A Post-2013 Drop-off in Total Ozone at Half of Global Ozonesonde Stations: ECC Instrument Artifacts?

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Key Points:

- We report a drop in ozonesonde total column O_3 of 2-8 % relative to independent measurements at nearly half of sites beginning around 2013
- Comparisons with satellite stratospheric O_3 profiles show the artifact loss peaking at 5-10 % or more in the middle and upper stratosphere

- Changes in the ozonesonde instrument are associated with the drop-off, but no single factor has been identified as a cause

Keywords: ECC Ozonesonde, Aura, OMI, MLS, Suomi-NPP, OMPS

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Abstract

An international effort to improve ozonesonde data quality and to reevaluate historical records has made significant improvements in the accuracy of global network data. However, during 2013-2016, ozonesonde total column ozone (TCO; O_3) at 17 of 37 regularly reporting stations exhibited a sudden drop-off relative to satellite measurements. The ozonesonde TCO drop is 2-8 % compared to satellite and ground-based TCO, and 5-10 % or more compared to satellite stratospheric O_3 profiles, compromising the use of recent data for trends, although they remain reliable for other uses. Hardware changes in the ozonesonde instrument appear to be a major factor in the O_3 drop-off, but no single property of the ozonesonde explains the findings. The bias remains in recent data. Research to understand the drop-off is in progress; this letter is intended as a caution to users of the data. Our findings underscore the importance of regular ozonesonde data evaluation.

Plain Language Summary

Balloon-borne ozonesondes provide accurate measurements of atmospheric ozone (O_3) from the surface to above 30 km with high vertical resolution. Dozens of global stations have regularly launched ozonesondes for decades, and they provide vital information for improving O_3-measuring satellite algorithms, tracking recovery of the stratospheric O_3 layer, and our understanding of surface to lower stratospheric O_3 changes in an evolving climate. We present the discovery of an apparent instrument artifact that has caused total column O_3 measurements

from about half of global stations to drop by 2-8 % starting in 2013-2016, limiting their suitability for calculating O_3 trends. Work is underway to solve the problem, but the exact cause of the drop is still unknown. This letter serves as a caution to the community of ozonesonde data users.

1 Background: The Ozonesonde Instrument and Data Quality Assurance

The electrochemical concentration cell (ECC) ozonesonde measures ozone (O_3) profiles from the surface through the mid-stratosphere (~5 hPa). Ozone is measured via a chemical reaction from bubbling ambient O_3 into two electrochemical cells containing a potassium iodide (KI) solution (sensing solution type or SST). The ECC is launched on a weather balloon in tandem with a radiosonde that transmits O_3 partial pressure simultaneously with pressure, temperature, humidity (PTU), and GPS-derived wind data to a ground station approximately once a second. With a 20-30 s response time, the effective vertical resolution of the O_3 signal is ~150 m.

Because each ozonesonde is a new instrument that must be prepared before launch, it is essential to standardize instrument preparation, operations, and the treatment of raw data. In the past decade, a panel of researchers have engaged in both individual and collective tests of instrumentation, meeting regularly to discuss quality assurance and to develop standard operating procedures (SOP) in an activity designated Assessment of SOP for Ozonesondes (ASOPOS). Current SOP were published in **Smit and ASOPOS (2014)**. The main sources of instrument variability are the instrument type (there are two major manufacturers of ECC instruments, which we call “Type1” and “Type2”), the composition of the SST, conditioning protocol, and post-processing; these parameters are given in the metadata for each record.

ASOPOS has also published guidelines for reprocessing sonde data records that may be affected by deliberate or inadvertent ECC preparation changes. Case in point: the ASOPOS recommendation is to deploy each ECC type with a different SST, even though the two types operate on the exact same measurement principle. If a station changes only one of these variables, the resulting step change in O_3 is considered an instrumental artifact. Reprocessing is carried out to compensate for such changes, and the data are said to be homogenized (**Smit and ASOPOS, 2012**; **Deshler et al., 2017**). Both the SOP and reprocessing guidelines are based on laboratory (**Smit et al., 2007**) and field tests (**Deshler et al., 2008**) in which different sensors are compared with a standard O_3 reference photometer. In the lab, tests are made with 2-4 ECC sensors operating in a closed chamber that simulates a standard profile over a 2-hr "flight." Field tests compare instruments on a single gondola launched with a special balloon.

During the period 2013 through 2017 data from more than 25 ozonesonde stations were reprocessed (**Tarasick et al., 2016; Van Malderen et al., 2016; Thompson et al., 2017; Witte et al., 2017; Sterling et al., 2018; Witte et al., 2019**). In general, reprocessed data show significant improvements in comparisons to independent total column ozone (TCO) measurements. Reprocessed data at 12 of 14 SHADOZ stations agree to within 2 % of satellite and ground-based TCO measurements (**Thompson et al., 2017**), compared to > 8 % offsets at half of the stations through 2004 in **Thompson et al. (2007)**. Improvements in tropical mid-stratospheric O_3 readings also led to better agreement with MLS profiles (2005-2017; **Witte et al., 2017**).

In spite of the reprocessing successes, the homogenized data for two tropical stations (Costa Rica and Hilo) displayed sharp 5 % drop-offs in TCO relative to satellite measurements after 2014; at Hilo a simultaneous discrepancy appeared relative to the Mauna Loa Dobson spectrometer (**Thompson et al., 2017; Sterling et al., 2018**). The drop-off was also observed in the original datasets, ruling out the reprocessing as the cause. Furthermore, NOAA's Boulder, CO, site, which used the same instrumentation and SST, did not appear to be similarly affected. Hypothesized causes for these findings, e.g., hardware changes in the 2011-2016 period (the company manufacturing Type1 ECCs changed ownership twice) or NOAA's non-standard SST used at the above-mentioned sites, were tested along with other variables in a new series of chamber tests (JOSIE; Jülich Ozonesonde Intercomparison Experiments) in late 2017. Initial results from the 80 chamber profiles in JOSIE-SHADOZ could not explain the drop-off behavior (**Thompson et al., 2019***)*, and the cause remained unsolved.

Because ozonesonde profiles are relied upon as the foundation for satellite O_3 retrievals and validation, we re-examine the agreement among sonde, satellite, and ground-based TCO with two more years of data from the SHADOZ and NOAA networks to determine if the drop-offs reported in **Thompson et al. (2017)** and **Sterling et al. (2018)** persist. We also extend these analyses to the global network during the Aura satellite era of October 2004 to present. We find that about half of these 37 stations exhibit an instrumental artifact drop-off in TCO after 2013, with a coincident decline in stratospheric O_3. Instrumental factors are investigated but no definitive explanation for these findings has yet emerged. In **Section 2** data sources and statistical methods are described. **Section 3** describes results and potential changes to the ECC instrument and factors that require further investigation. **Section 4** is a summary and recommendations for use of data affected by the ECC O_3 drop-off.

2 Data and Methods

2.1 ECC Ozonesonde Data

We selected a total of 37 global ECC ozonesonde sites based on the availability of consistent and up-to-date records during the Aura period of October 2004 to present (i.e. data available within the last few years; an exception is Watukosek which ended in October 2013) to analyze the recent drop in ECC TCO measurements. Currently, 28 of the sites launch Type1 ECCs, and nine launch Type2. Some sites have previously changed ECC types, SST, or both, so the most recent metadata are listed in **Table 1**. The primary evaluation of ozonesonde data is with NASA's Aura satellite; sample numbers listed in **Table 1** are from the Aura period only. None of the ozonesonde data here are normalized to a TCO measurement or an outside data source.

2.2 Satellite and Ground-Based Data

Satellite TCO measurements are from the Aura Ozone Monitoring Instrument (OMI; **McPeters et al., 2008**) and the Suomi-NPP Ozone Mapping Profiler Suite (OMPS; **McPeters et al., 2019**). Stratospheric O_3 profile measurements are from the Aura Microwave Limb Sounder (MLS; **Froidevaux et al., 2008**). To identify "coincident" satellite overpasses, we limit Level 2 TCO data to within 8 hours of the ozonesonde measurement. We use MLS v4.2 Level 2 O_3 data averaged within one day and 5° latitude and 8° longitude of the ozonesonde launch. MLS data are screened according to the v4.2 Level 2 MLS Data Quality document (**Livesey et al., 2018**). Sensitivity tests on our screening of coincident satellite TCO data by limiting comparisons based on cloud fraction or overpass distance to the ECC site had negligible effects on the statistics (less than 1 % change in overall OMI/ECC TCO agreement).

