

**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**.

### **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.

**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”)*”.

**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 Barnett, 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 Teixeira, 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<sub>3</sub> retrieval over the Sahara Desert [20 °N, 24 °E, 23 °N, 27 °E] region contains a larger fraction of the O<sub>3</sub> priori than a forest region [22 °N, 106 °E, 24 °N, 108 °E] at the same latitude. It means the AIRS O<sub>3</sub> retrievals over desert are highly determined by the O<sub>3</sub> 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<sub>3</sub> 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.

## **B. Technical issues**

**Comment 5:** Lines 9-12 and Lines 56-59: “Based on knowledge of the chemistry and transport of O<sub>3</sub> ...” The premise of the work is unclear to me. I recommend that the authors rephrase their argument for evaluating diurnal changes in O<sub>3</sub>, 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<sub>3</sub>. Further, an accurate assessment of O<sub>3</sub> variation is needed for a reliable and homogeneous long-term trend detection in the global O<sub>3</sub> distribution.

**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<sub>3</sub> 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.

**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*”.

**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<sub>3</sub> profiles*”.

**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*”.

**Comment 10:** Lines 75-79: Personally, I think this level of detail about the chemical reactions of O<sub>3</sub> (and its precursors) is irrelevant to the discussion here.

**Response 10:** We explained the chemical reactions of O<sub>3</sub> in detail in order to emphasize significant deviations between daytime and nighttime O<sub>3</sub> 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.

**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](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.

**Comment 13:** Line 140: What is considered a “small positive bias” in lower stratospheric MLS O<sub>3</sub> 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...*”.

**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*”.

**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<sub>3</sub> retrievals day-night differences*” and “*3.2.1 MLS O<sub>3</sub> profile*”.

**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*”.

**Comment 19:** Lines 186, 187, 189: “14 may” should be “14 May”.

**Response 19:** We modified this as you suggested.

**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).

**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<sub>3</sub> 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<sub>3</sub> becomes long enough to see significant differences between average daytime and nighttime concentrations.

**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.

**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<sub>3</sub> over equatorial Pacific revealed that both AIRS and MLS O<sub>3</sub> retrievals have biases in comparison to expected variations and changes. Our results show that maintaining the quality of the satellite observations of stratospheric O<sub>3</sub> is therefore highly relevant.*”