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1 **Quantifying the tropospheric ozone radiative effect and its temporal evolution in the satellite-era**

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21 **Key Points:**

- 22 • Using satellite data and model simulations, we quantify the long-term (2008-2017) global
23 average tropospheric ozone radiative effect to range between 1.21 and 1.28 W/m².
- 24 • Satellite/modelled long-term trends in the tropospheric ozone radiative effect have
25 remained stable (2008-2017) yielding no substantial changing influences on climate.
- 26 • Meteorological variability has been important in stabilising the global tropospheric ozone
27 radiative effect with time.

28 **Abstract:**

29 Using state-of-the-art satellite ozone profile products, and chemical transport model, we provide an
30 updated estimate of the tropospheric ozone radiative effect (TO₃RE) and observational constraint on
31 its variability over the decade 2008-2017. Previous studies have shown the short-term (i.e. a few
32 years) globally weighted average TO₃RE to be 1.17±0.03 W/m², while our analysis suggests that the
33 long-term (2008-2017) average TO₃RE to be 1.21-1.28 W/m². Over this decade, the
34 modelled/observational TO₃RE linear trends show negligible change (i.e. ±0.1%/year), so the
35 tropospheric ozone radiative contribution to climate has remained stable with time. Two model
36 sensitivity experiments fixing emissions and meteorology to one year (i.e. start year – 2008) show



37 that ozone precursor emissions (meteorological factors) have had limited (substantial) impacts on
38 the long-term tendency of globally weighted average TO_3RE . Here, the meteorological variability in
39 the tropical/sub-tropical upper troposphere is dampening any tendency in TO_3RE from other factors
40 (e.g. emissions, atmospheric chemistry).

41 **Plain Language Summary:**

42 Tropospheric ozone is a potent air pollutant and an important short-lived climate forcer (SLCF). It is a
43 secondary pollutant formed through chemical reactions of precursor gases and sunlight. As a SLCF, it
44 influences the incoming solar short-wave radiation and the outgoing long-wave radiation in the
45 upper troposphere (approximately at altitudes of 10-15 km) where the balance between the two
46 yields a net positive (i.e. warming) effect at the surface. The majority of previous estimates of the
47 tropospheric ozone radiative effect (TO_3RE) have been quantified from atmospheric chemistry
48 climate model simulations. However, satellite retrievals of tropospheric ozone in recent decades
49 have provided the opportunity to estimate these model TO_3RE estimates. In this study, we utilise
50 satellite ozone profile retrievals from the Infrared Atmospheric Sounding Interferometer (IASI), on-
51 board the MetOp-A satellite, to derive a long-term average TO_3RE estimate of 1.21-1.28 W/m^2 .
52 While this builds upon previous studies (e.g. TO_3RE estimates of $1.17 \pm 0.03 W/m^2$), the improved
53 spatial coverage and temporal record of IASI also allows for the assessment of TO_3RE variability and
54 tendencies on a decadal scale. Here, we find negligible trends in the TO_3RE (2008-2017) suggesting
55 that the contribution of tropospheric ozone to climate, via radiative properties, remained stable
56 over that period.

57 **1. Introduction**

58 Tropospheric ozone (TO_3) is a short-lived climate forcer (SLCF). It is the third most important
59 greenhouse gas (GHG; Myhre et al., 2013) and a hazardous air pollutant with adverse impacts on
60 human health (WHO, 2018) and the biosphere (e.g. agricultural and natural vegetation; Sitch et al.,
61 2007). Since the pre-industrial (PI) period, anthropogenic activities have increased the atmospheric
62 loading of ozone (O_3) precursor gases, most notably nitrogen oxides (NO_x) and methane (CH_4),
63 resulting in an increase in TO_3 of 25-50% since 1900 (Gauss et al., 2006; Lamarque et al., 2010; Young
64 et al., 2013). The PI to present day (PD) radiative forcing (RF) from TO_3 is estimated to be 0.4 (0.2-
65 0.6) Wm^{-2} (Myhre et al., 2013; Stevenson et al., 2013) based on model simulations.

66 While models provide a valuable framework to quantify the TO_3 RF, observations are required to
67 validate the models' representation of TO_3 and TO_3 RF. Observations are not available for the PI, but
68 multiple satellite products of TO_3 are readily available in the PD (e.g. Richards et al., 2008; Boynard
69 et al., 2018; Barret et al., 2020). The tropospheric ozone radiative effect (TO_3RE) is defined as the
70 radiative flux imbalance at the tropopause between incoming short-wave solar radiation and the
71 outgoing long-wave radiation due to the presence of TO_3 (Rap et al., 2015). Therefore, satellite
72 ozone profile datasets from infrared instruments, in combination with off-line ozone radiative
73 kernels to account for vertical sensitivity (e.g. Bowman et al., (2013); Rap et al., 2015), can be used
74 to quantify the PD TO_3RE and thus provide some constraint on modelled TO_3RE which is used to
75 derive the TO_3RF .

76 Several studies have previously used satellite data to derive short-term estimates of the TO_3RE (i.e.
77 from a few months of data). Joiner et al., (2009) used tropospheric column ozone (TCO_3) data based
78 on two satellite instruments: Ozone Monitoring Instrument (OMI) and Microwave Limb Sounder



79 (MLS) measurements, also known as OMI-MLS product for January and July 2005, to estimate the
80 resultant instantaneous TO₃RE at the tropopause to be 1.53 W/m². Worden et al., (2008) used ozone
81 profile data for 2006 from the Tropospheric Emissions Spectrometer (TES), on-board NASA's Aura
82 satellite, to estimate the average instantaneous long-wave TO₃RE at the top-of-the-atmosphere
83 (TOA) over the oceans (45°S-45°N) to be 0.48±0.14 W/m². Worden et al., (2011), using TES data for
84 August 2006, estimated the instantaneous long-wave TO₃RE at TOA to be 0.33 W/m². Later, Bowman
85 et al., (2013) also used TES data (averaged between 2005 and 2009) to constrain the simulated
86 instantaneous long-wave TO₃RE from an ensemble model average. They found that seasonally, TES
87 long-wave TO₃RE peaks in northern Africa/Mediterranean/Middle East in June-July-August over 1.0
88 W/m² with minimum values (0.0-0.2 W/m²) over the winter-time high-latitudes. Overall, the
89 ensemble average long-wave TO₃RE low bias was 0.12 W/m². Doniki et al., (2015) took this further
90 by calculating the instantaneous long-wave TO₃RE from the Infrared Atmospheric Sounding
91 Interferometer (IASI), though using a small subset of the data, and found estimates from Worden et
92 al., (2008), using TES, had a low bias of ~25%. Rap et al., (2015) also used TES satellite ozone profile
93 observations (2005-2008) in combination with the TOMCAT chemical transport model (CTM) and
94 provided the first robust satellite constraint on annual globally weighted resultant TO₃RE (after
95 stratospheric temperature adjustment) with a range of 1.17±0.03 W/m².

