Manuscript Number: amt-2020-194

Manuscript Name: Verification of the AIRS and MLS ozone algorithms based on retrieved daytime and nighttime ozone

We thank the reviewers for their constructive comments and useful suggestions.

Point-by-point response to the comments: For easier reading, our responses are written in **Blue Color** and detail changes in the revised manuscript are written in **Red Color**.

Referee #1

Comment 1: Line 25-26: significant threat of CFCs and other … (remove 2 'the's).

Line 47: Remove the leading 'The'.

Line 48: is the most used method… (add 'method').

Line 49: …occultation method… (add 'method').

Line 60: …day and night or issues with… (add "issues with").

Line 63: I suggest: "daytime values have a low bias due to errors in the retrieval method".

Line 65: Change profile to profiles.

Line 71: upper or lower stratosphere?

Line 72: add "the" to Chapman cycle.

Line 87: Hydrogen and molecular what?

Line 149: efficiency.

Line 220: replace 'that is why O3 low over" with "causing low O3 over".

Line 222: use "loss due to photochemical mechanisms".

Line 271-272. It is necessary … This sentence is confusing. Please rewrite.

Response 1: We agree with these grammatical changes. Besides, we have revised other grammar issues throughout manuscript including text and figures.

Line 27-28. Line 49. Line 50. Line 51. Line 65-67. Line 68. Line 71. Line 78. Line 80. Line 93. Line 154. Line 236. Line 238. Line 318-320.

Comment 2: Line 99: A reference to the recent Frith et al paper on diurnal changes in ozone would be useful.

Response 2: Results reported by Frith et al. (2020) are meaningful. We have added this paper in manuscript.

Line 75-77.

Comment 3: Lines 124-129. Why is version 5 being discussed? In line 200 you discuss the different MLS versions and state that there in not much difference. But there is no discussion of AIRS V5 vs V6 in the paper. Either add a discussion on those differences between V5 & V6 or remove all references to V5. BTW, V7 is coming out sometime soon.

Lines 162-165: see question from Line 124.

Line 248-250. Ah ha! There is the discussion about V5 AIRS. Please move up to Line 124 and add more information on where/when/why the ozone values were different and by how much. Or just drop the discussion all together.

Response 3: We generally introduced AIRS ozone retrievals accuracy in 'Data' section 2.1 (Lines 120-129 in original manuscript) and discussed improvements between versions in 'Result' section 3.1 (Lines 162-170 in original manuscript) and summarized in 'Conclusions' section 4 (Line 248-250 in original manuscript). And do the same for MLS in section 2.2 and 3.2. In order to make it more clear, we removed accuracy introduction of AIRS V5 specifically in 'Data' section 2.1 as you suggested.

Comment 4: Figure 1. Why were the scales changed in plot E? It is better to keep the same scale for all plots.

Response 4: We have changed Figure 1e scales to be the same as the others.

Line 505.

Comment 5: Figure 5 & section 3.3: By far, the most interesting feature in these plots has gone unnoticed (or undiscussed). Why does the difference between MLS and AIRS look so different in figure 5b and so similar in all the others?

Response 5: The differences of the monthly 14-year average daytime AIRS SCO and MLS SCO in 60°S-60°N (Figure 5b) have greater amplitudes than in the polar zones (Figure 5d and 5f). This associates with clouds and the surface type which affect the AIRS ozone retrievals. Seasonal or random changes of clouds and surface emissivity have more significant impact on each monthly AIRS SCO retrieval than on the MLS SCO retrieval. Compared with the 60°S-60°N region, surface types in polar zones are less diverse (snow or ice) and more stable. Therefore, the monthly 14-year average daytime AIRS SCO and MLS SCO show in the polar zones similar patterns. We have added this discussion for Figure 5b in the revised manuscript.

Line 224-232.

Comment 6: Figure 6 is interesting but confusing. How do you define "low ozone" for AIRS and MLS? There are not many points in fig 6d leading this reviewer to wonder if the MLS lines in 6f are meaningful. Could you please explain a bit more what you are trying to point out with these plots?

Response 6: According to the WMO, the Antarctic ozone hole is defined geographically as the area where the total columns of ozone are less than 220 DU (Fahey and Hegglin, 2011). Generally, the ozone hole is well known to appear in Antarctica. However, there also exist well-known low ozone regions outside of Antarctica. We used this phenomenon to illustrate the importance of the small biases in AIRS and MLS.

Figure 6b shows for MLS, the low ozone regions appear in large areas at night besides in tropical western Pacific. However, Figure 6d shows the occurrence frequency and intensity of daytime low ozone regions by MLS SCO retrievals drastically reduces and exists mainly in tropical western Pacific. The yearly and monthly averaged AIRS TCO and MLS SCO of the low ozone regions show no consistency and regularity in Figure 6e and 6f. The analysis of daytime MLS SCO of the low ozone regions is based on only a few observations. The evaluation of day-night differences in both MLS and AIRS has revealed the existence of biases in the satellite data. We cannot distinguish whether it is an algorithm problem or a chemical mechanism that caused this phenomenon. Therefore, our results show that maintaining the quality of the satellite observations of stratospheric ozone is highly relevant.

Line 249-254, 318-320.

Referee #2

Comment 1: Line 44: The BAMS SOTC report also includes annual updates every year beginning 2013 of tropospheric ozone including trends and effects from El Nino.

Response 1: We have added: "*The Bulletin of the American Meteorological Society (BAMS) annually publishes its "State of the Climate", which includes tropospheric* O_3 *trends and effects from El Nino-Southern Oscillation (ENSO)*".

Line: 44-45.

Comment 2: Lines 56-58: Central to your analysis is the assumption that true diurnal variability of atmospheric ozone affects mostly just the BL and the upper stratosphere/mesosphere, neither of which contributes much to either AIRS total column ozone or MLS stratospheric column ozone. This seems to be a valid assumption. In the Introduction you discuss details driving diurnal variability, especially for stratospheric and mesospheric ozone where you give some numbers. For tropospheric ozone diurnal variability, a study by Strode et al. (2019, Atmos.

Env.) using a photochemical transport model indicated that diurnal variability in global tropospheric column ozone appears very small, at most only ~1-2 DU in some regions such as central Africa, India, and east Asia (their Figure 11). Strode et al. provides at least some estimated numbers in DU for global tropospheric ozone diurnal variability that you might include for your paper. These are small diurnal changes in tropospheric column ozone which further reinforce your conclusion that the main issue seems to be an over determination of day-night differences in AIRS total ozone. I.e., as you discuss in Section 3.3, your Figure 5 for 60S-60N shows much larger inferred day-night AIRS minus MLS (TOR) differences of about 5 DU, with most of it coming from AIRS total ozone.

Response 2: We have added: "*Strode et al. (2019) simulated the global diurnal cycle in the tropospheric O_3 columns, their results indicated that the mean peak-to-peak magnitude of the diurnal variability in tropospheric O_3 is approximately 1 DU*". Results reported by Strode et al. (2019) supported our results in Figures S3 to S6 in the supplement that AIRS TCO retrieval artefacts dominate the day/night variability of tropospheric O_3 residuals (TOR = AIRS TCO – MLS SCO).

Line: 305-307.

Comment 3: Line 215: More specifically for MLS it is mostly for the SH Antarctic region and it seems to be very large. The abrupt change of about 30 DU in 2015 for MLS in Figure 5e for the Antarctic region suggests that something changed significantly with the MLS v4.2 retrieval (and for the better). In addition, there are huge day-night differences for MLS in Figure 5f that are greatest in September-October during the Antarctic ozone hole with numbers of 60-70 DU. It is also noteworthy that AIRS day versus night total ozone differences appear smaller at ~2-3 DU in both polar regions compared to ~5 DU for 60S-60N. (I may not have inferred these numbers very precisely since vertical scales are all different for the three regions.) In your paper you mention AIRS day-night differences associated with ocean scenes via cloud patterns and also mostly over dry land areas likely related to surface emissivity issues. This seems to be consistent with AIRS in Figure 5. Your Figure 5 is very interesting and you might discuss more about the features.

Response 3: We have added more discussion about Figure 5 as follows "*AIRS SCO retrievals show smaller day-night differences in the polar zones (1-2 DU) than in 60 °S-60 °N (4-5 DU). This is related to clouds and the surface type which affect the AIRS O_3 retrievals as mentioned above. Figure 5b shows monthly 14-year average daytime AIRS SCO and MLS SCO in 60 °S-60 °N for 2005-2018. Seasonal or random changes of clouds and the surface emissivity issues have more significant impact on each monthly AIRS SCO retrieval than on the MLS SCO retrieval. Compared with 60 °S-60 °N region, surface types in the polar zones are less diverse (snow or ice). Therefore, the monthly 14-year average daytime AIRS SCO and MLS SCO in Figure 5d and 5f show similar patterns. Figures 5c* *to 5f also confirm that MLS SCO has a polar bias when compared with AIRS SCO at high latitudes. In addition, for MLS SCO in Figure 5f, the biggest day-night differences (50-60 DU) occur in September and October during the Antarctic O_3 hole.*"

Line: 224-232.

Comment 4: Line 140: "…accuracy was estimated at ~40 or ppbv +5% (~20 140 ppbv or +20% at 215 hPa)." Please clarify sentence.

Response 4: We rephrased this sentence as follows "*Livesey et al. (2008) estimated the MLS O_3 accuracy as ~40 ppbv ±5% (~20 ppbv ±20% at 215 hPa)*".

Line: 144-145.

Comment 5: Line 255: "… (< 1 DU for the upper atmospheric SCO), expect in the upper stratosphere and mesosphere." Please clarify sentence. There are other typos and wording/sentence issues throughout the paper that you will find upon re-reading the current manuscript.

Response 5: We rephrased this sentence as follows "*MLS day-night differences in SCO and O_3 profiles show that day-night differences are only small (< 1 DU) and likely to be in the upper stratosphere and mesosphere*". Besides, we have revised other grammar issues throughout manuscript including text and figures.

Line: 313-315.

