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

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Key Points:

- Using satellite data and model simulations, we quantify the long-term (2008-2017) global average tropospheric ozone radiative effect to range between 1.21 and 1.28 W/m^2.
- Satellite/modelled long-term trends in the tropospheric ozone radiative effect have remained stable (2008-2017) yielding no substantial changing influences on climate.
- Meteorological variability has been important in stabilising the global tropospheric ozone radiative effect with time.

Abstract:

Using state-of-the-art satellite ozone profile products, and chemical transport model, we provide an updated estimate of the tropospheric ozone radiative effect (TO$_3$RE) and observational constraint on its variability over the decade 2008-2017. Previous studies have shown the short-term (i.e. a few years) globally weighted average TO$_3$RE to be 1.17±0.03 W/m^2, while our analysis suggests that the long-term (2008-2017) average TO$_3$RE to be 1.21-1.28 W/m^2. Over this decade, the modelled/observational TO$_3$RE linear trends show negligible change (i.e. ±0.1%/year), so the tropospheric ozone radiative contribution to climate has remained stable with time. Two model sensitivity experiments fixing emissions and meteorology to one year (i.e. start year – 2008) show

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

Plain Language Summary:

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

1. Introduction

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

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

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

(MLS) measurements, also known as OMI-MLS product for January and July 2005, to estimate the resultant instantaneous TO_3RE at the tropopause to be 1.53 W/m^2. Worden et al., (2008) used ozone profile data for 2006 from the Tropospheric Emissions Spectrometer (TES), on-board NASA's Aura satellite, to estimate the average instantaneous long-wave TO_3RE at the top-of-the-atmosphere (TOA) over the oceans (45°S-45°N) to be 0.48±0.14 W/m^2. Worden et al., (2011), using TES data for August 2006, estimated the instantaneous long-wave TO_3RE at TOA to be 0.33 W/m^2. Later, Bowman et al., (2013) also used TES data (averaged between 2005 and 2009) to constrain the simulated instantaneous long-wave TO_3RE from an ensemble model average. They found that seasonally, TES long-wave TO_3RE peaks in northern Africa/Mediterranean/Middle East in June-July-August over 1.0 W/m^2 with minimum values (0.0-0.2 W/m^2) over the winter-time high-latitudes. Overall, the ensemble average long-wave TO_3RE low bias was 0.12 W/m^2. Doniki et al., (2015) took this further by calculating the instantaneous long-wave TO_3RE from the Infrared Atmospheric Sounding Interferometer (IASI), though using a small subset of the data, and found estimates from Worden et al., (2008), using TES, had a low bias of ~25%. Rap et al., (2015) also used TES satellite ozone profile observations (2005-2008) in combination with the TOMCAT chemical transport model (CTM) and provided the first robust satellite constraint on annual globally weighted resultant TO_3RE (after stratospheric temperature adjustment) with a range of 1.17±0.03 W/m^2.

Following the methodology adopted in Rap et al. (2015), we exploit satellite ozone profile data from IASI, on the MetOp-A satellite, which has a long-term record and substantial spatial coverage, in combination with the TOMCT CTM, to improve the TO_3RE estimate and investigate its long-term variability and implications for climate. The satellite data, radiation model and CTM used are discussed in Section 2, our results are presented in Section 3 and Section 4 summarises our conclusions.

2. Observations and Model

2.1. Satellite Observations

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

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

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

2.2. Ozonesondes

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

$$\boldsymbol{sonde_{AK}} = \boldsymbol{AK}.(\boldsymbol{sonde_{int}} - \boldsymbol{apr}) + \boldsymbol{apr} \qquad (1)$$

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

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

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

2.3. TOMCAT

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

2.4. Radiative Transfer Model and Kernel

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

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

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

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

3. Results

3.1. Tropospheric Ozone Radiative Effect

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

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

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

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

TO_3 in the mid-troposphere where the radiative kernel has less sensitivity. Further to this, as the IASI ozone products only have approximately 1.0 DOFS, the harmonisation of the products using the ozonesondes can only be done on a tropospheric column level and thus the scaling of the satellite derived TO_3RE (i.e. even though the upper troposphere is the most sensitive region to ozone radiative properties, the scaling of the TO_3RE is applied based on the satellite-ozonesonde TCO_3 relative differences).