96 Following the methodology adopted in Rap et al. (2015), we exploit satellite ozone profile data from
97 IASI, on the MetOp-A satellite, which has a long-term record and substantial spatial coverage, in
98 combination with the TOMCT CTM, to improve the TO₃RE estimate and investigate its long-term
99 variability and implications for climate. The satellite data, radiation model and CTM used are
100 discussed in Section 2, our results are presented in Section 3 and Section 4 summarises our
101 conclusions.

102 2. Observations and Model

103 2.1. Satellite Observations

104 IASI is a Michelson interferometer with a nadir-viewing spectral range between 645 and 2760 cm⁻¹
105 with spectral sampling of 0.25 cm⁻¹ (Illingworth et al., 2011). It measures simultaneously in four fields
106 of view (FOV, each circular at nadir with a diameter of 12 km) in a 50 x 50km square which are
107 scanned across track to sample a 2200 km-wide swath (Clerbaux et al., 2009). IASI, on Eumetsat's
108 MetOp-A satellite, is in a sun-synchronous polar orbit with equator crossing local times of 9.30 (day)
109 and 21.30 (night).

110 The three IASI products we use in this study are the IASI-FORLI product (vn 20151001, IASI-FORLI,
111 2020; Boynard et al., 2018; Wespes et al., 2018), the IASI-SOFRID product (vn 3.5, IASI-SOFRID, 2022;
112 Barret et al., 2020) and the RAL IASI-IMS product (IASI-IMS, 2022; Pope et al., 2021; Pimlott et al.
113 ,2022) between 2008 and 2017 (i.e. period of consistent data coverage for all the IASI products). All
114 three products use an optimal estimation method (OEM, Rogers, 2000) to retrieve ozone. Both IASI-
115 SOFRID and IASI-IMS use the RTTOV radiative transfer model (Saunders et al. 1999), while the IASI-
116 FORLI product uses look-up tables to speed up its radiative transfer calculations (Hurtmans et al.,
117 2012). Meteorological inputs (pressure, water vapour, temperature and clouds) for IASI-FORLI come
118 from Eumetsat level-2 data, while IASI-SOFRID uses ECMWF operational analyses and IASI-IMS uses
119 ECMWF surface pressures and co-retrieves other meteorological and surface variables. For the
120 ozone apriori, IASI-FORLI and IASI-IMS use the ozone climatology of McPeters et al., (2007), while
121 IASI-SOFRID uses the dynamical ozone climatology described in Sofieva et al., (2014).



122 The IASI-FORLI level-2 data are filtered for a geometric cloud fraction < 0.2 , degrees of freedom > 2.0 ,
123 O_3 values > 0.0 , solar zenith angle $< 80.0^\circ$ and the surface to 450 hPa sub-column O_3 / total column
124 $O_3 < 0.085$. The IASI-SOFRID data were provided on a $1.0^\circ \times 1.0^\circ$ horizontal grid (i.e. level-3 product,
125 but daily temporal resolution – we used daytime retrievals only) with filtering already applied as in
126 Barret et al., (2020). Here, only O_3 values > 0.0 were used. For IASI-IMS level-2, the data are filtered
127 for a geometric cloud fraction < 0.5 , O_3 values > 0.0 , solar zenith angle $< 80.0^\circ$ and a cost function $<$
128 1000.0 . However, for IASI-IMS, we relaxed the geometric cloud fraction threshold to 0.5 as it retains
129 more data as the data product in this study has only been processed for 1 in 10 days and 1 in 4
130 pixels.

131 2.2. Ozonesondes

132 Despite the three IASI ozone profile products using the same radiance data, the three retrieval
133 schemes produced systematic differences between the products in the long-term TCO_3 average (e.g.
134 **Figures S2 and S3** from the **Supporting Information (SI)**). Though, the spatial structure in the three
135 products compares well. Therefore, to harmonise the three IASI TCO_3 data sets (i.e. absolute values
136 but not long-term variability) we use ozonesonde data from the World Ozone and Ultraviolet
137 Radiation Data Centre (WOUDC; WOUDC, 2023), the Southern Hemisphere Additional Ozonesondes
138 (SHADOZ; SHADOZ, 2023) project and the Global Monitoring Laboratory, National Oceanic and
139 Atmospheric Administration (NOAA; NOAA, 2023). Here, O_3 measurements were rejected if the O_3 or
140 pressure values were unphysical (i.e. < 0.0), if the O_3 partial pressure > 2000.0 mPa or the O_3 value
141 was set to 99.9, and whole ozonesonde profiles were rejected if least 50% of the measurements did
142 not meet these criteria. These criteria are similar to those applied by Keppins et al., (2018) and
143 Hubert et al., (2016). To allow for direct like-for-like comparisons between the two quantities,
144 accounting for the vertical sensitivity of the satellite, the instrument averaging kernels (AKs) are
145 applied the ozonesonde profiles as:

$$146 \quad \text{sonde}_{AK} = AK \cdot (\text{sonde}_{int} - \text{apr}) + \text{apr} \quad (1)$$

147 where sonde_{AK} is the modified ozonesonde sub-column profile, AK is the averaging kernel matrix,
148 sonde_{int} is the ozonesonde sub-column profile interpolated on the satellite pressure grid and apr is
149 the a priori for the satellite retrieval. For the application of the AKs to the ozonesonde profiles, the
150 full ozone profile is required which is not available from the ozonesondes (i.e. mid-stratosphere and
151 above). Therefore, the ozonesonde profile above its minimum pressure level is extended using the
152 apriori profile from the corresponding satellite product. The profile is smoothed vertically across the
153 joining pressure level to avoid a profile discontinuity.