Referee #3

A. Conceptual issues

Comment 1: The authors compare total column ozone from infrared nadir measurements (AIRS) with stratospheric column ozone from microwave limb measurements (MLS) without sufficient acknowledgement of the effects instrument differences will have on their results. Top of atmosphere infrared radiances (AIRS) are sensitive to stratospheric and (to a lesser extent) tropospheric ozone (Nalli et al., 2018 and references therein). AIRS radiances have almost no sensitivity to ozone in the lower troposphere and boundary layer. Moreover, infrared measurements have strong sensitivity to clouds, which dominate the signal in channels sensitive to tropospheric variability. Microwave limb measurements (MLS), on the other hand, are sensitive to stratospheric ozone down to ~200hPa, with almost no sensitivity to clouds in the upper troposphere (< 200hPa cloud top pressure). The authors compare AIRS total column ozone (troposphere + stratosphere) to MLS stratospheric column ozone (stratosphere only) and find that the former has higher diurnal variability. My sense, as reviewer, is that their results have limited value because they included tropospheric, and thus diurnal, variability into their AIRS values from the start. A scientifically more meaningful comparison would have been a comparison between stratospheric columns from both AIRS and MLS. One can easily calculate partial column totals from the AIRS Level 2 products, which are distributed as 100-layer profiles (Earth surface to top of atmosphere) for every retrieval scene.

Response 1: We compared yearly and monthly averaged stratospheric ozone columns (SCO) between AIRS (250 hPa – 1 hPa) and MLS (261 hPa – 0.02 hPa) for 2005-2018 in the revised manuscript as you suggested (see Figure 5). The day-night difference of MLS SCO is 0.79 DU in the mesosphere (10 hPa - 0.1 hPa) and 0.03 DU in the stratosphere (100 hPa - 10 hPa). The day-night difference of AIRS SCO is 1.51 DU in the mesosphere (10 hPa - 1 hPa) and 3.85 DU in the stratosphere (100 hPa - 10 hPa). Compared to the AIRS SCO day-night differences, the magnitude of MLS SCO day-night differences in the stratosphere and in the mesosphere are much smaller. It has been pointed out that errors in temperature profiles and water vapour mixing ratios will adversely affect the AIRS ozone retrievals. Significant biases (0 - 100%) may exist in the region between ~300 hPa and ~80 hPa (Wang et al., 2019; Olsen et al., 2017). AIRS ozone retrievals are insensitive to profile changes. Because all AIRS ozone channels are sensitive to both the surface as well atmospheric ozone and thus are insensitive to the entire ozone profile, the total ozone retrieval is compromised if the surface is not well characterized (Olsen et al., 2017).

There are a number of separate processes that may cause day/night differences in either AIRS or MLS. The first one is the diurnal ozone cycle chemistry – either tropospheric or stratospheric. AIRS total column ozone can be affected by both, MLS SCO down to 200 hPa only by stratospheric chemistry. The day-night MLS differences show that – after accounting for an MLS bug affecting day-night orbits – day/night MLS differences are confined to the mesosphere (1 hPa and higher), whose contribution to MLS SCO is negligible.

The strongest diurnal ozone effects occur over land in the boundary layer (nighttime surface deposition and daytime photochemical production in the presence of air pollution). In the marine boundary layer, the diurnal cycle is much weaker due to absence of air pollution and a general slow ozone destruction regime (~10%/day). Similarly, in the free troposphere, the diurnal ozone cycle is also weak due low production rates (generally low levels of ozone relevant pollution), and the diurnal ozone cycle in the free troposphere is even negligible above 750 hPa (Petetin et al., 2016). Overall, any tropospheric photochemical diurnal ozone cycle effect should resemble some correspondence with air pollution. The day-night differences in AIRS total column ozone clearly do not resemble patterns of surface air pollution.

There exists a range of processes that can cause day/night differences in AIRS and MLS ozone retrievals: clouds, emissivity, and averaging kernels. As discussed in the paper, we identify day/night AIRS total column ozone differences over oceans that resemble cloud patterns. The strongest diurnal cycles in cloud fraction are found in the tropics over land, following strong daytime heating (Noel et al., 2018). Over oceans, diurnal cycles in cloud fraction are weaker, but very broadly indicate reduced cloudiness during day compared to night, especially in the tropics and subtropics (Noel et al., 2018). In case of clouds, AIRS total column ozone appears to be larger during daytime compared to nighttime. This is consistent with the notion of increased cloudiness during the night, increasing the chance of shielding by undetected or unrecognized clouds in the AIRS retrieval. Over land, patterns in day/night differences appear to be dominated by the dryness of the surface, suggesting that emissivity may not be well represented or that reduced sensitivity to the lower troposphere during night compared to day over hot surfaces results in difference AIRS total column ozone. The spatial inhomogeneity of day-night AIRS total column ozone differences over drier regions points to the emissivity rather than the averaging kernels dominating these differences. Infrared satellite retrieval artefacts due to land surface emissivity is a well-known phenomenon (George et al., 2009; Zhou et al., 2013; Bauduin et al., 2017).

We modified the discussion section in the revised manuscript to include what is discussed above.

Line:216-224, 266-312.

Comment 2: The authors attempt to draw a distinction between ascending/descending (MLS) versus day/night (AIRS) but this remains confusing throughout the paper. I recommend that the authors limit AIRS and MLS values to the exact same latitudinal zones and pressure zones to legitimize their comparisons and clarify their results.

Response 2: We mention in section 2 that we change from ascending/descending to day/night. However, for discussion of the bias due to the AscDescMode bug (see lines 178-182 + section 3.2.2 in original manuscript) we have to return to the use of ascending/descending. We clearly explain that we use day/night only between 60S-60N, preferring to use ascending/descending for the polar regions, but apparently this still leads to some confusion. Therefore, we modified text (lines 178-182) and Figure 2, 3, 4 by using the brackets "*ascending ("daytime") and descending (''nighttime")*".

Line: 184, 510-515.

Comment 3: Infrared and microwave instruments have different observing capabilities for the same atmospheric variables. When comparing products from different instrument types, one has to account for inherent instrument limitations. E.g. ozone retrievals from AIRS will never have value in urban-scale air quality applications, because the AIRS infrared measurements lack sensitivity to boundary layer ozone. There is no retrieval algorithm that can extract boundary layer ozone from AIRS measurements because the signal is simply not there. Another example is that MLS ozone observations will have very limited cloud contamination (if any) because, by definition, microwave radiance measurements lack sensitivity to non-precipitating clouds. One has to acknowledge basic instrument capability when comparing products.

Response 3: The primary question that we address in this paper is what day-night differences in the AIRS TCO and the MLS SCO look like, as well as in MLS upper atmospheric ozone profiles, and trying to understand these differences. The MLS and AIRS measurements that allow for investigating these day-night differences have existed for quite some time, but differences in day-night observations of atmospheric ozone has remained a largely if not completely unexplored research area.

Our analysis confirms that spatio-temporal variations in day-night differences exist. We find evidence that they are likely related to instrumental capabilities and limitations therein, algorithm shortcomings, and data file artefacts (clouds, land surface infrared emissivity, and inconsistencies in supplementary information in data files). We added this to discussion section.

However, it is well beyond the scope of paper to discuss for which atmospheric processes the improvements in the satellite data might be beneficial.

Comment 4: The authors posit that one of the possible reasons for diurnal variability in AIRS total column ozone is due to a mis-characterization of surface emissivity. While this may be true for boundary layer temperature or water vapor, it should have minimal effect on ozone retrievals because AIRS radiance channels lack sensitivity to lower tropospheric ozone. By far a stronger effect on the retrieval product is the a-priori. AIRS V6 is an optimal estimation retrieval system that uses a non-linear regression as a-priori for temperature, water vapor and ozone (Milstein and Blackwell, 2016; Smith and Barnet, 2019, 2020; Susskind et al., 2014). This regression algorithm uses all available AIRS channels to retrieve a host of atmospheric variables simultaneously, thus propagating their spectral correlation into the retrieved products. In optimal estimation retrieval systems, the a-priori functions as a stabilization factor, such that wherever the radiance channels lack sensitivity, the a-priori will fill the result will default to the a-priori. My sense is that the diurnal variability observed in AIRS V6 total column ozone probably originates from the regression a-priori. The authors can test this because the a-priori (or first guess) values are distributed with the retrievals in the Level 2 file. The authors can also test their hypothesis that clouds affect total column ozone values by correlating AIRS ozone with cloud fraction and cloud top pressure, both retrieved from AIRS radiances and available in the Level 2 file (AIRS Science Team/Joao Texeira, 2013).

Response 4: As we mentioned in **Response 1**, because all AIRS ozone channels sense the surface as well atmospheric ozone and thus are insensitive to the shape of the entire ozone profile, the total ozone is affected if the surface is not well characterized (Olsen et al., 2017).

The ozone first guess is an observationally-based climatology, which is month-by-month on 10 °latitude bins from 80S to 80N. The ozone profile shape is mainly determined by a priori profile.

Figure S1(a) in the supplement shows the AIRS O_3 retrieval over the Sahara Desert [20 °N,24 °E,23 °N,27 °E] region contains a larger fraction of the O_3 priori than a forest region [22 °N,106 °E,24 °N,108 °E] at the same latitude. It means the AIRS O_3 retrievals over desert are highly determined by the O_3 priori and thus have little information content. The weak radiance information over deserts may relate to surface emissivity. For most desert areas, emissivities are less than 0.85 due to the strong quartz absorption feature between 8-9.6 μm range (9.6 μm band is used to retrieve AIRS TCO and O_3 profiles during both day and night), whereas the emissivity of forest, water and ice cover are generally greater than 0.95 and spectrally flat in the 3-12 μm spectral range (Olsen et al., 2017). Figure S1(b) indicates day-night differences of radiance information over deserts are also larger, which is consistent with large differences of AIRS TCO retrievals over deserts.

We also test our hypothesis that clouds affect total column ozone values by correlating AIRS ozone with cloud fraction and cloud top pressure. For ocean regions with persistent clouds during day and night (for example over ITCZ), Figure S2 in the supplement shows that the variety of cloud layer height has a greater impact on AIRS TCO day-night differences than cloud fraction.

Line: 271-278, 290-292.

B. Technical issues

Comment 5: Lines 9-12 and Lines 56-59: "Based on knowledge of the chemistry and transport of O3 …" The premise of the work is unclear to me. I recommend that the authors rephrase their argument for evaluating diurnal changes in O3, to clarify the scientific meaning of their results.

Response 5: This slight variation in diurnal total column ozone can serve as a natural test signal for remote sensing instruments and data retrieval techniques. We show how sensitive different space-based instruments are to the diurnal cycle of total column ozone. Any remaining difference in day and night ozone is used to distinguish potential biases from retrieval artefacts. Applying this day-night verification on the AIRS and the MLS data can access their capacities to characterize atmospheric O_3. Further, an accurate assessment of O_3 variation is needed for a reliable and homogeneous long-term trend detection in the global O_3 distribution.

