Dear Editor,

In addition to our revisions based on the comments from our two referees, we have added some introduction of other OMI ozone algorithms in the first paragraph of the introduction section, following the editor's suggestion of our companion paper (Huang et al., 2017).

We have revised it as:

"The Dutch-Finnish built Ozone Monitoring Instrument (OMI) on board the NASA Aura satellite 103 has been making useful measurements of trace gases including ozone and aerosols since October 2004. There are various retrieval algorithms to retrieve ozone profile and/or total ozone from OMI data (Bak et al., 2015), **including two independent operational total ozone algorithms (Bhartia and Wellemeyer, 2002; Veefkind et al., 2006) and two ozone profile algorithms. Of the two ozone profile algorithms, one is the operational algorithm (OMO3PR) developed at KNMI (van Oss et al., 2001), and the other one is a research algorithm developed at Smithsonian Astrophysical Observatory (SAO) by (Liu et al., 2010b). Both algorithms retrieve ozone profile 110 from the spectral region 270-330 nm using the optimal estimation method, but they differ significantly in implementation details including radiometric calibration, radiative transfer model simulation, a priori constraint, retrieval grids, and additional retrieval parameters.** The **SAO** ozone profile retrieval algorithm **was** initially developed at the Smithsonian Astrophysical Observatory (Liu et al., 2005) for Global Ozone Monitoring Experiment (GOME) data and was adapted to OMI data (Liu et al., 2010b)."

Huang, G., Liu, X., Chance, K., Yang, K., and Cai, Z.: Validation of 10-year SAO OMI Ozone Profile (PROFOZ) Product Using Aura MLS Measurements, Atmos. Meas. Tech. Discuss., 2017, 1-25, doi: 10.5194/amt-2017-92, 2017.

Response to Referee #1

We thank referee's helpful and constructive comments and review. The referee's comments are listed in *italics*, and our responses in black with revised texts in **bold black**.

Overview:

The authors have produced a carefully determined retrieval of ozone profiles, SOCs, and TOCs for over 10 years from an OMI profile retrieval algorithm (SAO). They compared retrieved SOC, TOC, and profile measurements extensively with global ozonesondes. These ozonesonde comparisons included filtering the OMI measurements for nearly clear-sky scenes, SZA < 75 degrees, and cross-track positions least affected by OMI row anomaly. The authors show from differences between pre and post-row anomaly periods that the current 10-year profile product (and derived columns) does not appear to be useful for evaluating decadal trends; however, the product at shorter timescales including daily from the analyses appears to be a useful C1 science product, particularly from the tropics to mid-latitudes. The paper appears good in current form and publishable with mostly just a few small comments that are listed below.

General comments:

* Lines 116-118: You might include in this reference list the paper by Yang et al. [2007] which used OMI and MLS to derive tropospheric ozone columns: Yang, Q., D. M. Cunnold, H. –J. Wang, L. Froidevaux, H. Claude, J. Merrill, M. Newchurch, and S. J. Oltmans, Midlatitude tropospheric ozone columns derived from the Aura Ozone Monitoring Instrument and Microwave Limb Sounder measurements, J. Geophys. Res., 112, D20305, doi:10.1029/2007JD008528, 2007.

Done.

* Line 165: Were the NCEP tropopause pressures for getting TOCs and SOCs determined from a PV-theta definition, or a lapse rate definition, or something else?

The tropopause pressures are defined by the lapse rate. We have added "(**defined based on the lapse rate**)" after the tropopause.

Section 4.1.3.: Is your derivation of cloud optical centroid pressure and your effective scene pressure the same as Joiner et al. (2009, ACP)? Is there any major difference with your effective cloud fraction and their radiative cloud fraction for determining effective scene pressure? Is effective scene pressure determined the same?

Joiner, J., M. R. Schoeberl, A. P. Vasilkov, L. Oreopoulos, S. Platnick, N. J. Livesey, and P. F. Levelt (2009), Accurate satellite-derived estimates of the tropospheric ozone impact on the global radiation budget, Atmos. Chem. Phys., 9, 4447-4465, doi:10.5194/acp-9-4447-2009.

Yes, we use the optical centroid cloud pressure (OCCP) from the OMI Raman cloud product by J. Joiner (Vasilkov et al., 2008), i.e., same as in Joiner et al. (2009). We directly use cloud pressure values meeting all the recommended quality flags. For pixels without qualified OCCP values, they are filled in by spatial interpolation on an orbital basis. Remaining empty values after the interpolation are filled in by a climatology derived from 7 years of OMI OCCPs. We use pixel-independent approximation for partial cloudy conditions, i.e., Lambertian clouds with OCCP and clear-sky conditions with surface pressure, and thus we do not use effective scene pressure. We use the effective cloud fraction from the same cloud product as initial value. We re-derive the effective cloud pressure from radiances at a weakly-absorbing wavelength (~347 nm) similar to the Raman cloud product. Typically, the effective cloud-top pressure is used in the radiative transfer calculation. Radiative cloud fraction, ratio of cloud radiance to total radiance, is derived from the effective cloud fraction.

We have added "**from the OMI Raman cloud product** (**Vasilkov et al., 2008**)" after "Our OMI ozone algorithm assumes clouds as Lambertian surfaces with optical centroid cloud pressure"

* Line 257: This appears to be a dead link.

The address is correct. But the line number of 258 is added between "discover-" and "aq" when copying the link or clicking it directly. We have corrected it the updated version.

* Line 274: There are more error sources than just the pump efficiency for the ozonesondes, but correct that pump efficiency errors are largest in higher altitudes.

We have changed "uncertainties in pump efficiency" to "**uncertainties mainly from pump efficiency**"

** Where "SOC" is first mentioned (including the Abstract) it might be useful to state clearly that SOC is not the generally inferred total stratospheric ozone column but instead the ozone column from the tropopause up to balloon burst pressure. In your second submitted joint validation paper that uses MLS, "SOC" probably refers to total stratospheric ozone column?

The SOC is first mentioned in the Abstract. We have revised it as:

"The MBs of the stratospheric ozone column (SOC, the ozone column from the tropopause pressure to the ozonesonde burst pressure) are within 2% with SDs of < 5% and the MBs of the tropospheric ozone column (TOC) are within 6% with SDs of 15%."

In addition, we have described the SOC as the ozone column from the tropopause pressure to the corresponding ozonesonde burst pressure in the Sect. 3. But to make it more clear, we have changed "The TOC is integrated from the surface to the tropopause and the SOC is integrated from the tropopause pressure to the ozonesonde burst pressure" to "The TOC is integrated from the surface to the tropopause. And the SOC is not the total stratospheric ozone column, but the

ozone column integrated from the tropopause pressure to the ozonesonde burst pressure."

* *Line 343: Should be ". . .stratosphere (UTLS). . . "* Done.

* Many of the figures, if intended as single column will have figure text that will be too small to read. The authors might specify to the journal that these figures should be printed double column, or perhaps instead increase some of the figure text.

Thank for the suggestion. We will contact the journal about this during the production process.

References:

Vasilkov, A. P., J. Joiner, R. Spurr, P. K. Bhartia, P. F. Levelt, and G. Stephens: Evaluation of the OMI cloud pressures derived from rotational Raman scattering by comparisons with satellite data and radiative transfer simulations, J. Geophys. Res., 113, D15S19, doi:10.1029/2007JD008689, 2008.

Response to Referee #2

We thank referee's helpful and constructive comments and review. The referee's comments are listed in *italics*, and our responses in black with revised texts in **bold black**.

The manuscript deals with the validation of nadir (partial) ozone profiles from 10 years of OMI data using balloon soundings. The quality and limitations of the products are well described showing the results of various tests and for different conditions.

General comments:

To me it would have made the narrative order more logic to first read about the tests on SZA, cloud fraction and cross-track position dependence before presenting the results of the final dataset resulting from the selected criteria (i.e. move section 4.1.1 to 4.1.4).

We agree that it is more logical to first discuss tests on SZA, cloud fraction and cross-track position dependence before presenting the results of the general results. However, we have already mentioned conducting the tests on these parameters and the order in Sect. 2.1 and in Sect. 3, and the discussion of Sects. 4.1.2-4.1.4 depends on results and discussion in Sect. 4.1.1. Therefore, we keep our original order to present the overall results first.

To make it clear, we have changed "We will use all OMI pixels of each filtering parameter when evaluating retrieval quality as a function of that specific parameter." To "**The selection and justification of these criteria will be discussed in Sects. 4.1.2-4.1.4, in which we** will use all OMI pixels of each filtering parameter when evaluating retrieval quality as a function of that specific parameter." in line 223 of Sect. 2.1 of the AMTD manuscript.

Also in section 3, we have changed "Although we filter OMI data based on cloud fraction, cross-track position, and SZA, we conduct the comparison as a function of these parameters using coincidences at all latitude bands to show how these parameters affect the retrieval quality" to "Although we filter OMI data based on cloud fraction, cross-track position, and SZA in the final evaluation of our retrievals against ozonesonde observations as shown in Sect. 4.1.1, we conduct the comparison as a function of these parameters using coincidences at all latitude bands to show how these parameters affect the retrieval section of these parameters using shown in Sects. 4.1.2-4.1.4."

Please mention version numbers throughout the text ('current', 'updated', etcetera are confusing).

Unfortunately, SAO OMI retrieval algorithm does not have any official version number. The first version is described in Liu et al. (2010) and the current (with minor updates) version that is used in this paper is briefly described in Kim et al. (2013). The future or next version refers to the version that we are currently working on.

For clarifying, we have revised the description of our current algorithm in Set. 2.1 as follows: "The **current** algorithm of our SAO OMI ozone product **that is used in this paper** was briefly described in Kim et al. (2013)."

Specific/technical comments:

References to Huang et al., 2016: I am not sure if you can specify 2016 if it is not yet published in ACP discussions.

This paper had been submitted to Atmospheric Measurement Techniques and has been published at AMTD. We have updated the reference and citation as:

"Huang, G., Liu, X., Chance, K., Yang, K., and Cai, Z.: Validation of 10-year SAO OMI Ozone Profile (PROFOZ) Product Using Aura MLS Measurements, Atmos. Meas. Tech. Discuss., 2017, 1-25, doi: 10.5194/amt-2017-92, 2017."

Page 5 line 105: remove second 'in' (.. retrieval errors in in the range.). Done.

Page 9 line 215: there seems to be a space missing in '2009and' Done.

Page 9 line 219, second criterion: rephrase, now it appear to be a contradiction that you select rows 4-27 but state that these have a worse quality and larger footprint as that is what this selection is avoiding.

We have rephrased it as:

"... 2) cross track positions between 4 and 27, due to **the** relatively worse quality and much larger footprint size of **the off-nadir pixels beyond this range**; ..."

Page 11 line 295 remove the second (double) comma after the first use of OMI Done.

Page 12 line 311 add 'above' to "~30 km" Done.

