

1 Tropospheric and Stratospheric Ozone Profiles during the 2019 2 TROPomi vaLIdation eXperiment (TROLIX-19)

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18 **Abstract** A TROPospheric Monitoring Instrument (TROPOMI) validation campaign was held in the
19 Netherlands based at the CESAR (Cabauw Experimental Site for Atmospheric Research) Observatory
20 during September 2019. The TROPomi vaLIdation eXperiment (TROLIX-19) consisted of active and
21 passive remote sensing platforms in conjunction with several balloon-borne and surface chemical (e.g.
22 ozone and nitrogen dioxide) measurements. The goal of this joint NASA-KNMI geophysical validation
23 campaign was to make intensive observations in the TROPOMI domain in order to be able to establish
24 the quality of the L2 satellite data products under realistic conditions, such as non-idealized conditions
25 with varying cloud cover and a range of atmospheric conditions at a rural site. The research presented
26 here focuses on using ozone lidars from NASA's Goddard Space Flight Center to better evaluate the
27 characterization of ozone throughout TROLIX-19. Results of comparisons to the lidar systems with
28 balloon, space-borne, and ground-based passive measurements are shown. In addition, results are
29 compared to a global coupled chemistry meteorology model to illustrate the vertical variability and
30 columnar amounts of both tropospheric and stratospheric ozone during the campaign period.

31 **1 Introduction**

32 In September 2019, a joint Royal Netherlands Meteorological Institute (KNMI) and the U.S. National
33 Aeronautics and Space Administration (NASA) field campaign was performed in the Netherlands, based
34 at the Cabauw Experimental Site for Atmospheric Research (CESAR, 51.97° N, 4.93° E), to provide the
35 scientific community with additional information to further understand and evaluate the Copernicus
36 Sentinel-5 Precursor mission (S-5P) TROPOspheric Monitoring Instrument (TROPOMI) instrument
37 (<https://sentinels.copernicus.eu/web/sentinel/missions/sentinel-5p>). The main objective of the
38 Copernicus Sentinel-5P mission is to perform atmospheric measurements with high spatio-temporal
39 resolution, to be used for scientific studies and monitoring of air quality and chemical transport
40 (https://www.esa.int/Applications/Observing_the_Earth/Copernicus/Sentinel-5P).

41 To properly support satellite evaluation, the 2019 TROPomi vaLIdation eXperiment (TROLIX-19)
42 campaign was designed to bring together many active and passive remote sensing platforms in
43 conjunction with several balloon-borne, airborne and surface measurements. Specifically, the
44 observations were established to provide geophysical verification in order to establish the quality of
45 TROPOMI Level 2 (L2) main data products under realistic non-idealized conditions with varying cloud
46 cover and a wide range of atmospheric conditions. Cabauw, using its comprehensive in-situ and remote
47 sensing observation program in and around the 213 m meteorological tower ([https://ruisdael-
48 observatory.nl/trolix19-tropomi-validation-experiment-2019/](https://ruisdael-observatory.nl/trolix19-tropomi-validation-experiment-2019/)) was the main site of the campaign with
49 focus on vertical profiling using lidar instruments for aerosols, clouds, water vapor, tropospheric and
50 stratospheric ozone, as well as balloon-borne sensors for nitrogen dioxide (NO₂) and ozone (Figure 1).
51 Although this work focuses primarily on the ozone lidar profiling during the study, the larger campaign
52 overview, background, and motivation can be found in Apituley *et al.* (2019; 2020) or Kreher *et al.*
53 (2020a).

54 One main goal of this work is also to understand ozone profile retrievals as they relate to upcoming
55 satellite endeavors. As NASA prepares to launch its first geostationary air quality satellite “Tropospheric

56 Emissions: Monitoring of Pollution” (TEMPO) this work also specifically establishes a paradigm of
57 evaluation for TEMPO-derived products such as tropospheric ozone columns and a 0-2km tropospheric
58 ozone product. An analogous geo-stationary air quality satellite, the Copernicus Sentinel-4 mission (S-
59 4), <https://sentinels.copernicus.eu/web/sentinel/missions/sentinel-4>), will provide hourly data on
60 tropospheric constituents over Europe and the CESAR site is directly within the satellite’s field of
61 regard. Due to the finer spatial footprint, increased temporal frequency and vertical extent of the
62 TEMPO tropospheric ozone retrievals, ozone lidars are an ideal platform to perform future evaluations
63 of the products, which builds from recent work done in Johnson *et al.*, 2018. Specifically, this work will
64 investigate the results from the combination of having a co-located NASA tropospheric (Sullivan *et al.*,
65 2014) and stratospheric ozone lidars (McGee *et al.*, 1991) in order to obtain an entire vertical profile of
66 ozone from ~0.2km to 50km.

67 For the first time, this transportable combination of lidars is able to explicitly derive diurnally varying
68 tropospheric and total ozone columns, which are compared directly to measurements obtained by
69 ground-based passive sensors, current satellite instrumentation and chemical transport models. In
70 Section 2 we present all available data and methods used in this work, across the various platforms
71 during the TROLIX-19 study. Section 3 focuses on comparisons of the tropospheric ozone retrievals of
72 the vertical profiles of ozone within the troposphere and columnar reductions of 0-10 km and 0-2 km.
73 Comparisons of lidar data with available complete ozone profiles (Sec 4) and columnar amounts (Sec 5)
74 from several platforms and chemical transport models are also presented to further understand the
75 quality of satellite derived ozone profiles during the TROLIX-19 period.

76

77 2 Data and Methods

78 Descriptions of the various observational and model data sets and used in this study are below, including
79 a summary table (Table 1).

80 2.1 NASA Ozone Differential Absorption Lidars (DIAL)

81 NASA deployed and operated two ozone lidars during TROLIX-19 at the Cabauw site near the CESAR
82 tower to observe temporal and vertical gradients in tropospheric and stratospheric ozone. This was the
83 first dual-deployment of these lidars, in which the tropospheric ozone lidar measured between the near
84 surface (about 0.2 km) to a height of about 18 km and the stratospheric lidar during night-time from 15
85 km upwards to nearly 50 km, providing complete hybrid ozone profiles for the campaign period.

86 Measurements were made during periods of mostly clear skies, although occasional cloud cover did
87 enter the measurement period.

88 The NASA GSFC Mobile Stratospheric Ozone Lidar Trailer Experiment (STROZ-LITE) has been a
89 participant in the Network for the Detection of Atmospheric Composition Change (NDACC) since its
90 inception and is housed in a 12.5m container allowing for transport around the world (McGee et al.,
91 1991). The lidar instrument transmits two wavelengths, 308 nm from a XeCl excimer laser, and 355 nm
92 from a ND:YAG laser to derive ozone number density profiles, which have historically served as an
93 intercomparison data set for other NDACC ozone lidars (recent intercomparison can be found at Wing *et*
94 *al.*, 2020; 2021).

95 The NASA GSFC TROPOZ has been developed in a transportable 13.5m trailer to take routine
96 measurements of tropospheric ozone near the Baltimore–Washington, D.C. area as well as various
97 campaign locations (Sullivan *et al.*, 2014; 2015,2019, Leblanc *et al.*, 2018). This instrument, which
98 utilizes a ND:YAG laser and Raman cell, has been developed as part of the ground-based Tropospheric
99 Ozone Lidar NETwork (TOLNet, <https://www-air.larc.nasa.gov/missions/TOLNet/>), which currently
100 consists stations across the North America (<http://www-air.larc.nasa.gov/missions/TOLNet/>). The
101 primary purposes of the instruments within TOLNet are to provide regular, high-fidelity profile
102 measurements of ozone within the troposphere for satellite and model evaluation. This lidar also
103 operates routinely for the Network for the Detection of Atmospheric Composition Change (NDACC).