Line: 12-15, 62-64, 72-74.

Comment 6: Line 59: The references listed here for ozone retrievals from infrared radiances, predate the launch of AIRS. Since this paper is about AIRS ozone retrievals, I recommend that the authors reference more recent papers.

Response 6: Line 59 "*Day-night inter-comparisons present a unique opportunity to assess the internal consistency of infrared O_3 instruments*" cited papers which used the method 'Day-night inter-comparisons'. However, few studies focused on AIRS night ozone retrievals recently. Alternatively, we added Pommier et al. (2012) analysed day/night differences of Infrared Atmospheric Sounding Interferometer (IASI) tropospheric ozone over the Arctic.
Line: 65.

Comment 7: Line 60: "calibration procedures between day and night …" This sentence implies that radiometric calibration varies diurnally for all instruments. This is not true, of course. Can the authors be more specific here?
Response 7: We rephrased this sentence as follows "*Systematic differences could potentially arise, for example, from temperature effects within the instrument, from differences in signal magnitude procedures between daytime and nighttime or from the retrieval algorithms*".
Line: 65-67.

Comment 8: Lines 64-65: "There are infrared satellite instruments, like AIRS and MLS …" MLS is not an infrared instrument.
Response 8: We rephrased this sentence as follows "*There are satellite instruments, like Atmospheric InfraRed Sounder (AIRS) and The Microwave Limb Sounder (MLS), that provide global daytime and nighttime TCO/SCO and O_3 profiles*".
Line: 69.

Comment 9: Line 69: "near the polar day terminator in the upper troposphere" Can the authors explain what they mean here?
Response 9: We rephrased this sentence as follows "*in the upper stratosphere during the polar day near 70 °N*".
Line: 76.

Comment 10: Lines 75-79: Personally, I think this level of detail about the chemical reactions of O_3 (and its precursors) is irrelevant to the discussion here.
Response 10: We explained the chemical reactions of O_3 in detail in order to emphasize significant deviations between daytime and nighttime O_3 are only expected either in the planetary boundary layer (PBL) and high in the stratosphere or mesosphere, having little effect on the total column of ozone.

Comment 11: Line 110: I would suggest that the authors write out "TCO" to make this title less cryptic.

Response 11: We rephrased the title and subtitle with complete spelling.

Line: 117, 132, 151.

Comment 12: Line 114: Can the authors provide a reference and perhaps doi number for the AIRS V6 level 3 data products?

Response 12: We added AIRS V6 L3 data web link: https://disc.gsfc.nasa.gov/datasets/AIRS3STD_006/summary, access date: August 27, 2020. AIRS Science Team/Joao Teixeira (2013), AIRS/Aqua L3 Daily Standard Physical Retrieval (AIRS-only) 1 degree x 1 degree V006, Greenbelt, MD, USA, Goddard Earth Sciences Data and Information Services Center (GES DISC), Accessed: August 27, 2020, doi:10.5067/Aqua/AIRS/DATA303.

Line: 124.

Comment 13: Line 140: What is considered a "small positive bias" in lower stratospheric MLS O3 data?

Response 13: According to Froidevaux et al. (2008), the average differences between MLS and other satellite ozone retrievals in the lower stratosphere often exhibit oscillations of a few percent in amplitude (e.g., with a positive notch at 22 hPa); while the MLS retrievals appear to generally be the source of such oscillations, the impact on most scientific investigations should be minimal.

Comment 14: Line 141: "Comparisons with expectations and other observations …" What do the authors mean here?

Response 14: We rephrased this to "*Expectations and comparisons with other observations…*".

Line: 145-146.

Comment 15: Lines 145-147: "… and the decline over land is larger than over oceans indicating differences in surface loss." Can the authors clarify this statement?

Response 15: We rephrased this sentence as "*The reduction of AIRS TCO over land at night is greater than over oceans differences depending on surface type*".

Line: 151-152.

Comment 16: Line 153: Can the authors give an example of what they mean by "atypical earth surface properties"?

Response 16: We have removed this sentence, since it is already better phrased in the previous sentence.

Comment 17: The titles for Section 3.2 and 3.2.1 are cryptic and almost exactly the same. I recommend the revise these to distinguish the two sections.

Response 17: We modified titles as follows "*3.2 MLS O_3 retrievals day-night differences*" and *"3.2.1 MLS O_3 profile*".

Line: 176, 177.

Comment 18: Line 184: "When this flag has a value of plus one or minus…" Rephrase.

Response 18: We rephrased this sentence as follows "*We counted the daily number of pixels at both poles when observation mode is ascending (AscDescMode = 1) and descending (AscDescMode = -1) respectively*".

Line: 189-190.

Comment 19: Lines 186, 187, 189: "14 may" should be "14 May".

Response 19: We modified this as you suggested.

Line: 192, 193, 194.

Comment 20: Line 198: "scientifically reliable values" Could the authors elaborate on what they mean here?

Response 20: Livesey et al. (2015) reported a high MLS v2.2 bias at 215 hPa had been observed in some comparisons versus ozonesonde and satellite datasets. Such high biases were reduced in versions v3.3x and v3.4x, with additional smaller reductions in the ozone values in MLS v4.2x (ozone accuracy was estimated at ~20 ppbv +10% at 261 hPa).

Line: 202-203.

Comment 21: Line 264: "Timescale becomes low enough". What do the authors consider a "low" timescale?

Response 21: As shown in Smith et al. (2015) the lifetime of O_3 due to chemistry is strongly altitude dependent (<20 min in the upper mesosphere above 0.01 hPa). Only in the mesosphere the loss timescale for O_3 becomes long enough to see significant differences between average daytime and nighttime concentrations.

Line: 302-304.

Comment 22: Line 265: "Figures S1 to S4" should be "Figures 1 to 4".

Response 22: "Figures S1 to S4" refers to the figures in the Supplement.

Comment 23: Line 266: “O3” should be a subscript “3”.

Response 23: We modified this mistake.

Line: 308.

Comment 24: Lines 266-267: “small day-night differences of tropospheric O3 are hard to discriminate comparing day/night TCO.” This sentence needs revision.

Response 24: We deleted this sentence after consideration.

Comment 25: Line 268-269: “we found that the frequency and intensity of low O3 regions between 60S and 60N was higher at night by AIRS and MLS” Line 270-273: “whether the more serious low region at night are due to the problem of the algorithm itself or the atmospheric physical and chemical factors different from that in the daytime, we compared both MLS and AIRS at day and at night. It is necessary to verify day-night differences by infrared TCO observations for retrieval aspect first. Our results show that maintaining the quality of the satellite observations of stratospheric O3 is therefore highly relevant.” What do the authors mean here?

Response 25: We rephrased this paragraph as follows “*A case study of day-night differences O_3 over equatorial Pacific revealed that both AIRS and MLS O_3 retrievals have biases in comparison to expected variations and changes. Our results show that maintaining the quality of the satellite observations of stratospheric O_3 is therefore highly relevant.*”

Line: 318-320.

Verification of the AIRS and MLS ozone algorithms based on retrieved daytime and nighttime ozone

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Abstract. Ozone (O_3) plays a significant role in weather and climate on regional to global spatial scales. Most studies on the variability in the total column of O_3 (TCO) are typically analysed using daytime data. Based on knowledge of the chemistry and transport of O_3, significant deviations between daytime and nighttime O_3 are only expected either in the planetary boundary layer (PBL) or high in the stratosphere or mesosphere, having little effect on the TCO. Hence, we expect the daytime and nighttime TCO to be very similar. However, a detailed evaluation of satellite measurements of daytime and nighttime TCO is still lacking, despite the existence of long records of both. Comparing daytime and nighttime TCOs thus provides a novel approach to verify the retrieval algorithms of for example the Atmospheric InfraRed Sounder (AIRS) and the Microwave Limb Sounder (MLS). In addition, such a comparison also helps in assessing the value of nighttime TCO for scientific research. Applying this verification on the AIRS and the MLS data we identified inconsistencies in observations of O_3 from both satellite instruments. For AIRS, daytime-nighttime differences were found over oceans resembling cloud cover patterns, and over land, mostly over dry land areas, likely related to infrared surface emissivity. These differences point to issues with the representation of both processes in the AIRS retrieval algorithm. For MLS, a major issue was identified with the "ascending-descending" orbit flag, used to discriminate nighttime and daytime MLS measurements. Disregarding this issue, MLS day-night differences were significantly smaller than AIRS day-night differences, providing additional support for retrieval method origin of AIRS day-night TCO differences. MLS day-night differences are dominated by the upper stratospheric and mesospheric diurnal O_3 cycle. These results provide useful information for improving infrared O_3 products and at the same time will allow study the day-night differences of stratospheric and mesospheric O_3.

1 Introduction

Atmospheric ozone (O_3) is a key factor in the structure and dynamics of the Earth's atmosphere (London 1980). The 1987 Montreal Protocol on Substances that Deplete the O_3 Layer formally recognized the significant threat of chlorofluorocarbons and other O_3-depleting substances (ODCs) to the O_3 layer and marks the start of joint international efforts to reduce and ultimately phase-out the global production and consumption of ODCs (Velders et al., 2007). Indeed, concerns about changes in O_3 due to catalytic chemistry involving man-made chlorofluorocarbons has become as an important topic for the scientific community, the general public and governments (Fioletov et al., 2002).

In response to this concern and associated environmental policies, during the last two decades a large number of studies have focused on estimating long-term variations and trends in stratospheric column of O_3 (SCO). A summary of the state of the science is frequently reported in the quadrennial O_3 Assessment Reports issued by the United Nations Environmental Program (UNEP) and the World Meteorological Organization (WMO). These reports are written in response to the global treaties aiming at minimizing emissions of ODSs. The signatories of these treaties ask for regular updates on the state of the science and knowledge. The most recent O_3 Assessment reports extensively discuss long-term variations and trends in stratospheric O_3 in relation to expected recovery (WMO, 2011, 2014, 2018). According to WMO (2018), Antarctic stratospheric O_3 has started to recover, while outside of the polar regions, upper stratospheric O_3 has also increased. On the other hand, no significant trend has been detected in global (60 °S-60 °N) total column O_3 over the 1997-2016 period with average values for the years since the last Assessment remaining roughly 2% below the 1964-1980 average. Moreover, recently a debate has emerged over the question as to whether lower stratospheric O_3 between 60 °S-60 °N has continued to decline despite decreasing O_3 depleting substances (Ball et al., 2018; Ball et al., 2019). In addition to the quadrennial O_3 Assessments, the Bulletin of the American Meteorological Society (BAMS) annually publishes its "State of the Climate", which since 2015 includes tropospheric O_3 trends and effects from El Nino-Southern Oscillation (ENSO) and a description of the relevant stratospheric events of the past year, the state of the Antarctic O_3 hole, as well as an annual update of global and zonal trends in stratospheric O_3. These regularly recurring reports and publications illustrate the continued attention and monitoring of the O_3 layer and its recovery, in which the long records of satellite observations play a crucial role. Establishing and maintaining the quality of the satellite observations of stratospheric O_3 is therefore highly relevant.