Page 12 lines 319-310. Why do you not use median/percentile values to deal with outliers? It is a good idea to use the median/percentile values to deal with outliers. But it is also common to remove outliers beyond the range of means plus and minus 3 times of standard deviations. Switching to the use of median/percentile values should not affect the overall conclusion of this paper.

Page 13 line 343: second troposphere should be stratosphere Done.

Page 13 lines 347-348: '.. differences .. can be reduced ..''. Unclear phrasing, do you mean that this could be an improvement in a future version or do you apply it here?

Yes. We mean an improvement in a future version. We have rephrased it as:

"Consequently, the SDs of OMI/sonde differences in the UTLS at mid- and high-latitudes can be reduced through reducing the retrieval uncertainties **in a future version of the algorithm that uses the TB climatology**."

Page 14 line 389: underestimate -> underestimates Done. We think you mean the "underestimate" at line 378.

Page 15 lines 400-401: I find it strange to read 'best agreement .. (except for the MBs) ...'. Given that the MBs are not closer to zero at all altitudes in the NH summer, please consider rephrasing 'best' (also in the conclusions section).

We refer "best agreement" to the smallest standard deviations. We have rephrased the sentence "The comparison results are clearly season-dependent with best agreement in the summer (except for the MBs) and the worst agreement in the winter" to "The comparison results are clearly season-dependent with different altitude-dependent bias patterns, and with the smallest SDs in the summer and the worst SDs in the winter"

In the conclusion, we have changed the original sentence "At northern mid-latitudes, the agreement is generally best (except for MBs) in the summer, with the best retrieval sensitivity and the smallest SDs as great as 20%, and the worst in the winter with the worst retrieval sensitivity and the largest SDs reaching 31%" to "At northern mid-latitudes, there are generally the best retrieval sensitivity and the smallest SDs as great as 20% in the summer, and the worst retrieval sensitivity and the largest SDs reaching 31% in the summer, and the worst retrieval sensitivity and the largest SDs reaching 31% in the winter".

Page 15 lines 407-410: Please clarify that you are referring to SDs only here: the non NH summer seasons show improvements in MBs over the a priori only above the 3-4 lowermost layers (not 2-3). Also, the statement on the NH summer season improvements 'at all tropospheric layers except for the bottom one' is not valid for the MBs.

Yes, we are referring to SDs only. We have revised it as:

"Also, the retrieval in the summer shows the most improvements **in terms of reduction in SDs** over the a priori in the lower troposphere at all tropospheric layers except for the bottom layer, while the retrievals during other seasons show the improvement over a priori only above the lowermost two/three layers."

Page 17 line 462: I guess you mean striping instead of stripping Yes.

Section 4.2.1 What pressure/altitude border do you use? How large is the contribution of the assumed profile above the burst altitude?

As mentioned in Sect. 3, the SOC is integrated from the tropopause pressure to the ozonesonde burst pressure. Based on the ozonesonde data with coincident total ozone measurements, ozone column above burst altitude constitutes 6-33% of stratospheric ozone column (14% on average).

Page 19 line 513: missing space in '1-3% except' Done.

Page 20 lines 546-547: why only at New Delhi, as Trivandrum also uses Indian sonde? What about the SD at Poona?

Although the bias at Trivandrum is small, there is a large SD at this station similar to that at New Delhi. Poona ozonesonde station is not included in this TOC comparison due to the small number (< 10) of ozonesonde-OMI pairs after applying our validation filters.

We have revised the sentence from "The large bias of >6 DU at New Delhi is likely associated with the large uncertainties of the Indian ozonesonde data" to "In addition, there is a large bias of > 6 DU at New Delhi. The poor comparisons at these two stations are likely associated with the large uncertainties of the Indian ozonesonde data."

Page 20 line 549: 'no much' \Box 'little' or 'not much'

Done.

Page 21 lines 590-591: add references for this statement. Sorry for the confusion. We meant evaluation in Sects. 4.1 and 4.2. Therefore, we have revised "Previous evaluation" to "**Comparisons in Sects. 4.1 and 4.2**"

Page 24 line 664: 'the comparison is seasonally dependent'. Rephrase (the results may depend on season but the comparison shouldn't) We have revised it **to** "The comparison **results are** seasonally dependent."

Figures 3 and 4: Label of the x-axis states 'A proiri' -> A priori Done.

Figure 3 (d, f, h): explain why sometimes the pre-/post RA number of collocations used (N) does not sum to the N for the full period (c, e, g).

This is because we applied the outlier removal process to each comparison, which may cause slight differences between the sum of the number of pre/post-RA collocations and the number collocations for the full period.

Figure 6: the colour bars are distinct from similar figures (5 and 7) – esthetic, not a real problem.

Thanks for pointing this out. We made them consistent.

Figures 8 to 12: these figures are quite fuzzy in the pdf, especially when zooming in to see the (colour of the) small dots – please check the figures' resolution for the final publication. Yes, we will upload our high-resolution figures for the final publication.

Figure 10: Consider that red versus green can be confusing for the colour blind. You might want to extend or shorten the vertical axis so that you get rid of the overlap between 0 and 80 DU.

We have changed the colors readable to everyone, as well as the vertical axis to get rid of the overlap between 0 and 80 accordingly. The new figure is shown as below:



Figure 11 caption states 'same as Figure 9' and Figure 12 caption states 'same as Figure 8'. Since the figures' setup refer to subsets and have a different orientation, I would rephrase this.

We have revised the captions of Figure 11 and 12 as:

"Figure11. Similar to panels in Fig. 9 but for different seasons at northern middle latitude during the 2004-2014 period.

Figure 12. Similar to panels in Fig. 9 but for comparison of lower tropospheric ozone columns during the 2004-2014 period. (a) Surface~550 hPa ozone column and (b) Surface~750 hPa ozone column in 30° N-60° N during the summer only, (c) and (d) same as (a) and (b) but for the tropics (30°S-30°N) during all year."

Figure 12 caption: is this for the full time series as for Figure 11? Yes, Figure 12 is for the full time series as for Figure 11. We have revised Figure 12 caption as shown above.

Figure 12a states '30N-60N JJA only' in the figure whereas it should be '30N-60N all year'. The original text in the figure is correct. Summer at mid-latitudes and all the seasons in the tropics are the times/locations when and where there are good retrieval sensitivities to lower tropospheric ozone.

Figure 13 caption: state use of AVK

We applied AVKs in this figure. We have added it a sentence "**OMI retrieval averaging** kernels are applied to ozonesonde data."

Figure 14b: for consistency with 14a add the years to the numeric information on the trends in the figure? Done.

References:

Kim, P. S., Jacob, D. J., Liu, X., Warner, J. X., Yang, K., Chance, K., Thouret, V., and Nedelec, P.: Global ozone–CO correlations from OMI and AIRS: constraints on tropospheric ozone sources, Atmos. Chem. Phys., 13, 9321-9335, doi: 10.5194/acp-13-9321-2013, 2013.

Liu, X., Bhartia, P. K., Chance, K., Spurr, R. J. D., and Kurosu, T. P.: Ozone profile retrievals from the Ozone Monitoring Instrument, Atmos. Chem. Phys., 10, 2521-2537, doi: 10.5194/acp-10-2521-2010, 2010.

1 Validation of 10-year SAO OMI Ozone Profile (PROFOZ)

2 Product Using Ozonesonde Observations

3

- 4 Guanyu Huang^{1,*}, Xiong Liu¹, Kelly Chance¹, Kai Yang², Pawan K. Bhartia³, Zhaonan Cai¹,
- 5 Marc Allaart⁴, <u>Gérard Ancellet⁵</u>, Bertrand Calpini⁶⁵, Gerrie J. R. Coetzee²⁶, Emilio Cuevas-
- 6 Agulló⁸⁷, Manuel Cupeiro⁹⁸, Hugo De Backer¹⁰⁹, Manvendra K. Dubey¹¹⁴⁰, Henry E.
- 7 Fuelberg¹²⁺⁺, Masatomo Fujiwara¹³⁺², Sophie Godin-Beekmann⁵⁺³, Tristan J. Hall¹²⁺⁺,
- 8 Bryan Johnson¹⁴, Everette Joseph¹⁵, Rigel Kivi¹⁶, Bogumil Kois¹⁷, Ninong Komala¹⁸, Gert
- 9 König-Langlo¹⁹, Giovanni Laneve²⁰, Thierry Leblanc²², Marion Marchand¹³,
- 10 Kenneth R. Minschwaner²³, Gary Morris²⁴, M<u>ichaelike</u> J. Newchurch²⁵, Shin-Ya Ogino²⁶,
- 11 Nozomu Ohkawara²⁷, Ankie J. M. Piters⁴, Françoise Posny²⁸, Richard Querel²⁹, Rinus Scheele⁴,
- 12 Frank -J. Schmidlin³, Russell C. Schnell¹⁴, Otto Schrems¹⁹, Henry Selkirk³⁰, Masato Shiotani³¹,
- 13 Pavla Skrivánková³², René Stübi⁶⁵, Ghassan Taha³⁰, David W. Tarasick³³, Anne M. Thompson³,
- 14 Valérie Thouret³⁴, Matt Tully³⁵, Roeland van Malderen¹⁰⁹, Geraint Vaughan³⁶, Holger Vömel³⁶³⁷,
- 15 Peter von der Gathen³⁷³⁸, Jacquelyn C. Witte³⁸³⁹, Margarita Yela³⁹⁴⁰
- 16 1. Harvard-Smithsonian Center for Astrophysics, Cambridge, MA, USA
- Department of Atmospheric and Oceanic Science, University of Maryland, College Park,
 Maryland, USA
- 19 3. NASA Goddard Space Flight Center, Greenbelt, Maryland, USA
- 20 <u>4.</u> Royal Netherlands Meteorological Institute (KNMI), De Bilt, the Netherlands
- 21 4.5. LATMOS-ISPL, Université Paris 6 Pierre-et-Marie-Curie, Paris, France
- 5.6. MeteoSwiss Aerological Station, Federal Office of Meteorology and Climatology
 MeteoSwiss, Payerne, Switzerland
- 24 6.7. South African Weather Service, Pretoria, South Africa
- 7.8. Izana Atmospheric Research Center, Meteorological State Agency of Spain, Santa Cruz de
 Tenerife, Spain
- 27 8.9. National Meteorological Service, Ushuaia, Tierra del Fuego, Argentina
- 28 9-10. Royal Meteorological Institute of Belgium, Brussel, Belgium
- 29 <u>10.11.</u> Los Alamos National Laboratory, Los Alamos, NM, USA