104 More than thirty NDACC ground-based Lidar instruments
105 (https://lidar.jpl.nasa.gov/ndacc/index_ndacc.php) deployed worldwide from Pole to Pole are
106 monitoring atmospheric ozone, temperature, aerosols, water vapor, and polar stratospheric clouds.
107 Both lidars collect backscattered radiation with a large primary telescope and a 10cm telescope for near
108 field channels. Spectral separation is accomplished using dichroic beam-splitters and interference filters.
109 For the stratospheric system, five return wavelengths are recorded: the two transmitted wavelengths, and
110 the nitrogen Raman scattered radiation from each of the transmitted beams 332 nm and 382 nm, and the
111 408 nm water vapor channel. In this arrangement for TROLIX-19, the tropospheric system pumped the
112 Raman cell with the fourth harmonic (266 nm), which resulted in conversion to 289 nm and 299 nm
113 using a single hydrogen/deuterium Raman cell. All of the signals are further split to improve the
114 dynamic range of the respective lidar optical detection chains and are then amplified, discriminated and
115 recorded using photon counting techniques.

116 During TROLIX-19, the STROZ-LITE was operated on cloud free nights, with measurements lasting
117 between 2-4 hours to obtain enough signal to properly retrieve the entire stratospheric ozone profile. The
118 TROPOZ was operated during daytime and night time to provide tropospheric ozone profiles. For
119 instances of TROPOMI overpasses, campaign ozonesondes, or coincident stratospheric ozone lidar
120 measurements, the TROPOZ reported data is averaged for 30 minutes, centered around the satellite
121 overpass or launch time. This temporal period of averaging has been optimized in several cases to avoid
122 cloud contamination. For all other times during the TROPOZ operation, the data has been averaged to
123 10 minutes, which is suitable under most clear sky conditions to retrieve ozone information within the
124 entire troposphere. A brief description and community standardized definitions of the uncertainty budget
125 of the lidar measurements presented in this paper can be found in Sullivan *et al.*, 2014, Leblanc *et al.*,
126 2016 and Leblanc *et al.*, 2018. The maximum statistical uncertainties for the two GSFC lidars vary from
127 night to night depending on atmospheric conditions and laser power fluctuations. They are mostly within
128 10-20% for 5 min and 5-8% for 30 min integrations throughout the atmosphere. Within overlapping

129 measurement regions in the upper troposphere/lower stratosphere, they are different at the same altitude
130 due to laser performance and telescope/detector efficiency differences, and are therefore joined
131 manually for this work based on appropriate signal to noise and uncertainty estimates.

132 **2.2 Ground Based Passive Sensors and Ozonesondes**

133 **2.2.1 Pandora Spectrometer Instrument**

134 A Pandora spectrometer instrument (#118) has been used to measure columnar amounts of trace gases in
135 the atmosphere at 3–5-minute resolution at the Cabauw site since 2016 and previously used for the
136 second Cabauw Intercomparison of Nitrogen Dioxide (CINDI-2) campaign (Kreher et al., 2020). Using
137 the theoretical solar spectrum as a reference, Pandora determines trace gas amounts using differential
138 optical absorption spectroscopy (DOAS). This attributes in principal these differences in spectra
139 measured by Pandora to the presence of trace gases within the atmosphere (*i.e.* the difference between
140 the theoretical solar spectrum and measured spectrum is caused by absorption of trace gas species). For
141 this study, L2 direct sun columnar values of ozone are used, although retrievals of nitrogen dioxide are
142 also operationally acquired. Data used passed the strictest QC/QA estimate (Flags = 10) and was
143 obtained from the Pandonia Global Network (<http://data.pandonia-global-network.org/>).

144 **2.2.2 Brewer MKIII Spectrophotometer**

145 A Brewer MKIII spectrometer instrument (#189) has been used to measure daily columnar amounts of
146 ozone in the atmosphere at the KNMI/De Bilt (30km NE of Cabauw, 52.10° N, 5.18° E). Brewer #189
147 has been operated continuously since 1 October 2006. It replaced Brewer #100 which provided
148 observations since 1 January 1994. De Bilt has the longest continuous record of ozone measured with an
149 MKIII instrument in the World Ozone and Ultraviolet Radiation Data Centre (WOUDC) database.
150 The Brewer is specifically designed to provide high accuracy measurement of spectrally resolved UV
151 for satellite evaluation, climatology monitoring and public health to international standards. Similar to

152 Pandora spectrometers, these measurements of total column of trace gases are compared to the measured
153 UV spectrum with the known solar output, and modeling the scattering properties of the atmosphere and
154 have been historically used to evaluate columnar satellite products (McPeters *et al.*, 2007; Wenig *et al.*,
155 2008; Garane *et al.*, 2019). The Brewer is the standard instrument used in the [World Meteorological](#)
156 [Organization](#) ozone monitoring network and for NDACC. This data was obtained at the NDACC
157 website (<https://www-air.larc.nasa.gov/missions/ndacc/data.html>).

158 **2.2.3 Ozonesondes**

159 Ozonesondes have been used to measure vertical profiles of ozone in the atmosphere at the KNMI/De
160 Bilt (30km NE of Cabauw) site since November 1992, and measurements are made weekly, historically
161 at 12 UTC on Thursdays. Description of the Electro Chemical Cell (ECC) details and metadata are
162 summarized in Malderen *et al.*, 2016, which also describes the importance of understanding and
163 reporting changes in ozonesonde operation procedures. During the campaign, in situ measurements of
164 ozone were made using a balloon-borne payload consisting of an ECC ozonesonde (Science Pump
165 Corporation, Serial Numbers: 6A35438, 6A35447, 6A35448, 6A35441) coupled with a radiosonde
166 (Vaisala RS41) and have been used to evaluate TROPOMI tropospheric ozone products in the tropics
167 (Hubert *et al.*, 2021). The ECC technique is widely used for the high vertical resolution measurements of
168 O₃. The ECC consists of two chambers with platinum electrodes immersed in potassium iodide (KI)
169 solutions at different concentrations. The accuracy in the O₃ concentration measured by an ECC
170 ozonesonde is $\pm 5\%$ – 10% up to an altitude of 30 km (Smit *et al.*, 2007; Smit and Thompson *et al.*,
171 2021). This data was obtained at the NDACC website ([https://www-](https://www-air.larc.nasa.gov/missions/ndacc/data.html)
172 [air.larc.nasa.gov/missions/ndacc/data.html](https://www-air.larc.nasa.gov/missions/ndacc/data.html)).

173 **2.3 Satellite Observations and Products**

174 Satellite data used in this work was selected based on the closest retrieval (*i.e.* column, profile) to the
175 CESAR station within ± 2.5 degrees latitude and ± 10 degrees in longitude.

176 **2.3.1 Ozone Mapping and Profiling Suite (OMPS) and MERRA-2 products**

177 The Ozone Mapping and Profiler Suite (OMPS) on the Suomi National Polar-orbiting Partnership (S-
178 NPP) platform consists of three sensors to measure the total column and the vertical distribution of
179 ozone with high spatial and vertical resolutions (Flynn et al., 2006). Daily total column ozone
180 overpasses over Cabauw station from the OMPS Nadir-Mapper (NM) instrument are used in this study.
181 The vertical distribution of ozone in the stratosphere and lower mesosphere is obtained from the OMPS
182 Limb-Profiler (LP) sensor on the Suomi-NPP satellite merging the UV (29.5-52.5 km) and VIS (12.5-
183 35.5 km) bands to provide a full profile from 12.5km to 52.5km (Kramarova *et al.*, 2018). Variations of
184 this merged OMPS-LP retrieval were considered, however the work shown in Arosio *et al.*, 2018,
185 indicates the same overall conclusions would be reached. Further work beyond this manuscript may
186 involve comparing this TROLIX-19 measurement data set to specific experimentally performed satellite
187 retrievals.