154 Once the ozonesondes had been co-located with the satellite data (i.e. within 6-hours and 500 km)
155 and the AKs applied, the two datasets were compared across the full 2008-2017 period. We typically
156 find a global annual TCO_3 systematic bias of 14.9%, 2.7% and 17.4% for IASI-FORLI, IASI-SOFRID and
157 IASI-IMS, respectively, which is consistent with Boynard et al., (2018), Barret et al., (2020) and
158 Pimlott et al., (2022). Here, we generated annual-latitude (30° bins) bias correction factors (BCF)
159 which were applied to the gridded satellite records (see **SI-2**) to harmonise the retrieved TCO_3 (i.e.
160 remove the systematic errors) and scale the derived TO_3RE . This is an important exercise as it
161 provides a more accurate absolute range in satellite retrieved TCO_3 (and the ozone values used to
162 derive the TO_3RE) but as the ozonesondes generally have poor spatial coverage, the global coverage
163 and spatial distribution of the satellite data is critical in our analysis. Note, that as a climatology was



164 used, the systematic biases in the satellite records were affected but their long-term temporal
165 variability retained.

166 2.3. TOMCAT

167 In this study, we use the 3D global chemical transport model TOMCAT (Chipperfield, 2006), which
168 has a detailed tropospheric chemistry scheme including 229 gas-phase reactions and 82 advected
169 tracers (Monks et al., 2017). Model heterogeneous chemistry uses size-resolved aerosol from the
170 GLOMAP module (Mann et al., 2010). The model was run between 2008 and 2017 at a $2.8^\circ \times 2.8^\circ$
171 spatial resolution with 31 vertical levels between the surface and 10hPa. The model is forced by
172 meteorological reanalyses (ERA-Interim) from the European Centre for Medium-Range Weather
173 Forecasts (ECMWF; Dee et al., 2011) including reanalysis cloud fields and mass fluxes (e.g. as in
174 Rowlinson et al., 2020, Pimlott et al., 2022). Annually varying anthropogenic emissions come from
175 the Coupled Model Intercomparison Project Phase 6 (CMIP6, Feng et al., 2020). Climatological
176 biogenic emissions are from the Chemistry-Climate Model Initiative (CCMI; Morgenstern et al., 2017)
177 but isoprene and monoterpene emissions are annually varying from the Joint UK Land Environment
178 Simulator (JULES, Pacifico et al., 2011) within the free-running UK Earth System Model (UKESM,
179 Sellar et al., 2019). Other natural emissions come from the Precursors of Ozone and their Effects in
180 the Troposphere (POET, Olivier et al., 2003) and biomass burning emissions from the Global Fire
181 Emissions Database (GFED) version 4 (van der Werf et al., 2017). For methane (CH_4), the model
182 tracer is scaled to the annually varying global averaged surface CH_4 value from NOAA (Dlugokencky,
183 2020). The model was spun up for 1-year (2007) and the model tracers output daily at 09:30 local
184 time (LT) globally to match the MetOp-A daytime overpass time. When comparing with IASI, the
185 satellite AKs are applied to the TOMCAT vertical ozone profiles in the same way as the ozonesondes
186 (i.e. **Equation 1**). Here, the TOMCAT ozone profile (already temporally co-located) is co-located from
187 the model grid box the retrieval sits in. To investigate the importance of emissions and meteorology
188 on TO_3 and TO_3RE , two sensitivity experiments were run between 2008 and 2017 using repeating
189 emissions and meteorology for 2008 (i.e. start of the time-series) annually in the model simulation
190 over the time period.

191 2.4. Radiative Transfer Model and Kernel

192 The TO_3RE was calculated using the SOCRATES off-line radiative transfer model (Edwards and Slingo,
193 1996) in combination with TOMCAT and the three IASI ozone products. SOCRATES has six bands in
194 the short-wave and nine in the long-wave. To account for stratospheric temperature adjustments,
195 Rap et al., (2015) used the dynamical heating approximation (Fels et al., 1980). This involved
196 accounting for changes in the stratospheric heating rate determined from the model due to the O_3
197 perturbation, which were applied to the temperature field, with the model run iteratively until
198 stratospheric temperatures reached equilibrium (Rap et al., 2015). This approach of using the
199 SOCRATES off-line radiative kernel with output from model simulations to derive the TO_3 radiative
200 effect has been used in several studies e.g. Rap et al., (2015), Scott et al., (2018), Iglesias-Suarez et
201 al. (2018) and Rowlinson et al., (2020).

202 To derive the satellite TO_3RE , the annual average IASI 3D ozone field is multiplied by the off-line
203 radiative kernel (grid box by grid box) and then summed from the surface to the tropopause
204 pressure. Here, the IASI ozone data is mapped onto the spatial resolution of the radiative kernel and
205 then interpolated vertically onto its pressure grid. The equation for each grid box is:



$$TO_3RE = \sum_{i=surf}^{trop} RK_i \times O_{3i} \times dp_i/100 \quad (2)$$

207 where TO_3RE is the tropospheric ozone radiative effect (W/m^2), RK is the radiative kernel
208 ($W/m^2/ppbv/100$ hPa), O_3 is the satellite ozone grid box value (ppbv), dp is the pressure difference
209 between vertical levels (hPa) and i is the grid box index between the surface pressure level and the
210 tropopause pressure. The tropopause pressure is based on the World Meteorological Organisation
211 (WMO) definition of “the lowest level at which the temperature lapse rate decreases to 2 K/km or
212 less” (Bethan et al., 1996).

213 3. Results

214 3.1. Tropospheric Ozone Radiative Effect

215 **Figure 1** shows the IASI derived TCO_3 , TO_3RE and normalised TO_3RE (NTO_3RE , i.e. the TO_3RE divided
216 by its TCO_3 as in Rap et al., (2015)). For the TCO_3 , all three harmonised IASI products have good
217 spatial agreement in the long-term (2008-2017) average with a background north-south hemisphere
218 gradient of approximately 30.0-40.0 to 15.0-25.0 DU. Peak TCO_3 (>40.0 DU) occurs over East Asia,
219 the Middle East and ozone outflow from central Africa (e.g. from lightning and biomass burning
220 precursor gases (Moxim & Levy, 2000)). The global average TCO_3 values for IASI-FORLI, IASI-SOFRID
221 and IASI-IMS are 32.6 DU, 29.9 DU and 29.9 DU, respectively (**Figure 1 left column**).