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- 30 HI.12. Earth, Ocean and Atmospheric Sciences, Florida State University, Tallahassee, FL, USA
- 31 <u>12.13.</u> Faculty of Environmental Earth Science, Hokkaido University, Sapporo, Japan
- 32 13. LATMOS-ISPL, Université Paris 6 Pierre et Marie Curie, Paris, France
- 33 14. NOAA/ESRL Global Monitoring Division, Boulder, CO, USA
- 34 15. Atmospheric Sciences Research Center, SUNY University at Albany, Albany, NY, USA
- 35 16. Finnish Meteorological Institute, Helsinki, Finland
- 17. The Institute of Meteorology and Water Management, National Research Institute, Warsaw,
 Poland
- 38 18. Indonesian Institute of Aeronautics and Space (LAPAN), Bandung, Indonesia
- 39 19. Alfred Wegener Institute for Polar and Marine Research, Bremerhaven, Germany
- 40 20. Earth Observation Satellite Images Applications Lab (EOSIAL), Università di Roma 'La
- 41 Sapienza', Rome, Italy
- 42 21. Danish Meteorological Institute, Copenhagen, Denmark
- 43 22. Jet Propulsion Laboratory, California Institute of Technology, Pasadena, CA, USA
- 44 23. Department of Physics, New Mexico Institute of Mining and Technology, Socorro, NM,
- 45 USA
- 46 24. St. Edward's University, Austin, TX, USA
- 47 25. Department of Atmospheric Science, University of Alabama in Huntsville, Huntsville, AL,
 48 USA
- 26. Department of Coupled Ocean-Atmosphere-Land Processes Research, Japan Agency for
 Marine-Earth Science and Technology, Yokosuka, Japan
- 51 27. Global Environment and Marine Department, Japan Meteorological Agency, Tokyo, Japan
- 52 28. Université de la Réunion, Saint Denis, France
- 53 29. National Institute of Water and Atmospheric Research, Lauder, Central Otago, New Zealand
- 54 30. Universities Space Research Association, Greenbelt, MD, USA
- 55 31. Research Institute for Sustainable Humanosphere, Kyoto University, Kyoto, Japan
- 32. Upper Air and Surface Observation Department, Czech Hydrometeorological Institute,
 Praha, Czech Republic
- 33. Air Quality Research Division, Environment & Climate Change Canada, Downsview, ON,
 Canada.

- 60 34. Laboratoire d'Aerologie, Université de Toulouse, Toulouse, France
- 61 35. Observations & Infrastructure Division, Bureau of Meteorology, Melbourne, Victoria,
 62 Australia
- 63 36. School of Earth, Atmosphere and Environmental Sciences, University of Manchester,
 64 Manchester, U.K.
- 65 <u>37.36.</u> Earth Observing Laboratory, National Center for Atmospheric Research, Boulder, CO,
 66 USA
- 67 <u>38.37.</u> Alfred Wegener Institute, Potsdam, Germany
- 68 <u>39-38.</u> Science Systems and Applications Inc. Greenbelt, MD, USA
- 69 40:39. Atmospheric Research and Instrumentation Branch, National Institute for Aerospace
- 70 Technology (INTA), Madrid, Spain
- 71 *Correspondence to: Guanyu Huang (guanyu.huang@cfa.harvard.edu)

72 Abstract

73 We validate the Ozone Monitoring Instrument (OMI) ozone-profile (PROFOZ) product from 74 October 2004 through December 2014 retrieved by the Smithsonian Astrophysical Observatory 75 (SAO) algorithm against ozonesonde observations. We also evaluate the effects of OMI Row 76 anomaly (RA) on the retrieval by dividing the data set into before and after the occurrence of 77 serious OMI RA, i.e., pre-RA (2004-2008) and post-RA (2009-2014). The retrieval shows good 78 agreement with ozonesondes in the tropics and mid-latitudes and for pressure < 50 hPa in the 79 high latitudes. It demonstrates clear improvement over the a priori down to the lower troposphere in the tropics and down to an average of ~550 (300) hPa at middle (high latitudes). In the tropics 80 81 and mid-latitudes, the profile mean biases (MBs) are less than 6%, and the standard deviations 82 (SDs) range from 5-10% for pressure $< \sim 50$ hPa to less than 18% (27%) in the tropics (mid-83 latitudes) for pressure > -50 hPa after applying OMI averaging kernels to ozonesonde data. The 84 MBs of the stratospheric ozone column (SOC, the ozone column from the tropopause pressure to 85 the ozonesonde burst pressure) are within 2% with SDs of < 5% and the MBs of the tropospheric ozone column (TOC) are within 6% with SDs of 15%. In the high latitudes, the profile MBs are 86 87 within 10% with SDs of 5-15% for pressure < ~50 hPa, but increase to 30% with SDs as great as 88 40% for pressure $> \sim$ 50 hPa. The SOC MBs increase up to 3% with SDs as great as 6% and the 89 TOC SDs increase up to 30%. The comparison generally degrades at larger solar-zenith angles 90 (SZA) due to weaker signals and additional sources of error, leading to worse performance at 91 high latitudes and during the mid-latitude winter. Agreement also degrades with increasing 92 cloudiness for pressure $> \sim 100$ hPa and varies with cross-track position, especially with large 93 MBs and SDs at extreme off-nadir positions. In the tropics and mid-latitudes, the post-RA 94 comparison is considerably worse with larger SDs reaching 2% in the stratosphere and 8% in the 95 troposphere and up to 6% in TOC. There are systematic differences that vary with latitude 96 compared to the pre-RA comparison. The retrieval comparison demonstrates good long-term 97 stability during the pre-RA period, but exhibits a statistically significant trend of 0.14-0.7%/year 98 for pressure < ~ 80 hPa, 0.7 DU/year in SOC and -0.33 DU/year in TOC during the post-RA 99 period. The spatiotemporal variation of retrieval performance suggests the need to improve 100 OMI's radiometric calibration especially during the post-RA period to maintain the long-term 101 stability and reduce the latitude/season/SZA and cross-track dependence of retrieval quality.

102 **1** Introduction

103 The Dutch-Finnish built Ozone Monitoring Instrument (OMI) on board the NASA Aura satellite 104 has been making useful measurements of trace gases including ozone and aerosols since October 105 2004. There are various retrieval algorithms to retrieve ozone profile and/or total ozone from 106 OMI data (Bak et al., 2015), including two independent operational total ozone algorithms 107 (Bhartia and Wellemeyer, 2002; Veefkind et al., 2006) and two ozone profile algorithms. Of the 108 two ozone profile algorithms, one is the operational algorithm (OMO3PR) developed at KNMI 109 (van Oss et al., 2001), and the other one is a research algorithm developed at Smithsonian 110 Astrophysical Observatory (SAO) by (Liu et al., 2010b). Both algorithms retrieve ozone profile 111 from the spectral region 270-330 nm using the optimal estimation method, but they differ 112 significantly in implementation details including radiometric calibration, radiative transfer model 113 simulation, a priori constraint, retrieval grids, and additional retrieval parameters. The SAO 114 ozone profile retrieval algorithm was initially developed at the Smithsonian Astrophysical 115 Observatory (Liu et al., 2005)-for Global Ozone Monitoring Experiment (GOME) data and was 116 adapted to OMI data (Liu et al., 2010b). Total ozone column (OC), Stratospheric Ozone Column 117 (SOC) and Tropospheric Ozone Column (TOC) can be directly derived from the retrieved ozone 118 profile with retrieval errors in-in the range of a few Dobson Units (DU) (Liu et al., 2006b; Liu et 119 al., 2010a). This algorithm has been put into production in the OMI Science Investigator-led 120 Processing System (SIPS), processing the entire OMI data record with approximately one-month 121 delay. The ozone profile product titled PROFOZ is publicly available at the Aura Validation 122 Data Center (AVDC) (http://avdc.gsfc.nasa.gov/index.php?site=2045907950). This long-term 123 ozone profile product, with high spatial resolution and daily global coverage, constitutes a useful 124 dataset to study the spatial and temporal distribution of ozone. 125 To effectively use the retrieval dataset, it is necessary to evaluate and understand its retrieval 126 quality and long-term performance. Although validation of the ozone profile product (mostly 127 earlier versions) has been partially performed against aircraft, ozonesonde, and Microwave Limb 128 Sounder (MLS) data, these evaluations are limited to certain time periods and/or spatial region 129 and/or to only portion of the product (e.g., total ozone columns (OC) or TOC only) (Bak et al., 130 2013a; Hayashida et al., 2015; Lal et al., 2013; Liu et al., 2010a; Liu et al., 2010b; Pittman et al., 131 2009; Sellitto et al., 2011; Wang et al., 2011; Yang et al., 2007; Ziemke et al., 2014).

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132 Additionally, the quality of ozone profile retrievals is very sensitive to the signal to noise ratio 133 (SNR) of the radiance measurements as well as their radiometric calibration, which may degrade 134 over time as shown in GOME and GOME-2 retrievals (Cai et al., 2012; Liu et al., 2007). 135 Although OMI's optical degradation is remarkably small to within 1-2% over the years, the SNR 136 and the number of good spectral pixels (not flagged as bad/hot pixels) have been gradually 137 decreasing over the years due to the expected CCD degradation (Claas, 2014). Furthermore, the 138 occurrence of RA, which affects level 1b data at all wavelengths for particular viewing directions 139 or cross-track positions and likely due to blocking objects in the optical path, started in June 140 2007 affecting a few positions. This effect abruptly worsened in January 2009 affecting $\sim 1/3$ of 141 the cross-track positions (Kroon et al., 2011). The impacts of RA not only evolve with time but 142 also vary over the duration of an orbit. Analysis indicates that radiances in the UV1 channels 143 (shorter than ~310 nm) used in our retrievals might have been affected at all positions (Personal 144 communication with S. Marchenko) and are not adequately flagged for RA. Therefore, we need 145 to evaluate the impacts of instrument degradation and especially row anomaly on the temporal performance of our ozone profile product. Currently, we are planning an update of the ozone 146 147 profile algorithm to maintain the long-term consistency of the product. The update will include 148 empirical correction of systematic errors caused by the instrument degradation and row anomaly 149 as a function of time. Such correction also requires us to evaluate the long-term retrieval quality 150 of our product.

151 To understand retrieval quality and the resulting spatial and temporal performance of our OMI 152 product, we evaluate our data from October 2004 through December 2014 against available 153 ozonesonde and MLS observations, respectively, in two papers. This paper evaluates our ozone 154 product including both ozone profiles and stratospheric and tropospheric ozone columns using 155 ozonesonde observations with a focus on retrieval quality in the troposphere. More than 27,000 156 ozonesonde profiles from both regular ozonesonde stations and field campaigns are used in this 157 study to provide a comprehensive and global assessment of the long-term quality of our OMI 158 ozone product. This paper is followed by the validation against collocated MLS data with a focus 159 on the retrieval quality in the stratosphere (Huang et al., 2017), also submitted to this special 160 issue).

161 This paper is organized as follows: Section 2 describes OMI retrievals and ozonesonde data. The 162 validation methodology is introduced in Section 3. Section 4 presents results, analysis and 163 discussions regarding the OMI and ozonesonde comparisons. Section 5 summarizes and 164 concludes this study.