The OMI and OMPS TCO measurements compare well with the series of Solar Backscatter Ultraviolet instruments and are suitable for TCO trends analysis (**McPeters et al., 2015; 2019**). Aura MLS O_3 measurements in the stratosphere exhibit little drift – the v3.3

measurements are stable to within 1.5 % per decade (**Hubert et al. 2016**; it is presumed the v4.2 data used here have similar stability). Thus, these three satellite instruments are suitable to detect significant changes in the ECC ozonesonde network. Our primary ECC comparisons are with OMI because of its > 15 year record. OMPS and MLS reinforce the OMI results.

A total 23 of the 37 ECC sites have a co-located ground-based TCO instrument (**Table 1**). Most sites have a Brewer or Dobson spectrophotometer (or both at Hilo and Tateno); Réunion uses a SAOZ UV-visible spectrometer. ECC TCO comparisons with all three ground-based instrument types are found in **Thompson et al. (2017)**.

2.3 Defining the ECC O_3 Drop-off: Example Sites

To characterize the O_3 drop-off, we separate the sites with unambiguous drops in TCO, which we call "affected" sites, from those called "reference" sites. Affected sites are defined as those recording drops of TCO relative to OMI of greater than 2 % after visually locating a downward step-change in the time series of comparisons with OMI. This does not mean there is no change at the reference sites; a < 2 % drop-off is assumed to be less significant. The TCO drop is calculated as the difference in mean bias compared to OMI TCO before (Oct. 2004 to drop-off date) and after the drop-off date (through the end of the site's ECC record). For example, **Figure 1a** displays a sudden TCO drop-off relative to OMI at Kelowna in March 2015. The ECC TCO averaged 4.0 % higher than OMI before the drop-off in March 2015 (564 samples), and -0.4 % lower than OMI after March 2015 (100 samples) – a 4.4 % drop, meeting the > 2 % criterion. The visual identification of the drop-off date is subjective, but objective analyses of ECC serial numbers follow in Section 3.3.

The drop-off emerged at Nairobi in June 2015, and at Lauder in September 2016 (**Figure 1b, c**). Nairobi and Lauder both exhibit drop-offs of 2.2 % relative to OMI. The percent differences between ozonesonde and MLS stratospheric O_3 in the top panels of **Figure 1** show that the drop in ECC O_3 relative to MLS is coincident with the TCO drop.

3 Results and Discussion

3.1 Sites Affected by the ECC O_3 Drop-off

Using the criterion of a > 2 % TCO drop relative to OMI, we find that 17 of 37 sites are affected by a sudden TCO drop-off. **Table 1** lists the affected sites in bold including the TCO drop relative to OMI. A map of all sites examined, with affected sites colored according to the magnitude of TCO drop-off, is shown on **Figure 2**. Defining the drop in TCO as relative to OMI is necessary considering that some sites previously exhibited a high bias compared to satellites, with the drop-off leading to better agreement with OMI (e.g. Kelowna in **Figure 1a**).

Dates of the first notable drop in TCO measurements range from August 2013 at San Cristóbal to January 2017 at American Samoa. All but one (Natal) of the affected sites are Type1 sites. The magnitude of the TCO drop-off varies considerably. The drop in TCO at Lauder is a relatively modest -2.2 %, whereas changes of -7.5 % and -8.2 % are observed at Churchill and Yarmouth.

Comparisons similar to **Figure 1** for the remaining 34 sites in **Table 1** are found in the Supplementary Material in **Figures S1a-n and S2a-t**. We note that sites show periods of high or low bias compared to OMI and MLS (e.g. Madrid's high bias for a portion of 2009; **Figure S2g**), but our focus is on sudden drops in O_3 that persist for more than 2 or 3 years in the most recent record.

The three Japanese stations examined do not exhibit a drop in ECC TCO. Out of 10 SHADOZ stations that are currently launching Type1 ECCs, only Réunion Island and Kuala Lumpur are not affected. Note that, for reasons unknown, Kuala Lumpur has measured consistently low O_3 since the beginning of the Aura record in late 2004. In summary, there is inconsistency in TCO drop-off amount, and the drop-off is not a universal problem.

3.2 Comparisons with Aura MLS Stratospheric O_3

A closer examination of ECC and MLS stratospheric O_3 comparisons is warranted given the coincidence between the OMI and OMPS TCO drop-off, and apparent MLS O_3 drop-off in **Figure 1**. **Figure 3** shows a composite of comparisons between MLS and ECC ozonesonde stratospheric O_3 at the 17 affected sites before and after the identified drop-off (dates in **Table 1**). Prior to the drop-off at the 17 affected ECC sites, stratospheric O_3 biases compared to MLS follow the zero line in **Figure 3** (blue colors). After the drop-off in TCO, the ECC measurements shift 5-10 % or more lower relative to MLS (red colors), occasionally reaching > 20 % low above 10 hPa (the 25th percentile value at the 6.81 hPa MLS level is -23.0 %). **Figure 3** shows that the stratospheric O_3 drop-off is the major contributor to the TCO offsets with OMI and OMPS. At this point, a similar drop-off in tropospheric O_3 has not been detected and is presumed to be insignificant. Exceptions are two stations, Costa Rica and Hilo, which may be reading low in recent years in the troposphere due to volcanic SO_2 interference (e.g. **Morris et al., 2010**). That is beyond the scope of our study.

3.3 Potential ECC Instrument Factors in the O_3 Drop-off

The ECC O_3 drop-off has been quantified against satellite TCO, satellite O_3 profiles, and ground-based instruments (**Thompson et al., 2017; Sterling et al., 2018**; ground-based comparisons to follow in Section 3.5). Thus, we rule out geophysical factors as a cause; the drop-off is an instrument artifact, so we consider potential instrumental contributions. Each ECC is built from a number of components that may change over time as the manufacturers' suppliers change. For example, the Type1 instrument changed manufacturer twice between 2011 and 2016. Components include the cells holding the SST, the ion bridge between the two cells, the air intake pump, the constant-speed motor, batteries, and the platinum electrodes. A 2-8 % change of response could be caused by loss of O_3 or of molecular iodine, losses through the internal resistance of the cell, or in-flight changes in the pump and motor efficiency. The SST composition and the radiosonde model (and interface) are additional considerations (Section 3.6). The ECC serial number is used to evaluate potential instrument or component changes over time.

Figure 4 shows histograms of ECC TCO offsets with OMI and OMPS separated by the 16 affected (red on **Figure 4**) and 12 reference (blue on **Figure 4**) Type1 sites. Each histogram displays statistics for every 1000 serial numbers (e.g. 24K = 24000-24999). The affected sites show a low bias for 25K serial numbers, abruptly dropping from a median TCO bias compared to OMI and OMPS of +0.7 % (24K), to -2.9 % (25K). The reference sites show no such drop, and, in fact, no recent serial number set since 24K has a median bias larger than -1.2 % (30K) for the 12 reference sites. The affected sites show significant negative biases for all serial numbers from 25K to 35K, with a maximum median low bias of -5.1 % for 31K serial numbers. Histograms for *all* serial numbers at affected (**Figure S3**) and reference (**Figure S4**) Type1 sites are found in the Supplementary Material. **Figure S3** shows the history of good ECC/satellite agreement at affected Type1 sites throughout the Aura record since October 2004 and prior to the 25K serial numbers, although there are indications of some low-biased measurements from serial numbers 20-22K. The largest deviation for reference Type1 sites is the +2.1 % median bias for 16K serial numbers (**Figure S4**). In summary, before the TCO drop-off at the affected sites, the ECC TCO comparisons with satellite measurements averaged within 1 or 2 %, and comparisons at reference sites remain, on average, within 1 or 2 %.

Reference and affected Type1 sites were both launching ECCs with similar serial numbers, so it is puzzling why they show such large discrepancies in their comparisons with satellite TCO. **Figure S5** shows a continuous time series of Type1 serial numbers and TCO comparisons with OMI and OMPS, which illustrates the consistent unbiased TCO values at reference sites (**Figure S5a**), and the large drop-off at affected sites (**Figure S5b**). This commingling of good and poorly-performing Type1 serial numbers, which appear to be distinguishable only by site, suggests that the ECC O_3 drop-off is not due to manufacturing issues for the Type1 ECC alone and that at least one additional secondary factor must play a role in the occurrence of this issue.