222 When the TO_3RE is calculated (**Figure 1 middle column**), peak values occur over the sub-tropics,
223 Africa and Australia ranging between approximately 2.0 and 2.5 W/m^2 consistently for each IASI
224 product. The minimum values are in the high latitudes ranging between 0.0 and 0.8 W/m^2 where TO_3
225 appears to have limited impact on the TO_3RE . The bottom panel of **Figure 1** supports this as the
226 zonally average profiles, weighted by the cosine of degrees latitude, show that TCO_3 is near-zero in
227 the high-latitudes, approximately 15.0-20.0 DU in the mid-latitudes, peaking at 28.0-33.0 DU in the
228 sub-tropics and then decreasing by several DU at the tropics. The corresponding TO_3RE profiles
229 follow a similar pattern with near-zero values at the high-latitudes, approximately 0.5-1.0 W/m^2 in
230 the mid-latitudes, peak at 1.5 W/m^2 in the sub-tropics and then decrease to 1.1-1.2 W/m^2 in the
231 tropics. Therefore, the sub-tropics have the largest contribution to the global TO_3RE . The global
232 weighted TO_3RE averages for IASI-FORLI, IASI-SOFRID and IASI-IMS are 1.23, 1.21 and 1.21 W/m^2 ,
233 respectively.

234 The NTO_3RE (**Figure 1 right column**) provides an estimate of where the TO_3RE is most sensitive to
235 changes in TCO_3 (i.e. the unit of TO_3RE per unit of TCO_3). Peak NTO_3RE (>45.0 $mW/m^2/DU$) occurs in
236 similar locations to the peak TO_3RE (e.g. sub-tropics, Africa and Australia), while the minimum values
237 (10.0-20.0 $mW/m^2/DU$) occur in the high-latitudes. However, while the South Pacific TCO_3 values
238 (23.0-30.0) are lower than other ocean regions (e.g. >30.0 DU), the NTO_3RE values are of similar
239 magnitude (approximately 50.0 $mW/m^2/DU$). Therefore, while the sub-tropical/mid-latitude oceans
240 have reasonable large TCO_3 and TO_3RE values, the South Pacific is more effective at contributing to
241 the TO_3RE , despite its lower TCO_3 values (i.e. more positive radiative effect per unit of TO_3).

242 Overall, the global weighted average NTO_3RE is 37.78, 40.43 and 40.60 $mW/m^2/DU$ for IASI-FORLI,
243 IASI-SORID and IASI-IMS, respectively. Based on the AKs, the tropospheric degrees of freedom of
244 signal (DOFS, between the surface and 170 hPa – approximate tropopause) is approximately 1.0 for
245 all three IASI products (not shown here). However, it is likely that differences in the IASI ozone
246 profiles are driving the contrasting globally averaged NTO_3RE values. As the IASI-FORLI NTO_3RE is
247 lower, while having the highest global average TCO_3 and TO_3RE , it suggests that IASI-FORLI has more



248 TO₃ in the mid-troposphere where the radiative kernel has less sensitivity. Further to this, as the IASI
249 ozone products only have approximately 1.0 DOFS, the harmonisation of the products using the
250 ozonesondes can only be done on a tropospheric column level and thus the scaling of the satellite
251 derived TO₃RE (i.e. even though the upper troposphere is the most sensitive region to ozone
252 radiative properties, the scaling of the TO₃RE is applied based on the satellite-ozonesonde TCO₃
253 relative differences).

254 TOMCAT allows for a further quantification of the TO₃RE in the satellite-era and the ability to run
255 sensitivity experiments to explore important processes. Therefore, the TOMCAT equivalent metrics
256 from **Figure 1** are presented in **Figure 2**. Evaluation of the model using the IASI products and
257 ozonesondes (see **SI-2, Figure S3 & S4**) shows the model generally captures the TCO₃ spatial pattern
258 and absolute values. In the tropics (mid/high-latitudes), the model underestimates (overestimates)
259 by approximately 10-20% on average. These biases are comparable with other modelling studies
260 evaluating models against satellite TO₃ observations (e.g. Archibald et al., 2020; Monks et al., 2017;
261 Nassar et al., 2009; Young et al., 2013), indicating that TOMCAT is a suitable modelling framework in
262 this study.

263 The globally mean TCO₃ from TOMCAT (2008-2017) with the three sets of AKs applied (**Figure 2 left**
264 **column**) ranges between 31.6 and 32.5 DU, so it slightly larger than the IASI data sets in **Figure 1**.
265 When translated into TO₃RE, the peak values from TOMCAT (with AKs applied) ranges between 2.0
266 and >2.5 W/m² over Africa, Australia and the sub-tropics. The globally weighted TO₃RE for TOMCAT
267 with the IASI-FORLI and IASI-SOFRID AKs applied is 1.28 W/m² and thus moderately higher than IASI
268 (1.21-1.23 W/m²) but comparable overall. However, the globally weighted TO₃RE for TOMCAT with
269 IASI-IMS AKs applied is larger at 1.34 W/m². As TOMCAT has a positive TCO₃ bias with the
270 observations in the sub-tropics, where the TO₃RE influence is most pronounced, this probably
271 explains the larger model TO₃RE values. In the bottom panel of **Figure 2**, the zonal profiles (weighted
272 by cosine of degree latitude) for TCO₃ (TO₃RE) are consistent with IASI as high-latitude values are
273 near-zero, mid-latitude values range between 10.0 and 20.0 DU (0.5 to 1.0 W/m²) and sub-tropical
274 values range between 30.0 and 38.0 DU (1.5 and 1.7 W/m²). There is a decrease to approximately
275 25.0 DU (1.0-1.3 W/m²) in the tropics. In terms of the NTO₃RE, the TOMCAT (with AKs applied) global
276 weighted values range between 39.4 and 42.4 mW/m²/DU, which is similar to IASI. The peak NTO₃RE
277 values are over the oceans (50.0-60.0 mW/m²/DU) and over Africa/Australia (>60.0 mW/m²/DU).
278 Like for IASI, the TCO₃ values over the South Pacific are lower than the other ocean values but the
279 NTO₃RE values are similar, again showing that despite the lower TO₃, the South Pacific region is
280 important for the global TO₃RE given its greater sensitivity (i.e. more radiative effect per unit of TO₃).