165 2 OMI and Ozonesonde Datasets

166 2.1 OMI and OMI Ozone Profile Retrievals

OMI is a Dutch-Finnish built nadir-viewing pushbroom UV/visible instrument aboard the NASA 167 Earth Observing System (EOS) Aura satellite that was launched into a sun-synchronous orbit in 168 169 July 2004. It measures backscattered radiances in three channels covering the 270-500 nm 170 wavelength range (UV1: 270-310 nm, UV2: 310-365 nm, visible: 350-500 nm) at spectral 171 resolutions of 0.42-0.63 nm (Levelt et al., 2006). Measurements across the track are binned to 172 60 positions for UV2 and visible channels, 30 positions for the UV1 channels due to the weaker 173 signals. This results in daily global coverage with a nadir spatial resolution of 13 km \times 24 km 174 (along \times across track) for UV2 and visible channels, and 13 km \times 48 km for the UV1 channel.

175 The SAO OMI ozone profile algorithm was adapted from the GOME ozone profile algorithm 176 (Liu et al., 2005) to OMI and was initially described in detail in Liu et al. (2010b). Profiles of 177 partial ozone columns are retrieved at 24 layers, ~2.5 km for each layer, from the surface to ~60 178 km using OMI radiance spectra in the spectral region 270-330 nm with the optimal estimation 179 technique. In addition to the OC, SOC and TOC can be directly derived from the retrieved ozone 180 profile with the use of tropopause (defined based on the lapse rate) from the daily National 181 Center for Environmental Protection (NCEP) reanalysis data. The retrievals are constrained with 182 month- and latitude-dependent climatological a priori profiles derived from 15-year ozonesonde 183 and SAGE/MLS data (McPeters et al., 2007) with considerations of OMI random-noise errors. 184 OMI radiances are pre-calibrated based on two days of average radiance differences in the 185 tropics between OMI observations and simulations with zonal mean MLS data for pressure less 186 than 215 hPa and climatological ozone profile for pressure greater than 215 hPa. This "soft 187 calibration" varies with wavelength and cross-track positions but does not depend on space and 188 time.

189 The updated-current algorithm of our SAO OMI ozone product that is used in this paper was 190 briefly described in Kim et al. (2013). The radiative transfer calculations have been improved 191 through the convolution of simulated radiance spectra at high resolutions rather than effective 192 cross sections, which is done by interpolation from calculation at selected wavelengths assisted 193 by weighting function. In addition, four spatial pixels along the track are coadded to speed up 194 production processes at a nadir spatial resolution of 52 km \times 48 km. Meanwhile, minimum 195 measurement errors of 0.4% and 0.2% are imposed in the spectral ranges 270-300 nm and 300-196 330 nm, respectively, to stabilize the retrievals. The use of floor errors typically reduces the 197 Degree of Freedom for Signals (DFS) and increases retrieval errors. Compared to the initial 198 retrievals, the average total, stratospheric, and tropospheric DFS decrease by 0.49, 0.27, and 199 0.22, respectively, and the mean retrieval errors in OC, SOC, and TOC increase by 0.6, 0.5, and 200 1.2 DU, respectively. The corresponding changes to the retrievals are generally within retrieval 201 uncertainties except for a systematic increase in tropospheric ozone at SZA larger than ~75°, 202 where the TOC increases to ~12 DU. Validation against ozonesonde data indicates that this TOC 203 increase at large SZA makes the retrieval worse. Therefore retrieved tropospheric ozone at such 204 large SZA should not be used, but the retrieved total ozone still shows good quality (Bak et al., 205 2015).

206 For current products, retrievals contain ~5.5-7.4 DFS, with 4.6-7.3 in the stratosphere and 0-1.2 207 in the troposphere. Vertical resolution varies generally from 7-11 km in the stratosphere to 10-208 14 km in the troposphere, when there is adequate retrieval sensitivity to the tropospheric ozone. Retrieval random-noise errors (i.e., precisions) typically range from 0.6–2.5 % in the middle 209 210 stratosphere to approximately 12% in the lower stratosphere and troposphere. The solution 211 errors, dominated by smoothing errors, vary generally from 1-7% in the middle stratosphere to 7-212 38% in the troposphere. The solution errors in the integrated OC, SOC, and TOC are typically in 213 the few DU range. Errors caused by the forward model and forward model parameter 214 assumptions are generally much smaller than the smoothing error (Liu et al., 2005). The main 215 sources of these errors include systematic errors in temperature and cloud-top pressure. 216 Systematic measurement errors are the most difficult to estimate, mostly due to lack of full 217 understanding of the OMI instrument calibration.

218 Certain cross track positions in OMI data have been affected by RA since June 2007 (Kroon et 219 al., 2011). Loose thermal insulating material in front of the instrument's entrance slit is believed 220 to block and scatter light, causing measurement error. The anomaly affects radiance 221 measurements at all wavelengths for specific cross-track viewing directions that are imaged to 222 CCD rows. Initially, the anomaly only affected a few rows. But since January 2009, the anomaly 223 has spread to other rows and shifted with time. The RA also shows slight differences among 224 different spectral channels, and varies during the duration of an orbit. Pixels affected by the RA 225 are flagged in the level 1b data. The science team suggested that they are not be used in research. 226 For data before 2009, the RA flagging is not applied in the processing. Pixels seriously affected 227 by RA will typically show enhanced fitting residuals. The algorithm was updated to use RA 228 flagging in the UV1 channel and was used to process the data starting from 2009. If a pixel is 229 flagged as a row anomaly then it is subsequently not retrieved to speed up the processing except 230 that the cross-track position 24 is still retrieved due to reasonably good fitting. It should be noted 231 that the retrieval quality of those non-flagged pixels may still be affected by the RA, because of 232 the different RA flagging in the UV1 and UV2, the lack of RA flagging before 2009_and 233 inadequacy of the RA flagging.

234 To screen out OMI profiles for validation, we only use OMI ozone profiles meeting the 235 following criteria based on three filtering parameters: 1) nearly clear-sky scenes with effective 236 cloud fraction less than 0.3; 2) cross track positions between 4 and 27, due to the relatively worse 237 quality and much larger footprint size of the off-nadir pixels beyond this rangefor these greater 238 off nadir positions; 3) SZA should be less than 75° due to very limited retrieval sensitivity to 239 tropospheric ozone and the aforementioned positive biases. The selection and justification of 240 these criteria will be discussed in Sects. 2.1.2-4.1.4, in which wWe will use all OMI pixels of 241 each filtering parameter when evaluating retrieval quality as a function of that specific 242 parameter. The fitting quality of each retrieval is shown in the fitting RMS (root mean square of 243 the fitting residuals relative to the assumed measurement errors). The mean fitting RMS 244 including both UV1 and UV2 channels has been increasing with time as shown in Figure 1. This 245 is primarily due to the increase of fitting residuals in UV1 caused by the instrument degradation 246 and RA since the fitting residuals of UV2 only slightly increase with time. As aforementioned, 247 the retrieval information of stratospheric and tropospheric ozone mainly comes from UV1 and

248 UV2, respectively. Consequently, retrievals in the troposphere, the focus of this paper, are less 249 impacted by the increasing fitting RMS. However, to apply consistent filtering in validation 250 against both ozonesonde in this study and MLS data in the companion paper-<u>(Huang et al.,</u> 251 <u>2017)(Huang et al., 2016, submitted to the same special issue)</u>, we set the RMS threshold based 252 on the overall fitting RMS and select retrievals with fitting RMS smaller than the sum of 253 monthly mean RMS and its 2σ (i.e., Standard Deviations (SDs) of fitting RMS).

254 2.2 Ozonesondes

The balloon-borne ozonesonde is a well-established technique to observe the ozone profile from 255 256 the surface to ~35 km with vertical resolution of ~100-150 m and approximately 3-5% precision 257 and 5-10% accuracy (Deshler et al., 2008; Johnson, 2002; Komhyr, 1986; Komhyr et al., 1995; 258 Smit et al., 2007). Ozonesonde data have been widely used in the studies of stratospheric ozone, climate change, tropospheric ozone and air quality, as well as the validation of satellite 259 260 observations (Huang et al., 2015; Kivi et al., 2007; Thompson et al., 2015; Wang et al., 2011). 261 However, the accuracy of ozonesonde observations depends on data processing technique, sensor 262 solution, and instrument type and other factors. Consequently, station-to-station biases may 263 occur in ozonesonde measurements and could be as great as 10% (Thompson et al., 2007c; 264 Worden et al., 2007).

265 A decade (2004-2014) of global ozonesonde data with locations shown in Figure 2, are utilized 266 in this study to validate our OMI ozone profile product. Most of our ozonesonde data were 267 obtained from the Aura Validation Data Center (AVDC) archive. It contains routine launches 268 from ozonesonde stations, mostly weekly and occasionally 2-3 times a week at some stations. It 269 also collects launches from field campaigns, for instance, IONS 06 (INTEX-B Ozone Network 270 2006), ARCIONS (Arctic Intensive Study Ozonesonde Network Study) 271 (http://croc.gsfc.nasa.gov/arcions/) (Tarasick et al., 2010; Thompson et al., 2008). Data not 272 available at AVDC are obtained from other archives such as the World Ozone and Ultraviolet 273 Radiation Data Center (WOUDC) (http://woudc.org/), the Southern Hemisphere Additional 274 Ozonesondes (SHADOZ) (Thompson et al., 2007a; Thompson et al., 2007b), as well as archives 275 of recent field campaigns including DISCOVER-AQ (Deriving Information on Surface 276 Conditions from Column and Vertically Resolved Observations Relevant to Air Quality,

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http://discover-ag.larc.nasa.gov/) (Thompson et al., 2015) and SEACR⁴S (Studies of Emissions 277 278 and Atmospheric Composition, Clouds and Climate Coupling by Regional Surveys, 279 https://espo.nasa.gov/home/seac4rs) (Toon et al., 2016). Almost all of the ozonesonde data in 280 this study were obtained from electrochemical concentration cell (ECC) ozonesondes, which is 281 based on the oxidation reaction of ozone with potassium iodide (KI) in solution. The exceptions 282 are Hohenpeissenberg station in Germany that uses Brewer-Mast (BM) ozonesondes, the New 283 Delhi, Poona, and Trivandrum stations that use Indian ozonesondes, and four Japanese stations 284 (i.e., Sapporo, Tsukuba, Naha and Syowa) that switched from KC ozonesondes to ECC 285 ozonesondes during late 2008 and early 2010. These types of ozonesondes have been reported to 286 have larger uncertainties than ECC ozonesondes (Hassler et al., 2014; Liu et al., 2013; WMO, 287 1998).