3.4 Stations with Type2 ECCs

We examined nine Type2 ECC ozonesonde sites for a drop-off and sudden low TCO bias. Histograms of the TCO offset between reference Type2 ECCs and OMI and OMPS are shown in **Figure S6** with the serial numbers grouped by 1000 as in **Figure 4**. Note that the similar serial numbers between Type1 and Type2 ECCs are purely a coincidence. The Type2 histograms show no abrupt downward shift in agreement with satellite TCO as seen at the affected Type1 sites in **Figure 4 and Figure S3**. An exception is at Natal.

3.5 ECC Comparisons with Ground-Based TCO Measurements

Of the 37 sites analyzed here, 23 have ground-based TCO measurements to compare against the ECCs (**Table 1**). Example time series of the comparisons between ECCs and the Brewer at Churchill, and the Brewer and Dobson at Hilo are shown in **Figure S7**. The ground-based TCO measurements near Hilo are taken at Mauna Loa (3405 m), which explains why the ECC TCO is higher than the Brewer and Dobson prior to the August 2014 drop-off. Histograms similar to **Figure 4** and **Figures S3, S4, and S6** for the ground-based TCO comparisons are shown in **Figures S8-S10**. The ECC TCO drop-off relative to the ground-based instruments is ~3-4 % after Type1 24K serial numbers in **Figure S8**. The ground-based comparisons with reference Type1 and Type2 sites (**Figures S9 and S10**) are quite variable, but no sustained drop-off is apparent as observed in **Figure S8**.

3.6 Possible Sources of the Drop-Off

Around 2010-2012, most of the affected ozonesonde sites examined here switched from the Vaisala RS-80 to RS-92 radiosonde, or from RS-80 to the InterMet iMet radiosonde. The radiosonde pressure measurements affect the ECC O_3 calculation and altitude registration, so a change from non-GPS RS-80 to GPS-enabled RS-92 and iMet radiosondes can lead to pressure measurement changes, which translate to O_3 changes (**Stauffer et al., 2014**). Some sites (e.g. Lauder in 2015) switched radiosondes again from RS-92 to the RS-41. An example of an RS-80 to iMet transition at Hilo is shown in **Figure S11**. There is a shift in mid-stratospheric pressure and temperature measurements with the transition to iMet in 2011-2012, but this change occurs two years before the Hilo low O_3 bias in August 2014. Similar mismatches between radiosonde changes and the ECC drop-off are found at other sites. Costa Rica switched from RS-80 to iMet radiosondes in 2012-2013, but the drop-off did not occur until January 2016 (**Thompson et al., 2017**). Nairobi switched from RS-80 to RS-92 radiosondes in 2010, but there was no drop-off until June 2015. We therefore rule out radiosonde changes as the primary cause of the ECC O_3 drop-off.

The drop-off is found at sites that use a variety of SSTs (**Table 1**) and three different radiosonde types (RS-92 or 41 and iMet). Sites that are seemingly unaffected, e.g. Trinidad Head, Boulder, and Huntsville, all use the same 1.0 % KI with $1/10^{th}$ buffer SST and iMet radiosonde combination as Hilo and Costa Rica (**Figure S1h, S1i**). We have not fully explored the effects of different SSTs on the O_3 drop-off, but given that all three SSTs currently in use are affected (**Table 1**), it does not appear that SST is a major factor.

The ASOPOS 2.0 panel will perform additional experiments and analyses to identify possible sources of the O_3 drop-off. Candidate tests include examining the different radiosonde interface boards and batteries used on Type1 ECC sondes, reviewing site ECC preparation procedures, testing ECC pump performance in flight and in vacuum chambers, and experiments with older Type1 ECCs manufactured before the drop-off began. Both Type1 and Type2 ozonesondes, four different sensing SSTs, and varying preparation procedures were tested in the

2017 JOSIE-SHADOZ experiment (**Thompson et al., 2019**). In-depth analysis of the 80 profiles from JOSIE-SHADOZ should help identify the causes and magnitudes of contributing factors like SST to the ECC O_3 drop-off.

4 Summary and Recommendations for Affected Data

Since 2013-2016, we observed a drop-off in ECC ozonesonde TCO and stratospheric O_3 at 17 ECC global ozonesonde sites, 16 of which launch Type1 ECC ozonesondes. The TCO drop is 2-8 % compared to OMI TCO measurements, and the stratospheric O_3 drop can be greater than 10 % compared to MLS O_3 profiles in the mid-stratosphere. The low bias is notably absent at almost half of the 28 Type1 sites that we examined. Except for Natal, there is no significant drop-off or change in bias for Type2 ECC ozonesondes during similar years. Because the drop-off varies greatly from site-to-site, it seems likely that it is influenced by station-specific procedures yet to be identified; the ECC O_3 drop-off probably has more than one single cause.

Affected data archives such as SHADOZ (https://tropo.gsfc.nasa.gov/shadoz/), the World Ozone and Ultraviolet Data Centre (WOUDC.org), and the Network for the Detection of Atmospheric Composition Change (NDACC; ndaccdemo.org) are posting caveats and flagging affected profiles. Ongoing research is directed at identifying the cause of the low O_3 bias.

We emphasize that all reprocessed data are more accurate than unhomogenized data. For affected sites, data before the drop-off are highly reliable and even affected data are accurate for satellite validation and algorithms, process studies, and model evaluation because the apparent drop-off averages less than 5 %. However, the affected data are not appropriate for calculations of TCO or above-50-hPa stratospheric trends or satellite drift.

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References

Deshler, T., et al. (2008), Atmospheric comparison of electrochemical cell ozonesondes from different manufacturers, and with different cathode solution strengths: The Balloon Experiment on Standards for Ozonesondes, *J. Geophys. Res*., 113, D04307, doi:10.1029/2007JD008975.

Deshler, T., R. Stuebi, F. J. Schmidlin, J. L. Mercer, H. G. J. Smit, B. J. Johnson, R. Kivi, R., and B. Nardi (2017), Methods to homogenize electrochemical concentration cell (ECC) ozonesonde measurements across changes in sensing solution concentration or ozonesonde manufacturer, *Atmos. Meas. Tech*., 10, 2021–2043, doi:10.5194/amt-10-2021-2017.

Froidevaux, L., et al. (2008), Validation of Aura Microwave Limb Sounder stratospheric ozone measurements, *J. Geophys. Res*., 113, D15S20, doi:10.1029/2007JD008771.

Hubert, D., et al. (2016), Ground-based assessment of the bias and long-term stability of 14 limb and occultation ozone profile data records, *Atmos. Meas. Tech*., 9, 2497–2534, https://doi.org/10.5194/amt-9-2497-2016.

Livesey, N. J., et al. (2018), Version 4.2x-3.1 Level 2 data quality and description document, JPL D-33509 Rev. B. [Available at https://mls.jpl.nasa.gov/data/v4-2_data_quality_document.pdf.]

McPeters, R., Kroon, M., Labow, G., Brinksma, E., Balis, D., Petropavlovskikh, I., Veefkind, J. P., Bhartia, P. K., and Levelt, P. F. (2008), Validation of the Aura Ozone Monitoring Instrument total column ozone product, *J. Geophys. Res*., 113, D15S14, doi:10.1029/2007JD008802.

McPeters, R. D., Frith, S., and Labow, G. J. (2015), OMI total column ozone: extending the long-term data record, *Atmos. Meas. Tech.*, 8, 4845–4850, https://doi.org/10.5194/amt-8-4845-2015.

McPeters, R., Frith, S., Kramarova, N., Ziemke, J., and Labow, G. (2019), Trend quality ozone from NPP OMPS: the version 2 processing, *Atmos. Meas. Tech.*, 12, 977–985, https://doi.org/10.5194/amt-12-977-2019.

Morris, G. A., W. D. Komhyr, J. Hirokawa, J. Flynn, B. Lefer, N. Krotkov, F. Ngan (2010), A balloon sounding technique for measuring SO_2 plumes, *J. Atmos. Oceanic Technol.*, 27, 1318–1330. doi: 10.1175/2010JTECHA1436.1

Smit, H. G. J., et al. (2007), Assessment of the performance of ECC-ozonesondes under quasi-flight conditions in the environmental simulation chamber: Insights from the Jülich Ozone Sonde Intercomparison Experiment (JOSIE), *J. Geophys. Res.*, 112, D19306, doi:10.1029/2006JD007308.

Smit, H. G. J., and the Panel for the Assessment of Standard Operating Procedures for Ozonesondes (ASOPOS) (2012), Guidelines for homogenization of ozonesonde data, SI2N/O3S-DQA activity as part of “Past changes in the vertical distribution of ozone assessment”. [Available at http://www-das.uwyo.edu/%7Edeshler/NDACC_O3Sondes/O3s_DQA/O3S-DQA-Guidelines%20Homogenization-V2-19November2012.pdf.]