281 **3.2. Temporal Evolution of the Tropospheric Ozone Radiative Effect**

282 As IASI has daily global coverage (Clerbaux et al., 2009), we are able to derive annual average 3D
283 ozone fields between 2008 and 2017, thus providing the first assessment of temporal variability and
284 tendency in satellite derived TO₃RE. **Figure 3** shows the annual TO₃RE time series for all three IASI
285 products. First thing to note, is that the Eumetsat meteorological data used to retrieve ozone for the
286 IASI-FORLI product is subject to inhomogeneities (Boynard et al., 2018; Wespes et al., 2018). As a
287 result, we include long-term analysis of the IASI-FORLI data for the full time period (2008-2017) and
288 then a sub-time period (2011-2017) given the large inhomogeneity in September 2010 reported by
289 Boynard et al., (2018) and Wespes et al., (2018). Here, we can derive the TO₃RE to quantify the
290 absolute values (e.g. are they generally similar year to year) and how they compare between



291 products over the two time periods. In the near future, a new consistent IASI-FORLI ozone climate
292 data record will be available using homogeneous level-2 Eumetsat meteorological data.

293 For IASI-SOFRID and IASI-IMS, the annual TO₃RE values range between 1.19 and 1.24 W/m² across
294 the 2008-2017 time period. IASI-FORLI has somewhat larger values at the start of the record (1.26-
295 1.28 W/m²) before tending to that of IASI-SOFRID/IASI-IMS from 2011 onwards. Correlations
296 (squared) in the annual TO₃RE time-series between IASI-FORLI and IASI-SOFRID (IASI-IMS) are poor at
297 R²=0.148 (R²=0.132). However, IASI-SOFRID and IASI-IMS have a much stronger agreement with
298 R²=0.591 (significant at the 95th confidence level, CL95%) sharing nearly 60% of the temporal
299 variability. We also calculate the coefficient of variation (CoV, i.e., time series standard deviation
300 divided by its mean) to assess the inter-annual variability. For IASI-SOFRID and IASI-IMS, this is 1.1%,
301 but for IASI-FORLI it is 2.5%. Therefore, there is more year-to-year variability in the IASI-FORLI TO₃RE
302 record. However, when focussing on IASI-FORLI data for 2011-2017, the CoV drops to 1.2% in-line
303 with IASI-SOFRID and IASI-IMS. The correlation (squared) values are now R²_{FORLI-SOFRID}=0.496
304 (significant at the CL95%) and R²_{FORLI-IMS}=0.137, which shows improved agreement between IASI-
305 FORLI and IASI-SOFRID, but slightly surprisingly not with IASI-IMS. Using ordinary least squares fit
306 regression, IASI-FORLI, IASI-SOFRID and IASI-IMS have global average weighed TO₃RE linear trends
307 of -0.6%/year (CL95%), 0.0%/year (non-significant) and -0.1%/year (non-significant). As the IASI-
308 FORLI product has known inhomogeneities (hence the larger CoV), the insignificant IASI-SOFRID and
309 IASI-IMS trends are more robust. This is supported by IASI-FORLI when only considering 2011-2017
310 with an insignificant linear trend of -0.2%/year. Therefore, this suggests negligible change in the
311 contribution of TO₃ to the tropospheric radiative effect and thus climate over the recent past (i.e.
312 2008-2017).

313 TOMCAT global average weighed TO₃RE (without AKs applied) ranges between 1.24 and 1.29 W/m²
314 between 2008 and 2017. The CoV is 1.5% for TOMCAT, so it is larger than both IASI-SOFRID and IASI-
315 IMS. When the IASI AKs are applied to TOMCAT, there is a substantial shift in the modelled absolute
316 TO₃RE values. TOMCAT with IASI-SOFRID and IASI-FORLI AKs applied ranged between 1.28 and 1.30
317 W/m². And for TOMCAT with the IASI-IMS AKs applied, the TO₃RE values peak at 1.33 to 1.34 W/m²
318 between 2008 and 2017. As well as the increase in TO₃RE values, the application of the AKs squashes
319 the TOMCAT inter-annual variability with corresponding CoV values between 0.4 and 0.6%, which is
320 smaller than the original CoV of 1.5%. Interestingly, without the application of the AKs, the TOMCAT
321 TO₃RE time-series has similar temporal variability (e.g. peaks in 2008, 2010 and 2017 and troughs in
322 2009 and 2014). Overall, all the TOMCAT TO₃RE time-series (with and without AKs applied have
323 insignificant linear trends ranging between -0.1%/year and 0.1%/year. Therefore, even with the
324 influence of the IASI AKs on the TOMCAT TO₃RE time-series, there appears to be a negligible trend in
325 the modelled TO₃RE, supporting that of the IASI records. As a result, between 2008 and 2017, there
326 has been limited change in TO₃ and TO₃RE, thus the impact of TO₃ on climate has remained stable.

327 To investigate the importance of emissions and meteorology on the long-term TO₃RE trends,
328 TOMCAT was run using repeating emissions and repeating meteorology for 2008 (i.e. start of the
329 time-series) in two sensitivity experiments for the full time-period. Here, we find that in absolute
330 terms, using fixed emissions reduces the TO₃ burden and the TO₃RE as the time-series drops to 1.22
331 to 1.28 W/m² (i.e. minima in 2014 and 2015 more pronounced). However, the trend in TO₃RE (-
332 0.2%/year) remains insignificant and that emissions are only moderately important in driving long-
333 term tendencies in TO₃RE. On the other hand, meteorological factors, while not dramatically altering
334 the absolute simulated TO₃RE values, are more important as fixing the meteorology yields a steady



335 and significant increase (0.3%/year). Thus, without year-to-year variability in meteorology, temporal
336 variability in TO₃ would likely have a more substantial impact on the present day climate.