288 To avoid using anomalous profiles, we screen out ozonesondes that burst at pressure exceeding 289 200 hPa, ozone profiles with gaps greater than 3 km, more than 80 DU TOC or less than 100 DU 290 SOC. In the SOC comparison, we also filter measurements that do not reach 12 hPa. Some 291 ozonesonde data used in this paper (e.g. WOUDC data) are provided with a correction factor 292 (CF) derived by normalizing the integrated ozone column (appended with ozone climatology 293 above burst altitude) to the coincident total ozone column measured by a Dobson or Brewer 294 instrument to account for uncertainties mainly from the in pump efficiency especially near the 295 top of the profiles. The CF is also included in our screening processes. If the CF is available, we select ozonesonde profiles with the CF in the range of 0.85 to 1.15 to filter profiles that require 296 297 too much correction, and apply the correction. Finally, a small number of obviously erroneous profiles are visually examined and rejected. 298

299 **3** Comparison Methodology

Previous studies on the validation of satellite observations used a range of coincidence criteria. Wang et al. (2011) set a 100 km radius and 3 hour time difference as coincidence criteria. Kroon et al. (2011) applied coincidence criteria of $\pm 0.5^{\circ}$ for both latitude and longitude and 12 hours. In this paper, we determine our coincident criteria based on the balance between finding most coincident OMI/ozonesonde pairs to minimize differences due to spatiotemporal samplings and finding a sufficient number of pairs for statistical analysis. For each screened ozonesonde profile, 306 we first select all filtered OMI data within $\pm 1^{\circ}$ latitude, $\pm 3^{\circ}$ longitude and ± 6 hours and then 307 find the nearest OMI retrieval within 100 km from the ozonesonde station to perform the 308 validation on the individual profile basis.

309 Ozonesondes have much finer vertical resolution than OMI retrievals. To account for the 310 different resolutions, ozonesonde profiles are first integrated into the corresponding OMI vertical 311 grids and then degraded to the OMI vertical resolution by using the OMI retrieval Averaging 312 Kernels (AKs) and *a priori* ozone profile based on the following equation:

$$\hat{x} = x_a + A(x - x_a), \tag{1}$$

314 where x is the ozonesonde profile integrated into the OMI grid, \hat{x} is the retrieved ozone profile if 315 the ozonesonde is observed by OMI, A is the OMI AK matrix, and x_a is the OMI *a priori* ozone profile. We refer to this retrieval as "convolved ozonesonde profile", which is a reconstruction of 316 317 ozonesonde profile with OMI retrieval vertical resolution and sensitivity. Missing ozone profiles 318 above ozonesonde burst altitude are filled with OMI retrievals. The convolution process 319 essentially removes OMI smoothing errors and the impacts of a priori from the comparison so 320 that OMI/ozonesonde differences are mainly due to OMI/ozonesonde measurement precision, 321 spatiotemporal sampling differences and other errors. However, in the regions and altitudes 322 where OMI has low retrieval sensitivity, the comparisons can show good agreement because 323 both the retrieval and convolved ozonesonde approach the a priori profile. To overcome the 324 limitation of such a comparison, we also compare with unconvolved ozonesonde profiles since it 325 indicates how well the retrievals can represent the actual ozonesonde observations (i.e., 326 smoothing errors are included as part of retrieval errors). In addition, we also compare OMI a 327 priori and convolved/unconvolved ozonesonde profiles to indicate the retrieval improvement 328 over the a priori.

For consistent calculations of TOC and SOC from the OMI/ozonesonde data, the tropopause pressure included in the OMI retrieval and ozonesonde burst pressure (required to be less than 12 hPa or <u>above</u>~30 km) are used as the proper boundaries. The TOC is integrated from the surface to the tropopause. <u>Aand the SOC is not the total stratospheric ozone column, but the ozone</u> <u>column</u> integrated from the tropopause pressure to the ozonesonde burst pressure. 334 The relative profile difference is calculated as (OMI- Sonde) / OMI a priori ×100% in the present 335 comparison with ozonesonde and with MLS in the companion paper. Choosing OMI a priori 336 rather than MLS/ozonesonde is to avoid unrealistic statistics skewed by extremely small values 337 in the reference data especially in the MLS retrievals of upper troposphere and lower 338 stratosphere ozone (Liu et al., 2010a). Unlike the profile comparison, ozonesonde/OMI 339 SOC/TOC values are used in the denominator in the computation of relative difference. To 340 exclude remaining extreme outliers in the comparison statistics, values that are exceeding 3σ 341 from the mean differences are filtered.

After applying the OMI/ozone filtering and coincident criteria, approximately 10,500 342 343 ozonesonde profiles are used in the validation. We performed the comparison for five latitude bands: northern high latitudes (60° N-90° N), northern mid-latitudes (30° N-60° N), tropics (30° 344 S-30° N), southern mid-latitudes (60° S- 30° S), and southern high latitudes (90° S- 60° S) to 345 346 understand the latitudinal variation of the retrieval performance. We investigated the seasonal 347 variations of the comparisons mainly at northern mid-latitudes where ozone retrieval shows distinct seasonality and there are adequate coincidence pairs. To investigate the RA impacts on 348 349 OMI retrievals, we contrasted the comparison before (2004-2008, i.e., pre-RA) and after (2009-350 2014, i.e., post-RA). Although we filter OMI data based on cloud fraction, cross-track position, 351 and SZA in the final evaluation of our retrievals against ozonesonde observations as shown in 352 Sect. 4.1.1., we conduct the comparison as a function of these parameters using coincidences at 353 all latitude bands to show how these parameters affect the retrieval quality as shown in the Sects. 354 4.1.2 - 4.1.4. In these evaluations, the filtering of OMI data based on cloud fraction, cross-track 355 position, and SZA are switched off, respectively. Approximately 15,000 additional ozonesonde profiles are used in this extended evaluation. To evaluate the long-term performance of our 356 357 ozone profile retrievals, we analyze the monthly mean biases (MBs) of the OMI/ozonesonde 358 differences as a function of time using coincidences in the 60° S-60° N region and then derive a 359 linear trends over the entire period as well as the pre-RA and post-RA periods.

360 4 Results and Discussions

361 4.1 Comparison of Ozonesonde and OMI profiles

362 4.1.1 Ozone Profile Differences

Comparisons of ozone profiles between OMI/a priori and ozonesondes with and without 363 364 applying OMI AKs for the 10-year period (2004-2014) are shown in the left panels of Figure 3. 365 The MBs and SDs vary spatially with altitude and latitude. Vertically, the SD typically 366 maximizes in the upper troposphere and lower troposphere stratosphere (UTLS) in all latitude bands due to significant ozone variability and a priori uncertainty. Bak et al. (2013b) showed that 367 368 the use of Tropopause-Based (TB) ozone profile climatology with NCEP Global Forecast 369 System (GFS) daily tropopause pressure can significantly improve the a priori, and eventually 370 reduce the retrieval uncertainty. Consequently, the SDs of OMI/sonde differences in the UTLS at 371 mid- and high-latitudes can be reduced through reducing the retrieval uncertainties in a future 372 version of the algorithm that uses the TB climatology. Latitudinally, the agreement is better in 373 the tropics and becomes worse at higher latitudes. The patterns are generally similar in the 374 northern and southern hemispheres. The MBs between OMI and ozonesonde are within ~6% 375 with AKs and 10% without AKs in the tropics and the middle latitudes. Large changes in the 376 biases between with and without AKs occur in the tropical troposphere where the bias 377 differences reach 10%. The MBs increase to 20-30% at high latitudes consistently with large 378 oscillation from $\sim 20-30\%$ at ~ 300 hPa to +20% near the surface both with and without the 379 application of AKs. At pressure < 50 hPa, the SDs for comparisons with OMI AKs are typically 5-10% at all latitudes except for the 90° S-60° S region. For pressure > 50 hPa, the SDs are 380 381 within 18% and 27% in the tropics and middle-latitudes, respectively, but increase to 40% at 382 higher latitudes. The SDs for comparison without applying OMI AKs, i.e., including OMI 383 smoothing errors in the OMI/ozonesonde differences, typically increase up to 5% for pressure < 50 hPa, but increase up to 15-20% for pressure $> \sim$ 50hPa. The smoothing errors derived from 384 root square differences of the MBs with and without OMI AKs are generally consistent with the 385 386 retrieval estimate from the optimal estimation.

387 The improvements of OMI over the climatological (a priori) profiles can be reflected in the 388 reduction of MBs and SDs in the comparisons between ozonesondes and OMI retrievals, and 389 between ozonesondes and a priori. The retrieval improvements in the MBs are clearly shown in 390 the tropics and at ~ 100 hPa pressure in the middle latitudes. At high latitudes, the MBs and 391 corresponding oscillations in the troposphere are much larger than these in the a priori 392 comparison, suggesting that these large biases are mainly caused by other systematic 393 measurements errors at high latitudes (larger SZAs and thus weaker signals). As can be seen from the reduction of SDs, OMI retrievals show clear improvements over the a priori at pressure 394 395 < 300 hPa. For pressure > 300 hPa, the retrieval improvements vary with latitudes. There are 396 consistent retrieval improvements throughout the surface - 300hPa layer in the tropics and only 397 the 550 - 300 hPa layer at middle latitude, while there is no retrieval improvement over the a 398 priori for > 300 hPa at high latitudes. The failure to improve the retrieval over a priori in part of the troposphere at middle and high latitudes is caused by several factors. They are the inherent 399 400 reduction in retrieval sensitivity to lower altitudes at larger SZAs as a result of reduced photon 401 penetration into the atmosphere, unrealized retrieval sensitivity arising from retrieval 402 interferences with other parameters (e.g., surface albedo) as discussed in Liu et al. (2010b) and 403 the use of floor-noise of 0.2% that underestimates the actual OMI measurement SNR. In 404 addition, the a priori ozone error in the climatology is quite small since the SDs of the 405 differences between the a priori and ozonesonde without AKs are typically less than 20% in the lower troposphere for middle and high latitudes, which also makes it more difficult to improve 406 407 over the a priori comparison.

408 The right column of Figure 3 shows the comparisons between OMI retrievals and ozonesondes 409 convolved with OMI AKs in the pre-RA and post-RA periods, respectively. In the tropics and 410 mid-latitudes, the pre-RA comparison is better than the post-RA comparison, with SDs smaller 411 by up to ~8% at most altitudes especially in the troposphere. The pre-RA comparison also shows smaller biases near ~300 hPa at middle latitudes while the post-RA comparison exhibits negative 412 413 biases reaching 8-12%. At high latitudes, the pre-RA period does not show persistent 414 improvement during the post-RA period. The pre-RA comparison shows slightly smaller SDs at 415 most altitudes and smaller negative biases by 10% around 300 hPa in the northern high latitudes, 416 and smaller positive biases by 20% near the surface in the southern high latitudes. The worse

417 results during the post-RA period are caused by increasingly noisy OMI measurements with 418 smaller SNR and the additional radiometric biases made by the RA, which vary with space and 419 time. The smaller SDs at some altitudes of high latitudes may reflect a combination of ozone 420 variation, uneven distribution of ozonesondes with varying uncertainty at different stations, and 421 cancellation of radiometric errors by the RA.