TOMCAT allows for a further quantification of the TO_3RE in the satellite-era and the ability to run sensitivity experiments to explore important processes. Therefore, the TOMCAT equivalent metrics from **Figure 1** are presented in **Figure 2**. Evaluation of the model using the IASI products and ozonesondes (see **SI-2**, **Figure S3** & **S4**) shows the model generally captures the TCO_3 spatial pattern and absolute values. In the tropics (mid/high-latitudes), the model underestimates (overestimates) by approximately 10-20% on average. These biases are comparable with other modelling studies evaluating models against satellite TO_3 observations (e.g. Archibald et al., 2020; Monks et al., 2017; Nassar et al., 2009; Young et al., 2013), indicating that TOMCAT is a suitable modelling framework in this study.

The globally mean TCO_3 from TOMCAT (2008-2017) with the three sets of AKs applied (**Figure 2 left column**) ranges between 31.6 and 32.5 DU, so it slightly larger than the IASI data sets in **Figure 1**. When translated into TO_3RE, the peak values from TOMCAT (with AKs applied) ranges between 2.0 and >2.5 W/m^2 over Africa, Australia and the sub-tropics. The globally weighted TO_3RE for TOMCAT with the IASI-FORLI and IASI-SOFRID AKs applied is 1.28 W/m^2 and thus moderately higher than IASI (1.21-1.23 W/m^2) but comparable overall. However, the globally weighted TO_3RE for TOMCAT with IASI-IMS AKs applied is larger at 1.34 W/m^2. As TOMCAT has a positive TCO_3 bias with the observations in the sub-tropics, where the TO_3RE influence is most pronounced, this probably explains the larger model TO_3RE values. In the bottom panel of **Figure 2**, the zonal profiles (weighted by cosine of degree latitude) for TCO_3 (TO_3RE) are consistent with IASI as high-latitude values are near-zero, mid-latitude values range between 10.0 and 20.0 DU (0.5 to 1.0 W/m^2) and sub-tropical values range between 30.0 and 38.0 DU (1.5 and 1.7 W/m^2). There is a decrease to approximately 25.0 DU (1.0-1.3 W/m^2) in the tropics. In terms of the NTO_3RE, the TOMCAT (with AKs applied) global weighted values range between 39.4 and 42.4 mW/m^2/DU, which is similar to IASI. The peak NTO_3RE values are over the oceans (50.0-60.0 mW/m^2/DU) and over Africa/Australia (>60.0 mW/m^2/DU). Like for IASI, the TCO_3 values over the South Pacific are lower than the other ocean values but the NTO_3RE values are similar, again showing that despite the lower TO_3, the South Pacific region is important for the global TO_3RE given its greater sensitivity (i.e. more radiative effect per unit of TO_3).

3.2. Temporal Evolution of the Tropospheric Ozone Radiative Effect

As IASI has daily global coverage (Clerbaux et al., 2009), we are able to derive annual average 3D ozone fields between 2008 and 2017, thus providing the first assessment of temporal variability and tendency in satellite derived TO_3RE. **Figure 3** shows the annual TO_3RE time series for all three IASI products. First thing to note, is that the Eumetsat meteorological data used to retrieve ozone for the IASI-FORLI product is subject to inhomogeneities (Boynard et al., 2018; Wespes et al., 2018). As a result, we include long-term analysis of the IASI-FORLI data for the full time period (2008-2017) and then a sub-time period (2011-2017) given the large inhomogeneity in September 2010 reported by Boynard et al., (2018) and Wespes et al., (2018). Here, we can derive the TO_3RE to quantify the absolute values (e.g. are they generally similar year to year) and how they compare between

