Tropical Tropospheric Ozone Trends (1998 to 2023): New Perspectives from SHADOZ, IAGOS and OMI/MLS Observations Revisions-TC-Iuly 2025

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Abstract. Tropospheric ozone trends are important indicators of climate forcing and surface pollution yet relevant satellite observations are too uncertain for assessments. The assessment project TOAR-II has used multi-instrument, ground-based data for global trends over 2000-2022 (Van Malderen et al., 2025a,b). For the tropics trends are derived from SHADOZ ozonesonde profiles (Thompson et al., 2021, "T21"; Stauffer et al., 2024) or combinations of satellite, SHADOZ and IAGOS aircraft measurements [Gaudel et al., 2024]. We extend T21 that covered 1998-2019, analyzing SHADOZ data at 5 sites with a Multiple Linear Regression (MLR) model for 1998-2023, and reporting trends for two free tropospheric (FT) segments, lowermost stratosphere and total tropospheric column (TrCO<sub>sonde</sub>). Trends for the Aura period, 2005-2023, are computed from OMI/MLS TrCO<sub>satellite</sub>. We find: (1) Extending SHADOZ analyses four years shows little change from T21; TrCO<sub>sonde</sub> trends are small (0.5-1 DU/dec) except over SE Asia. (2) Annual trends for TrCO<sub>sonde</sub>, and OMI/MLS TrCO<sub>satellite</sub> agree within uncertainties at 4 of 5 sites. Sensitivity tests show: (1) adding thousands of FT IAGOS profiles to SHADOZ yields little change in trends; SHADOZ sampling is sufficient. (2) QR and MLR median trends are both near-zero but QR captures extremes (5%-ile, 95-%ile) with changes up to +1 DU/decade (p < 0.10). (3) Twelve-year analyses for trends lead to uncertainty changes too large for an assessment. This study and Van Malderen et al. (2025a;b) provide the most reliable TOAR-II trends to date: over the past ~25 years, tropical FT ozone changes are modest, ~(-3-+3)%/decade, except over SE Asia.

### 1 Introduction

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The importance of tropical tropospheric ozone in atmospheric composition and climate variability has long been known. Although the thickness of total column ozone (TrCO) in the tropics (~250-325 Dobson Units, DU; 1 DU=2.69x10<sup>16</sup>cm<sup>-2</sup>) is much less than in the extra-tropics (350-450 DU), the latitude band from -30° to +30° covers roughly 1/3 of the Earth's surface. In this region tropospheric ozone is a major source of global OH (hydroxyl radical), key to Earth's oxidizing capacity (Thompson et al., 1992), controlling the lifetimes of countless biogenic and anthropogenic species. Global OH also controls the lifetime of methane, a powerful greenhouse gas with both natural and anthropogenic sources (Khalil, 2000). Methane (CH<sub>4</sub>) increases alone add ozone to the troposphere and methane's oxidation by OH to carbon monoxide (CO), that also affects the amount of OH, establishes a feedback cycle among O<sub>3</sub>-OH-CH<sub>4</sub>-CO (Thompson et al., 1986; 1990). Regional variability in factors controlling the cycle derives from local levels of the shorter-lived nitrogen oxides and reactive volatile organic compounds (VOC). The same latitude band (-30° to +30°) is where the "tropical pipe" (Plumb, 1996) introduces ozone and ozone-destroying or creating trace species into the lowermost stratosphere (LMS). Trends in tropical tropospheric and LMS ozone are of interest for several reasons. First, free tropospheric (FT) ozone is an important greenhouse gas. There is a potential for significant changes in FT ozone because parts of the tropics are in areas of rapid changes in emissions. These may be caused by economic development (Zhang et al., 2016) and/or variations in land-use and fire activity (Christensen et al., 2022; Tsivlidou et al., 2023). Second, with relatively low ozone amounts relative to the extra-tropics, ozone in the tropical troposphere is highly sensitive to dynamical interactions. Recent analyses of ozone profiles over the past ~25 have shown regional meteorological changes propagating to seasonal ozone increases. Stauffer et al. (2024) verified that a suspected decline in convective activity