A variety of techniques exist to measure the O_3 column and stratospheric O_3. UV absorption spectroscopy with the sun or stars as sources of UV light is the most used method to derive O_3 (Weeks et al., 1978; Fussen et al., 2000). In addition to the UV occultation method, the absorption of infrared radiation has also been used to detect O_3 profiles throughout the column (Gunson et al., 1990; Brühl et al., 1996). Another technique is the detection of the molecular oxygen dayglow emissions (Mlynczak and Drayson, 1990; Marsh et al., 2002). Some ground-based instruments use O_3 emissions in the microwave region to infer the O_3 density in the mesosphere (Zommerfelds et al., 1989; Connor et al., 1994). Infrared emission measurements overcome the limitations in the local time coverage of solar occultation and dayglow technique and their altitude resolution is significantly higher compared with microwave measurements (Kaufmann et al., 2003). The strongest O_3 infrared absorption centres near 9.6 um.

Based on knowledge of chemistry and transport of O_3, significant deviations between daytime and nighttime O_3 are only expected either in the planetary boundary layer (PBL) and high in the stratosphere or mesosphere, having little effect on the total column of O_3 (TCO). Hence, we expect that the daytime and nighttime TCO to be very similar. This slight variation in diurnal TCO can serve as a natural test signal for remote sensing instruments and data retrieval techniques. We need to clarify how sensitive different space-based instruments are to TCO slight changes and to distinguish potential biases from retrieval artefacts. Day-night inter-comparisons present a unique opportunity to assess the internal consistency of infrared O_3 instruments (Brühl et al., 1996; Pommier et al., 2012; Parrish et al., 2014). Systematic differences could potentially arise, for example, from temperature effects within the instrument, from differences in signal magnitude between daytime and nighttime or from the retrieval algorithms. The Stratosphere Aerosol and Gas Experiment (SAGE) applied day-night differences to validate O_3 profiles and found daytime values have a low bias due to errors in the retrieval method since the magnitude of the difference was much less in a photochemical model (Cunnold et al., 1989). There are satellite instruments, like Atmospheric InfraRed Sounder (AIRS) and The Microwave Limb Sounder (MLS), that provide global daytime and nighttime TCO/SCO and O_3 profiles. Although their daytime O_3 retrievals have been validated (Livesey et al., 2008; Sitnov and Mokhov, 2016), day-night differences in TCO and SCO are still largely unexplored. Applying this day-night verification on the AIRS and the MLS data can assess their capacities to characterize atmospheric O_3. Further, an accurate assessment of O_3 variation is needed for a reliable and homogeneous long-term trend detection in the global O_3 distribution.

The O_3 diurnal cycle depends on latitude, altitude, weather and time. The variations of the diurnal cycle are less than 5% in the tropics and subtropics and increase to more than 15% in the upper stratosphere during the polar day near 70 °N (Frith et al., 2020). There exist diurnal variations in atmospheric O_3 at certain altitudes. There are two distinct O_3 maxima in the typical vertical profile of the O_3 volume mixing ratio, one in the lower stratosphere and one in the mesosphere. The secondary maximum in the mesosphere is present during both day and night (Evans and Llewellyn, 1972; Hays and Roble, 1973). Chapman (1930) revealed the photochemical scheme in the mesosphere. The reactions of the Chapman cycle are important for us to understand diurnal O_3 variation.

$$O_2 + hv \rightarrow 2O(\lambda < 240nm) \, , \qquad (1)$$

$$O + O_2 + M \rightarrow O_3 + M, \text{ (in which M stands for an air molecule)} \qquad (2)$$

$$O_3 + O \rightarrow 2O_2 \, , \qquad (3)$$

$$O_3 + hv \rightarrow O_2 + O(\lambda < 1140nm) \, , \qquad (4)$$

In the daytime mesosphere, catalytic O_3 depletion by odd hydrogen has to be considered in addition to the Chapman cycle. The anti-correlation of O_3 and temperature is mainly due to the temperature dependence of the chemical rate coefficients (Craig and Ohring, 1958; Barnett et al., 1975). Huang et al. (2008) and Huang et al. (1997) found midnight O_3 increases in the mesosphere, based on SABER and MLS data, respectively. Zommerfelds et al. (1989) surmised that eddy transport may explain this increase, while Connor et al. (1994) stated that atmospheric tides are expected to cause systematic day-night variations.

During daytime, photolysis is the major loss process. The main nighttime O_3 source in the mesosphere is atomic oxygen, while its sinks are atomic hydrogen and atomic oxygen (Smith and Marsh, 2005). In addition to O_3 chemical reactions with active hydrogen and molecular, the turbulent mass transport also plays an important role in the explanation of the secondary O_3 maximum (Sakazaki et al., 2013; Schanz et al., 2014).

Tropospheric O_3 is mainly produced during chemical reactions when mixtures of organic precursors (CH_4 and non-methane volatile organic carbon, NMVOC), CO, and nitrogen oxides (or NO_x), are exposed to the UV radiation in the troposphere (Simpson et al., 2014). At night, in the absence of the sunlight, there is no O_3 production, but surface O_3 deposition and dark reactions transform the NO_x-VOC mixture and remove O_3. The dark chemistry affects O_3 and its key ingredients mainly depend on the reactions of two nocturnal nitrogen oxides, NO_3 (the nitrate radical) and N_2O_5 (dinitrogen pentoxide). NO_3 oxidizes VOC at night, while reaction of N_2O_5 with aerosol particles containing water removes NOx. Both processes remove O_3 as well at night (Brown et al., 2006).

The diurnal cycle of O_3 in the middle stratosphere had generally been considered small enough to be inconsequential, with known larger variations in the upper stratosphere and mesosphere (Prather, 1981; Pallister and Tuck, 1983). Later studies have highlighted observed and modelled peak-to-peak variations of the order of 5% or more in the middle stratosphere between 30 and 1 hPa (Sakazaki et al., 2013; Parrish et al., 2014; Schanz et al., 2014).

In terms of dynamics, vertical transport due to atmospheric tides is expected to contribute to diurnal O_3 variations at altitudes where background O_3 levels have a sharp vertical gradient (Sakazaki et al., 2013). The Brewer/Dobson circulation transports air upwards in the tropics, polewards and downwards at high latitudes, with stronger transport towards the winter pole (Chipperfield et al., 2017).

The main objective of this paper is to analyse day-night differences in the AIRS TCO and the MLS SCO, as well as in MLS upper atmospheric O_3 profiles. Section 2 discusses the data used. Section 3 presents results for AIRS, MLS, the comparison of AIRS with MLS, and an application of AIRS TCO data over the Pacific low O_3 regions to highlight how day-night differences affect use and interpretation of TCO data. Finally, section 4 ends the paper with a brief summary and conclusions.

2 Data

2.1 AIRS total column of O_3 retrievals

The AIRS satellite instrument was the first in a new generation of high spectral resolution infrared sounder instruments flown aboard the National Aeronautics and Space Administration (NASA) Earth Observing System (EOS) Aqua satellite (Divakarla et al., 2008). The AIRS radiance data at 9.6 μm band are used to retrieve column O_3 and O_3 profiles during both day and night (including the polar night) (Pittman et al., 2009; Fu et al., 2018). The AIRS V6 level 3 standard TCO products (2003-2018) comprising daily averaged measurements on the ascending and descending branches of an orbit with the quality indicators 'best' and 'good' and binned into 1°×1° (latitude × longitude) grid cells are used here (https://disc.gsfc.nasa.gov/datasets/AIRS3STD_006/summary access date: August 27, 2020). Outside of the polar zones (60°N-90°N and 90°S-60°S), ascending and descending correspond respectively to daytime (13:30 in local solar time) and nighttime (01:30 in local solar time). Hereafter we refer to “day” and “night” rather than ascending and descending over 60°S-60°N. In the polar zones, it is inappropriate to use ascending/descending mode to define daytime/nighttime, therefore, we just compare differences between ascending and descending mode. AIRS TCO measurements agree well with the global Brewer/Dobson Network station measurements with a bias of less than 4% and a root mean squared error (RMSE) difference of approximately 8% (Divakarla et al., 2008). Analysis of AIRS TCO monthly maps revealed that its retrievals depict seasonal trends and patterns in concurrence with OMI and SBUV/2 observations (Divakarla et al., 2008).

2.2 MLS stratospheric column of O_3 and O_3 profile retrievals

The MLS instrument on-board Aura satellite, which was launched on 15 July 2004 and placed into a near-polar Earth orbit at 705 km with an inclination of 98 °, uses the microwave limb sounding technique to measure vertical profiles of chemical constituents and dynamical tracers between the upper troposphere and the lower mesosphere (Waters et al., 2006). Its orbital ascending mode is at 13:42 (local solar time) and the orbital descending mode at 01:42 (local solar time) over 60 °S-60 °N. In this study, we use the MLS v4.2x standard O_3 product during 2005-2018 (https://mls.jpl.nasa.gov/products/o3_product.php access date: August 27, 2020). Its retrieval is using 240-GHz radiance, providing near-global spatial coverage (82 °S to 82 °N latitude), with each profile spaced 1.5 degrees or ~165 km along the orbit track. This O_3 product includes the O_3 profile on 55 pressure surfaces and the recommended useful vertical range is from 261 to 0.02 hPa. In addition, it contains an O_3 column, which is the integrated stratospheric column down to the thermal tropopause calculated from MLS measured temperature (Livesey et al., 2015). Jiang et al. (2007) found the MLS stratospheric O_3 data between 120 and 3 hPa agreed well with ozonesonde measurements, within 8% for the global daily average. Froidevaux et al. (2008) reported MLS stratospheric O_3 uncertainties of the order of 5%, with values closer to 10% (and occasionally 20%) at the lowest stratospheric altitudes. Livesey et al. (2008) estimated the MLS O_3 accuracy as ~40 ppbv ± 5% (~20 ppbv ± 20% at 215 hPa). Expectations and comparisons with other observations show good agreements for the MLS O_3 product, generally consistent with the systematic errors quoted above.