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

For IASI-SOFRID and IASI-IMS, the annual TO_3RE values range between 1.19 and 1.24 W/m^2 across the 2008-2017 time period. IASI-FORLI has somewhat larger values at the start of the record (1.26-1.28 W/m^2) before tending to that of IASI-SOFRID/IASI-IMS from 2011 onwards. Correlations (squared) in the annual TO_3RE time-series between IASI-FORLI and IASI-SOFRID (IASI-IMS) are poor at R^2=0.148 (R^2=0.132). However, IASI-SOFRID and IASI-IMS have a much stronger agreement with R^2=0.591 (significant at the 95th confidence level, CL95%) sharing nearly 60% of the temporal variability. We also calculate the coefficient of variation (CoV, i.e., time series standard deviation divided by its mean) to assess the inter-annual variability. For IASI-SOFRID and IASI-IMS, this is 1.1%, but for IASI-FORLI it is 2.5%. Therefore, there is more year-to-year variability in the IASI-FORLI TO_3RE record. However, when focussing on IASI-FORLI data for 2011-2017, the CoV drops to 1.2% in-line with IASI-SOFRID and IASI-IMS. The correlation (squared) values are now $R^2_{FORLI-SOFRID}$=0.496 (significant at the CL95%) and $R^2_{FORLI-IMS}$=0.137, which shows improved agreement between IASI-FORLI and IASI-SOFRID, but slightly surprisingly not with IASI-IMS. Using ordinary least squares fit regression, IASI-FORLI, IASI-SORFRID and IASI-IMS have global average weighed TO_3RE linear trends of -0.6%/year (CL95%), 0.0%/year (non-significant) and -0.1%/year (non-significant). As the IASI-FORLI product has known inhomogeneities (hence the larger CoV), the insignificant IASI-SOFRID and IASI-IMS trends are more robust. This is supported by IASI-FORLI when only considering 2011-2017 with an insignificant linear trend of -0.2%/year. Therefore, this suggests negligible change in the contribution of TO_3 to the tropospheric radiative effect and thus climate over the recent past (i.e. 2008-2017).

TOMCAT global average weighed TO_3RE (without AKs applied) ranges between 1.24 and 1.29 W/m^2 between 2008 and 2017. The CoV is 1.5% for TOMCAT, so it is larger than both IASI-SOFRID and IASI-IMS. When the IASI AKs are applied to TOMCAT, there is a substantial shift in the modelled absolute TO_3RE values. TOMCAT with IASI-SOFRID and IASI-FORLI AKs applied ranged between 1.28 and 1.30 W/m^2. And for TOMCAT with the IASI-IMS AKs applied, the TO_3RE values peak at 1.33 to 1.34 W/m^2 between 2008 and 2017. As well as the increase in TO_3RE values, the application of the AKs squashes the TOMCAT inter-annual variability with corresponding CoV values between 0.4 and 0.6%, which is smaller than the original CoV of 1.5%. Interestingly, without the application of the AKs, the TOMCAT TO_3RE time-series has similar temporal variability (e.g. peaks in 2008, 2010 and 2017 and troughs in 2009 and 2014. Overall, all the TOMCAT TO_3RE time-series (with and without AKs applied have insignificant linear trends ranging between -0.1%/year and 0.1%/year. Therefore, even with the influence of the IASI AKs on the TOMCAT TO_3RE time-series, there appears to be a negligible trend in the modelled TO_3RE, supporting that of the IASI records. As a result, between 2008 and 2017, there has been limited change in TO_3 and TO_3RE, thus the impact of TO_3 on climate has remained stable.

To investigate the importance of emissions and meteorology on the long-term TO_3RE trends, TOMCAT was run using repeating emissions and repeating meteorology for 2008 (i.e. start of the time-series) in two sensitivity experiments for the full time-period. Here, we find that in absolute terms, using fixed emissions reduces the TO_3 burden and the TO_3RE as the time-series drops to 1.22 to 1.28 W/m^2 (i.e. minima in 2014 and 2015 more pronounced). However, the trend in TO_3RE (-0.2%/year) remains insignificant and that emissions are only moderately important in driving long-term tendencies in TO_3RE. On the other hand, meteorological factors, while not dramatically altering the absolute simulated TO_3RE values, are more important as fixing the meteorology yields a steady

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and significant increase (0.3%/year). Thus, without year-to-year variability in meteorology, temporal variability in TO_3 would likely have a more substantial impact on the present day climate.