Smit, H. G. J., and the Panel for the Assessment of Standard Operating Procedures for Ozonesondes (ASOPOS) (2014), Quality assurance and quality control for ozonesonde measurements in GAW, World Meteorological Organization, GAW Report 201. [Available at http://www.wmo.int/pages/prog/arep/gaw/documents/FINAL_GAW_201_Oct_2014.pdf.]

Stauffer, R. M., G. A. Morris, A. M. Thompson, E. Joseph, G. J. R. Coetzee, and N. R. Nalli (2014), Propagation of radiosonde pressure sensor errors to ozonesonde measurements, *Atmos. Meas. Tech.*, 7, 65–79, doi:10.5194/amt-7-65-2014.

Sterling, C. W., Johnson, B. J., Oltmans, S. J., Smit, H. G. J., Jordan, A. F., Cullis, P. D., et al. (2017). Homogenizing and estimating the uncertainty in NOAA's long term vertical ozone profile records measured with the electrochemical concentration cell ozonesonde. *Atmos. Meas. Tech*., 11, 3661-3687, https://doi.org/10.5194/amt-2017-397

Tarasick, D. W., J. Davies, H. G. J. Smit, and S. J. Oltmans (2016), A re-evaluated Canadian ozonesonde record: Measurements of the vertical distribution of ozone over Canada from 1966 to 2013, *Atmos. Meas. Tech.*, 9, 195–214, doi:10.5194/amt-9-195-2016.

Thompson, A. M., J. C. Witte, H. G. J. Smit, S. J. Oltmans, B. J. Johnson, V. W. J. H. Kirchhoff, and F. J. Schmidlin (2007), Southern Hemisphere Additional Ozonesondes (SHADOZ) 1998–2004 tropical ozone climatology: 3. Instrumentation, station-to-station variability, and evaluation with simulated flight profiles, *J. Geophys. Res*., 112, D03304, doi:10.1029/2005JD007042.

Thompson, A. M., Witte, J. C., Sterling, C., Jordan, A., Johnson, B. J., Oltmans, S. J., et al. (2017). First reprocessing of Southern Hemisphere Additional Ozonesondes (SHADOZ) profiles (1998-2016). 2. Comparisons with satellites and ground-based instruments, *J. Geophys. Res. Atmos.*, 122, 13000-13025, https://doi.org/10.1002/2017JD27406

Thompson, A. M., H. G. J. Smit, J. C. Witte, R. M. Stauffer, B. J. Johnson, G. Morris, et al. (2019), Ozonesonde quality assurance: The JOSIE-SHADOZ (2017) Experience, *Bull. Amer. Met. Soc.*, 100 (1), 155-171, https://doi.org/10.1175/BAMS-D-17-0311.1

Van Malderen, R., Allaart, M. A. F., De Backer, H., Smit, H. G. J., and De Muer, D. (2016). On instrumental errors and related correction strategies of ozonesondes: Possible effect on calculated ozone trends for the nearby sites Uccle and De Bilt. *Atmos. Meas.Tech*., 9, 3793–3816. https://doi.org/10.5194/amt-9-3793-2016.

Witte, J. C., Thompson, A. M., Smit, H. G. J., Fujiwara, M., Posny, F., Coetzee, G. J. R., et al. (2017). First reprocessing of Southern Hemisphere ADditional Ozone-sondes (SHADOZ) profile records (1998–2015): 1. Methodology and evaluation. *J. Geophys. Res. Atmos*, 122, 6611–6636. https://doi.org/10.1002/2016JD026403.

Witte, J. C., Thompson, A. M., Schmidlin, F. J., Northam, E. T., Wolff, K. R., and Brothers, G. B. (2019). The NASA Wallops Flight Facility digital ozonesonde record: Reprocessing, uncertainties, and dual launches. Journal of Geophysical Research: Atmospheres, 124, 3565–3582. https://doi.org/10.1029/2018JD030098

Table 1. ECC type, total samples, lat/lon, KI solution type (SST), the 25th percentile, mean, and 75th percentile TCO differences with OMI (October 2004-present), date and amount of drop-off if applicable, and ground-based instrument if applicable are listed. Sites with a > 2 % drop in TCO relative to OMI (Section 2.3) are in bold. Type1 is EnSci (Westminster, CO, USA) and Type2 is Science Pump Corporation (SPC; Camden, NJ, USA).

Site	ECC	N	Lat (°)	Lon (°)	KI SST	OMI 25th (%)	OMI μ (%)	OMI 75th (%)	Drop-Off	TCO Drop (%)	Ground TCO
Alert	**Type1**	**645**	**82.49**	**-62.34**	**1.0%, Full**	**-0.6**	**1.0**	**3.1**	**02/2016**	**-3.6**	**Brewer**
Eureka	**Type1**	**922**	**79.98**	**-85.94**	**1.0%, Full**	**-0.4**	**1.9**	**4.5**	**01/2016**	**-2.4**	**Brewer**
Resolute	**Type1**	**540**	**74.7**	**-94.96**	**1.0%, Full**	**-4.8**	**-2.2**	**0.6**	**03/2015**	**-4.4**	**Brewer**
Churchill	**Type1**	**417**	**58.74**	**-94.07**	**1.0%, Full**	**-1.1**	**0.7**	**3.3**	**06/2016**	**-7.5**	**Brewer**
Edmonton	**Type1**	**674**	**53.54**	**-114.1**	**1.0%, Full**	**-2.9**	**-0.4**	**2.9**	**08/2016**	**-3.3**	**Brewer**
Goose Bay	Type1	663	53.31	-60.36	1.0%, Full	-1.9	0.7	3.4	N/A	N/A	Brewer
De Bilt	Type2	736	52.1	5.18	1.0%, Full	-0.6	1.3	2.9	N/A	N/A	Brewer
Uccle	Type1	2140	50.8	4.35	0.5%, Half	-1.5	0.0	2.0	N/A	N/A	Brewer
Kelowna	**Type1**	**664**	**49.93**	**-119.4**	**1.0%, Full**	**1.4**	**3.4**	**5.9**	**03/2015**	**-4.4**	**N/A**
Payerne	Type1	2191	46.49	6.57	0.5%, Half	-2.5	-0.7	0.9	N/A	N/A	N/A
Yarmouth	**Type1**	**616**	**43.87**	**-66.11**	**1.0%, Full**	**-0.2**	**2.4**	**5.3**	**04/2016**	**-8.2**	**N/A**
Sapporo	Type1	373	43.06	141.33	0.5%, Half	1.0	2.7	4.4	N/A	N/A	Dobson
Trinidad Head	Type1	772	40.8	-124.16	1.0%, 1/10	-2.1	-0.2	1.6	N/A	N/A	N/A
Madrid	Type2	680	40.47	-3.58	1.0%, Full	-2.1	-0.3	1.6	N/A	N/A	Brewer
Boulder	Type1	816	40	-105.25	1.0%, 1/10	-2.1	-0.3	2.0	N/A	N/A	Dobson
Wallops Island	Type2	773	37.93	-75.48	1.0%, Full	-2.5	-0.3	1.8	N/A	N/A	Dobson
Tateno	Type1	430	36.06	140.13	0.5%, Half	0.8	2.6	4.3	N/A	N/A	Dobson, Brewer
Huntsville	Type1	759	34.72	-86.64	1.0%, 1/10	-1.6	0.0	1.9	N/A	N/A	N/A
Naha	Type1	403	26.21	127.69	0.5%, Half	0.2	1.7	3.5	N/A	N/A	Dobson
Hong Kong	Type2	690	22.31	114.17	1.0%, Full	-7.0	-4.6	-2.1	N/A	N/A	N/A
Hanoi	**Type1**	**264**	**21.01**	**105.8**	**0.5%, Half**	**-4.1**	**-1.8**	**0.5**	**11/2014**	**-2.6**	**N/A**
Hilo	**Type1**	**711**	**19.43**	**-155.04**	**1.0%, 1/10**	**-3.7**	**-1.9**	**0.2**	**08/2014**	**-3**	**Dobson, Brewer**
Costa Rica	**Type1**	**605**	**9.94**	**-84.04**	**1.0%, 1/10**	**-3.1**	**-0.8**	**1.9**	**01/2016**	**-5.5**	**N/A**
Paramaribo	Type2	517	5.8	-55.21	1.0%, Full	-5.0	-2.5	-0.1	N/A	N/A	Brewer
Kuala Lumpur	Type1	264	2.73	101.27	0.5%, Half	-7.3	-4.5	-1.3	N/A	N/A	N/A
San Cristobal	**Type1**	**168**	**-0.92**	**-89.62**	**1.0%, 1/10**	**-4.9**	**-0.8**	**2.4**	**08/2013**	**-5.2**	**N/A**
Nairobi	**Type1**	**596**	**-1.27**	**36.8**	**0.5%, Half**	**-3.7**	**-2.1**	**-0.4**	**06/2015**	**-2.2**	**N/A**
Natal	**Type2**	**400**	**-5.42**	**-35.38**	**1.0%, Full**	**-3.6**	**-1.5**	**1.0**	**09/2013**	**-2.7**	**Dobson**
Watukosek	Type1	115	-7.5	112.6	2.0%, None	-3.4	-1.9	0.4	N/A	N/A	N/A
Ascension	**Type1**	**394**	**-7.58**	**-14.24**	**0.5%, Half**	**-6.0**	**-2.8**	**0.4**	**03/2016**	**-3.3**	**N/A**
Samoa	**Type1**	**474**	**-14.23**	**-170.56**	**1.0%, 1/10**	**-3.0**	**-1.2**	**0.9**	**01/2017**	**-3.1**	**Dobson**
Fiji	**Type1**	**200**	**-18.13**	**178.4**	**1.0%, 1/10**	**-3.1**	**-0.5**	**2.2**	**12/2015**	**-4**	**N/A**
Réunion	Type1	449	-21.06	55.48	0.5%, Half	-2.0	0.3	2.4	N/A	N/A	SAOZ
Irene	Type2	212	-25.9	28.22	1.0%, Full	-1.3	1.3	4.3	N/A	N/A	Dobson
Broadmeadows	Type2	667	-37.69	144.95	1.0%, Full	-0.9	0.6	2.7	N/A	N/A	Dobson
Lauder	**Type1**	**705**	**-45**	**169.68**	**0.5%, Half**	**-3.3**	**-1.3**	**0.8**	**09/2016**	**-2.2**	**Dobson**
Macquarie	Type2	675	-54.5	158.95	1.0%, Full	-4.6	-2.4	0.1	N/A	N/A	Dobson