188 The Modern-Era Retrospective analysis for Research and Applications, Version 2 (MERRA-2) provides
189 data beginning in 1980 and since August 2004 assimilates NASA's satellite ozone profile observations
190 from Aura Microwave Limb Sounder (MLS) (Livesey et al, 2008) to more comprehensively characterize
191 stratospheric ozone abundance. A residual tropospheric ozone product (Ziemke *et al.*, 2019) is derived
192 using the OMPS NM total column ozone minus the co-located MERRA-2 stratospheric column ozone.
193 Tropopause pressure is derived from MERRA-2 potential vorticity (2.5 PVU) and potential temperature
194 (380 K).

195 **2.3.3 MLS**

196 NASA's Aura Microwave Limb Sounder (MLS) uses microwave emission to measure stratospheric
197 and upper tropospheric constituents, such as ozone. Ozone data (v5) used in this study is binned on
198 various vertical grids and are converted from volume mixing ratio to number density using the

199 coincident MERRA-2 atmosphere state parameters. Both daytime and nighttime data are used in this
200 study and the corresponding closest profile is utilized for comparison.

201 **2.3.4 TROPOMI**

202 In October 2017, the Sentinel-5 Precursor (S5P) mission was launched, carrying the TROPOspheric
203 Monitoring Instrument (TROPOMI), which is a nadir-viewing 108° Field-of-View push-broom grating
204 hyperspectral spectrometer. Starting in August 2019, Sentinel-5P TROPOMI along-track high spatial
205 resolution (approximately 5.5 km at nadir) has been implemented and total ozone columns values used
206 in this work are subsetted from the NASA GES DISC
207 (https://tropomi.gesdisc.eosdis.nasa.gov/data/S5P_TROPOMI_Level2/S5P_L2__O3_TOT_HiR.1/) to
208 provide the Offline 1-Orbit L2 (S5P_L2__O3_TOT_HiR), which is based on the Direct-fitting algorithm
209 (S5P_TO3_GODFIT), comprising a non-linear least squares inversion by comparing the simulated and
210 measured backscattered radiances.

211 Tropospheric Ozone vertical profiles were retrieved using the TOPAS (Tikhonov regularized Ozone
212 Profile retrieval with SCIATRAN) algorithm and were applied to the TROPOMI L1B spectral data
213 version 2, using spectral data between 270 and 329 nm for the retrieval (Mettig *et al.*, 2021). This data
214 set will cover the TROLIX-19 period from 09 September until 28 September; however, it is available
215 outside of this work for specific weeks between June 2018 and October 2019. Since the ozone profiles
216 are very sensitive to absolute calibration at short wavelengths, a re-calibration of the measured radiances
217 is required using comparisons with simulated radiances with ozone limb profiles from collocated
218 satellites used as input. The a priori profiles for ozone are taken from the ozone climatology of Lamsal
219 *et al.* (2004) and the calibration correction spectrum is determined using the radiances modelled with
220 ozone information from collocated MLS/Aura measurements as described in depth throughout Mettig *et*
221 *al.*, 2021.

222

223 2.4 Coupled Chemistry and Meteorology Model

224 The GEOS Composition Forecasting (GEOS-CF, https://gmao.gsfc.nasa.gov/weather_prediction/GEOS-
225 [CF/](https://gmao.gsfc.nasa.gov/weather_prediction/GEOS-CF/), Keller *et al.*, 2021, Knowland *et al.*, 2021) system was chosen to serve as a comparison simulation
226 for this effort, based on its altitude coverage (up to 80 km) and implications for future geostationary
227 satellite use. The system produces global, three-dimensional distributions of atmospheric composition
228 with a spatial resolution of 25km. Using meteorological analyses from other GEOS systems, the GEOS-
229 CF products include a running atmospheric replay to provide near-time estimates of surface pollutant
230 distributions and the composition of the troposphere and stratosphere. Individual case study evaluations
231 using ozone lidar of the GEOS-CF meteorological replay have recently been performed in Dacic *et al.*
232 (2020), Gronoff *et al.* (2021) and Johnson *et al.* (2021). These results will also be used to better evaluate
233 the GEOS-CF as the source of a priori ozone profiles for use in the TEMPO tropospheric ozone
234 retrievals. Model output for this work is used from the closest GEOS-CF model grid cell to the CESAR
235 observatory.

236 3 Tropospheric Ozone Comparisons

237 3.1 Vertical Profiles

238 Example tropospheric ozone profile observations are presented in **Figure 2** for 7 individual observation
239 periods during the TROLIX-19 campaign. Each of the panels show the cloud screened TROPOZ lidar
240 retrievals (top panels) and the corresponding GEOS-CF model output (bottom panels). Pink dots are
241 overlaid to indicate the simulated tropopause altitude based on a blended estimate (TROPPB) which
242 meets criteria of the lowest altitude bin corresponding with either a pressure level above the thermal
243 tropopause (380K) or dynamical (3 PVU) tropopause.

244 In general, the observations and simulations agree quite well in characterizing the broad features that
245 impacted the CESAR site during the TROLIX-19 campaign. However, in each panel there are ozone

246 laminae within the lower troposphere that are not replicated in the model simulation, most notably the
247 underestimation of ozone during the September 20-21 period from 3-5km (Figure 2d-f, black dashed
248 box). However, the model does simulate well the lowered tropopause height and abundance of lower
249 stratospheric ozone observed in the 2 October observations (see Fig. 2g), which is an indication of the
250 model representing the dynamical variability that affects the lowering of the tropopause height. This
251 suggests the model is appropriately capturing the complex dynamics during this period near the upper
252 troposphere, but may not have been initialized with the correct boundary conditions or is too spatially
253 coarse to allow for simulation of the layer emphasized with the black dashed box. However, this is an
254 important altitude region for identifying long range transport of aged stratospheric air and inter-
255 continental transport that may be downward mixing towards the surface layer and will be explored in
256 more detail below.

257 To bring in additional platforms and to better understand these differences throughout the campaign at
258 discrete altitudes, **Figure 3** shows the ozone number density values for the TROPOZ lidar, GEOS-CF
259 model, TROPOMI and ECC ozonesondes at the average 4 km vertical level for the entire TROLIX-19
260 campaign period. Within this layer, the platforms are all characterizing the general ozone features
261 throughout the campaign at an altitude that frequently is associated with aged transported laminae. There
262 is a noticeable difference between the observations and model during the previously described 20-21
263 September period. On 21 September at 12 UT, the lidar and ECC sonde quantify an elevated layer (1.2-
264 1.3×10^{21} molecules m^{-3}) into the region that is not simulated by model ($0.75-0.9 \times 10^{21}$ molecules m^{-3}),
265 resulting in an approximately 30% underestimation in ozone abundance within the layer.

266 **3.2 Columnar Data Reduction**

267 There continues to be a need within the atmospheric and satellite community to understand the
268 variability of ozone as it pertains to both the tropospheric column (*i.e.* the Earth's surface to the
269 tropopause height) and the 0-2km tropospheric column (*i.e.* the Earth's surface to the 2 km height). The

270 0-2 km region is of particular interest as it is projected to be delivered hourly from the North American
271 geo-stationary satellite: Tropospheric Emissions: Monitoring of Pollution (TEMPO). Due to the
272 increased temporal frequency and vertical extent of TEMPO's tropospheric ozone retrievals, ozone
273 lidars, such as those from TOLNet (<https://www-air.larc.nasa.gov/missions/TOLNet/>) used in this work,
274 are an ideal platform to perform future evaluations of the products.