Figure 4 shows the horizontal and vertical impact of the two sensitivity experiments on TOMCAT O_3 radiative effect (note the different colour bar scales). Consistent with **Figures 1** and **2**, the TOMCAT control TO_3RE has peak values (>2.50 W/m^2) over northern Africa and throughout the sub-tropics (approximately 2.0 W/m^2, **Figure 4a**). Vertically, the TOMCAT peak ozone radiative effect (>0.25 W/m^2) is in the upper troposphere (**Figure 4b**) with the largest impact in the sub-tropics of both hemispheres (500-200 hPa). Similar values extend through the hemispheric mid-latitudes but in a smaller pressure range (400-300 hPa). As shown in **Figure 3**, the fixed meteorological run imposes a significant TO_3RE trend on the modelled tendency between 2008 and 2017. From **Figure 4c**, the difference between the fixed meteorology and control runs shows mainly positive TO_3RE differences of 0.1 to >0.2 W/m^2 throughout the tropics and sub-tropics, though there is considerable spatial variation due to changes in the global circulation. In the high and mid-latitudes, there are smaller scale negative differences ranging between -0.1 and 0.0 W/m^2 (though some differences up to -0.15 W/m^2 in the sub-tropical Pacific). In the upper troposphere (**Figure 4d**), the zonal average O_3 radiative effect is consistent with positive differences of up to 0.02 W/m^2 at approximately 200 hPa in the tropics and sub-tropics. The positive differences (approximately 0.01 W/m^2) filter down to 600 hPa in the same latitudinal range. In the mid-latitudes, the peak negative differences are approximately -0.02 to -0.015 W/m^2 at 300 hPa, with a reach down to 500 hPa at -0.005 W/m^2. Overall, as shown in **Figure 3**, the fixed meteorology run increases the global average TO_3RE. While this could be a specific signal related to the 2008 meteorology (i.e. it is conducive to TO_3 formation), it clearly shows that the upper tropospheric tropical and sub-tropical regions predominantly control the global TO_3RE average and its temporal variability (i.e. the region where the meteorological inter-annual variability is buffering underlying increases in TO_3RE). With fixed emissions, there is a general increase (decrease) in TO_3RE in the tropics/sub-tropics (northern mid-latitudes) by 0.02 (-0.02) W/m^2. However, over tropical Asia, Indonesia and Australia, the decrease in TO_3RE is more substantial at -0.05 to -0.04 W/m^2 (**Figure 4e**). Vertically, there are decreases (increases) in the O_3 radiative effect of -0.005 (0.003) W/m^2 in the tropics/sub-tropics (northern mid-latitudes) between 600 and 200 (800 and 400) hPa (**Figure 4f**). Overall, the meteorological variability, in comparison to the long-term emission changes in O_3 precursor gases, has substantially more influence on the inter-annual variability of the global TO_3RE over this decade.

4. Conclusions

By using state-of-the-art satellite ozone profile retrievals from the Infrared Atmospheric Sounding Interferometer (IASI), on-board MetOp-A, in combination with the TOMCAT chemical transport model (CTM), we provide an updated estimate of the tropospheric ozone radiative effect (TO_3RE) and provide the first observational constraint on its variability over the decade 2008-2017. Building upon the previous study of Rap et al., (2015), who quantified the globally weighed average TO_3RE to be 1.17±0.03 W/m^2 (based on data between 2005 and 2008), we find the long-term average TO_3RE, between 2008 and 2017, to range from 1.21 and 1.28 W/m^2. Secondly, neither the modelled, nor the observed TO_3RE suggest any substantial change during this period. Therefore, the tropospheric ozone contribution to climate, through its infrared radiative properties, has remained stable with time during 2008-2017. Investigations of the importance of ozone precursor emissions and meteorology, through targeted sensitivity experiments repeating emissions and meteorology for 2008 (i.e. year at start of time-series), suggest that emissions have a limited impact on the globally

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weighted average TO_3RE. Meanwhile, fixing the meteorology to a specific year (i.e. 2008) introduces a significant positive trend in global TO_3RE, indicating that the meteorological variability in the tropical/sub-tropical upper troposphere has been important in stabilising the tropospheric ozone contribution to climate, via radiative properties, in the recent past (i.e. satellite-era).

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Data Availability

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

Author Contributions

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

Conflicts of Interest

The authors declare no conflicts of interest.