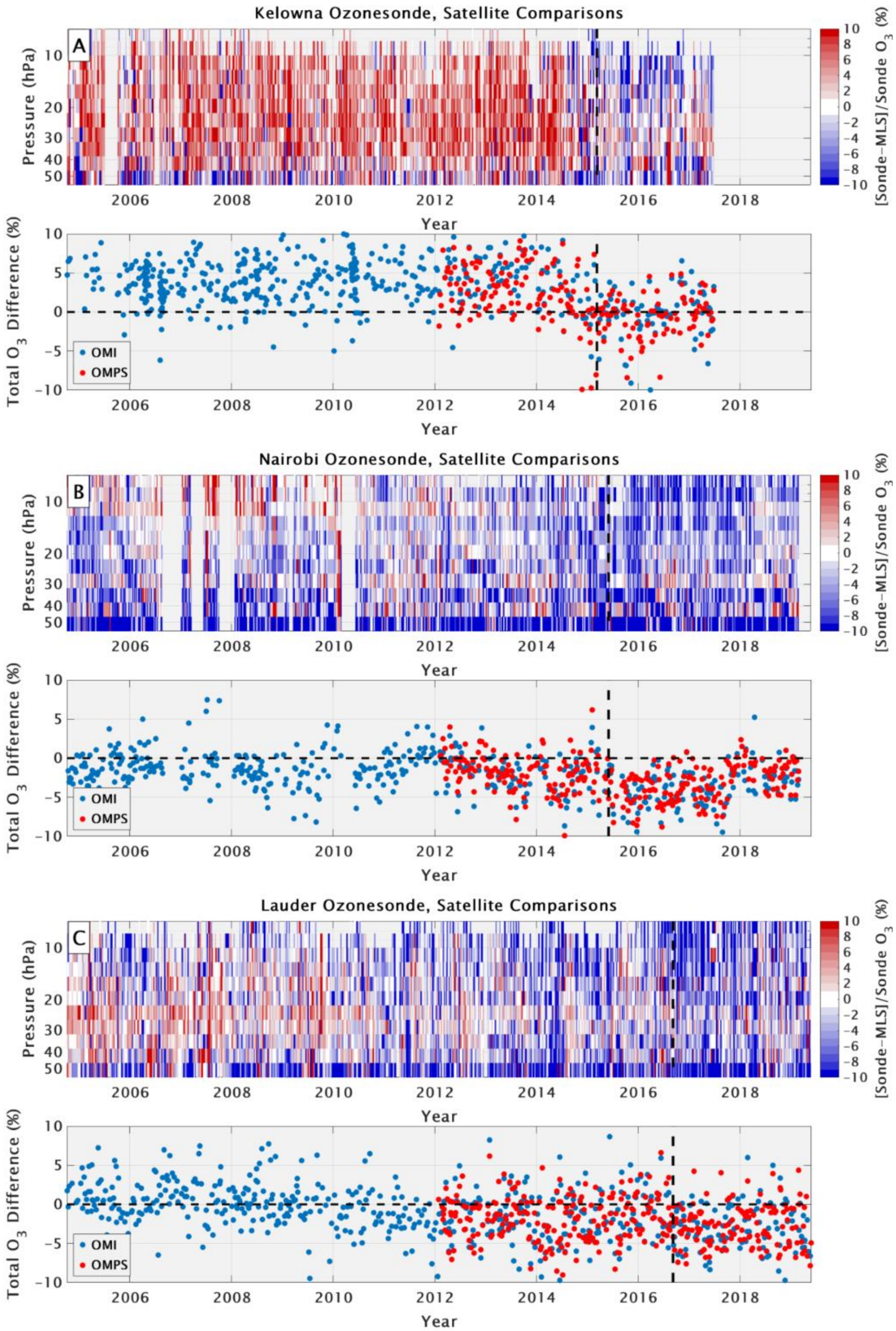


Figure 1. Time series of comparisons at Kelowna (A; data end in June 2017), Nairobi (B), and Lauder (C) between ECC ozonesondes and Aura MLS stratospheric O_3 profiles (top panels), and OMI (blue dots) and OMPS (red dots) TCO (bottom panels). Red or blue colors on the top panels indicate where the ECC O_3 is greater or less than MLS. Horizontal dashed lines indicate the 0 % line for TCO comparisons. Vertical dashed lines indicate the beginning of the low bias at each site (see Table 1 for dates), marked by a sudden drop in O_3 relative to satellite measurements.

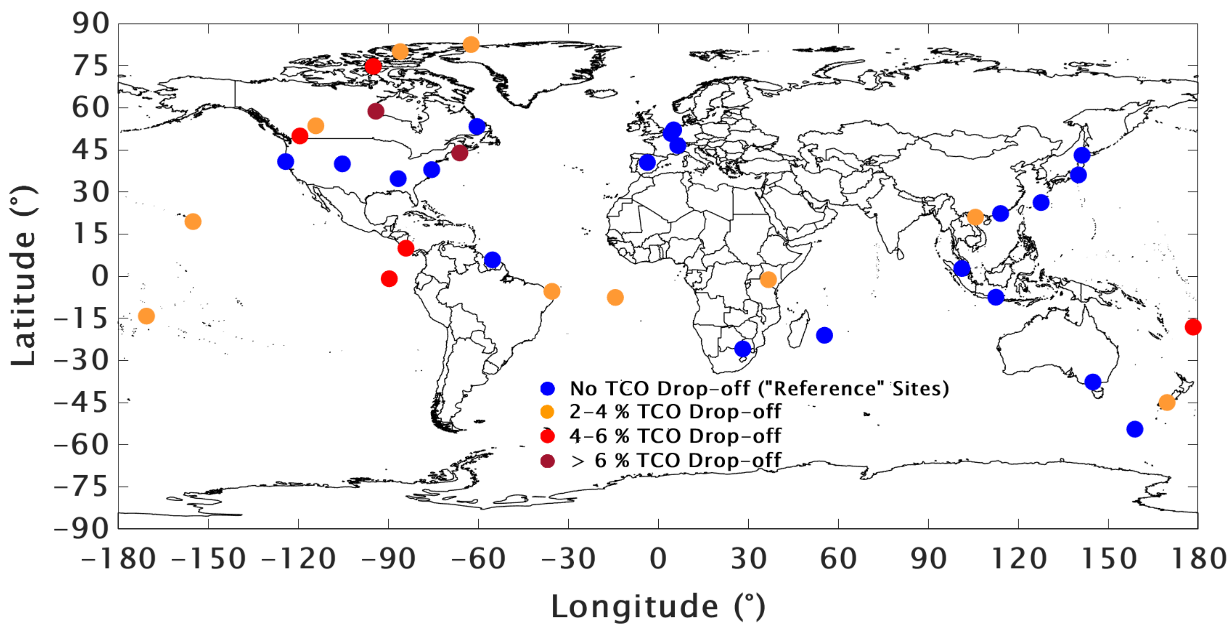


Figure 2. Map of all 37 ECC ozonesonde sites considered in this study. The blue dots indicate sites that show no detectable TCO drop-off relative to OMI TCO. We call these sites "reference" sites. The orange, red, and purple dots indicate sites that exhibit drops of 2-4 %, 4-6 %, and over 6 % relative to OMI TCO. The method for computing the values shown on this figure and in Table 1 are explained in Section 2.3.