3 Results

3.1 AIRS O_3 retrievals day-night differences

Figure 1 shows spatial variations in the differences between the AIRS day and night measurements. Generally, 90% of the world's AIRS TCO is smaller during nighttime compared to daytime. The reduction of AIRS TCO over land at night is greater than over oceans differences depending on surface type. Seasonal averaged O_3 day-to-night relative difference shown in Figures 1a to 1d reveal that AIRS TCO day and night difference variations in Asia, Europe and North America during winter in the Northern Hemisphere (DJF) are smaller than during summer-time (JJA), in line with the efficiency of photochemical production between seasons in the Northern Hemisphere. The Sahara Desert shows maximum difference value during winter-time when there are large day-night temperature differences. The same phenomenon is observed in Western Australia during summer-time.

In Figure 1e shows for the annual mean large differences of AIRS TCO retrievals over deserts, the Intertropical Convergence Zone (ITCZ) with persistent clouds and Arctic regions. These spatial patterns over land mimic regions with low IR surface emissivity and/or regions where IR surface emissivity exhibits large seasonal variations (Feltz et al., 2018). Figure 1f shows significant TCO changes at the land-ocean interface. All these effects are important parameters for the retrieval algorithm but bear no physical relation with total O_3. Hence, the differences shown in Figure 1 provide strong indications that the largest AIRS day-night TCO differences are dominated by retrieval artefacts. As such changes are unphysical, it confirms the hypothesis that clouds and the surface type (land/desert/vegetation/snow or ice) affects the AIRS TCO retrievals.

The AIRS emissivity retrieval uses the NOAA regression emissivity product as a first guess over land. The NOAA approach is based on clear radiances simulated from the European Centre for Medium-Range Weather Forecasts (ECMWF) forecast and a surface emissivity training data set (Goldberg et al., 2003). The training data set used for the AIRS V4 algorithm has a limited number of soil, ice, and snow types and very little emissivity variability in the training ensemble. In the AIRS V5 version, the regression coefficient set has been upgraded using a number of published emissivity spectra (12 spectra for ice/snow, 14 for land) blended randomly for land and ice (Zhou et al., 2008). These improvements generated a better emissivity first guess for use with the AIRS V5, and improved retrievals over the desert regions (Divakarla et al., 2008). In AIRS V6, a surface climatology was constructed from the 2008 monthly MODIS MYD11C3 emissivity product, and extended to the AIRS IR frequency hinge points using the baseline-fit approach described by Seemann et al. (2008). Nevertheless, using of day-night differences for evaluation of the AIRS V6 O_3 product suggest that further refinements for better surface emissivity retrievals are required and cloud covers is another problem that needs to be solved.

3.2 MLS O_3 retrievals day-night differences

3.2.1 MLS O_3 profile

In order to better understand day-night differences in TCO, we also study day-night changes in the vertical profile of O_3 using MLS O_3 profile measurements. Figure 2a shows that the global (60 °S-60 °N) differences between day and night MLS O_3 profile occur in the mesosphere (10 hPa - 0.1 hPa). The O_3 mixing ratios are about an order of magnitude larger during night in the mesosphere, which was revealed by Huang et al. (2008) previously. Different latitude bands (30 degree) between 60 °S and 60 °N all display similar results.

We also find an unexpected polar bias at high latitudes in Figure 2d and 2g. On the one hand, the larger differences between ascending (“daytime”) and descending (“nighttime”) MLS O_3 profile at high latitude extend from the stratosphere to the mesosphere. On the other hand, ascending O_3 is smaller than descending O_3 at 10 hPa over 60 °N-90 °N in Figure 2d, which is in contrast with the result of other latitude bands.

3.2.2 MLS O_3 retrievals in 90°S-60°S and 60°N-90°N

The MLS O_3 profile polar bias mentioned above turns out to be related to an inconsistency in the 'AscDescMode' flag of MLS v4.2x standard O_3 product in 90°S-60°S and 60°N-90°N. We counted the daily number of pixels at both poles when observation mode is ascending (AscDescMode = 1) and descending (AscDescMode = -1) respectively. Figures 3a and 3c show there is a clear change on 14 May 2015 in the daily number of ascending/descending pixels, consistent with the change of MLS SCO in Figure 3b and 3d. Before 14 May 2015, there are very large differences (about 500 pixels) in the number of pixels between ascending and descending mode, as well as the differences in MLS SCO. After 14 May 2015, the ascending and descending MLS SCO are much closer with smaller differences (about 20 pixels) of ascending and descending pixels.

For the MLS O_3 profile in Figure 4, differences between ascending and descending MLS O_3 profiles at high latitudes for 2016-2018 are much smaller and more realistic compared to the differences for 2005-2014. The large differences in the stratosphere disappear in polar regions with the correct 'AscDescMode' flag for 2016-2018. For 60°N-90°N, ascending mode O_3 also becomes larger than descending mode O_3 at 10 hPa in Figure 4b. This indicates that the MLS 'AscDescMode' flag is correct for 2016-2018.

The O_3 retrieval algorithm adopted by the MLS v2.2 products has been validated to be highly accurate using multiple correlative measurements and the data have been used widely (Jiang et al., 2007; Froidevaux et al., 2008). The MLS v3.3 and v3.4, O_3 profile was reported on a finer vertical grid and the bottom pressure level with scientifically reliable values (MLS O_3 accuracy was estimated at ~20 ppbv +10% at 261 hPa) increases from 215 to 261 hPa (Livesey et al., 2015). The latest MLS v4.2x O_3 profile used in this study, released in February 2015, were in general similar to the previous version. One of the major improvements of MLS v4.2x was the handling of contamination from cloud signals in trace gas retrievals that resulted in significant reduction in the number of spurious MLS profile in cloudy regions and a more efficient screening of cloud-contaminated measurements. Furthermore, the MLS O_3 products have been improved through additional retrieval phases and reduction in interferences from other species (Livesey et al., 2015). We find no indications that changes in instrument or algorithm are responsible for this 'AscDescMode' flag inconsistency. This flag inconsistency is not present between 60°S and 60°N.

3.3 Comparison between AIRS and MLS O_3 retrievals

Figure 5 presents comparison of yearly and monthly averaged SCO for 2005-2018 observed by AIRS and MLS three latitude bands. Figure 5a shows the 14-year average daytime AIRS SCO (250 hPa – 1 hPa) and MLS SCO (261 hPa – 0.02 hPa) in 60°S-60°N for 2005-2018. The time average MLS SCO column is 260.62 DU and AIRS SCO is 264.24 DU. The average MLS SCO day-night differences for 2005-2018 (0.88 DU) is smaller than the AIRS SCO day-night differences observed for the same time period (5.24 DU). The day-night difference of MLS SCO is 0.79 DU in the mesosphere (10 hPa - 0.1 hPa) and 0.03 DU in the stratosphere (100 hPa - 10 hPa). The day-night difference of AIRS SCO is 1.51 DU in the mesosphere (10 hPa - 1 hPa) and 3.85 DU in the stratosphere (100 hPa - 10 hPa). Compared to the AIRS SCO day-night differences, the magnitude of MLS SCO day-night differences in the stratosphere and in the mesosphere are much smaller. It has been pointed out that errors in temperature profiles and water vapour mixing ratios will adversely affect the AIRS O_3 retrieval. Significant biases (0 - 100%) may exist in the region between ~300 hPa and ~80 hPa (Wang et al., 2019; Olsen et al., 2017). AIRS O_3 retrievals do not distinguish portions of the O_3 profile as being of different qualities, because all AIRS O_3 channels sense the surface as well atmospheric O_3. Thus AIRS O_3 retrievals are compromised if the surface is not well characterized (Olsen et al., 2017). In addition, AIRS SCO retrievals show smaller day-night differences in the polar zones (1-2 DU) than in 60°S-60°N (4-5 DU). This is related to clouds and the surface type which affect the AIRS O_3 retrievals as mentioned above. Figure 5b shows the monthly 14-year average daytime AIRS SCO and MLS SCO in the 60°S-60°N for 2005-2018. Seasonal or random changes of clouds and the surface emissivity have more significant impact on each monthly AIRS SCO retrieval than on the MLS SCO retrieval. Compared with the 60°S-60°N region, surface types in polar zones are less diverse (snow or ice). Therefore, the monthly 14-year average daytime AIRS SCO and MLS SCO in Figure 5d and 5f show similar patterns. Figures 5c to 5f also confirm that MLS SCO has a polar bias when compared with AIRS SCO at high latitudes. In addition, for MLS SCO in Figure 5f, the biggest day-night differences (50-60 DU) occur in September and October during the Antarctic O_3 hole.

3.4 Day-night difference of equatorial Pacific low O_3 regions

Generally, the Pacific low O_3 region (TCO < 220 DU) exist all year round and its size is larger at night than during the day, unlike the seasonal O_3 hole which occurs over Antarctica during the Southern Hemisphere polar winter. On the one hand, there are limited direct NO_x emissions causing low O_3 over oceans compared to land. On the other hand, the low O_3 over the tropical western Pacific can be attributed to tropospheric O_3 loss in this area. Its presence is related to a pronounced minimum in the tropospheric column of O_3 over the west Pacific, which loss is due to photochemical mechanism with higher air temperatures and higher water concentrations for O_3. In addition, high sea surface temperatures also favour strong convective activity in the tropical West Pacific, which can lead to low O_3 mixing ratios in the convective outflow regions in the upper troposphere in spite of the increased lifetime of odd oxygen (Kley et al., 1996; Rex et al., 2014). A further reduction in the tropospheric O_3 burden through bromine and iodine emitted from open-ocean marine sources has been postulated by numerical models (Vogt et al., 1999; von Glasow et al., 2002; von Glasow et al., 2004; Yang et al., 2005) and observations (Read et al., 2008).