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(Thompson et al., 2021; hereafter T21) was a driving factor in 1998-2022 ozone increases over equatorial southeast Asia. A new report on decreasing tropical cloud cover may indicate changes in convection (Tselioudis et al., 2025). At Réunion a combination of shifting anticyclones caused an increase in free tropospheric ozone from 1998-2023 (Millet et al., 2025). Recurring influences of climate oscillations on FT and LMS ozone are well-documented in ozonesonde and satellite data (Thompson et al., 2001; Ziemke et al., 2003; Ziemke et al., 2006; Ziemke et al., 2019; Lee et al., 2010; Randel and Thompson, 2011; Thompson et al., 2011)

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# 1.1 The TOAR Project. Challenges in Assessing Tropospheric Ozone Trends

Context for this study comes from the International Global Atmospheric Chemistry/Tropospheric Ozone Assessment Report (IGAC/TOAR) that is completing its second phase, TOAR II, initiated in 2020. The first TOAR, designated here as TOAR I and published as a collection of 11 papers in *Elementa*, 2017-2020; https://online.ucpress.edu/elementa/toar) included an assessment of surface ozone changes (Chang et al., 2017) based on a vast set of surface ozone measurements from 7 continents (Schultz et al., 2017). However, because the FT is the region of greatest radiative forcing by ozone, the trends community needs profile data. Ozone observations for the FT are relatively sparse. A TOAR I evaluation by Tarasick et al. (2019) pointed out the uneven geographic coverage of ozone profiles from soundings (~60 publicly available station records since the early 1990s) and aircraft landing and takeoff profiles to ~250 hPa that are used for FT ozone, analyses. Tarasick et al. (2019) also questioned the suitability of all ground-based (sonde, aircraft, lidar, passive spectrometers) for monitoring FT ozone using illustrations from multi-decadal records that include obsolete techniques, evolving versions of instruments and/or inconsistent absolute calibration. Nonetheless, several follow-on studies to TOAR I employed sonde (T21) and commercial aircraft data (from the In-service Aircraft for a Global Observing System, IAGOS; https://iagos.org), with 5-10% accuracy or better, to estimate trends from the 1990s to ~2018 (Gaudel

Efforts to fill gaps with tropospheric ozone estimates from satellite data, preferred over in-situ methods for their even global coverage, have been mixed. For TOAR I Gaudel et al. (2018) pointed out that trends derived from five satellite products covering tropics and mid-latitudes for the 2005-2016 period differed from one another not only in magnitude but in sign. The newer TOAR II evaluation of six satellite products for 2015-2019 over the tropics (Gaudel et al., 2024), where satellite estimates tend to be most reliable (T21), exhibited a range of values. Not only were uncertainties among the products highly variable; comparisons of monthly mean satellite columns with sonde and IAGOS profiles up to 270 hPa exhibited  $r^{(2)}$  correlations as low as 0.27.

Other TOAR II studies also reveal a persistent uncertainty in the application of satellite data for trends analyses. Pope et al. (2023) published global trends 2005-2017 OMI total column ozone data that

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are too high, because of a drift in OMI that was corrected in Gaudel et al. (2024). The latter study led to a satellite-based estimate for tropical ozone trends for 2005-2019 of zz % or DU in TrCO (tropospheric column ozone). Trends determined from non-uv satellite instruments have been disappointing (Gaudel et al., 2018). A TOAR II contribution by Froidevaux et al. (2025) examined changes in tropical ozone (+ 26 deg latitude) over the 2005 to 2020 period using measurements from the three lowest levels of MLS (Microwave Limb Sounder). Unfortunately, examination of the zonal structure of MLS data at the 146 hPa and 213 hPa levels (Fig. 1 in Froidevaux et al., 2025) shows that the prominent wave-one feature (Thompson et al., 2003: Thompson et al., 2017) in tropical ozone is absent, i.e., MLS does not capture regional differences as OMI-based products do. Froidevaux et al. (2025: Figs. 1, 2, 5) compare the MLS global structure to ozone output at 215 and 146 hPa from 3 models, Whole Atmosphere Community Climate Model (WACCM6) and two variants of the Community Atmosphere Model with Chemistry (CAMchem), each variant using different anthropogenic emissions. The models are likewise zonally uniform in upper tropospheric ozone so they do not result in meaningful tropical upper tropospheric ozone trends either.