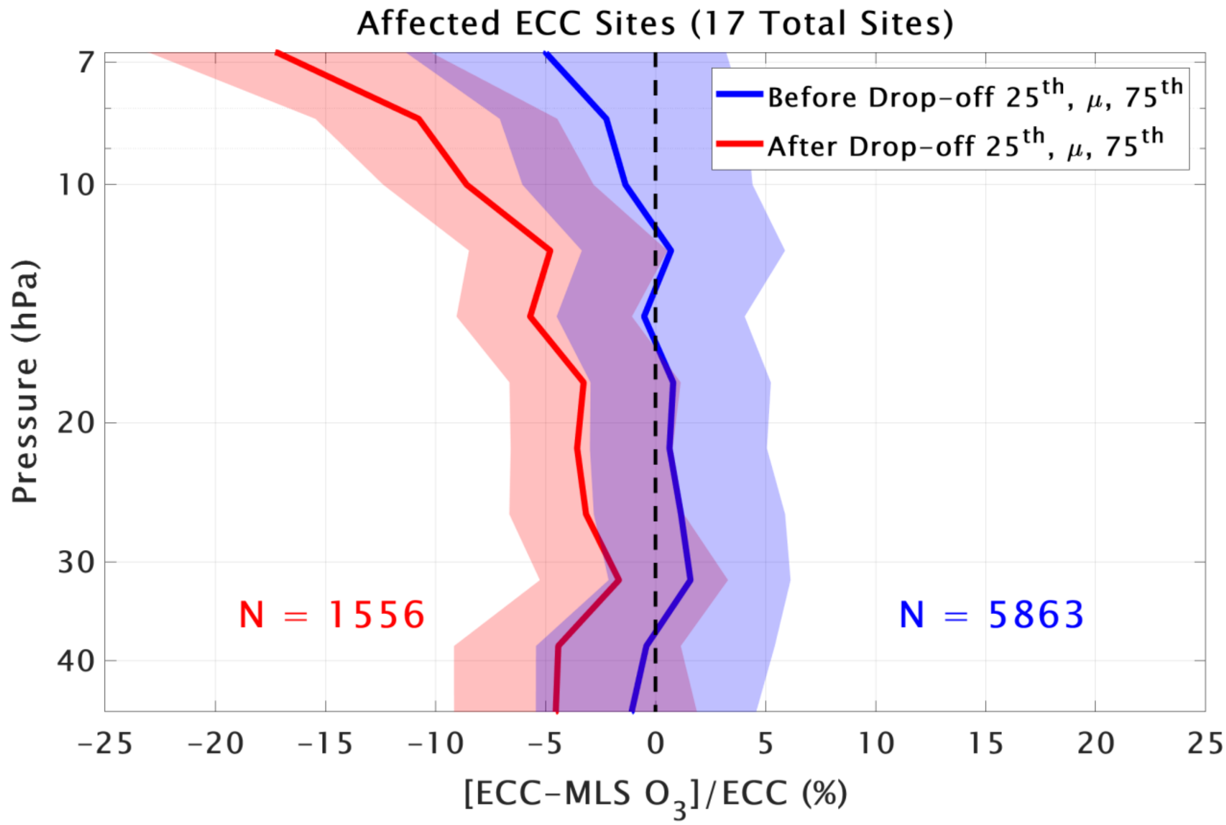


Figure 3. A composite of comparisons between ECC ozonesonde and Aura MLS stratospheric O_3 profiles from before the drop-off at each site (blue; dates of drop-off are in Table 1), and during the period after the drop-off (red). The shading indicates the 25th to 75th percentile, with mean values shown by the solid lines. ECC sonde sample numbers are shown for each period in the lower portion of the figure.

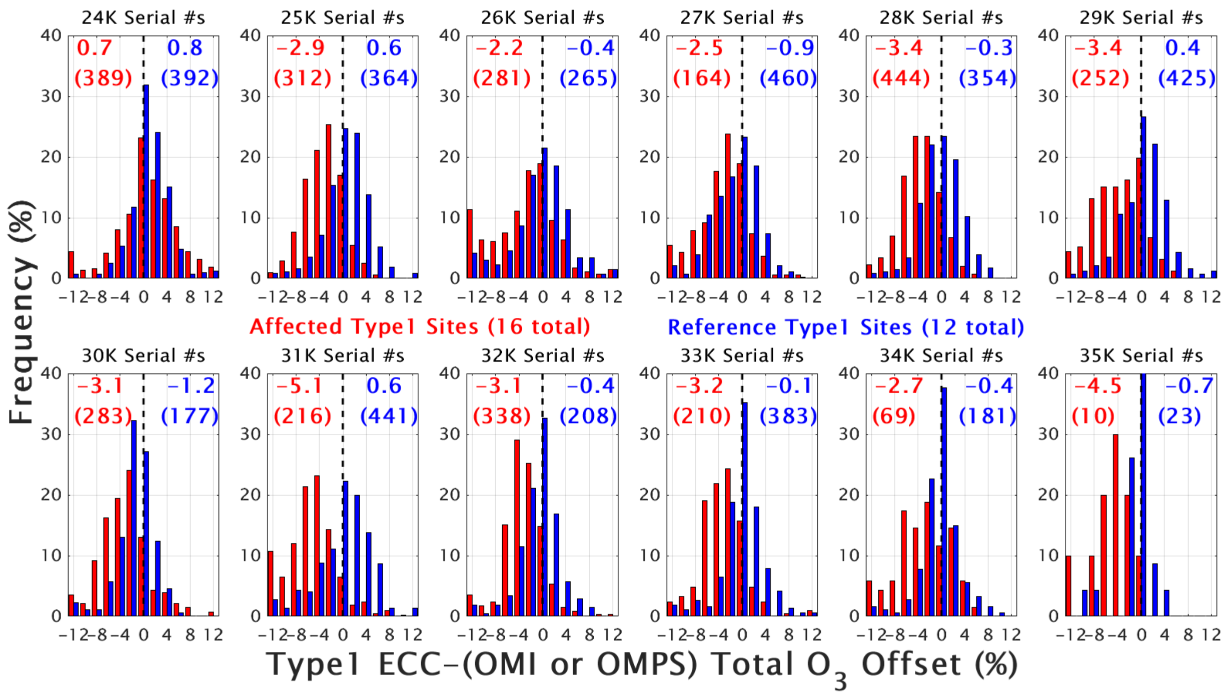
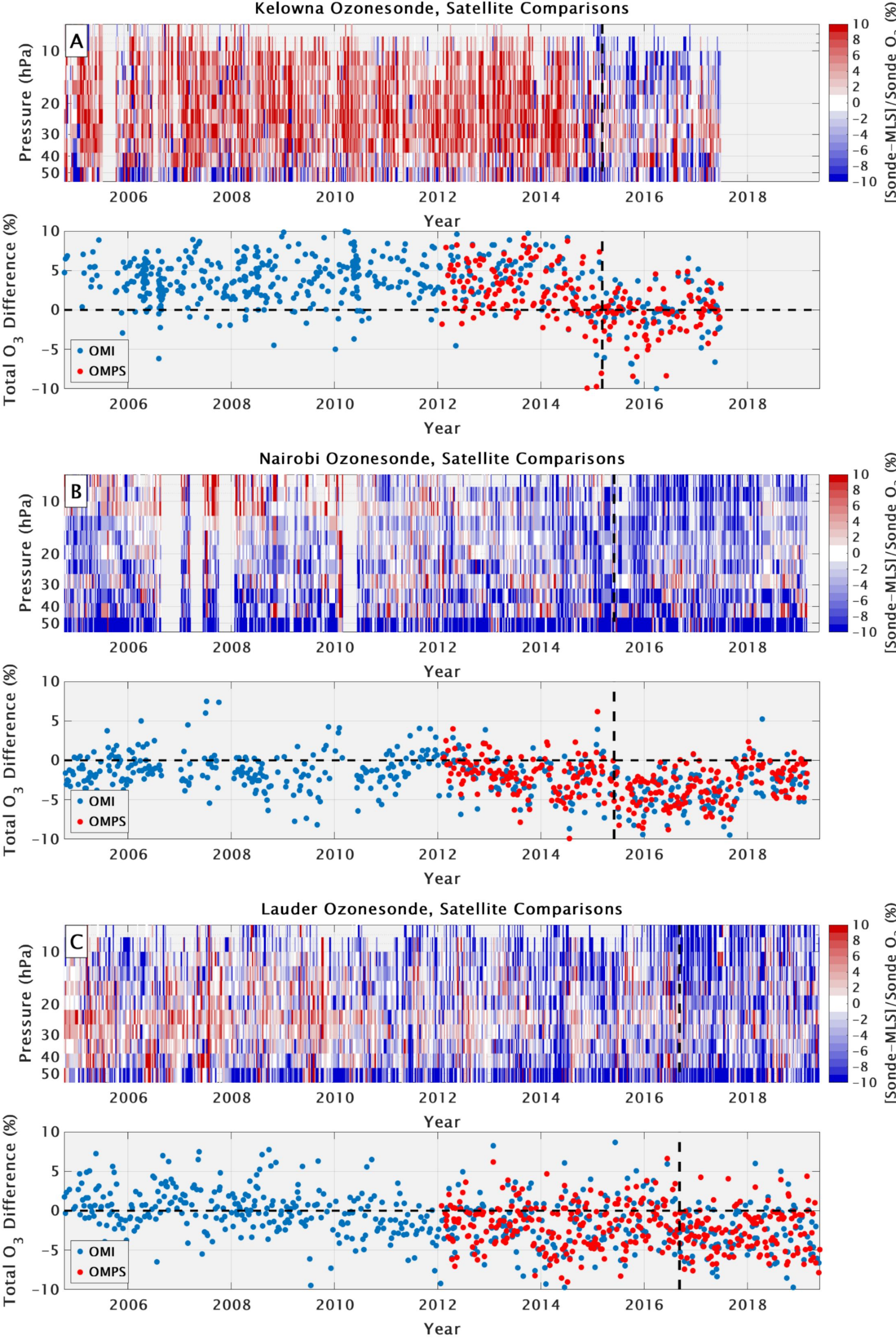