Figure 6a and 6c show the low O_3 region is mainly located over the western Pacific by AIRS. Rajab et al. (2013) investigated similar low TCO in Malaysia using AIRS data. They found the highest O_3 concentration occurred in April and May and the lowest O_3 concentration occurred during November and December, which is consistent with our results in Figure 6f. They also found that O_3 concentrations exhibited an inverse relationship with rainfall, but was positively correlated with temperature. Figure 6b shows for MLS, the low O_3 regions appear in large areas at night besides in tropical western Pacific. However, Figure 6d shows the occurrence frequency and intensity of daytime low O_3 regions by MLS SCO retrievals drastically reduces and exists mainly in tropical western Pacific. In Figure 6e and 6f, yearly and monthly averaged AIRS TCO and MLS SCO of the low O_3 regions show no consistency and regularity. The analysis of daytime MLS SCO of the low O_3 regions is based on only a few observations. We cannot distinguish whether it is an algorithm problem or a chemical mechanism that caused this phenomenon. For AIRS, clouds over oceans may have greater impact on the AIRS TCO retrievals at night. For MLS, more active chemical reactions may occur in these low O_3 regions at night.

For past, current and future monitoring of atmospheric phenomena like the Pacific tropospheric low O_3 area, it is important that observations are sufficient accurate. The evaluation of day-night differences in both MLS and AIRS has revealed the existence of biases in the satellite data that are sufficiently large in comparison to expected variations and changes in atmospheric O_3 that they may hamper the use of these satellite data studying them.

4 Conclusions

Comparison of daytime and nighttime AIRS TCO has revealed small but not insignificant biases in AIRS TCO. The differences are likely related to surface type (land/desert/vegetation/snow or ice) and infrared surface emissivity, especially over regions that exhibit smaller infrared emissivity or large seasonal variability in infrared emissivity. Differences typically were of the order of a few percent, which is significant given that long term changes in TCOs related to anthropogenic emissions of stratospheric O_3 depleting substances outside of polar regions are also of the order of a few percent.

Over land, patterns in day/night differences appear to be dominated by the dryness of the surface, suggesting that emissivity may not be well represented or that reduced sensitivity to the lower troposphere during night compared to day over hot surfaces results in a different AIRS TCO. The spatial inhomogeneity of day/night AIRS TCO differences over drier regions points to the emissivity rather than the averaging kernels dominating these differences. Infrared satellite retrieval artefacts due to land surface emissivity is well-known phenomenon (Zhou et al., 2013; George et al., 2015; Bauduin et al., 2017). On the other hand, Figure S1(a) in the supplement shows the AIRS O_3 retrieval over desert region contains a larger fraction of the O_3 priori than a forest region at the same latitude. It means the AIRS O_3 retrievals over desert are highly determined by the O_3 priori (derived from an observationally-based climatology) and thus have little information content. The weak radiance information over deserts may relate to surface emissivity. For most desert areas, emissivities are less than 0.85 due to the strong quartz absorption feature between 8-9.6 μm range (9.6 μm band is used to retrieve AIRS TCO and O_3 profiles during both day and night), whereas the emissivity of forest, water and ice cover are generally greater than 0.95 and spectrally flat in the 3-12 μm spectral range (Olsen et al., 2017). Figure S1(b) indicates that the day-night differences of radiance information over deserts are also larger, which is consistent with large differences of AIRS TCO retrievals over deserts.

There were major changes to the surface emissivity retrieval in AIRS V6 compared to previous versions resulting in a very significant improvement in yield and accuracy for surface temperature and emissivity over land and ice surfaces compared to previous versions. Nevertheless, our results indicate that the AIRS V6 TCO still can be further improved with regard to the representation of infrared emissivity. In addition, AIRS TCO differences over oceans bear a clear cloud cover signature which is likely related to uncertainties in the representation of clouds in the retrieval algorithm. The latter may also impact AIRS

TCO retrievals over land, although detection of cloud features in AIRS TCO day-night differences is difficult due to the presence of the land surface emissivity related bias.

The strongest diurnal cycles in cloud fraction are found in the tropics over land, following strong daytime heating (Noel et al., 2018). Over oceans, diurnal cycles in cloud fraction are weaker, but very broadly indicate reduced cloudiness during day compared to night, especially in the tropics and subtropics (Noel et al., 2018). In case of clouds, AIRS TCO appears to be larger during daytime compared to nighttime. This is consistent with the notion of increased cloudiness during the night, increasing the chance of shielding by undetected or unrecognized clouds in the AIRS retrieval. For ocean regions with persistent clouds during day and night (for example over ITCZ), Figure S2 in the supplement shows that variations of cloud layer height have a greater impact on AIRS TCO day-night differences than of the cloud fraction.

Our results do not provide much evidence of another possible causes of day/night differences in AIRS TCO: the photochemical diurnal O_3 cycle in the lower troposphere and upper atmosphere. The strongest diurnal O_3 effects occur in the boundary layer over land due to nighttime surface deposition and daytime photochemical O_3 production in the presence of air pollution. In the marine boundary layer, the diurnal O_3 cycle is much weaker due to absence of air pollution and a general slow O_3 destruction regime (~10%/day). Similarly, in the free troposphere, the diurnal O_3 cycle is also weak due to low O_3 production rates (generally low levels of pollution relevant for O_3 production). Hence, the diurnal O_3 cycle in the free troposphere above 750 hPa is negligible (Petetin et al., 2016). In summary, any tropospheric photochemical diurnal O_3 cycle effect should resemble some correspondence with air pollution. The day-night differences in AIRS TCO clearly do not resemble patterns of surface air pollution (Figure 1). MLS day/night differences are confined to the mesosphere (1 hPa and higher). As shown in Smith et al. (2014) the lifetime of O_3 due to chemistry is strongly altitude dependent (<20 min in the upper mesosphere above 0.01 hPa). Only in the mesosphere the loss timescale for O_3 becomes long enough to see significant differences between average daytime and nighttime concentrations. While its contribution to MLS SCO is negligible. The mesospheric diurnal O_3 cycle thus will also have a negligible effect on day/night AIRS TCO differences. In addition, Strode et al. (2019) simulated the global diurnal cycle in the tropospheric O_3 columns, their results indicated that the mean peak-to-peak magnitude of the diurnal variability in tropospheric O_3 is approximately 1 DU. Figures S3 to S6 in the supplement also show that the AIRS TCO retrieval artefacts dominate the day/night variability of tropospheric O_3 residuals (TOR = AIRS TCO – MLS SCO).

In summary, our analysis has identified evidence and indications that clouds, land surface infrared emissivity, and sensitivity of satellite measurements to the lower troposphere, influence AIRS satellite TCO observations, pinpointing to areas and processes for algorithm improvement.

The MLS v4.2x was very useful for verification of daytime and nighttime SCO and O_3 profile between 60 °S-60 °N. MLS day-night differences in SCO and O_3 profiles show that day-night differences are only small (< 1 DU) and likely to be in the upper stratosphere and mesosphere. However, an inconsistency was found in the 'AscDescMode' flag in 60°N-90 °N and in 90 °S-60 °S, resulting in inconsistent profiles in these regions before 14 May 2015. In processor version v4.22 and later versions this issue has been fixed, but since it is a relatively small issue, the MLS data set before 2016 has not been reprocessed.

A case study of day-night differences O_3 over equatorial Pacific revealed that both AIRS and MLS O_3 retrievals have biases in comparison to expected variations and changes. Our results show that maintaining the quality of the satellite observations of stratospheric O_3 is therefore highly relevant.

Data availability

Satellite data sets used in this research can be requested from public sources. AIRS total ozone column data are available online (https://giovanni.gsfc.nasa.gov/giovanni/). The MLS Level 2 data can be obtained from the NASA Goddard Space Flight Center Data and Information Services Center (GSFCDISC, https://disc.gsfc.nasa.gov/datasets/ML2O3_004/summary?keywords=ML2O3_004).

Author contributions

WNW and JL provided satellite data, tools, and analysis. RA, JL and THC undertook the conceptualization and investigation. WNW prepared original draft. RA and JL carried out review and editing. JW checked the English language. All authors discussed the results and commented on the paper.

Competing interests

The authors declare that they have no conflict of interest.

Acknowledgements

The support provided by China Scholarship Council (CSC) during a visit of Wannan Wang to Royal Netherlands Meteorological Institute (KNMI) is acknowledged.

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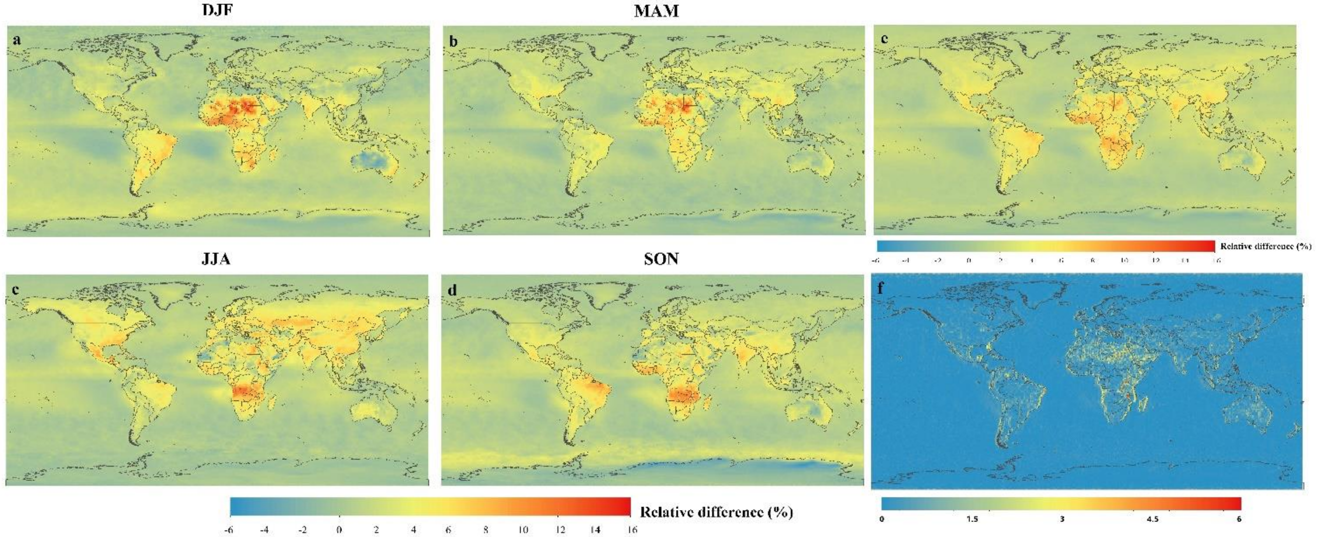

Figure 1: AIRS TCO averaged day-to-night relative difference for 2003-2018. The relative difference is calculated as: 100 × (daytime - nighttime) / daytime (in percent, %). (a) DJF. (b) MAM. (c) JJA. (d) SON. (e) 16 years averaged. (f) longitude gradient value using absolute difference between two pixels adjacent at the same latitude in (e).