337 **Figure 4** shows the horizontal and vertical impact of the two sensitivity experiments on TOMCAT O₃
338 radiative effect (note the different colour bar scales). Consistent with **Figures 1** and **2**, the TOMCAT
339 control TO₃RE has peak values (>2.50 W/m²) over northern Africa and throughout the sub-tropics
340 (approximately 2.0 W/m², **Figure 4a**). Vertically, the TOMCAT peak ozone radiative effect (>0.25
341 W/m²) is in the upper troposphere (**Figure 4b**) with the largest impact in the sub-tropics of both
342 hemispheres (500-200 hPa). Similar values extend through the hemispheric mid-latitudes but in a
343 smaller pressure range (400-300 hPa). As shown in **Figure 3**, the fixed meteorological run imposes a
344 significant TO₃RE trend on the modelled tendency between 2008 and 2017. From **Figure 4c**, the
345 difference between the fixed meteorology and control runs shows mainly positive TO₃RE differences
346 of 0.1 to >0.2 W/m² throughout the tropics and sub-tropics, though there is considerable spatial
347 variation due to changes in the global circulation. In the high and mid-latitudes, there are smaller
348 scale negative differences ranging between -0.1 and 0.0 W/m² (though some differences up to -0.15
349 W/m² in the sub-tropical Pacific). In the upper troposphere (**Figure 4d**), the zonal average O₃
350 radiative effect is consistent with positive differences of up to 0.02 W/m² at approximately 200 hPa
351 in the tropics and sub-tropics. The positive differences (approximately 0.01 W/m²) filter down to 600
352 hPa in the same latitudinal range. In the mid-latitudes, the peak negative differences are
353 approximately -0.02 to -0.015 W/m² at 300 hPa, with a reach down to 500 hPa at -0.005 W/m².
354 Overall, as shown in **Figure 3**, the fixed meteorology run increases the global average TO₃RE. While
355 this could be a specific signal related to the 2008 meteorology (i.e. it is conducive to TO₃ formation),
356 it clearly shows that the upper tropospheric tropical and sub-tropical regions predominantly control
357 the global TO₃RE average and its temporal variability (i.e. the region where the meteorological inter-
358 annual variability is buffering underlying increases in TO₃RE). With fixed emissions, there is a general
359 increase (decrease) in TO₃RE in the tropics/sub-tropics (northern mid-latitudes) by 0.02 (-0.02)
360 W/m². However, over tropical Asia, Indonesia and Australia, the decrease in TO₃RE is more
361 substantial at -0.05 to -0.04 W/m² (**Figure 4e**). Vertically, there are decreases (increases) in the O₃
362 radiative effect of -0.005 (0.003) W/m² in the tropics/sub-tropics (northern mid-latitudes) between
363 600 and 200 (800 and 400) hPa (**Figure 4f**). Overall, the meteorological variability, in comparison to
364 the long-term emission changes in O₃ precursor gases, has substantially more influence on the inter-
365 annual variability of the global TO₃RE over this decade.

366 4. Conclusions

367 By using state-of-the-art satellite ozone profile retrievals from the Infrared Atmospheric Sounding
368 Interferometer (IASI), on-board MetOp-A, in combination with the TOMCAT chemical transport
369 model (CTM), we provide an updated estimate of the tropospheric ozone radiative effect (TO₃RE)
370 and provide the first observational constraint on its variability over the decade 2008-2017. Building
371 upon the previous study of Rap et al., (2015), who quantified the globally weighed average TO₃RE to
372 be 1.17±0.03 W/m² (based on data between 2005 and 2008), we find the long-term average TO₃RE,
373 between 2008 and 2017, to range from 1.21 and 1.28 W/m². Secondly, neither the modelled, nor the
374 observed TO₃RE suggest any substantial change during this period. Therefore, the tropospheric
375 ozone contribution to climate, through its infrared radiative properties, has remained stable with
376 time during 2008-2017. Investigations of the importance of ozone precursor emissions and
377 meteorology, through targeted sensitivity experiments repeating emissions and meteorology for
378 2008 (i.e. year at start of time-series), suggest that emissions have a limited impact on the globally



379 weighted average TO₃RE. Meanwhile, fixing the meteorology to a specific year (i.e. 2008) introduces
380 a significant positive trend in global TO₃RE, indicating that the meteorological variability in the
381 tropical/sub-tropical upper troposphere has been important in stabilising the tropospheric ozone
382 contribution to climate, via radiative properties, in the recent past (i.e. satellite-era).

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389 Data Availability

390 The IASI-FORLI and IASI-SOFRID data can be obtained from <https://iasi.aeris-data.fr/O3> and
391 <https://iasi-sofrid.sedoo.fr/>. The IASI-IMS data is available via the NERC Centre for Environmental
392 Data Analysis (CEDA) Jasmin platform subject to data requests. However, the IASI-IMS data and
393 TOMCAT simulations used in this study are available from
394 <https://homepages.see.leeds.ac.uk/~earrjpo/to3re/>. The ozonesonde data for WOUDC, SHADOZ and
395 NOAA is available from <https://woudc.org/>, <https://tropo.gsfc.nasa.gov/shadoz/> and
396 <https://gml.noaa.gov/ozwv/ozsondes/>.

397 Author Contributions

398 RJP conceptualised, planned and undertook the research study. AR provided the SOCRATES radiative
399 kernel. BB, ELF, BJK, RS, BGL, LJV, AB and CW provided the IASI ozone data and advice on using the
400 products. MAP performed the TOMCAT model simulations with support from MPC and WF. CR
401 provided advice and help during RP's ESA CCI fellowship. RJP prepared the manuscript with
402 contributions from all co-authors.