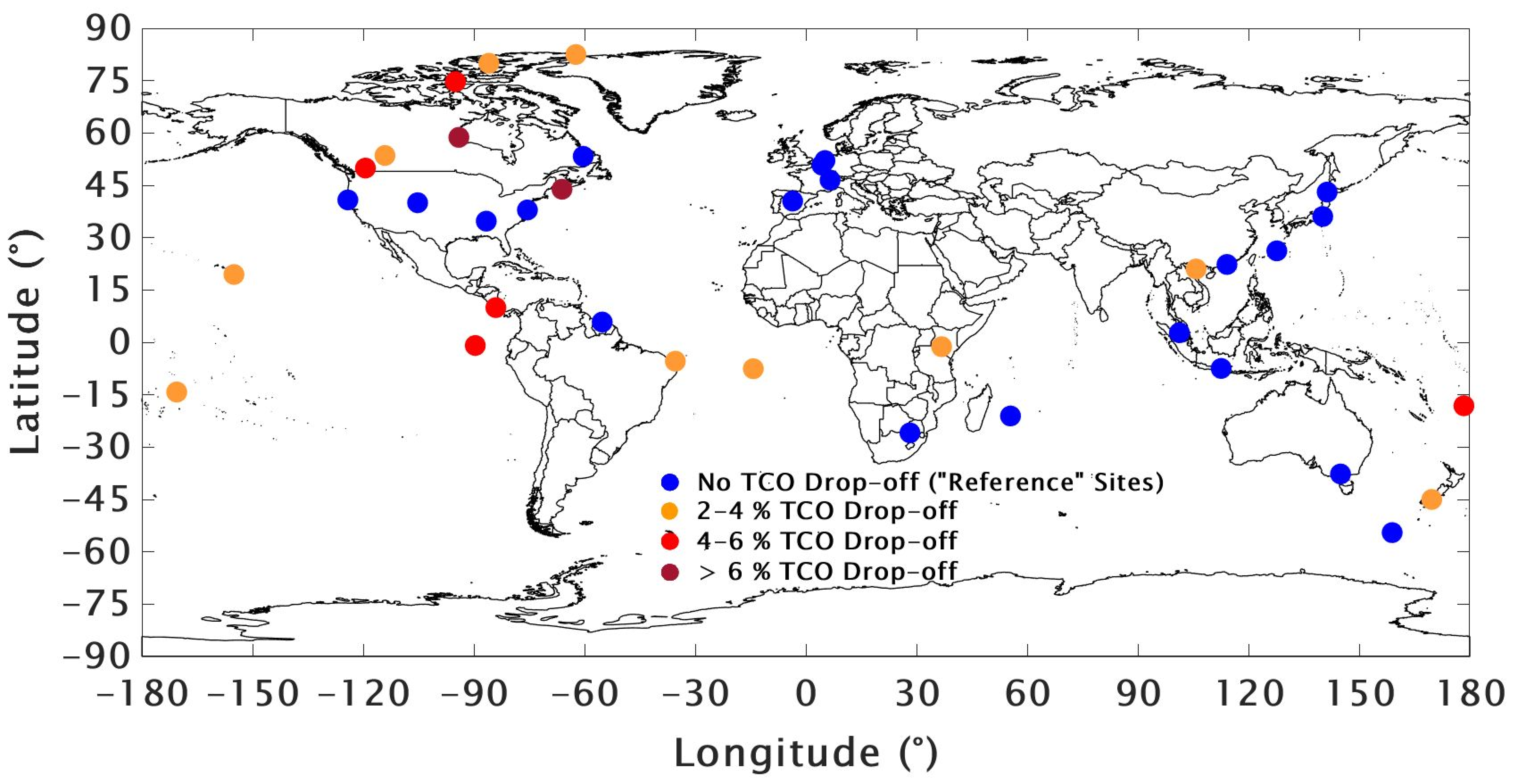
Figure 4. Histograms of Type1 ECC ozonesonde comparisons with OMI and OMPS TCO in percent difference (ECC-satellite/ECC). Comparisons are separated into every 1000 serial numbers from 24000 to 35000, and by sites affected (red) and reference (blue) by the ozonesonde drop-off. Data are binned every 2 % from -12 to 12 %. The median percent difference for each set is listed at top of the panels, with the total number of comparisons with OMI and OMPS in parentheses. For example, Type1 31000s serial numbers at affected sites have a median bias of -5.1 % compared to OMI and OMPS, with 216 total ECC/satellite comparisons (sondes are double-counted for comparison to both OMI and OMPS overpasses). Reference site Type1 31000s serial numbers have a median bias of +0.6 %, with 441 total ECC/satellite comparisons.

Figure1.



Kelowna Ozonesonde, Satellite Comparisons
A
Pressure (hPa)
[Sonde−MLS]/Sonde O_3 (%)
Year
Total O_3 Difference (%)
OMI
OMPS
Nairobi Ozonesonde, Satellite Comparisons
B
Lauder Ozonesonde, Satellite Comparisons
C

Figure2.



90
75
60
45
30
15
0
−15
−30
−45
−60
−75
−90
Latitude (°)
−180
−150
−120
−90
−60
−30
0
30
60
90
120
150
180
Longitude (°)
No TCO Drop-off ("Reference" Sites)
2-4 % TCO Drop-off
4-6 % TCO Drop-off
> 6 % TCO Drop-off

Figure3.