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Figures:

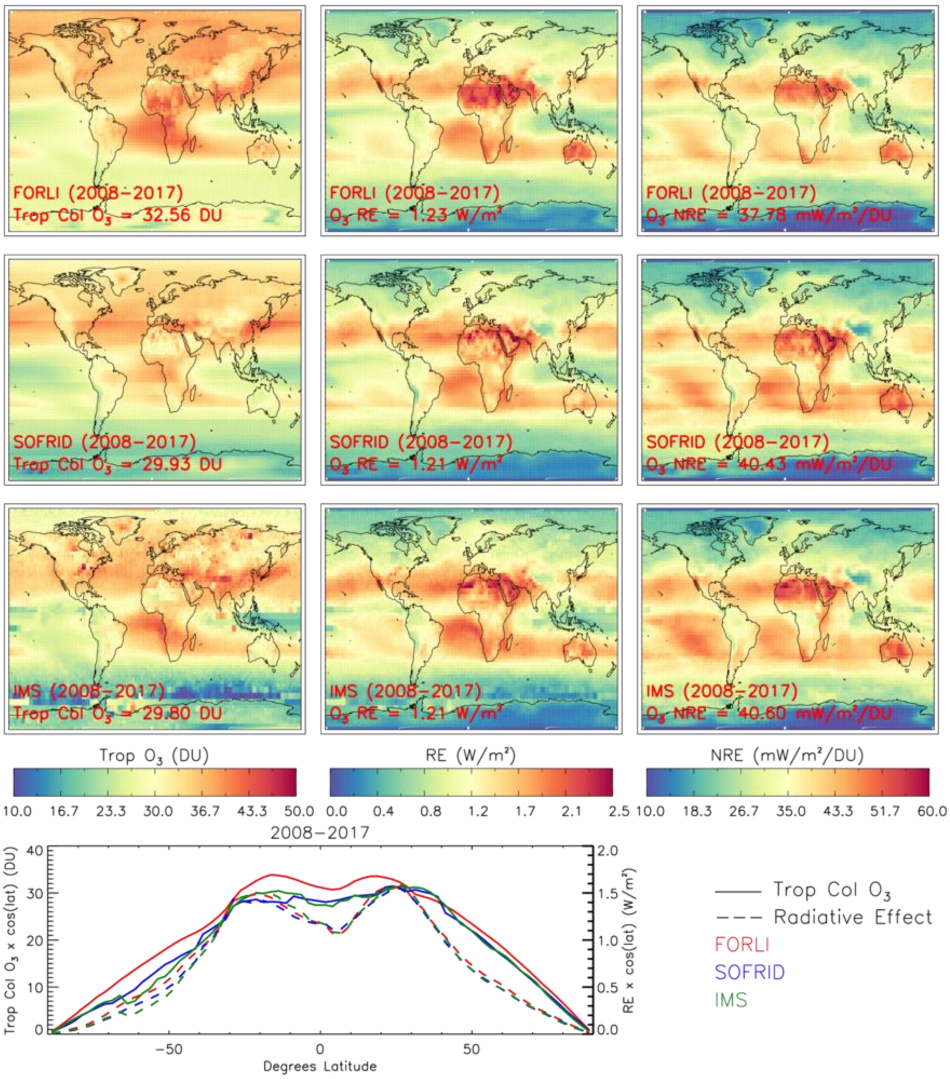
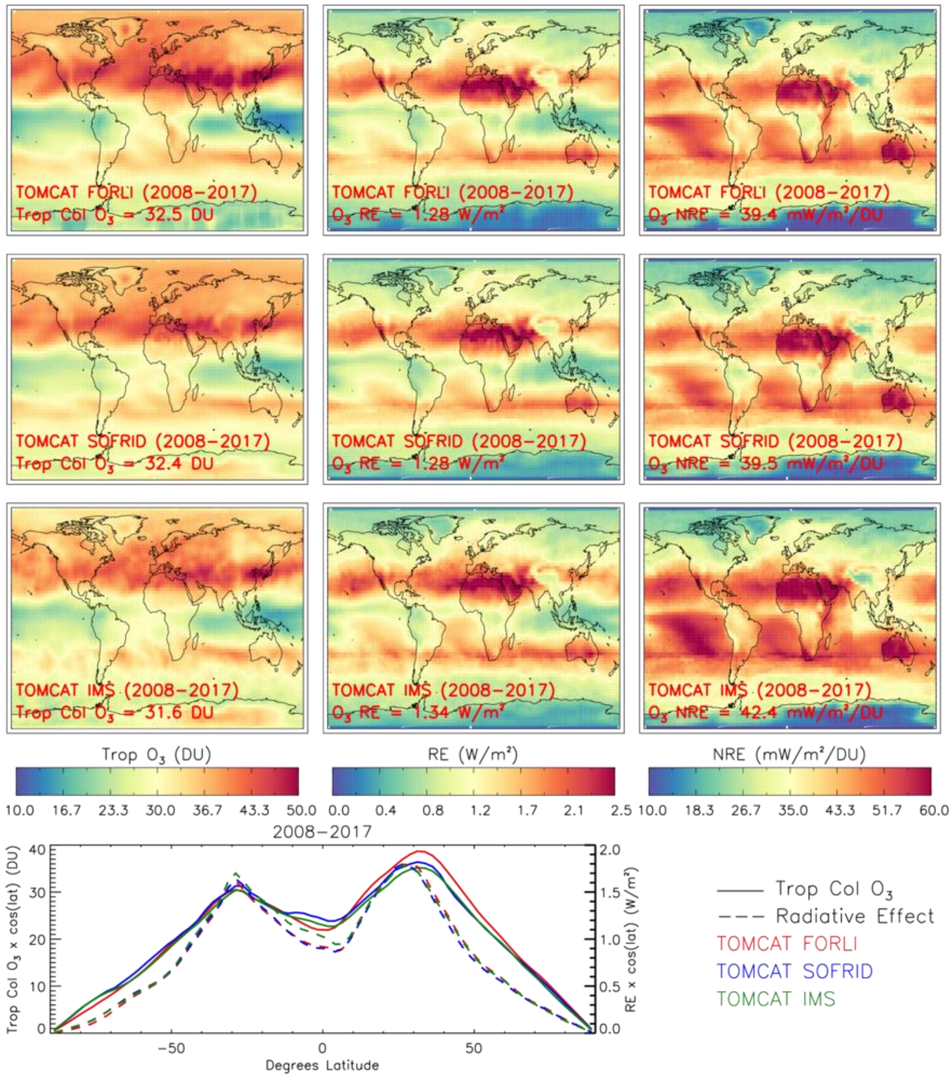