403 Conflicts of Interest

404 The authors declare no conflicts of interest.

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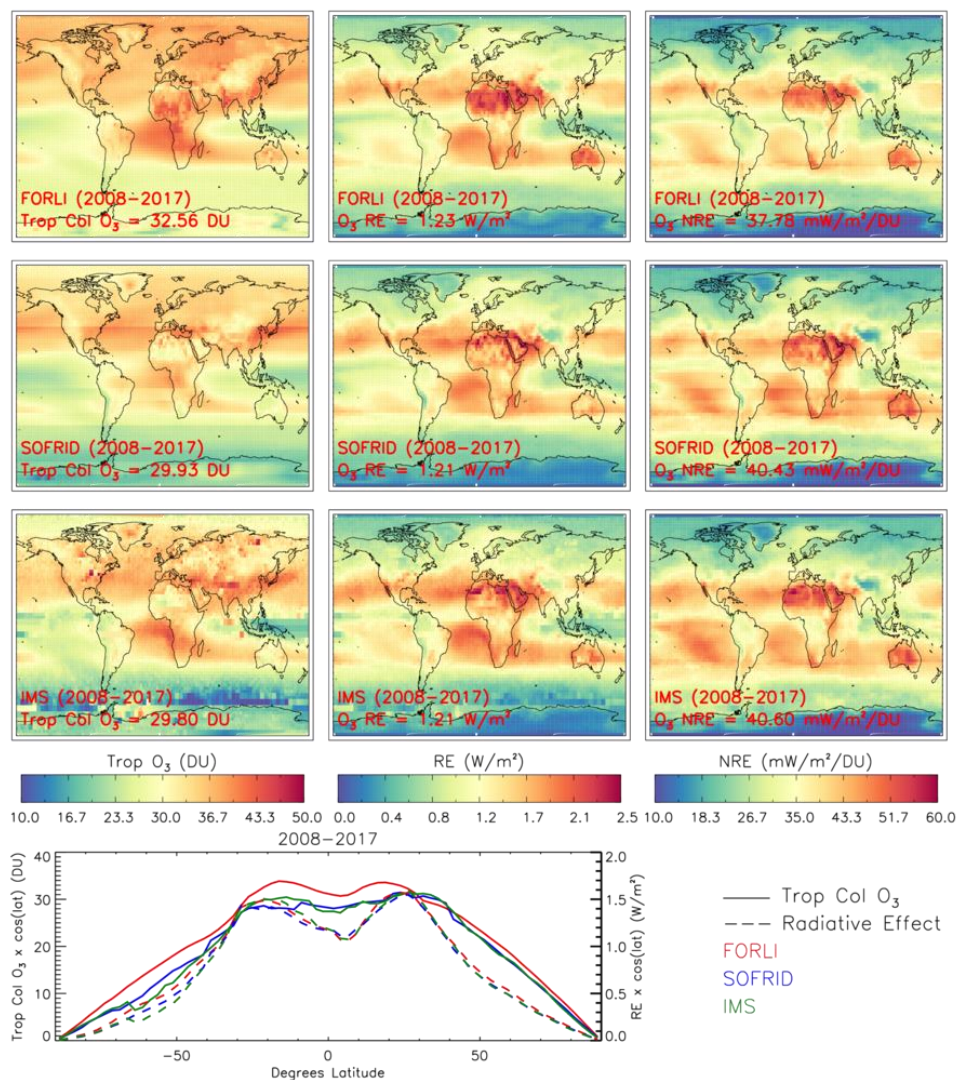
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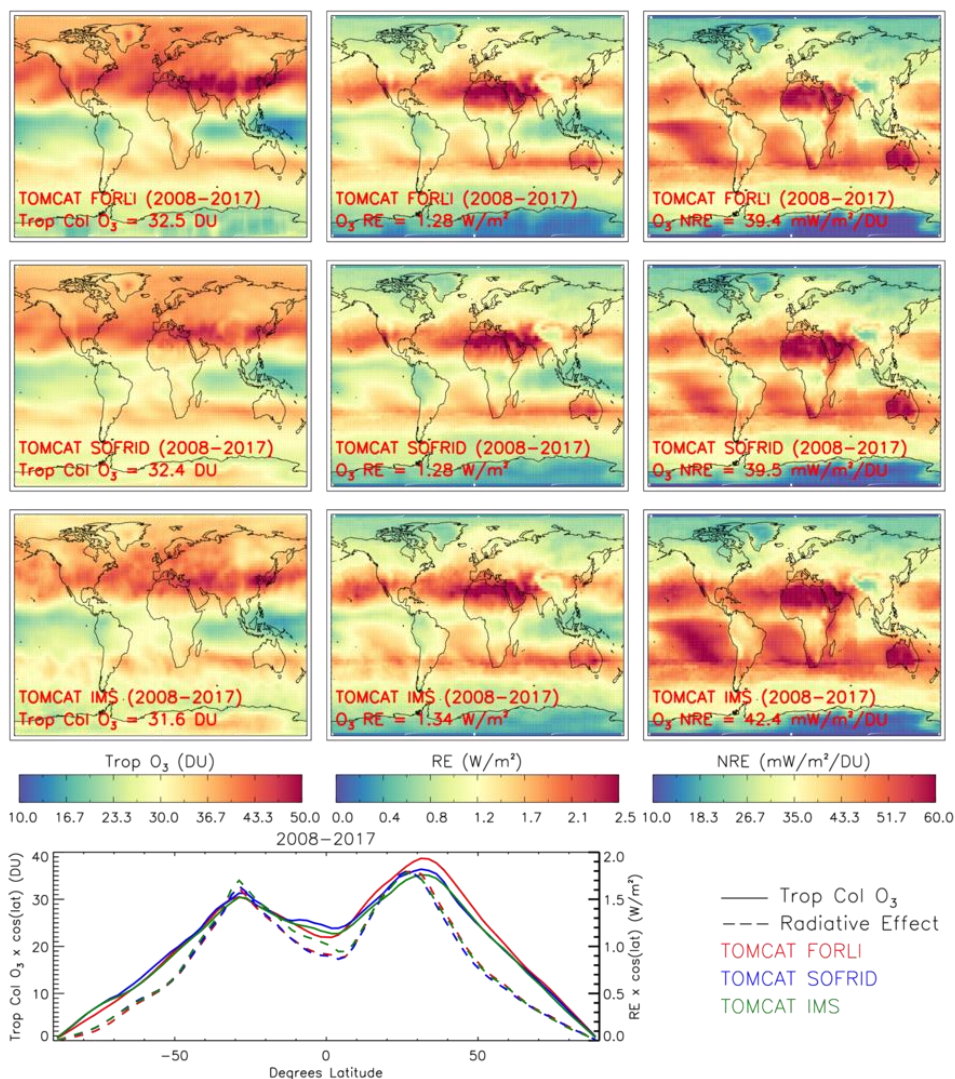
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574 **Figures:**



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 576 **Figure 1:** Tropospheric column O₃ (TCO₃, DU), tropospheric O₃ radiative effect (TO₃RE, W/m²) and
 577 normalised TO₃RE (NTO₃RE, mW/m²/DU) averaged for 2008 to 2017 for IASI-FORLI (top row), IASI-
 578 SOFRID (middle row) and IASI-IMS (bottom row). Zonal averages of TCO₃ (DU, solid lines) and TO₃RE
 579 (W/m², dashed lines), both weighted by cosine of latitude, is shown in the bottom panel from all the
 580 IASI instruments.