275 Full tropospheric columns (**Figure 4, top panel**) are consistently calculated from each platform using
276 the blended tropopause height (TROPPB) produced by the GEOS-CF and described above (*c.f.* pink dots
277 in **Figure 2**) and are then converted to Dobson Units (DU). The tropospheric and 0-2km columns are
278 calculated explicitly by integrating the ozone number density from the lowest data bin of usable data to
279 the TROPPB or 2km layer height produced in the nearest model temporal output. The exception to this
280 is the OMPS/MERRA-2 tropospheric column using the residual method described above (subtracting
281 the MERRA-2 stratospheric column from the OMPS-NM total ozone column).

282 For the full tropospheric column (Figure 4, top panel), the campaign variability ranges from
283 approximately 20-55 DU based on the lidar observations. The model, lidar, and ECC sonde
284 observations agree quite well throughout the 12 Sep to 23 Sep time frame when looking at day-to-day
285 variability. However, when assessing the variability on a single day for 21 Sep, full tropospheric
286 columns reduced from the lidar observations are some of the largest observed during this TROLIX-19
287 period (reaching 46 DU), but mostly staying between 34-40 DU. During this time, the model mainly
288 ranges between 35-37 DU, resulting in differences within 10% for most of the observations (albeit closer
289 to 30% for the peak on this day).

290 When looking at Figure 2d-f, the lower ozone values just below the tropopause during this period are not
291 simulated in the model, which may be an indication that the mesoscale ozone transport in the frontal
292 system is not very well resolved by the model for this specific event. Since the model correctly
293 simulated many other ozone features during this time period within the upper tropospheric region, this
294 may also be attributed to aged transport into the domain that was not available during model

295 initialization. Back-trajectories were performed to better identify the sources of these air masses,
296 however nothing conclusive can be reported. The layer is not associated with any increase in lidar
297 attenuated backscatter within the associated altitude, suggesting it was not urban in origin and therefore
298 more likely aged stratospheric air mixing down to the lower free troposphere. Outside of this Sep 21
299 period, there is generally good agreement between the observations (including the OMPS-MERRA2
300 product) and model, indicating the combination of observations and modeling is able to represent the
301 rural conditions and ozone perturbations at the CESAR site.

302 When assessing these tropospheric column values from the TROPOMI ozone profile
303 observations, it is important to mention the vastly different vertical resolution or averaging kernel
304 schemes as compared to the independent observations near the tropopause. The ECC samples an
305 instantaneous observation with a vertical resolution generally less than 100m, while the lidar is
306 averaging over 500-750m of atmosphere for each data point near the tropopause. However, the vertical
307 resolution near the tropopause for TROPOMI using the TOPAS algorithm (Mettig *et al.*, 2021) is nearly
308 6 km, indicating it is not able to completely represent sharp gradients that may occur near the tropopause
309 layer and the lower stratosphere (where ozone content sharply increases). This lack of degrees of
310 independent information is evident in the relatively higher TROPOMI tropospheric column ozone values
311 as compared to the other independent measurements presented in this work. This suggests ground-based
312 profiling observations are still critically needed to confirm large deviations from a priori and
313 climatology in order to evaluate the atmospheric chemistry models, especially in the upper tropospheric
314 region and within the boundary layer.

315 There exists both diurnal and day-to-day variability of the 0-2 km ozone, ranging from 4-10 DU (Figure
316 4, bottom panel). In the 0-2 km ozone reduction, the lidar and model are critically needed to understand
317 ozone variability on a continuous scale. For instance, on 15 Sep the 0-2 km ozone column was near 9
318 DU at 03 UT and finished near 5.5 DU at 16 UT, resulting in a -60% change in DU within 13 hours. The
319 need for continuous measurements during highly variable days are further emphasized by the fact that

320 this gradient in 0-2km ozone for this single day (15 Sep, 5.5 DU – 9 DU) was comparable to the
321 variance of 0-2 km ozone values throughout the entire campaign.

322 In summary, we find that the ozone columns evaluated in this study generally reproduced the structure
323 of the TROLIX-19 ozone lidar observations for N=835 coincidences. For the full tropospheric column,
324 the lidar calculated median was 30.9 ± 4.7 DU, compared to 33.4 ± 3.9 DU for the GEOS-CF. This
325 indicates a difference of 2.5 DU or 7.9 %, which is well within the lidar uncertainty of around 10 %
326 throughout the tropospheric column, and as we described above is likely driven by select days rather
327 than an overall bias between the measurements. For the 0-2 km tropospheric column, the lidar calculated
328 median was $5.8 \text{ DU} \pm 0.9 \text{ DU}$, compared to $7.8 \text{ DU} \pm 0.7 \text{ DU}$ for the corresponding GEOS-CF
329 measurements. For the TROLIX-19 campaign, a 0-2 km tropospheric column accounts for
330 approximately 20% of the tropospheric column as detailed in Figure 4 (top panel), indicating
331 measurements above the surface are critically needed at understanding ozone variability at rural sites
332 such as Cabauw, NL, where free tropospheric ozone features dominate the column.

333 **4 Full Profile Ozone Comparisons**

334 **4.1 Hybrid Tropospheric/Stratospheric Ozone Comparisons**

335 To better understand differences in ozone retrievals from multiple platforms, it is important to assess the
336 entire vertical distribution of ozone. To characterize the vertical distribution throughout the entire
337 troposphere and stratosphere, hybrid ozone profiles were created from longer (integrations of 60-120
338 minutes vs 10 minutes in Sec 3) temporal retrievals from the closed co-located daytime/nighttime
339 TROPOZ and nighttime STROZ lidar data, which were then interpolated to the GEOS-CF model
340 vertical grid levels. **Figure 5** compares these results to the GEOS-CF, OMPS-LP, TROPOMI, MLS and
341 the ECC ozonesonde profiles for 12 Sep, 17 Sep, 19 Sep, and 21 Sep 2019. These days were selected as
342 days within the campaign that had an ECC launch from De Bilt, NL (30 km from Cabauw).

343 For each observation period in **Figure 5**, all platforms manage to characterize a similar shape and extent
344 of the ozone maxima between 2.5-4.5 molecules m⁻³ throughout the vertical layer between 20-25 km. In
345 each case, there are differences between the platforms in characterizing the vertical variability and
346 extent of the ozone maxima, which will be quantified in the following section. One notable feature that
347 emphasizes the cross-platform ability to illustrate ozone variability in the stratosphere is from the 19 and
348 21 Sep profiles. A dual ozone maximum is observed quite remarkably by the merged lidar, ECC, MLS,
349 OMPS-LP and simulated by the GEOS-CF centered around 20 km and then again at 25km. The wind
350 observations from the ozonesonde payload (not shown) indicate a wind shear within the two ozone
351 layers, suggesting this feature was dynamically driven. The TROPOMI retrieval is not able to retrieve
352 this vertical features due to its coarser vertical resolution and appears to average through the layers.