Figure 1: *Tropospheric column O_3 (TCO_3, DU), tropospheric O_3 radiative effect (TO_3RE, W/m^2) and normalised TO_3RE (NTO_3RE, $mW/m^2/DU$) averaged for 2008 to 2017 for IASI-FORLI (top row), IASI-SOFRID (middle row) and IASI-IMS (bottom row). Zonal averages of TCO_3 (DU, solid lines) and TO_3RE (W/m^2, dashed lines), both weighted by cosine of latitude, is shown in the bottom panel from all the IASI instruments.*



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582 **Figure 2**: *TCO_3 (DU), TO_3RE (W/m^2) and NTO_3RE (mW/m^2/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 TCO_3 (DU, solid lines) and TO_3RE (W/m^2, 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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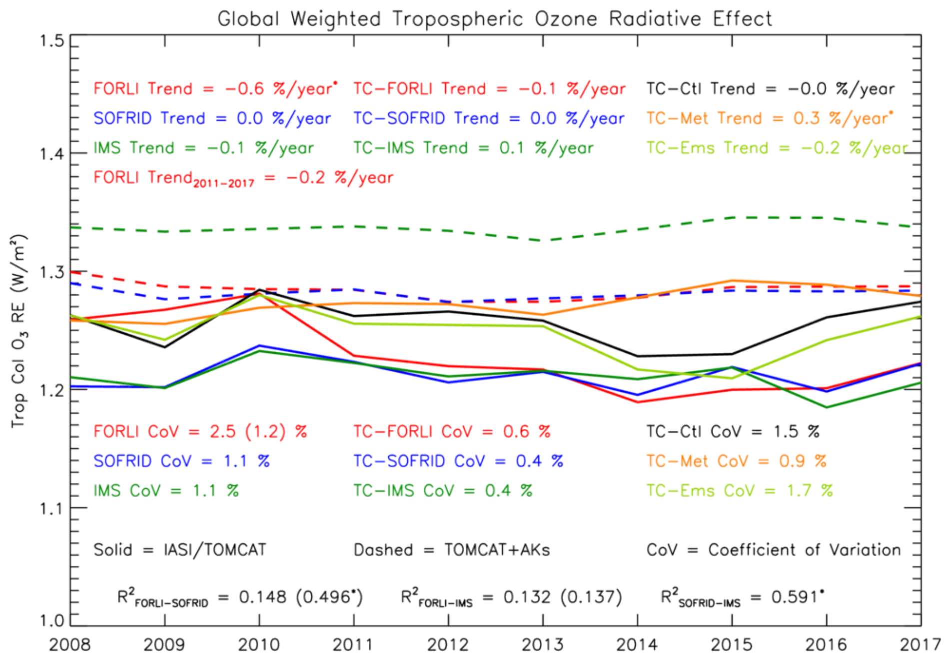

