPA are shown as dotted and solid lines. See text for detail.

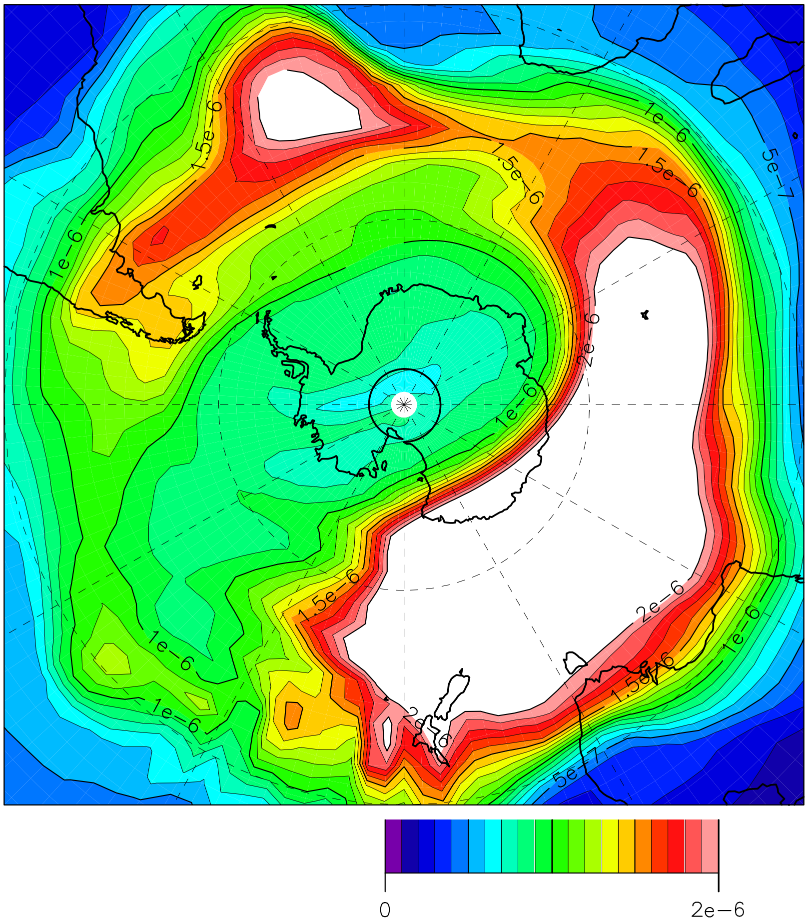


Figure S3. O_3 mixing ratios at 80 hPa computed using MIROC-CTM for October 2, 2009. The mixing ratio ranges from 0 to 2 ppmv.

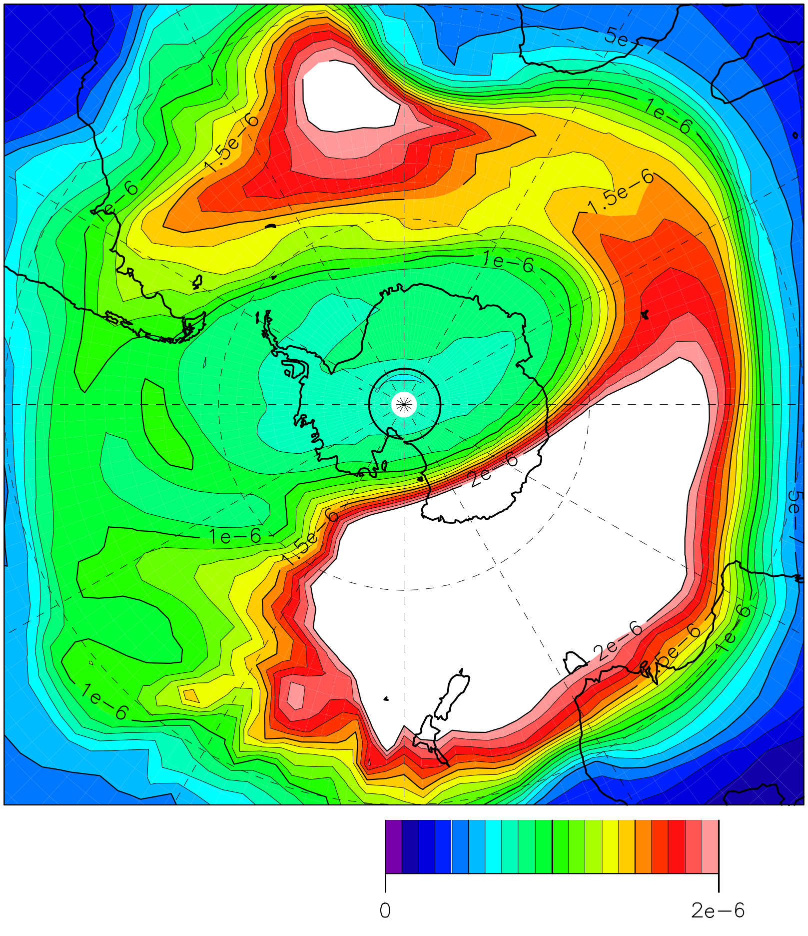


Figure S4. O_3 mixing ratios at 80 hPa computed using MIROC-CTM for October 3, 2009. The mixing ratio ranges from 0 to 2 ppmv.

Table S1. Distances computed from air-parcel trajectory analysis from the OAPA site at the 83 hPa level.

mid time of obs. (yyyy/mm/dd hh:mm)	duration (hh:mm)	distance (km)
2009/09/06 02:07	4:20	633
2009/09/09 04:19	2:31	361
2009/09/13 01:36	5:23	959
2009/09/14 02:18	5:39	809
2009/09/17 04:38	3:25	671
2009/09/18 03:55	3:24	618
2009/09/26 03:58	5:45	643
2009/10/03 04:26	4:05	831
2009/10/05 05:00	4:29	559
2009/10/07 03:40	3:56	455
2009/10/08 02:09	4:58	539
2009/10/12 05:36	2:24	235
2009/10/15 04:02	3:21	226
2009/10/17 01:53	4:06	622
2009/10/19 03:28	2:54	309
2009/10/27 03:48	2:34	250
2009/11/01 02:17	3:06	347
2009/11/05 03:27	2:35	283
2009/11/09 03:25	2:54	419
2009/11/13 06:31	2:58	508
2009/11/14 04:47	4:41	739
2009/11/23 03:47	5:11	779
2009/11/24 04:51	4:12	399