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A TOAR II-related investigation (Boynard et al., 2025) uses IR-based retrievals from the MetOP IASI satellite instrument to determine trends for the period 2008-2023. As in Gaudel et al. (2018) the newer IASI tropospheric ozone climatology differs from that of uv-based products; IASI's negative tropical trends continue to diverge with ozone increases from uv-based products. Boynard et al. (2025) offer little explanation for the ongoing discrepancies. They note that their 12-yr period trends (2008-2019), roughly half as long as the sonde or aircraft trends in T21 or Gaudel et al. (2024) may be too short for a statistically robust result. Pennington et al. (2025) does provide some insight into long-term changes of 3 satellite IR products (TROPESS CrIS, AIRS, AIRS+OML) compared to ozonesondes and finds that the global tropospheric ozone satellite-sonde bias is approximately one third the magnitude of trends in global tropospheric ozone reported by TOAR-L.

Keppens et al. (2025) addressed the question of whether satellite data harmonization for nadir ozone profile and column products improves satellite data consistency for both their mean distributions and long-term changes. They concluded that their harmonization methods reduce inter-product dispersion by about 10% when comparing to the HEGIFTOM global ozonesonde datasets from 43 sites, but there is a significant meridional dependence, and the dispersion reduction is not consistent in space or time. This implies that a substantial part of the inter-product differences is instrument and/or retrieval-specific and the harmonization methods have limitations for TOAR-II. An alternate method of combining the residual and profile products with a column fill-in method is expected to be published as the TOAR-II "satellite assessment."

# 1.2 TOAR II Studies with Ground-based Measurements. Statistical Issues

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TOAR II has engaged a more globally representative set of researchers than TOAR I and reports data and analyses from a larger set of observations. Dozens of TOAR-II-related publications can be reviewed at https://copernicus.org/articles/special issue10 1256.html. Given the persistent uncertainty of the satellite records, a number of TOAR II contributors formed a community project, the Harmonization and Evaluation of Ground-based Instruments for Free-Tropospheric Ozone Measurements (HEGIFTOM), to apply newly standardized ozone measurement and processing protocols for sondes, aircraft and other ground-based (GB) instruments: FTIR, tropospheric ozone lidar and Umkehr retrievals from Dobson spectrometers. The rationale is that GB networks, with stable operations at fixed sites and wellcalibrated instruments, e.g., as in the Network for Detection of Atmospheric Composition Change (NDACC; De Mazière et al., 2018), provide suitable time-series at dozens of sites over 7 continents and pole to pole. HEGIFTOM has two objectives: (1) harmonize data from ~80 long-term stations (1990s to 2023) in four GB networks using the most up-to-date reprocessing techniques with each record referenced to absolute standards; (2) calculate trends for the 2000 to 2022 period with harmonized data, reporting station trends with uncertainty. The trends for 55 individual stations are tabulated and illustrated in Van Malderen et al. (2025a; referred hereafter to HEGIFTOM-1). Regional trends based on merging selected stations in densely sampled areas appear in Van Malderen et al. (2025b).

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Early in the TOAR II study period we analyzed ozone profiles in the tropics collected in the Southern Hemisphere Additional Ozonesondes (SHADOZ) network (Thompson et al., 2003; Thompson et al., 2017) to compute FT and LMS ozone trends. The results in T21 are based on data from 8 combined SHADOZ stations within ± 15 degrees latitude; the Goddard Multiple-Linear Regression (MLR) model calculated trends from 1998 through 2019 from the surface to 20 km. Changes in layers between 5 and 15 km were relatively small, ~(1-3)%/decade at most, except over the equatorial SE Asian stations at Kuala Lumpur and Watukosek, Indonesia (Table 1). In T21 LMS ozone LMS (defined as within 15-20 km) computed with the MLR model displayed a seasonal loss (July through September) up to 10%/decade or 3%/decade, annually averaged. The loss maximized at ~18 km. However, at the same time of year, a positive trend in tropopause height (TH), derived from the SHADOZ radiosondes, was detected. Redetermining LMS ozone changes in the 5 km column above the tropopause zeroed out the apparent trend.