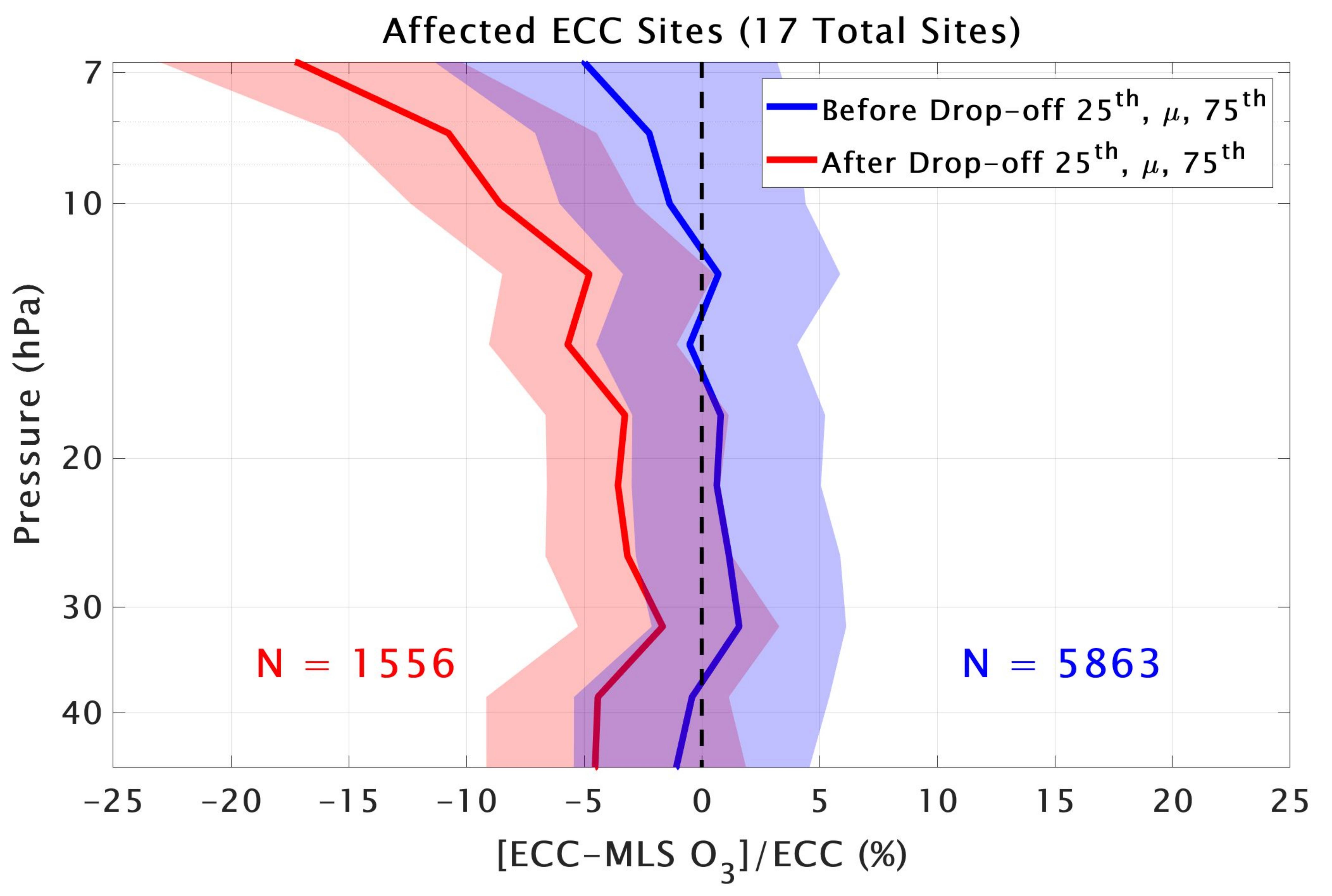
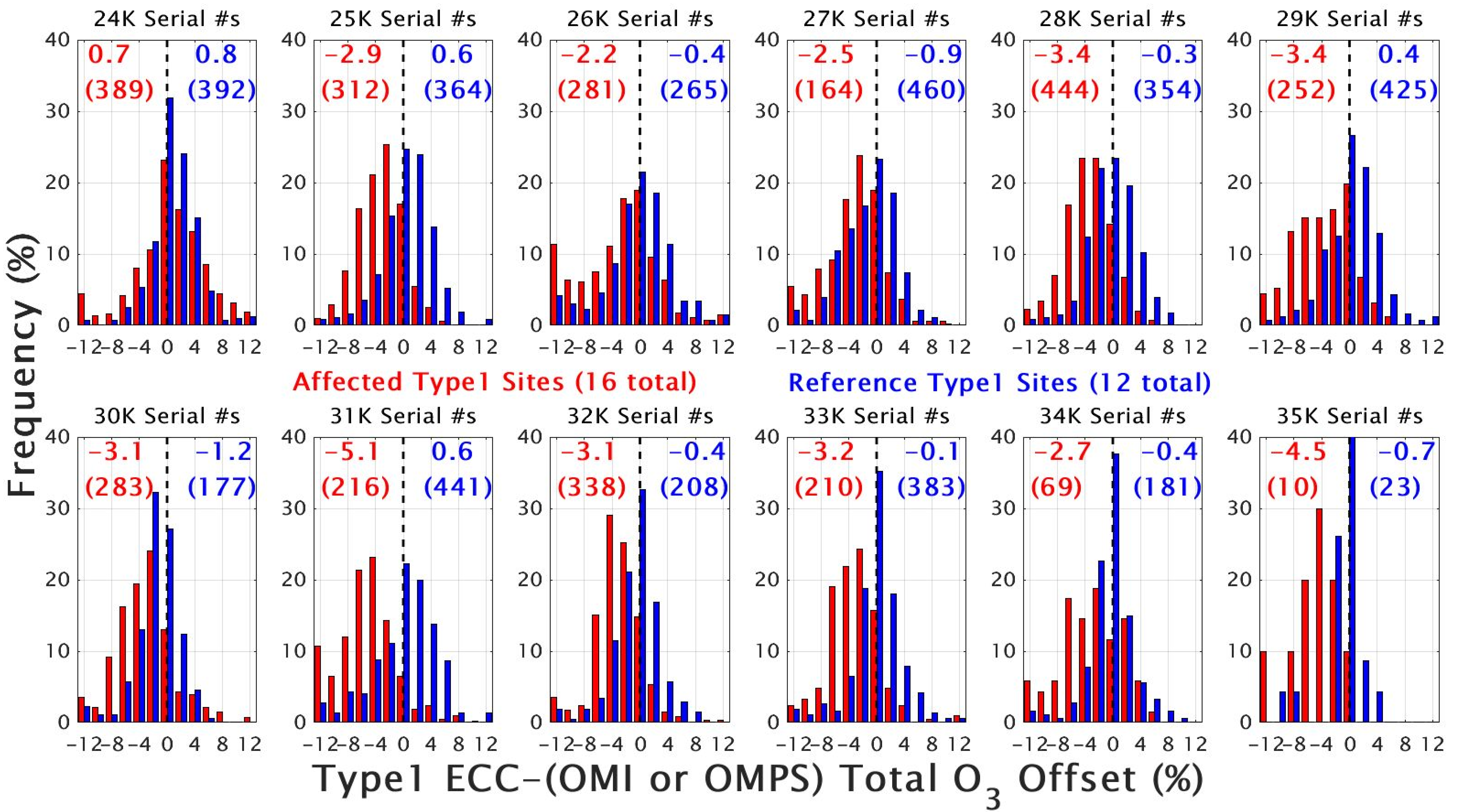

Affected ECC Sites (17 Total Sites)
Before Drop-off 25th, μ, 75th
After Drop-off 25th, μ, 75th
N = 1556
N = 5863
Pressure (hPa)
7
10
20
30
40
-25
-20
-15
-10
-5
0
5
10
15
20
25
[ECC-MLS O3]/ECC (%)

Figure4.



24K Serial #s
0.7
(389)
0.8
(392)
25K Serial #s
−2.9
(312)
0.6
(364)
26K Serial #s
−2.2
(281)
−0.4
(265)
27K Serial #s
−2.5
(164)
−0.9
(460)
28K Serial #s
−3.4
(444)
−0.3
(354)
29K Serial #s
−3.4
(252)
0.4
(425)
Affected Type1 Sites (16 total)
Reference Type1 Sites (12 total)
30K Serial #s
−3.1
(283)
−1.2
(177)
31K Serial #s
−5.1
(216)
0.6
(441)
32K Serial #s
−3.1
(338)
−0.4
(208)
33K Serial #s
−3.2
(210)
−0.1
(383)
34K Serial #s
−2.7
(69)
−0.4
(181)
35K Serial #s
−4.5
(10)
−0.7
(23)
Frequency (%)
Type1 ECC−(OMI or OMPS) Total O3 Offset (%)