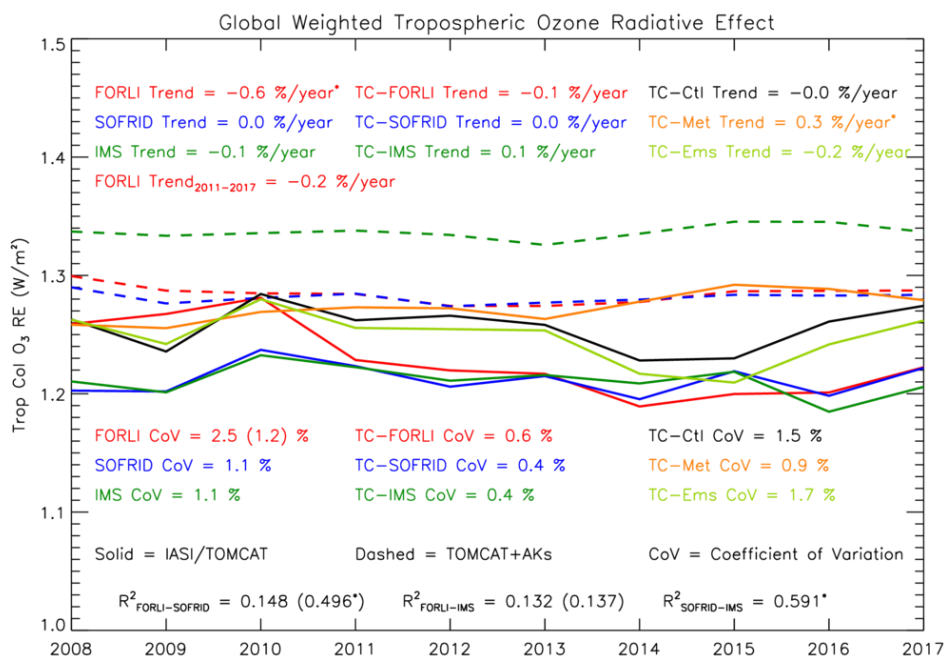


581
 582 **Figure 2:** TCO₃ (DU), TO₃RE (W/m²) and NTO₃RE (mW/m²/DU) averaged for 2008 to 2017 for
 583 TOMCAT with the averaging kernels (AKs) applied from IASI-FORLI (top row), IASI-SOFRID (middle
 584 row) and IASI-IMS (bottom row). Zonal averages of TCO₃ (DU, solid lines) and TO₃RE (W/m², dashed
 585 lines), both weighted by cosine of latitude, is shown in the bottom panel from all the IASI
 586 instruments.

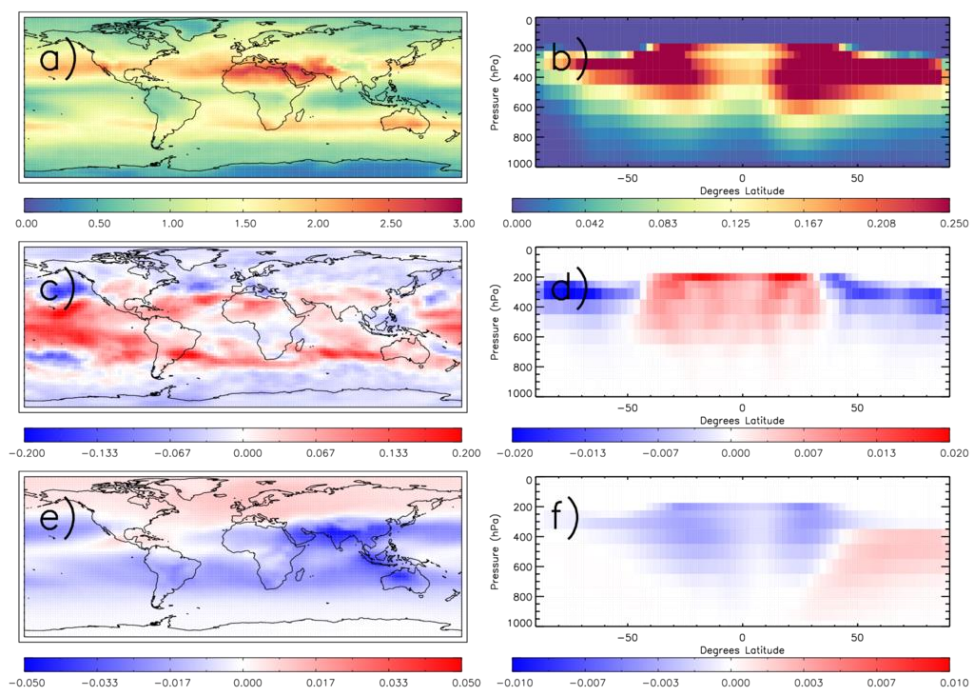
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590
 591 **Figure 3:** Annual global mean time-series of TO_3RE (W/m^2), between 2008 and 2017, for IASI-FORLI
 592 (red-solid), IASI-SOFRID (blue-solid) and IASI-IMS (green-solid). TOMCAT with the IASI-FORLI (red-
 593 dashed), IASI-SOFRID (blue-dashed) and IASI-IMS (green-dashed) AKs applied, original TOMCAT
 594 simulation (black-solid), TOMCAT with fixed emissions (lime-solid) and TOMCAT with fixed
 595 meteorology (orange-solid) are also shown. The linear trend (%/year) is shown as well as the
 596 percentage coefficient of variation (CoV). The correlation between IASI time-series are shown by the
 597 R^2 values. Significant linear trends and correlations in the TO_3RE are shown by an *. TC represents
 598 TOMCAT. The IASI-FORLI trend for 2011 to 2017 is also shown as well as the CoV and R^2 in brackets in
 599 addition to the statistical metrics over the full time period due to record inhomogeneities prior to
 600 2011 (Boynard et al., 2018).



601

602 **Figure 4:** a) TOMCAT control run TO₃RE (W/m²), b) TOMCAT control run zonal average grid box O₃
603 radiative effect (W/m²), c) TOMCAT fixed meteorology – TOMCAT control TO₃RE difference (W/m²),
604 d) TOMCAT fixed meteorology – TOMCAT control zonal average grid box O₃ radiative effect
605 difference (W/m²), e) TOMCAT fixed emissions – TOMCAT control TO₃RE difference (W/m²), f)
606 TOMCAT fixed emissions – TOMCAT control zonal average grid box O₃ radiative effect difference
607 (W/m²).

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