353 **4.2 Difference Profiles**

354 To quantitatively compare the ozone retrievals and simulations, **Figure 6** displays the ozone
355 values for the TROLIX-19 time period from the hybrid lidar dataset (**Figure 6a**), GEOS-CF (**Figure**
356 **6b**), OMPS-LP (**Figure 6c**), MLS (**Figure 6d**) and TROPOMI (**Figure 6e**). This double ozone maxima,
357 starting after 20 September serves as a geophysical marker to visually compare the ozone products. The
358 lidar, model, and OMPS-LP all capture this feature, but with varying ozone abundances and altitudes.
359 From **Figure 6d**, it appears as if TROPOMI retrievals are not able to resolve this feature. The percent
360 differences, as compared to the lidar observations, are displayed in **Figure 7a-d**. These percent
361 differences are calculated using (1)

$$(1) \text{ Percent Difference} = \frac{(E_1 - E_2)}{\frac{1}{2}(E_1 + E_2)} \times 100$$

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363
364
365
366 where E₂ are the lidar observations and E₁ are the respective ozone values from the various platforms in
367 **Figure 6**.

368 The percent differences in **Figure 7a** indicate the GEOS-CF model from 20-45 km generally represents
369 the lidar observations, but are generally 0-10 % lower in abundance. The percent differences in **Figure**
370 **7b** indicate OMPS-LP is also representing the ozone maxima and altitude above 25 km. There are larger
371 differences below 20 km, which indicates the OMPS-LP retrieval worsens (in both directions) as
372 compared to the ozone abundance below 20 km as shown in the profiles in **Figure 5**. The percent
373 differences in **Figure 7c** indicate the MLS data, especially that within the 20-40km region, perform
374 quite well as compared to the lidar observations. The percent differences in **Figure 7d** indicate the
375 TROPOMI retrieval is generally over representing the ozone concentrations throughout the atmosphere,
376 which worsens within the troposphere and has been discussed earlier for the tropospheric ozone column
377 as a result of a much larger vertical resolution in this region. In all cases, the most variability in the
378 differences occur within the active region from 10-20 km that is driven by the dynamical tropopause
379 height and lower stratospheric ozone abundance. Within each satellite dataset, we find larger biases in
380 the lower stratosphere and upper troposphere below 18km, which has been previously described in the
381 literature for the OMPS-LP dataset in Kramarova *et al.*, 2018 and were improved in the updated version
382 2.5 algorithm used in this work.

383 **5 Total Column Ozone**

384 Similar to the troposphere, to better understand to what extent the vertical distribution of ozone impacts
385 the atmospheric column, **Figure 8 (top panel)** shows the various platforms and their retrieved total
386 column ozone. For this analysis, the GEOS-CF, lidar, OMPS-NM, TROPOMI (GODFIT) are shown, in
387 addition to local ground-based measurements from a Pandora instrument and Brewer. The total column
388 values range from 230-300 DU throughout the campaign period, with the median total column ozone of
389 271 DU. With the previous analyses from Sec 3.2, this indicates the median total tropospheric column of
390 33 DU and 0-2km boundary layer column of 6 DU result in percentages of the entire ozone column of
391 12% and 2.3%, respectively. Similar to the full tropospheric ozone columns, larger total ozone columns

392 were observed towards the end of the TROLIX-19 period, suggesting this variability was partly due to a
393 larger abundance of ozone in the lower stratosphere.

394 **Figure 8 (bottom panel)** shows the various platforms as a percent differences from the model. In
395 general, the various platforms are all mostly within 5 % of each other, with most differences being
396 within $\pm 3\%$. This analysis emphasizes the stability and maturity of the Pandora and Brewer systems for
397 monitoring the total column ozone amounts. Interestingly, the double maxima feature in vertical ozone
398 distribution in the stratosphere (with local minima between) described in Sec 4.1 on 21 Sep does not
399 severely impact the total column ozone.

400 **6 Conclusions**

401 This work has highlighted the various differences in retrieved ozone quantities during the TROLIX-19
402 campaign. This has emphasized the importance of ground-based ozone lidars and other measurements in
403 understanding the vertical variability of ozone and how it relates to the column reduction. This work
404 also shows the first effort to directly resolve both tropospheric columns and 0-2km ozone columns from
405 the NASA TROPOZ lidar. Other TOLNet lidars are able to perform this data reduction and future work
406 will be to expand this effort to the other TOLNet locations. This work indicates the level of performance
407 of the GEOS-CF modeling system as compared to the other platforms, which ultimately performs
408 extremely well both in the stratosphere (Figure 6 and Figure 7) and within the troposphere (Figure 2 and
409 Figure 4).

410 One takeaway message or point of caution for future efforts is that although there are situations
411 identified where the vertical profile and the model disagree in a certain altitude range (Figure 3), when
412 the data is reduced to a columnar product, compensating over/under-estimations may cancel out and
413 produce a more accurate value when only looking at the resultant as compared to observations. For this
414 reason, it is essential when doing data columnar reduction for the troposphere, and even more so in the

415 0-2km column or planetary boundary layer, that observations of the vertical profile be used to evaluate
416 the representativeness of the model and auxillary data sets.

417 In looking towards the NASA TEMPO, this work indicates that the GEOS-CF, with its global coverage,
418 hourly resolution, and adequate vertical information to resolve most atmospheric features, is an
419 appropriate choice for the a priori profiles for the TEMPO ozone retrievals. Continued investigations are
420 needed with high resolution observations, as presented in this work, to better evaluate the GEOS-CF,
421 especially in these common transport regions of the atmosphere. Although the GEOS-CF performed
422 well in reproducing the ozone downward transport throughout the upper troposphere and lower
423 stratosphere, the model did fail to resolve some high-resolution laminae deeper into the lower
424 troposphere related to specific mesoscale ozone transport in this region as evidenced in Figure 2 and
425 Figure 3.

426 This work shows the TROPOMI TOPAS ozone profile algorithm products are able to accurately
427 reproduce ozone quantities in the lower troposphere at various atmospheric levels. In particular, **Figure**
428 **3** and Figure 4 show promising results that indicate the TROPOMI satellite observations compare well
429 with the observations from ground-based measurements (lidar, sonde) of specific elevated ozone
430 features. However, there is an observed overestimate of the TROPOMI retrieval in the upper
431 troposphere and lower stratosphere (between 10 and 15 km) associated with a larger vertical resolution
432 that needs to also be further evaluated to better understand the representativeness of the retrieval in this
433 region.

434

435 Figure 7 was presented as a quantitative resultant figure to illustrate both the temporal (*i.e.*, throughout
436 the course of the TROLIX-19 campaign) and vertical differences observed in the retrievals from each
437 observational platform. This serves as a rare opportunity to cross-evaluate multiple satellited based
438 observations, a global chemical transport model, ozonesondes and a high-resolution ozone lidar suite.

439 The authors feel that this figure has served to point out the strengths of each platform and present careful

440 considerations for areas of under/over estimation, Furthermore, we feel reducing these comparisons
441 down to a specific percentage may underserve the community push for supporting the vertical profiling
442 needed for these types of efforts.

443 The CESAR Observatory continues to be a critical landmark for campaigns that revolve around
444 atmospheric composition measurements for satellite validation and evaluation beyond this effort, such as
445 CINDI and CINDI-2 (Kreher *et al.*, 2020; Wang *et al.*, 2020; Tirpitz *et al.*, 2021). As the European
446 Commission (EC) in partnership with the European Space Agency (ESA) continues to launch
447 tropospheric composition satellites including the upcoming geo-stationary Sentinel-4 satellite, we expect
448 this observatory will continue to host and maintain critical atmospheric sampling for future validation
449 efforts.

