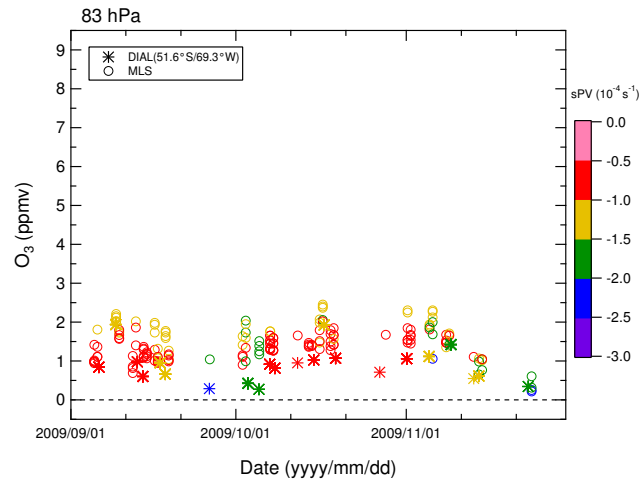


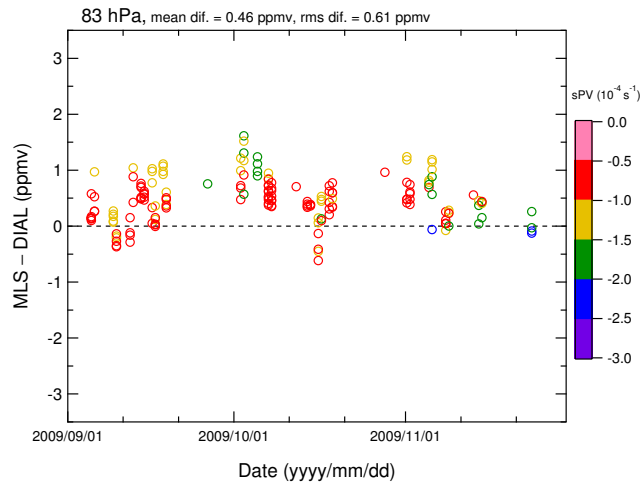
Supplement to the paper

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 $-1.6 \times 10^{-4} \text{ s}^{-1}$. 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 80 hPa. 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., $2.8^\circ \times 2.8^\circ \times 1$ 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.

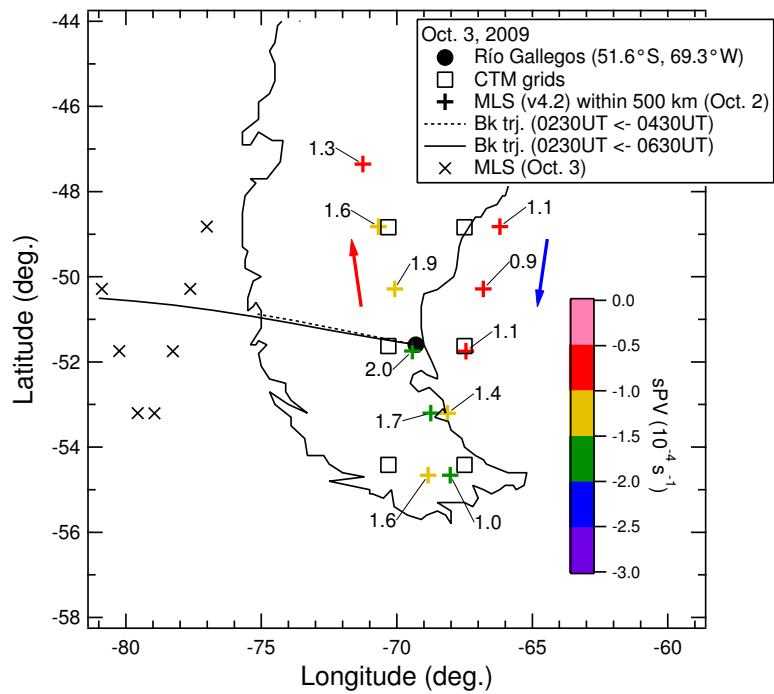


Figure S2. Locations of DIAL and MLS measurements on October 3, 2009. MLS data are color-scaled based on sPV values. Numbers are O_3 mixing ratios in ppmv. The six MIROC-CTM grids are also shown. The air mass trajectories from OAPA are shown as dotted and solid lines. See text for detail.

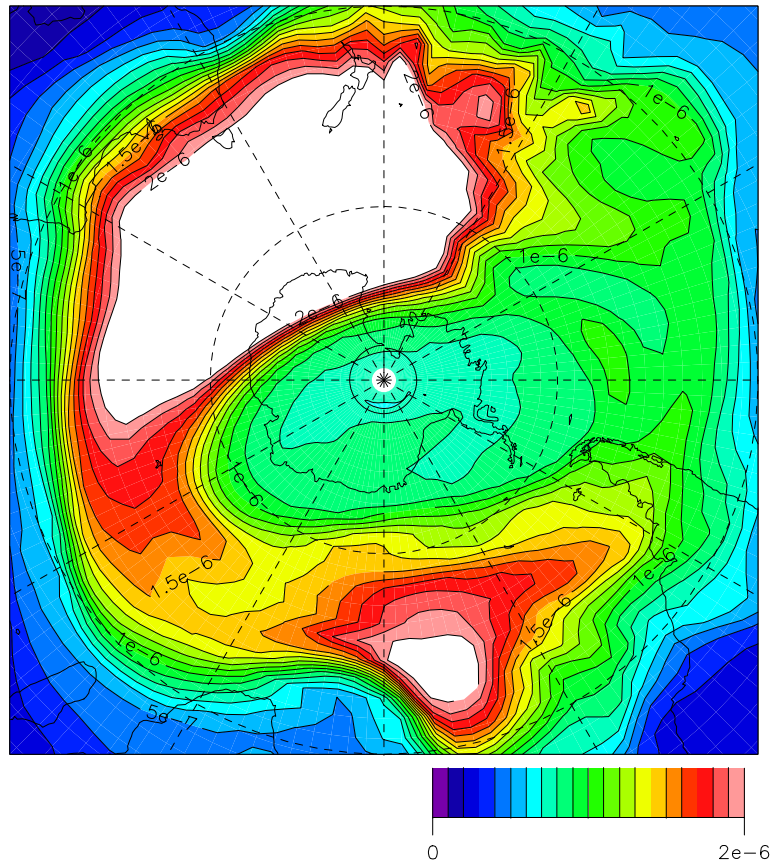


Figure S4. O₃ mixing ratios at 80 hPa computed using MIROC-CTM for October 3, 2009. The figure is oriented so that 180° longitude is at the top. The mixing ratio ranges from 0 to 2 ppmv.