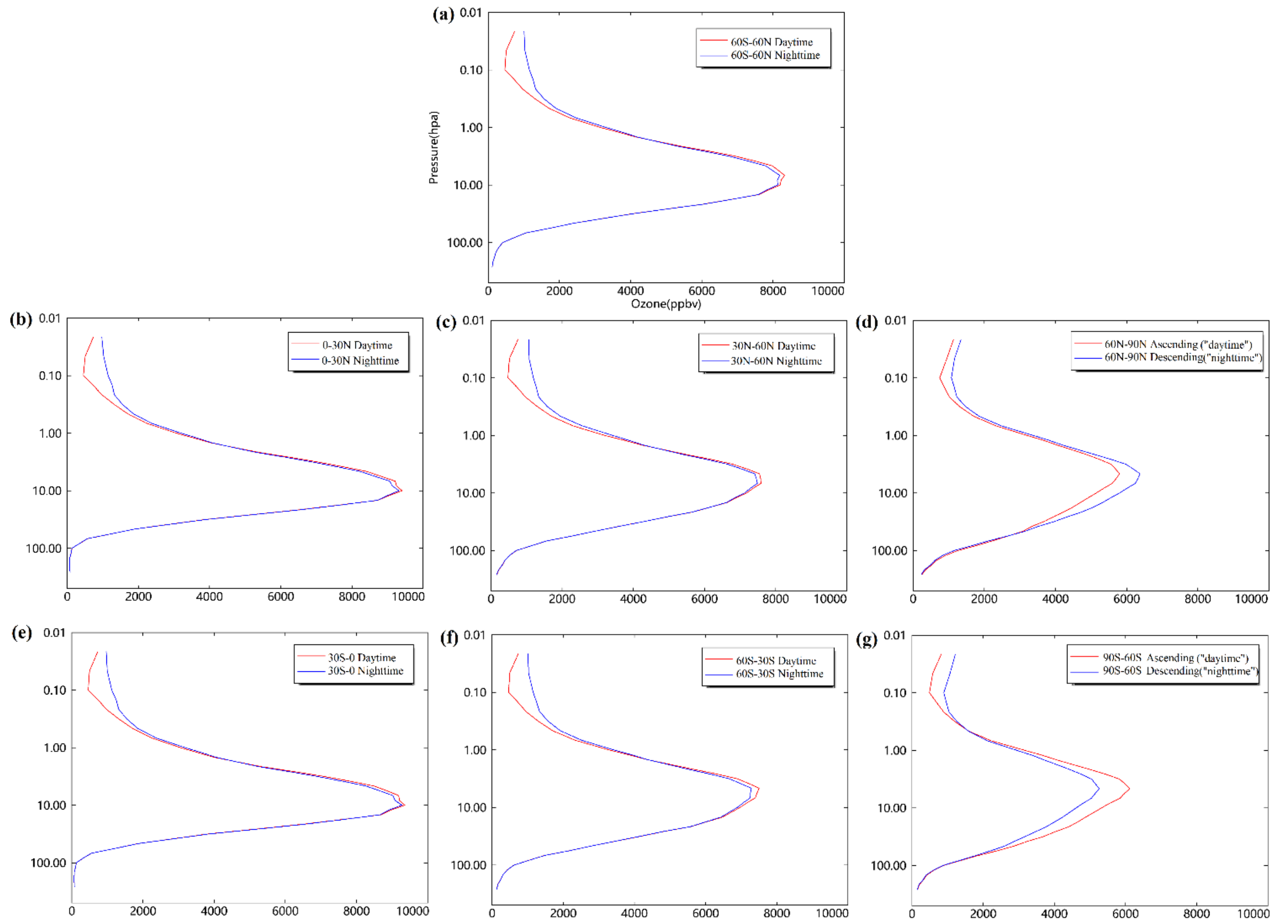

Figure 2: Averaged MLS ozone profile between 261 hPa and 0.02 hPa per latitude band (30 degree) for 2005-2018. (a) 60 °S-60 °N. (b) 0-30 °N. (c) 30 °N-60 °N. (d) 60 °N-90 °N. (e) 30 °S-0. (f) 60 °S-30 °S. (g) 90 °S-60 °S.

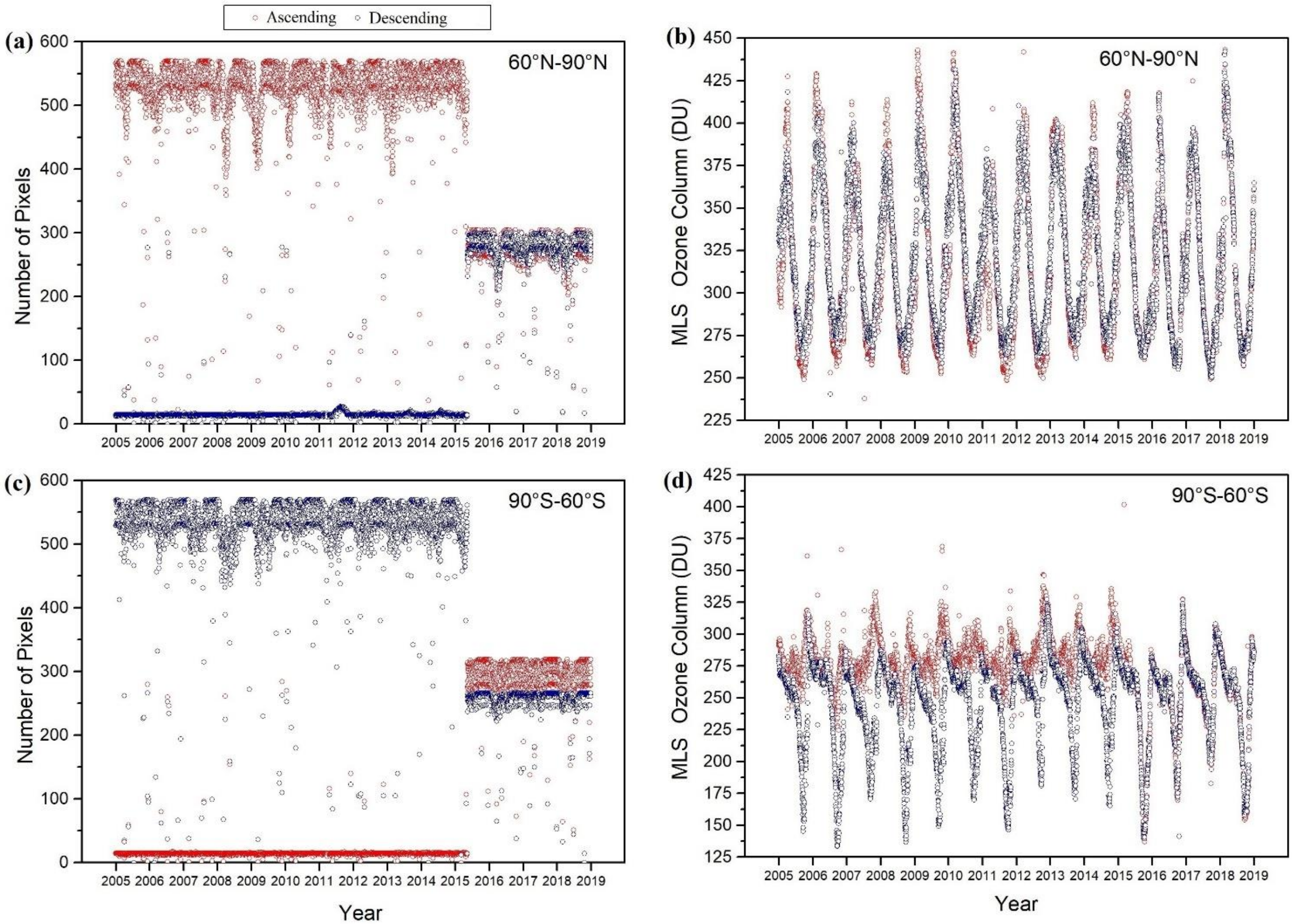

Figure 3: (a) Time series of daily number of Ascending ("daytime") and Descending ("nighttime") pixels in 60 °N-90 °N. (b) Time series of daily average Ascending and Descending MLS SCO in 60 °N-90 °N. (c) Same as (a), but in 90 °S-60 °S. (d) Same as (b), but in 90 °S-60 °S.