450 **Data Availability.**

- 451 1. MLS ozone profiles can be downloaded from the NASA Goddard Space Flight Center Earth
452 Sciences Data and Information Services Center (GES DISC; Schwartz *et al.*, 2020,
453 <https://doi.org/10.5067/Aura/MLS/DATA2516>, last access: 29 March 2022).
- 454 2. The Pandora data is available at the Pandonia Global Network Archive ([http://data.pandonia-](http://data.pandonia-global-network.org/Cabauw/)
455 [global-network.org/Cabauw/](http://data.pandonia-global-network.org/Cabauw/), last access 29 March 29, 2022).
- 456 3. The OMPS LP version 2.5 ozone profiles can be downloaded from the NASA Goddard Space
457 Flight Center Earth Sciences Data and Information Services Center (GES DISC;
458 at <https://doi.org/10.5067/X1Q9VA07QDS7> (Deland, 2017, last access: 29 March 2022).
- 459 4. The tropospheric ozone lidar data used in this publication were obtained from the Cabauw
460 Experimental Site for Atmospheric Research (CESAR) as part of a campaign involving the
461 Network for the Detection of Atmospheric Composition Change (NDACC) and NASA's
462 Tropospheric Ozone Lidar Network (TOLNet) and are publicly available ([https://www-](https://www-air.larc.nasa.gov/cgi-bin/ArcView.1/TOLNet?NASA-GSFC=1)
463 [air.larc.nasa.gov/cgi-bin/ArcView.1/TOLNet?NASA-GSFC=1](https://www-air.larc.nasa.gov/cgi-bin/ArcView.1/TOLNet?NASA-GSFC=1), last access: 29 March 2022).
- 464

- 465 5. The ozonesonde and Brewer data used in this publication were obtained from the De Bilt, NL
466 site as part of a campaign involving the Network for the Detection of Atmospheric Composition
467 Change (NDACC) and are publicly available (<ftp://ftp.cpc.ncep.noaa.gov/ndacc/station/debilt/>,
468 last access: 29 March 2022).
- 469 6. The stratospheric ozone lidar data used in this publication were obtained from the Cabauw
470 Experimental Site for Atmospheric Research (CESAR) as part of a campaign involving the
471 Network for the Detection of Atmospheric Composition Change (NDACC) and are publicly
472 available (<ftp://ftp.cpc.ncep.noaa.gov/ndacc/station/cabauw/>, last access: 29 March 2022).
- 473 7. The TROPOMI TOPAS Ozone Profile data and source codes are available upon request from
474 Nora Mettig (mettig@iup.physik.uni-bremen.de) or Mark Weber (weber@uni-bremen.de). The
475 L1B version of the S5P data is available upon request to the S5P Validation Team.
- 476 8. The Tropospheric Ozone Column from OMPS-NM/MERRA-2 Daily measurements data are
477 available upon request from Jerry Ziemke (Jerald.r.ziemke@nasa.gov).
- 478 9. The NASA GEOS-CF simulations are available at the data sharing portal
479 (<https://portal.nccs.nasa.gov/datashare/gmao/geos-cf/v1/forecast/>, last access 29 March 2022).

480

481 Author contributions. JS drafted the original manuscript. JS, LT, GS, and TM deployed and operated the
482 NASA ozone lidars and provided expertise on use of measurements. NM, AR, and MW provided
483 TOPAS ozone profile data and guidance on how best to use the measurements. AA and KK provided
484 overall context as principal investigators of the TROLIX-19 campaign and coordinated science team
485 meetings to foster this collaboration. KEK provided GEOS-CF data and insight on its use in this work.
486 MA, AP, MvR, and PV provided expertise and data for the ground-observations for ozonesondes,
487 Brewer, and historical data for the Cabauw site. JZ provided data for the OMPS-MERRA-2 tropospheric
488 column data. NK provided Aura MLS, OMPS-LP merged data and further insight into the use of the
489 data.

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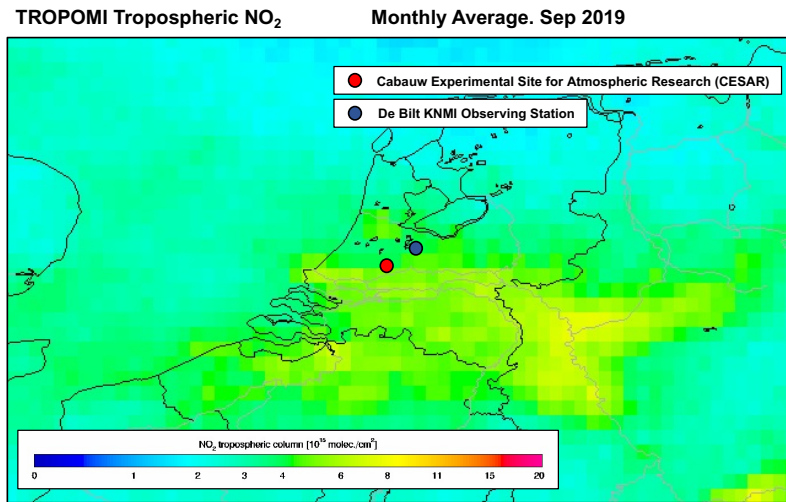
736 **Table 1: Instrument platforms, associated products, and short description used in this work during the TROLIX-19**
737 **campaign.**

738

Instrument	Products	Platform	Description
GSFC TROPOZ [NASA]	Profiles [0.2 – 18 km]	Ground-based Lidar	10 min integration; 30-90-min avg around ECC or Satellite Overpass
GSFC STROZ [NASA]	Profiles [15 - 48 km]	Ground-based Lidar	~2-4-hr avg between (20-23 UT)
ECC Ozonesondes [KNMI]	Profiles [0 – 33 km]	Balloonborne	Balloonborne, Launched at 12 UT from De Bilt (~30 km from Cabauw) on 4 days
Pandora [NASA/KNMI]	Column [TCO]	Spectrometer	L2 Pandora 118s, Data Used has QC/QA Flags = 10
Brewer [KNM]	Column [TCO]	Spectrophotometer	L2 Brewer #189m, MKIII, Located in De Bilt
S5P/TropOMI [ESA]	Column [TCL]	Satellite	L2 TOPAS Product, Overpass between 12-14 UT (5.5x3.5 km, nadir)
S5P/TropOMI [KNMI]	Column [TCO]	Satellite	L2 GODFIT v4 TO3 Product, Overpass between 12-14 UT (5.5x3.5 km, nadir)
OMPS [NASA]	Column [TCO]	Satellite	L3 NM Product, Version 2, Daily Overpass between 12-14 UT (50x50 km, nadir)
OMPS-LP [NASA]	Profiles [12-60km]	Satellite	Merged L2 v2.5 Daily Merged Product, Overpass between 12-14 UT (1km vertical bins)
OMPS/MERRA-2 [NASA]	Trop. Columns	Satellite/Assimilation	L4 Derived Product, OMPS-NM daily Overpass, MERRA-2
AURA MLS [NASA]	Profiles [12-60km]	Satellite	Merged L2 v5 Daily Daytime/Nighttime Products, Overpass between 12-14 UT (1km vertical bins) and 01-03 UT.
GEOS-CF [NASA]	Profiles [0-80km]	Global 3-D CCMM	1-Hr, 72 lev, Met. Replay, (25x25km)