Figure 3: *Annual global mean time-series of TO_3RE (W/m^2), between 2008 and 2017, for IASI-FORLI (red-solid), IASI-SOFRID (blue-solid) and IASI-IMS (green-solid). TOMCAT with the IASI-FORLI (red-dashed), IASI-SOFRID (blue-dashed) and IASI-IMS (green-dashed) AKs applied, original TOMCAT simulation (black-solid),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

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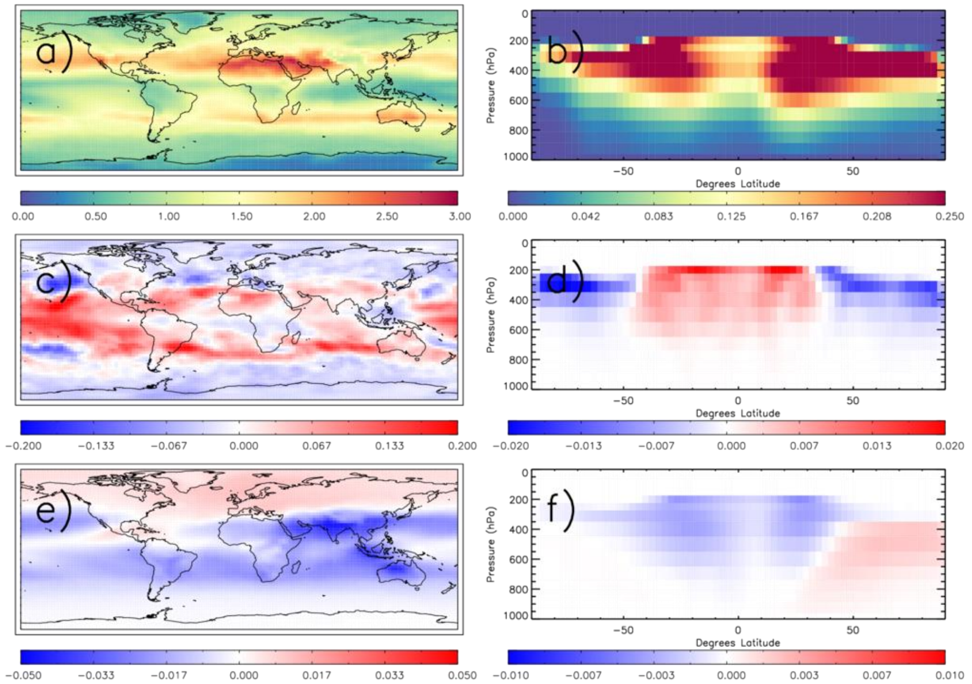

Figure 4: a) *TOMCAT control run TO_3RE (W/m^2), b) TOMCAT control run zonal average grid box O_3 radiative effect (W/m^2), c) TOMCAT fixed meteorology – TOMCAT control TO_3RE difference (W/m^2), d) TOMCAT fixed meteorology – TOMCAT control zonal average grid box O_3 radiative effect difference (W/m^2), e) TOMCAT fixed emissions – TOMCAT control TO_3RE difference (W/m^2), f) TOMCAT fixed emissions – TOMCAT control zonal average grid box O_3 radiative effect difference (W/m^2).*