422 As seen from the number of OMI/ozonesonde coincidences shown in Figure 3, the northern mid-423 latitudes and the tropics have sufficient coincidences to validate the retrievals as a function of 424 season. In the tropics, the retrieval comparison does exhibit little seasonality as expected (not 425 shown). Figure 4 shows the comparison similar to Figure 3(c) for each individual season at 426 northern middle latitudes. The comparison results are clearly season-dependent with different 427 altitude-dependent bias patterns, best agreement and with the smallest SDs -in the summer 428 (except for the MBs) and the worst SDsagreement-in the winter. This indicates the general best 429 retrieval sensitivity to lower tropospheric ozone during the summer as a result of small SZAs and 430 stronger signals and worst retrieval sensitivity during the winter as a result of large SZAs and 431 weaker signals. The MBs for with and without AKs at 300 hPa vary from ~12% in the winter to -432 10% in the summer. The overall MBs are the smallest during the spring, within 6%; but the MBs 433 at pressure < 50 hPa are the best during the summer. The maximum SDs vary from 31% in the 434 winter to 20% in the summer. Also, the retrieval in the summer shows the most improvements in 435 terms of reduction in SDs -over the a priori in the lower troposphere at all tropospheric layers 436 except for the bottom layer, while the retrievals during other seasons show the improvement over 437 a priori only above the lowermost two/three layers. The seasonal variation of retrieval quality is partially caused by the seasonal variations of the retrieval sensitivity and ozone variability. Bak 438 439 et al. (2013b) showed that the use of TB ozone climatology with daily NCEP GFS tropopause 440 pressure can significantly reduce the seasonal dependence of the comparison with ozonesondes. 441 In addition, radiometric calibration errors such as those caused by stray light and RA also 442 contribute to the seasonal variation of retrieval quality.

443 4.1.2 Solar Zenith Angle Dependence

444 The SZA of low earth orbit (LEO) satellite observation varies latitudinally and seasonally; 445 therefore the SZA dependence of the retrieval can cause latitudinal and seasonal dependent retrieval biases. SZA is one of the main drivers that affect retrieval sensitivity especially to tropospheric ozone. At large SZA, the measured backscattered signal becomes weak due to weak incoming signal and long path length; the retrieval sensitivity to the tropospheric ozone decreases due to reduced photon penetration to the troposphere. In addition, measurements are subject to relatively larger radiometric errors such as those from stray light and as a result of weaker signal, and radiative transfer calculations can lose accuracy at larger SZA (Caudill et al., 1997).

453 Figure 5 gives the MBs and SDs of differences between OMI and ozonesondes (with OMI AKs) 454 in a function of SZAs. We can see that retrieval performance generally becomes worse at large 455 SZA. The SD typically increases with SZA especially at pressure > 300 hPa. At SZA larger than 456 75°, the SD at ~300 hPa increases to greater than ~45%. The variation of MBs with SZA is more 457 complicated. We see generally larger positive biases at larger SZA in the troposphere with > 458 20% biases at SZA larger than 75°. The MBs near \sim 30 hPa becomes more negative at larger 459 SZAs. There is a strip of positive biases of $\sim 10\%$ that slightly decreases in pressure from ~ 50 460 hPa at low SZA to ~10 hPa at large SZA; it might be due to some systematic radiometric biases 461 that can affect ozone at different altitudes varying with SZA. Because of the clear degradation of the retrieval quality at large SZA, we set the SZA filtering threshold of 75° to filter OMI data. 462

463 **4.1.3 Cloud Fraction Dependence**

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464 The presence of cloud affects retrieval sensitivity since clouds typically reduce sensitivity to 465 ozone below clouds and increase sensitivity to ozone above clouds. The accuracy of ozone 466 retrievals is sensitive to the uncertainties of cloud information and cloud treatment (Antón and 467 Loyola, 2011; Bak et al., 2015; Liu et al., 2010a). Our OMI ozone algorithm assumes clouds as 468 Lambertian surfaces with optical centroid cloud pressure from the OMI Raman cloud product 469 (Vasilkov et al., 2008), and partial clouds are modeled using independent pixel approximation 470 such that the overall radiance is the sum of clear and cloudy radiances weighted by the effective 471 cloud fraction. The cloud albedo is assumed to be 80% and is allowed to vary (>80%) with the 472 effective cloud fraction.

473 Figure 6 gives the influences of effective cloud fraction on the comparisons between OMI and

ozonesonde observations convolved with OMI AKs. The MBs and SDs do not change much with

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475 cloud fraction for pressure < 100 hPa, and typically increase with the increase of cloud fraction 476 for pressure > 100 hPa. The MBs at pressure > 100 hPa, especially greater~300 hPa, increase to 477 more than 10% with cloud fraction greater than ~0.3. This indicates that the cloud fractions have 478 small impacts on the stratospheric retrievals but large impacts on the tropospheric retrievals as 479 expected. Some of the variation with cloud fraction such as negative biases near ~300 hPa at 480 cloud fraction of ~0.4 and the decreases of positive biases at ~ 50 hPa for cloud fraction greater 481 than ~ 0.8 may be partially related to the uncertainties of the cloud parameters. The chosen 482 filtering threshold of 0.3 in cloud fraction is a tradeoff between validating OMI data with 483 adequate retrieval sensitivity to tropospheric ozone and finding adequate number of 484 OMI/ozonesonde coincidences.

485 4.1.4 Cross-Track Position Dependence

486 The OMI swath is divided into 30 cross-track pixels at the UV1 spatial resolution of our product. 487 Each cross-track position is measured by a different part of the CCD detector, i.e., essentially a 488 different instrument. Radiometric calibration coefficients of the instrument are characterized 489 during pre-launch only at selected CCD column pixels and then interpolated to other columns, 490 causing variation in the radiometric calibration performance across the CCD detector. This in turn causes cross-track dependent biases in the calibrated radiance (Liu et al., 2010b), which 491 492 therefore causes stripping in almost all the OMI data products if no de-stripping procedure is 493 applied. Our retrieval algorithm has included a first-order empirical correction independent of 494 space and time to remove the cross-track variability (Liu et al., 2010b). However, residual 495 dependence on cross-track position remains and the radiometric calibration at different position can degrade differently with time (e.g., the RA impact). In addition, the viewing zenith angle 496 497 ranges from $\sim 0^{\circ}$ to $\sim 70^{\circ}$ and the footprint area increases by approximately an order of magnitude 498 from nadir to the first/last position. So the varying viewing zenith angle causes the variation of 499 retrieval sensitivities and atmospheric variabilities within varying footprint areas may also cause 500 additional cross-track dependence in the retrieval performance.

Figure 7 provides the MBs and SDs of the differences between OMI and ozonesonde convolved with OMI AKs as a function of cross-track position for pre-RA and post-RA periods, respectively. It clearly exhibits cross-track dependence especially with large positive/negative 504 MBs and large SDs at the first/last several extreme off-nadir positions. This is why we select 505 cross-track positions of 4-27 in the validation to avoid positions with large biases. The enhanced 506 biases/SDs at positions 24 (RA flagging not applied) and 27 (flagged as RA in UV2 since June 507 25, 2007 but not flagged/applied in UV1) are due to the RA impact during the post-RA period. 508 Cross-track positions 1-10 show consistent bias patterns with negative biases in ~300- 50 hPa 509 layer and positive biases in ~surface - 300 hPa layer, and large standard deviation around ~ 300 510 hPa although the magnitude decreases with increasing cross-track position. This pattern occurs during both pre-RA and post-RA periods although the values are larger during the post-RA 511 512 period. For other cross-track positions, the variation is relatively smaller but we can still see 513 small striping patterns.

514 **4.2 Comparison of Partial Ozone Columns**

515 We investigate and validate OMI partial ozone columns, including SOCs, TOCs, and surface-516 550 hPa and surface-750 hPa ozone columns in this section. We define the lowermost one and 517 two layer as surface-750 hPa and surface-550 hPa in this paper, respectively, for conveniences. 518 Similarly, we also analyze the validation results of SOCs and TOCs during pre-RA and post-RA, 519 respectively, to test the impacts of RA on OMI partial ozone columns. In addition, we validate ozone columns from the surface to ~550 hPa (bottom two layers) and ~ 750 hPa (bottom one 520 layer) against ozonesonde observations in the tropics and mid-latitude summer where there is 521 522 better retrieval sensitivity to these quantities.

523 4.2.1 Comparison of Stratospheric Ozone Columns (SOCs)

524 The left column of Figure 8 shows the MBs and SDs of the comparisons of OMI and ozonesonde 525 SOCs for each of the five latitude bands during 2004-2014. In all regions, the OMI SOCs have 526 excellent agreement with ozonesonde SOCs regardless of whether ozonesonde data are 527 convolved with OMI AKs. The application of OMI AKs to ozonesonde SOCs only slightly 528 improves the comparison statistics. The MBs with OMI AKs are within 1.8% except for a 529 negative bias of 3% at northern high latitudes, while the SDs are within 5.1% except for 5.7% at high latitudes. The correlation coefficient is greater than 0.95 except for 0.90 in the tropics due to 530 the smaller SOC range. The SDs are typically larger than the comparisons with MLS data (Liu et 531

al., 2010a) due to worse coincidence criteria, relatively larger uncertainty in the ozonesondestratospheric ozone columns compared to MLS data, and different altitude ranges of integration.

The middle and right columns of Figure 8 show comparison results during the pre-RA and post-RA periods, respectively. The comparison is typically better during the pre-RA with SDs smaller by 0.2-0.6% and larger correlation coefficients although the MBs are generally smaller during the post-RA period. One exception is at southern high-latitudes where the post-RA comparison statistics are significantly better except for the MB, consistent with Figure 3, likely due to a combination of ozone variation between these two periods, uneven distribution of ozonesondes

540 at different stations, and cancellation of various calibration errors.

541 **4.2.2** Comparison of Partial Ozone Columns in the Troposphere

542 The left column of Figure 9 shows the comparison of OMI and ozonesonde (with and without 543 OMI AKs) TOCs for each of the five latitude bands during 2004-2014. Without applying OMI 544 AKs, the MBs are within 1-3% except for 9% at northern high latitudes; The SDs are within 20% in the tropics and mid-latitudes and increase to ~30-40% at high-latitudes. The correlation 545 546 coefficient ranges from 0.83 in the tropics to ~0.7 at middle latitudes, and 0.5-0.6 at high-547 latitudes. The linear regression slopes are in the range 0.6-0.8 typically smaller at high latitudes 548 due to reduced retrieval sensitivity to the lower troposphere. After applying the OMI AKs to 549 ozonesonde data to remove smoothing errors, we see significant improvement in the comparison 550 statistics except for MBs, which are within 6% at all latitudes. The SDs are reduced to within 15% in the tropics and middle latitudes and ~30% (5.5-8.1 DU) at high latitudes; the correlation 551 552 improves by 0.04-0.12 and the slope significantly increases by 0.12-0.23 to the range 0.8-1.0 at 553 different latitude bands due to accounting for inadequate retrieval sensitivity to the lower and 554 middle troposphere.

555 The middle and right columns of Figure 9 show comparisons during pre-RA and post-RA, 556 respectively. The comparison between OMI and ozonesondes with OMI AKs TOCs during the 557 pre-RA period is significantly better than these during the post-RA period in the tropics and mid-558 latitudes with SDs smaller by 3.4-5.5% and greater correlation. The MBs during the post-RA 559 period is smaller by ~2 DU at mid-latitudes, but larger by ~1 DU in the tropics. However, the 560 post-RA comparison is similar to the pre-RA comparison at northern high-latitudes and is even 561 better at southern high latitudes probably due to the aforementioned ozonesonde issues.