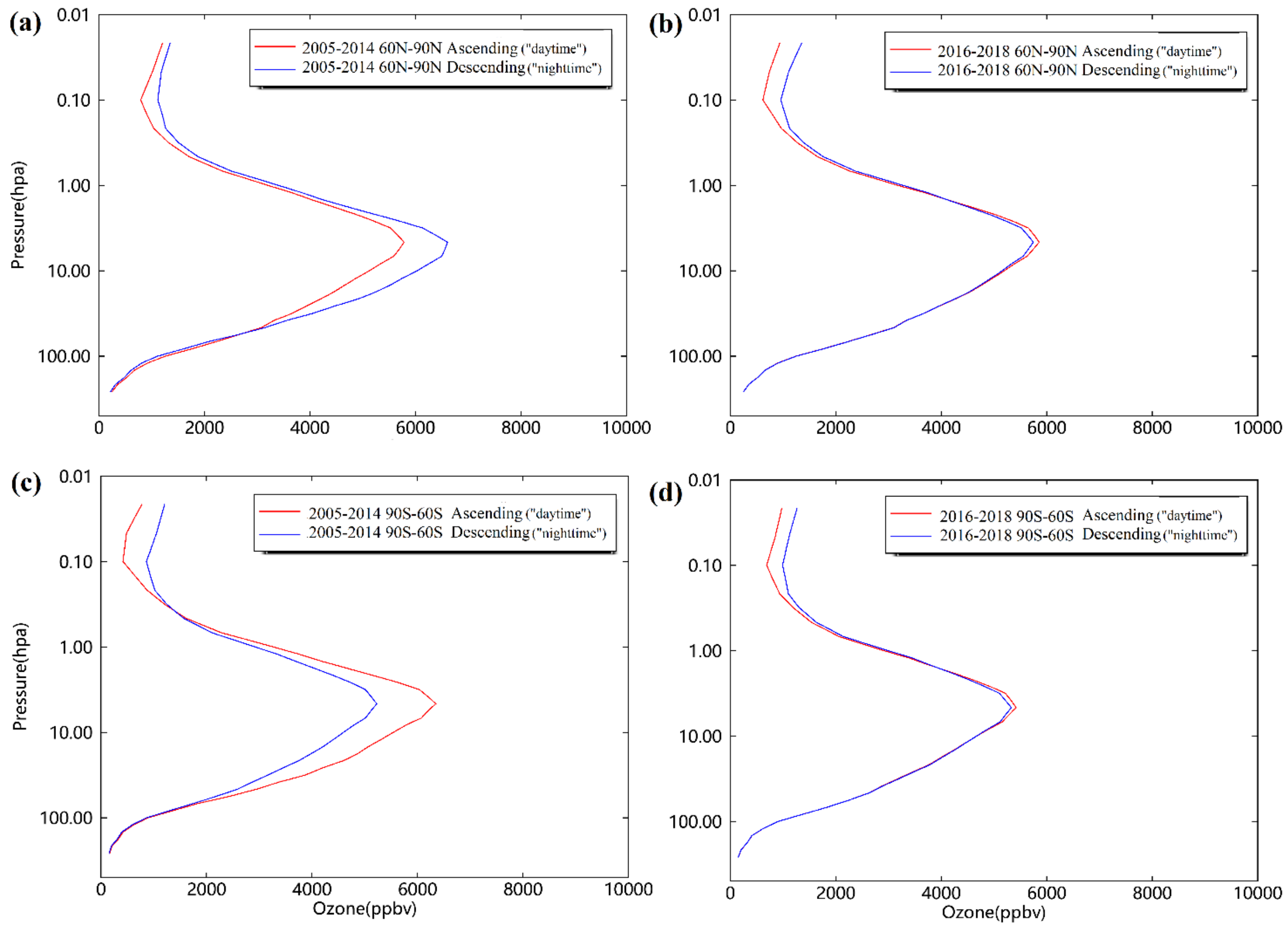

Figure 4: (a) Averaged MLS ozone profile between 261 hPa and 0.02 hPa for 2005-2014 in 60 °N-90 °N. (b) Averaged MLS ozone profile between 261 hPa and 0.02 hPa for 2016-2018 in 60 °N-90 °N. (c) Same as (a), but in 90 °S-60 °S. (d) Same as (b), but in 90 °S-60 °S.

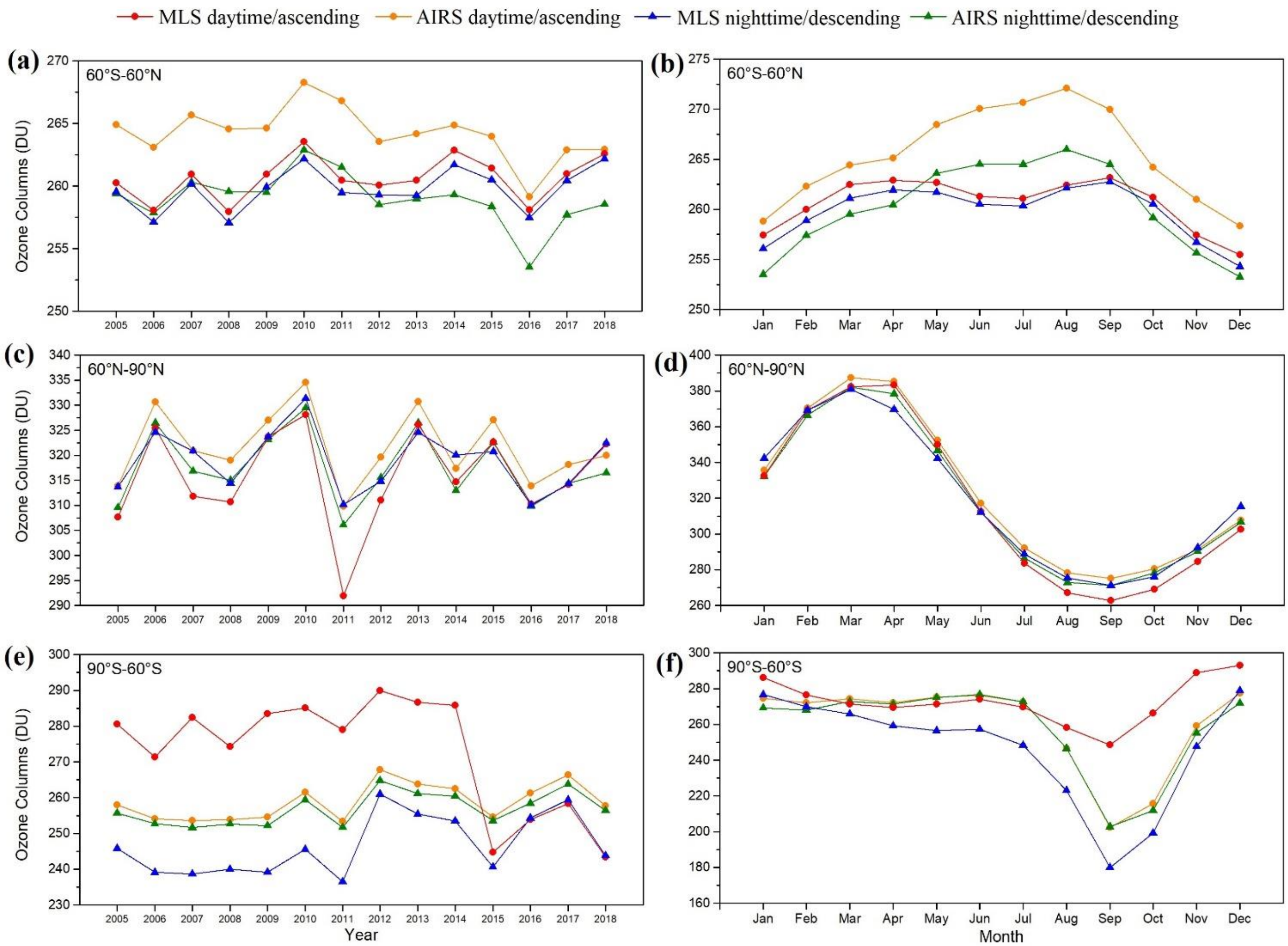

Figure 5: Yearly and monthly averaged AIRS SCO and MLS SCO for 2005-2018. AIRS SCOs are calculated from 250 hPa to 1 hPa.

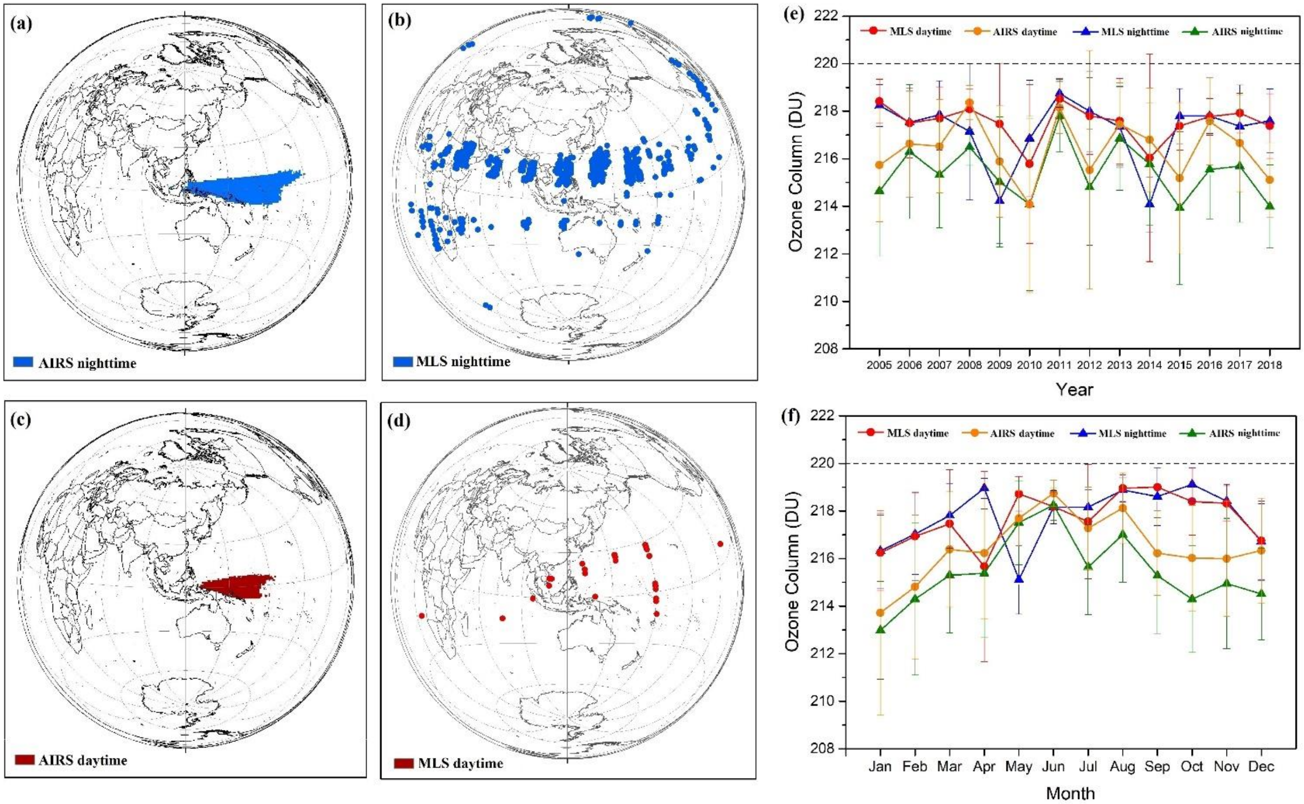

Figure 6: Spatial and temporal distribution of the low ozone. (a) Location (composite pixel) of the yearly nighttime low ozone from 2005 to 2018 for AIRS TCO. (b) Same as a but for MLS SCO. (c) Location (composite pixel) of the yearly daytime low ozone from 2005 to 2018 for AIRS TCO. (d) Same as c but for MLS SCO. (e) Yearly averaged AIRS TCO and MLS SCO of the low ozone regions for 2005-2018. (f) Monthly averaged AIRS TCO and MLS SCO of the low ozone regions for 2005-2018. Uncertainties represent the standard deviation of the measured values.