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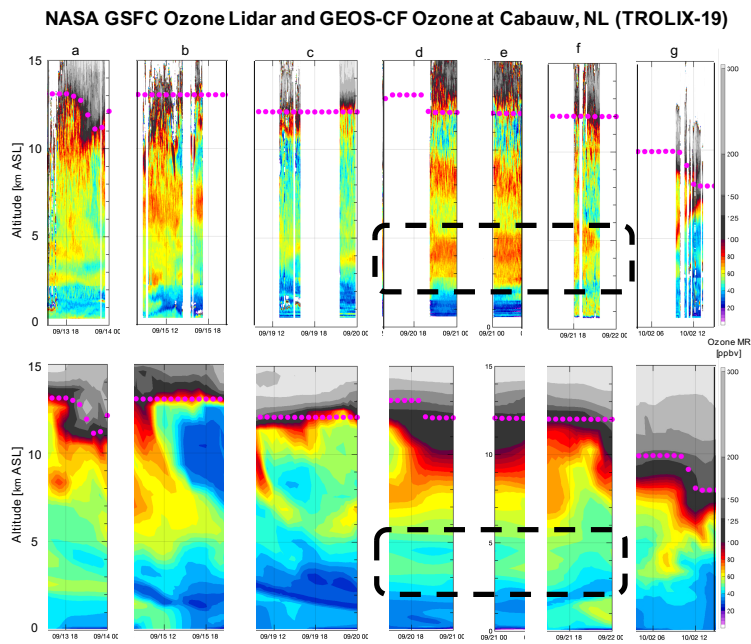


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763 **Figure 1: TROPOMI monthly-mean tropospheric NO₂ column (version 1.0) for September 2019. The CESAR and De**
764 **Bilt, NL sites are indicated in the image.**

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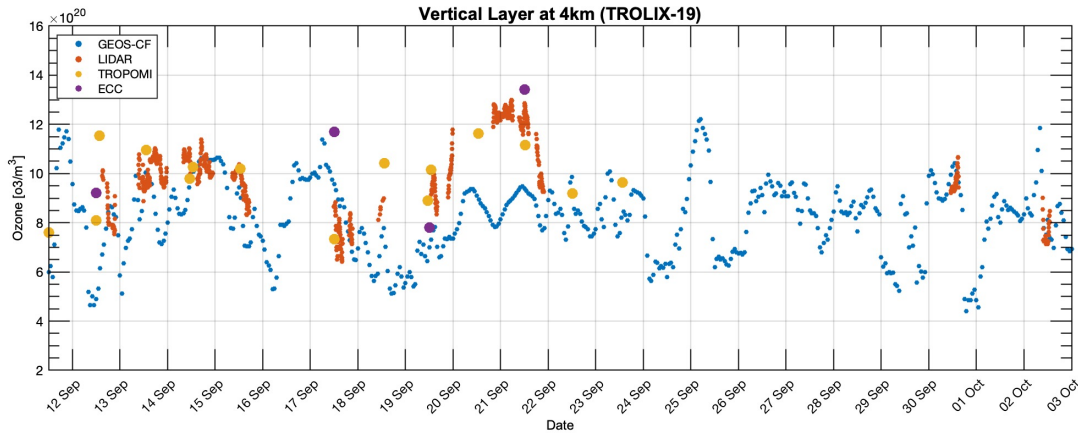


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795 **Figure 2: Cloud screened TROPOZ lidar retrievals (top panel) and the corresponding GEOS-CF model output (bottom**
796 **panel) from the closest model grid cell to the CESAR observatory during TROLIX-19 for a) 13 Sep 14-00 UTC, b) 15**
797 **Sep 09-21 UTC, c) 19 Sep 10-00 UT, d) 20 Sep 16-00 UT, e) 21 Sep 0-3 UT, f) 21 Sep 16-00UT, and g) 02 Oct 04-14 UT.**
798 **Pink dots are overlaid to indicate the simulated tropopause altitude based on a blended estimate (TROPPB).**

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819 **Figure 3: Ozone number density values for the TROPOZ lidar, GEOS-CF mode, TROPOMI and electro-chemical cell**
820 **(ECC) ozonesondes at the 4km layers/levels. The layer was calculated to match the closest representative vertical layer**
821 **of the GEOS-CF for consistent intercomparison. Data is averaged in a 500m layer from 3.94 km to 4.44 km AGL.**

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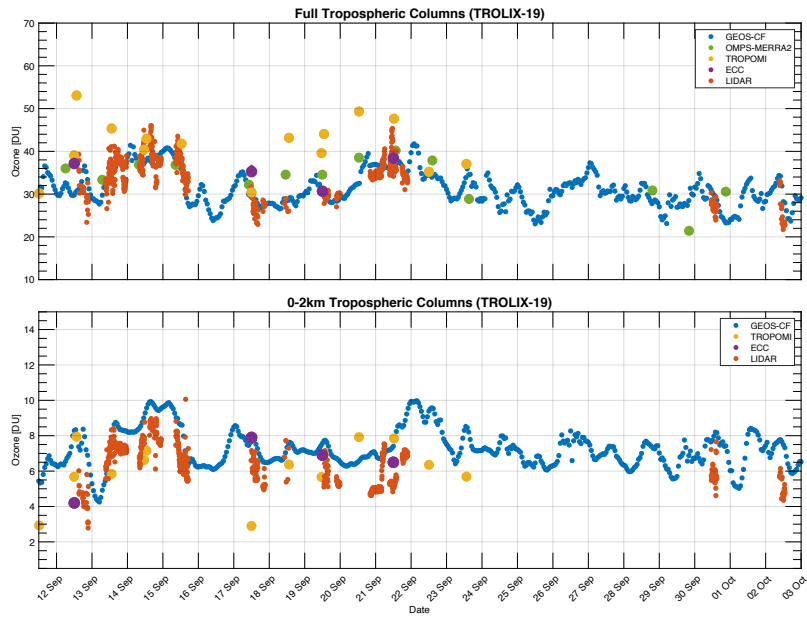
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841 **Figure 4: Full tropospheric columns (top panel) and 0-2km tropospheric columns (bottom panel) calculated from**
842 **GEOS-CF, OMPS-MERRA2 (full column only), TROPOMI, Lidar and ECC. Data where reflectivity was greater than**
843 **0.6 was excluded to remove cloud interference.**

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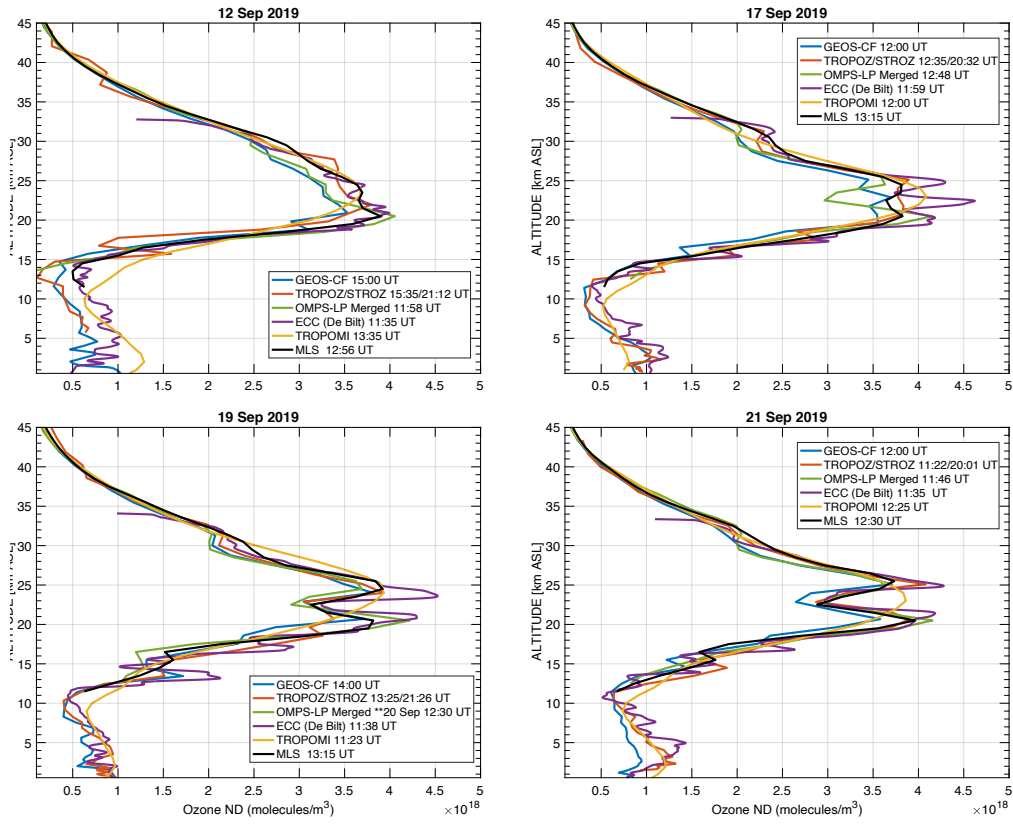
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869 **Figure 5: GEOS-CF, Lidar, OMPS-LP, ECC, TROPOMI, and MLS ozone profile comparisons for 12 Sep, 17 Sep, 19**
870 **Sep, and 21 Sep 2019. These days were selected as days within the campaign that had an ECC launch from De Bilt.**