562 Figure 10 shows examples of time series when comparing individual OMI and ozonesondes (with OMI AKs) TOCs and their corresponding differences at six selected stations, one for each 563 latitude region of 90° N-60° N, 60° N-30° N, 30° N-0°, 0°-30° S, 30° S-60° S and 60° S-90° S. 564 565 OMI TOC shows good agreement with ozonesondes at these stations with overall MBs \leq 3 DU 566 and SDs less than 5.1 DU. The comparison is also good even in the high latitude regions partially 567 because the Summit and Neymayer stations only have ozonesonde launches during local 568 summer. Seasonal dependent biases are clearly seen at Payerne, and bias trends can be seen at 569 several stations with positive trends at Summit and Neumayer and a negative trend at Naha. In 570 the pre-RA and post-RA periods, the MBs are typically within 2 DU and the SDs are typically 571 smaller during the pre-RA period except for Naha. The better comparison (both mean bias and 572 standard deviation) during the post-RA period at Naha is likely due to the switch to ECC 573 ozonesondes beginning on November 13, 2008 from KC ozonesonde that have greater 574 uncertainty (WMO, 1998).

Figure 2 also shows the MBs and SDs of the TOC differences between OMI and ozonesonde 575 576 convolved with OMI AKs at each station/location where there are at least 10 coincident 577 OMI/ozonesonde pairs. OMI data generally exhibit good agreement with ozonesondes at most of 578 the stations, with MBs of ≤ 3 DU and SDs of ≤ 6 DU. In the tropics (30° S-30° N), very large 579 SDs (>11 DU) occur at the two Indian stations (New Delhi, and Trivandrum). In addition, there 580 is a large bias of > 6 DU at New Delhi. The large bias of >6 DU at New Delhi is likely 581 associated with the large uncertainties of the Indian ozonesonde data. The poor comparisons at 582 these two stations are likely associated with the large uncertainties of the Indian ozonesonde 583 data. Hilo has large biases of ~4.5 DU with 3.2 and 6.2 DU for pre-RA and post-RA, 584 respectively. Java also has a large bias of ~5 DU but shows no much-little difference between 585 pre-RA and post-RA. Consistent ~2% and ~5% underestimates of OC by ozonesondes compared 586 to OMI total ozone are found in Hilo and Java, respectively (Thompson et al., 2012). These OC 587 underestimates may partly explain the large TOC biases in Hilo and Java. However, the reason 588 for underestimates of ozonesonde-derived OC is unknown. In the middle latitudes, noticeably 589 large SDs and/or biases occur at a few stations such as Churchill, Sable Islands,

590 Hohenpeissenberg, and Parah. Three Japanese stations, Sapporo, Tateno, and Naha, exhibit 591 relatively large biases of 2-3 DU and even larger biases before switching from KC to ECC 592 sondes. Almost half of the 11 northern high latitude stations (60° N-90° N) and two of the 6 593 southern high-latitude stations have large SDs/biases. In addition to retrieval biases from the 594 OMI data, some of the large biases or SDs might be partially related to ozonesonde type with 595 different biases and uncertainties due to different types (e.g., Indian sonde stations, Brewer-Mast 596 ozonesonde at Hohenpeissenberg, three KC sonde stations), manufacturers (e.g., SP vs. ENSCI 597 for ECC sonde), sensor solution or related to individual sonde operations, which was shown in 598 the validation of GOME ozone profile retrievals (Liu et al., 2006a).

599 Figure 11 shows the comparison for each season at northern mid-latitudes. Consistent with 600 profile comparison, the TOC comparison is season-dependent. When applying OMI AKs, the 601 mean bias varies from 3 DU in winter to -1.5 DU in summer. The SDs are within 6.8 DU with 602 the smallest value during fall due to less ozone variability. The regression slopes are very close, within 0.04 around 0.67. The retrieval sensitivity is smallest during the summer as seen from the 603 604 greatest correlation and slope and relatively small standard deviation, and is the worst during the 605 winter. With OMI AKs applied to ozonesonde profiles, the MBs only slightly change (varying 606 from 3.5 DU to -1.3 DU), but the SDs are significantly reduced to within 5.2 DU, the slopes 607 significantly increase by ~ 0.2 to 0.8-1.0, and the correlation improves significantly during the 608 winter and spring.

Figure 12 compares the surface~550 hPa and surface~750 hPa ozone columns with ozonesonde 609 610 data in the middle latitudes during summer and the tropics. Compared to the TOC comparisons in Figure 9 and Figure 11, the comparisons of these lower tropospheric ozone columns exhibit 611 612 smaller regression slopes and correlations that are a result of reduced retrieval sensitivity. In the 613 tropics, the slopes decrease from 0.78 in TOC to 0.65 in the surface~550 hPa ozone column and 614 ~ 0.50 in the surface ~ 750 hPa column, with corresponding correlation from 0.83 to 0.74 in the 615 surface-~550 hPa column, and 0.66 in the surface-~750 hPa column. This indicates that the 616 retrievals in the surface~550 hPa/750 hPa can capture ~65%/50% of the actual ozone change 617 from the a priori. During the middle latitude summer, the slope decreases from 0.71 in the TOC comparisons to 0.42 in the surface-~550 hPa comparisons and 0.32 in the surface-~750 hPa 618 619 comparisons, with corresponding correlation coefficients from 0.74 to 0.5 and 0.46. Thus, the retrievals in the surface~550 hPa and ~750 hPa only capture ~40%/30% of the actual ozone change from the a priori. The MBs are generally small within 0.5 DU (5%) with SDs of ~3.6 DU (20-28%) in the surface~550 hPa ozone column and ~2.5 DU (25-36%) in the surface~750 hPa ozone column. After applying OMI AKs to account for inadequate retrieval sensitivity and removing smoothing errors, the slope significantly increases to approach 1 (as expected). SDs are reduced to ~10% in the middle latitudes and ~15% in the tropics.

626 4.3 Evaluation of Long-term Performance

627 Comparisons in Sects 4.1 and 4.2 Previous evaluation-indicated systematic differences between 628 pre-RA and post-RA periods and generally worse performance during the post-RA periods. To 629 further illustrate the long-term stability of our ozone profile product and understand the quality 630 of OMI radiometric calibration as a function of time, we analyze monthly MBs of 631 OMI/ozonesonde differences with OMI retrieval AKs in ozone profiles, SOCs, and TOCs. Due 632 to the lack of OMI observations during some months at high-latitudes, we focus the evaluation 633 by using coincidence pairs in 60° S-60° N. Monthly MBs are calculated only if there are more 634 than 5 OMI-ozonesonde pairs in a given month. Linear regression trend is on the MBs for the 635 entire period (2004-2014) and/or for the pre-RA and post-RA periods, respectively. The trend is 636 considered statistically significant if its P value is less than 0.05.

637 The linear trends of monthly mean ozone biases for each OMI layer between 60° S-60° N are 638 plotted in Figure 13 for each of the three periods. During 2004-2014, marked in black, ozone 639 biases at layers above 50.25 hPa show significant positive trends of 0.06-0.17 DU/year (0.17-640 0.52%/year), while ozone biases between 290 hPa and 110 hPa exhibit significant negative 641 trends of 0.1-0.19 DU/year (1-2%/year). The positive trends in the stratosphere are generally 642 consistent with those shown in OMI-MLS comparisons (Huang et al., 2017). In the lowermost 643 three OMI layers, ozone differences are more stable but with several large spikes during the post-644 RA periods likely due to the RA evolution or instrument operation. The derived trends for the 645 pre-RA period are generally more flat and insignificant at all layers indicating good stability of our product as well as the OMI radiometric calibration. During the post-RA period, the derived 646 647 trends are positive above 75 hPa with statistical significance. These positive trends in the stratosphere are generally similar to those over the entire period, suggesting the dominant 648

contribution of the post-RA period to the overall trend. In the altitude range 214 – 108 hPa, the
post-RA trends are also flat similar to the pre-RA trends, but the values are systematically
smaller during the post-RA period, causing significantly negative trends over the entire period.

652 The SOC biases exhibit small positive trend of 0.14±0.09 DU/year in 2004-2014 with no 653 statistical significance (Figure 14(a)). This slight positive trend is a result of trend cancellation 654 by the positive trends above 80 hPa and negative trends between 220 hPa and 80 hPa The TOC biases reveal a significant negative trend of -0.18 ± 0.05 DU/year (Figure 14(b)), mostly from 655 656 layers in the upper troposphere. In the pre-RA and post-RA periods, both trends of both SOC and TOC biases are relatively flat during the pre-RA period, while the SOC trend in the post-RA 657 period is 0.77 ± 0.20 DU/year with significance. It is noticeable that the P value of TOC trend in 658 659 the post-RA period is 0.06.

660 The significant trends of ozone biases at different layers as well as in SOC and TOC suggest that 661 the current ozone profile product is not suitable for trend studies especially during the post-RA period. The relatively flat bias trends during the pre-RA periods and statistically significant 662 663 trends during the post-RA period confirm that the better stability of our product during the pre-664 RA period and more temporal variation of the retrieval performance during the post-RA period 665 are likely associated with the RA evolution. In previous sections, the validation of our retrievals revealed latitudinal/seasonal/SZA and cross-track dependent biases even during the pre-RA 666 667 period. This indicates the need to remove signal dependent errors and the calibration 668 inconsistency across the track. To maintain the spatial consistency and long-term stability of our ozone profile product, we need to further improve OMI's radiometric calibration especially 669 during the post-RA period. Preferably, the calibration improvement should be done in the level 670 671 0-1b processing. If this option is not possible, we can perform soft calibration similar to Liu et al. 672 (2010b) but derive the correction as a function of time and latitude/SZA. In addition, it should be 673 noted that the trend calculation might be affected by factors such as the availability of correction 674 factors with ozonesondes (Morris et al., 2013), station-to-station variability and the uneven spatiotemporal distribution of the ozonesondes, which can introduce considerable sampling 675 676 biases (Liu et al., 2009; Saunois et al., 2012).

677 5 Summary and Conclusion

678 We conducted a comprehensive evaluation of the quality of OMI ozone profile (PROFOZ) 679 products produced by the SAO algorithm, including their spatial consistency and long-term performance using coincident global ozonesonde observations during the decade 2004-2014. To 680 681 better understand retrieval errors and sensitivity, we compared the retrieved ozone profiles and a 682 priori profile at individual layers with ozonesondes before and after being degraded to the OMI 683 vertical resolution with OMI retrieval average kernels (AKs). We also compared the integrated 684 SOC, TOC, and surface-~550/~750 hPa ozone columns with ozonesonde data. To understand the 685 spatial distribution of retrieval performance, the validations are grouped into five latitude ranges: 686 northern/southern high/middle latitudes, and the tropics. To investigate the impacts of the OMI 687 row anomaly (RA) on the retrievals, we contrasted the comparison before and after the 688 occurrence of major OMI RA in January 2009, i.e., pre-RA (2004-2008) and post-RA (2009-689 2014) periods. In addition, we quantified the dependence of retrieval performance on seasonality and several key parameters including solar zenith angle (SZA), cloud fraction, and cross-track 690 691 position. Finally, we analyzed the monthly mean variation of the mean biases (MBs) to examine 692 the long-term stability of the PROFOZ product.

693 The comparison between OMI and ozonesonde profiles varies in altitude, with maximum 694 standard deviations (SDs) in the Upper Troposphere and Lower Stratosphere (UTLS) due to 695 significant ozone variability, and varies with latitude similarly in the northern and southern 696 hemispheres. There is good agreement throughout the atmosphere in the tropics and mid-697 latitudes. With the application of OMI AKs to ozonesonde data, the MBs are within 6%, and the SDs increase from 5-10% for pressure $< \sim 50$ hPa to within 18%(27%) in the tropics/mid-698 699 latitudes for pressure $> \sim 50$ hPa. In the high latitudes, the retrievals agree well with ozonesondes 700 only for pressure < 50 hPa with MBs of < 10% and SDs of 5-15% for pressure < 50 hPa, but 701 with MBs reaching 30% and SDs reaching 40% for pressure $> \sim 50$ hPa. The comparison results 702 are-is seasonally dependent. At northern mid-latitudes, there are generally the best retrieval 703 sensitivity and the smallest SDs as great as 20% in the summer, and the worst sensitivity and the 704 largest SDs reaching 31% in the winter. the agreement is generally best (except for MBs) in the 705 summer, with the best retrieval sensitivity and the smallest SDs as great as 20%, and the worst in the winter with the worst retrieval sensitivity and the largest SDs reaching 31%. The MBs near 706

707 300 hPa vary from 12% in the winter to -10% in the summer. The post-RA comparison is 708 generally worse in the tropics and mid-latitudes than the pre-RA comparison, with SDs larger by 709 up to 8% in the troposphere and 2% in the stratosphere, and with larger MBs around ~300 hPa in 710 the mid-latitudes. But at high latitudes, the pre-RA comparison does not show persistent improvement over the post-RA comparison, with smaller biases and larger SDs at some altitudes, 711 712 especially at southern high latitudes. The retrieval improvement over a priori can be determined 713 from the SD reduction of the retrieval comparison from the a priori comparison. The retrievals 714 demonstrate clear improvement over the a priori down to the surface in the tropics, but only 715 down to ~750 hPa during mid-latitude summer, ~550 hPa during the other seasons of mid-716 latitudes and ~ 300 hPa at high latitudes.

717 Retrieval performance typically becomes worse at large SZA, especially at SZA larger than 75°, 718 where the MBs in the troposphere are >20% and the SDs near ~ 300 hPa are > 45%. The worse 719 performance at larger SZA is due to a combination of weaker signal and greater influence by 720 radiometric calibration errors such as due to stray light, and radiative transfer calculation errors. 721 The variation of SZA is likely responsible for the majority of the retrieval dependence on latitude 722 and season. The retrieval quality for pressure $> \sim 100$ hPa degrades with increasing cloudiness in 723 terms of MBs and SDs, with MBs greater than 10% at cloud fraction > 0.3. The retrieval 724 performance also varies with cross-track position, especially with large MBs and SDs at the 725 first/last extreme off-nadir positions (e.g., 1-3 and 28-30). The dependence is stronger during the 726 post-RA period.

727 The integrated SOCs and TOCs also exhibit good agreement with ozonesondes. With the convolution of OMI AKs to ozonesonde data, the SOC MBs are within 2% with SDs within 728 729 $\sim 5.1\%$ in the tropics and mid-latitudes. These statistics do not change much even without the 730 applications of OMI AKs. The comparison becomes slightly worse at high latitudes, with MBs 731 up to 3% and SDs up to 6%. The pre-RA comparison is generally better with smaller SDs of 0.2-732 0.6% except for southern high latitudes, although with slightly larger MBs. The TOC MBs and SDs with OMI AKs are within 6%, with SDs of <~15% in the tropics and mid-latitudes but reach 733 734 30% at high latitudes. The pre-RA TOC comparison is also better in the tropics and mid-latitudes 735 with SDs smaller by 3.4-5.5% but worse values at southern high latitudes. The TOC comparison 736 at northern mid-latitudes varies with season, with MBs of 11%. There are worse correlation

737 during winter and MBs of -3% and best correlation in summer. The TOC comparison also shows 738 noticeable station-to-station variability in similar latitude ranges with much larger MBs and/or 739 SDs at the two Indian stations and larger MBs at several Japanese stations before they switched 740 from KC ozonesondes to ECC ozonesondes. This demonstrates the impacts of ozonesonde 741 uncertainties due to sonde types, manufacturers, sensor solution and operations. Without 742 applying OMI AKs, the TOC correlation with ozonesondes typically becomes worse at higher 743 latitudes, ranging from 0.83 in the tropics to 0.5-0.6 at high latitudes. The linear regression slope 744 is within 0.6-0.8, typically smaller at higher latitudes, reflecting the smaller retrieval sensitivity 745 down to the troposphere at higher latitudes mainly resulting from larger SZA. The convolution of 746 AKs significantly improves the correlation and slope. The impact of retrieval sensitivity related 747 to SZA is also reflected in the seasonal dependence of the comparison at mid-latitudes.

The surface-~550/750 hPa ozone columns in the tropics during mid-latitude summer compare quite well with ozonesonde data, with MBs of < 5% and SDs of 20-25%/28-36% without OMI AKs. The correlation and slope decrease with decreasing altitude range due to reduced retrieval sensitivity down to the lower troposphere. These columns capture ~65%/50% of the actual ozone change in the tropics and ~40%/30% in the troposphere. Convolving ozonesonde data with OMI AKs significantly increases the slope to ~1 and reduce the SDs to 10-15%.

The contrast of pre-RA and post-RA comparisons indicates generally worse post-RA performance with larger SDs. Linear trend analysis of the OMI/ozonesonde monthly MBs further reveals additional RA impact. The temporal performance over 60° S-60° N is generally stable with no statistically significant trend during the pre-RA period, but displays a statistically significant trend of 0.14-0.7%/year at individual layers for pressure < ~80 hPa, 0.7 DU/year in SOC and -0.33 DU/year in TOC during the post-RA period. Because of these artificial trends in our product, we caution against using our product for ozone trend studies.

This validation study demonstrates generally good retrieval performance of our ozone profile product especially in the tropics and mid-latitudes during the pre-RA period. However, the spatiotemporal variation of retrieval performance suggests that OMI's radiometric calibration should be improved, especially during the post-RA period, including the removal of signaldependent errors, calibration inconsistency across the track and with time to maintain the longterm stability and spatial consistency of our ozone profile product.

767

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Figure 1 Variation of monthly mean OMI RMS (defined as Root Mean Square of the ratio of
radiance residuals to assumed radiance errors). The dashed and solid lines represent respectively
the monthly mean RMS, and the sum of monthly mean plus its two standard deviations that is set
as the RMS threshold for data screening.





Figure 2 The distribution of ozonesonde stations in this study. The color represents the mean biases 984 between OMI and ozonesonde tropospheric ozone columns (TOCs) at each station (if the number of OMI and ozonesonde pairs is more than 10), and the dot size represents the standard deviation.





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Figure 3 Mean relative biases in ozone (line with circles) and corresponding standard deviations (solid lines) between OMI retrieval/a priori and ozonesondes with and without applying OMI retrieval averaging kernels (i.e., with AKs, and W/O AKs in red and green for comparing retrievals and in blue and yellow for comparing a priori) for five different latitude bands. The left panels

- show the comparison using 10 years of OMI data (2004-2014), and the right panels show the comparison between OMI retrieval and ozonesonde with OMI AKs for before and after the occurrence of serious OMI row anomaly (RA), i.e., pre-RA (2004-2008) in black and post-RA (2009-2014) in gray, respectively. The number (N) of OMI/ozonesonde coincidences used in the
- comparison is indicated in the legends.





1001 Figure 4 Same as Figure 3c but for each individual season at 30° N-60° N.



1004Figure 5 Mean relative biases in ozone (a) and standard deviations (b) of the differences between1005OMI and ozonesonde convolved with OMI AKs as a function of Solar Zenith Angle using all1006OMI/ozonesonde coincidences during 2004-2014.





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1010 Figure 6 Same as Figure 5 but as a function of cloud fraction.

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1015Figure 7 Same as Figure 5 but as a function of cross-track position for (left) pre-RA (2004-2008)1016and (right) post-RA (2009-2014) periods, respectively.



1019Figure 8. Scattering plots of OMI Stratospheric Ozone Columns (SOCs) vs. ozonesonde SOCs1020without (black) and with (red) average kernels for five different latitude bands during 2004-20141021(left), the pre-row anomaly (RA) period (i.e., 2004-2008, middle) and the post-RA period (i.e., 2009-10222014, right), respectively. Comparison statistics including mean biases and standard deviations in

1023both DU and %, the linear regression and correlation coefficients in DU, and the number of1024coincidences are shown in the legends.



1027 Figure 9. Similar to Figure 8, but for comparison of Tropospheric Ozone Columns (TOCs).





Figure 10. (Left) Time series of OMI tropospheric ozone columns (TOCs) as green dots and ozonesonde TOCs (with OMI AKs applied) in Summit (38.48° W, 72.57° N), Payene (6.57° E, 46.49°
N), Naha (127.69° E, 26.21° N), La Réunion (55.48° E, 21.06° S), Broadmeadows (144.95° E, 58.74°
S) and Neumayer (8.27° W, 70.68° S), and (Right) their corresponding differences, including the mean biases and standard deviations in 2004-2014, pre-RA (2004-2008) and post-RA (2009-2014)
periods, respectively, in the legends.



1038Figure 11. Similar to Fig. 9Same as Figure 9-but for different seasons at northern middle latitude1039during the 2004-2014 period.







1044 columns <u>during the 2004-2014 period</u>. (a) Surface~550 hPa ozone column and (b) Surface~750 hPa 1045 ozone column in 30° N-60° N during the summer, (c) and (d) same as (a) and (b) but for the tropics.



1049Figure 13. Monthly mean variation of OMI and ozonesonde mean biases in 60° N-60° S at each1050OMI layer. OMI retrieval averaging kernels are applied to ozonesonde data. The black, red and1051green lines represent the linear ozone bias trends in 2004-2014, pre-RA (2004-2008) and post-RA1052(2009-2014), respectively. The average altitude of each layer is marked on the left corner of each1053grid. The trends in DU/yr or % yr and P value for each time period are indicated in the legends.





1057Figure 14. Same as Figure 13 but for Stratospheric Ozone Columns (SOCs) and Tropospheric1058Ozone Columns (TOCs).