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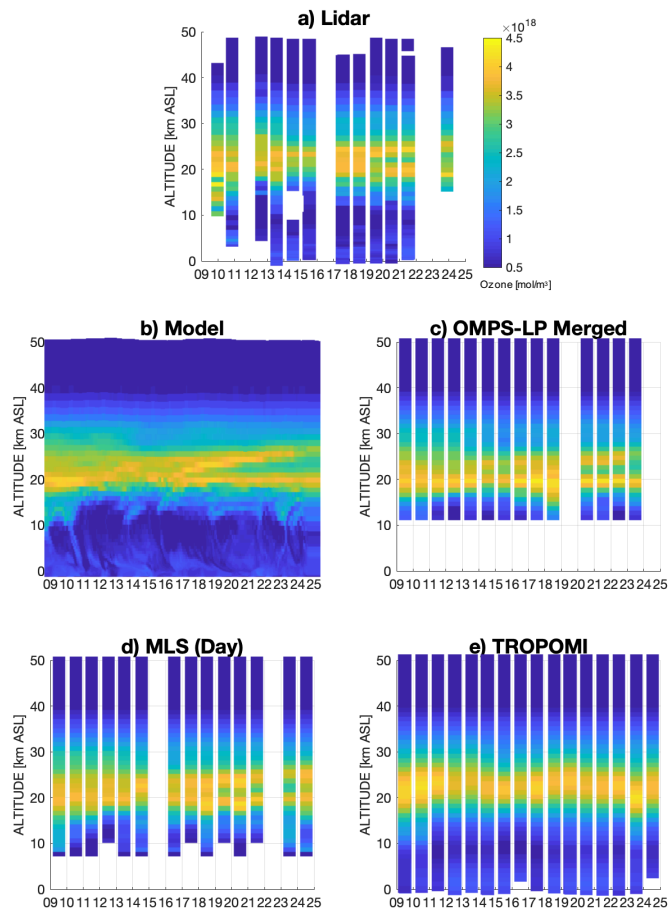
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883 **Figure 6: Ozone number densities across all platforms for the TROLIX-19 time period from the hybrid lidar dataset**

884 **(Figure 6a), GEOS-CF (Figure 6b), OMPS-LP (Figure 6c), MLS (Figure 6d), TROPOMI (Figure 6e). The x-axis as**

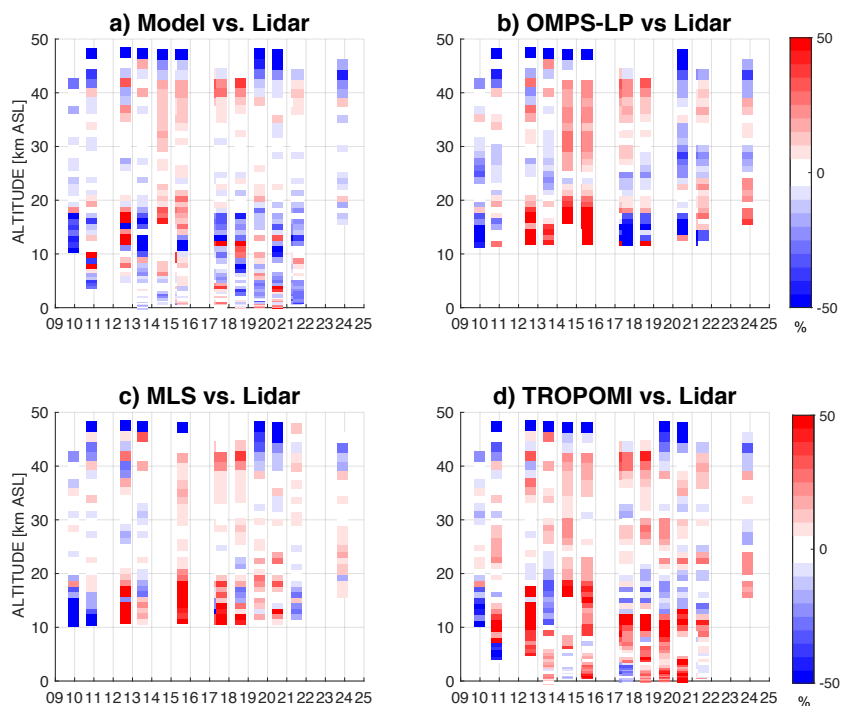
885 **day of September 2019.**

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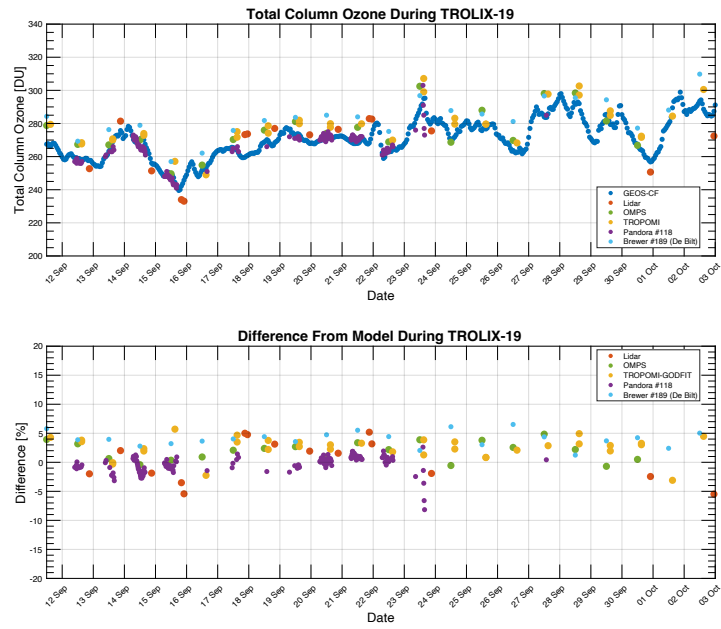
899 **Figure 7: Differences in ozone number densities across all platforms for the TROLIX-19 time period for Model (Figure**
900 **7a), OMPS-LP (Figure 7b), MLS (Figure 7c), and TROPOMI (Figure 7d). The x-axis as day of September 2019.**

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911 **Figure 8: Total Ozone columns (top panel) and percent differences (bottom panel) as compared to the model**
912 **observations for GEOS-CF, lidar, OMPS, TROPOMI, Pandora, and Brewer.**

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