More recently, the Stauffer et al. (2024, referred to as S24) paper demonstrated that over the 25-yr period 1998-2022, early year (February through April/May) FT ozone increases in the SHADOZ record are associated with declining convection, most pronounced over SE Asia but observed to a lesser degree at the other stations. With the newest OMI/MLS-based satellite estimates of total tropospheric column ozone (TrCO), Gaudel et al. (2024) showed that during the Aura era (2005-2019) trends from satellite, SHADOZ and IAGOS aircraft profiles were in good agreement over SE Asia, similar to S24, but more

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divergent over the equatorial Americas and Africa. HEGIFTOM-1 trends (2000-2022) used data from 327 328 SHADOZ stations only below 300 hPa. 329 1.3 This study Deleted: S We use the Goddard MLR model to calculate trends in monthly mean SHADOZ data for the period 330 331 1998 through 2023, addressing the following questions: Compared to T21, that reported on the 1998-2019 period, what do FT and LMS ozone 332 trends look like in the equatorial zone (-15 $^{\circ}$  to +15 $^{\circ}$ ) with four additional years of SHADOZ 333 profiles? Because the extension covers 2020 to 2023, the comparisons are looking for impacts 334 of COVID-19 (Steinbrecht et al., 2021; Ziemke et al., 2022). 335 Deleted: Ziemke ref/? 336 How do trends in tropospheric ozone for 1998-2023 compare to trends for 2000 to 2023? In other words, is the 26-year trend biased by SHADOZ starting at the end of the 1997-1998 ENSO 337 338 that produced strong perturbations to tropical ozone (Thompson et al., 2001)? Deleted: /T 1999. Deleted: ; Ziemke? Japanese refs) How do SHADOZ total tropospheric column changes (TrCO<sub>sonde</sub>) compare to OMI/MLS 339 Formatted: Not Highlight 340 column ozone trends over the 2005-2023 period? Do the satellite data capture the seasonality of sonde-derived trends as noted in T21 and S24? 341 342 343 In addition to updated trends for equatorial SHADOZ sites, we have used SHADOZ profiles to to 344 investigate three statistical issues raised in HEGIFTOM-1 and several other TOAR II studies. 345 HEGIFTOM-1 calculated median trends for sondes and all other GB data using both Quantile 346 Regression (QR) and MLR models. Within the associated uncertainties of each the two methods 347 gave identical results for annually averaged trends. Here we explore special features of QR and MLR to learn more about the nature of the trends, e.g., seasonality (MLR), changes in the highest 348 349 and lowest quantile (QR). Deleted: ML 350 Second, we use MLR to evaluate the sensitivity of trends on sample size as raised in TOAR II Deleted: Deleted: ] papers (key Chang/Cooper /Gaudel et al. 2024) and HEGIFTOM-1 by augmenting equatorial 351 Deleted: papers 352 SHADOZ data with tropical IAGOS profiles for the appropriate region. These calculations are 353 carried out for FT ozone, i.e., in the region where radiative forcing is most effective and both 354 sondes and aircraft sample (700-300 hPa, roughly 5-10 km). Satellite records are variable in length, with the most frequently used tropospheric products 355 356 starting after 2004 and a number of them merging measurements from multiple instruments. We 357 examine the sensitivity of trends to length of the observation period using QR by comparing the Deleted: /MLR? 26-yr SHADOZ ozone trend to a recalculation that coincides with the IASI period 2008-2019 358 359 (Boynard et al., 2025). 360 Data and analysis methods appear in Section 2 with Results and Discussion in Section 3. Section 4 Deleted: ¶

presents a Summary and Conclusions.

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# 2. Data and Methods of Analysis

### 2.1 Datasets for ozone

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Three datasets are used in our study: ozonesonde profiles from the SHADOZ network; partial ozone profiles from the IAGOS commercial aircraft network; monthly-averaged OMI/MLS tropospheric ozone column estimates from 2005 through 2023.

# 2.1.1 SHADOZ ozonesonde observations

Fig. 1a displays the SHADOZ network stations, italicized with coordinates in Table 1. As of 2025
Quito (Cazorla, 2016) and Palau (Müller et al., 2024) soundings since 2014 have been added to the archive at https://tropo.gsfc.nasa.gov/shadoz. The ozone profiles are obtained from electrochemical concentration cell ozonesondes coupled to standard radiosondes as described in earlier publications, e.g., Thompson et al., (2003); Thompson et al., (2007); Thompson et al., (2019). The profiles are archived with ozone uncertainties calculated with each individual ozone partial pressure available as separate files at the SHADOZ archive (Witte et al., 2018; WMO/GAW Rep. 268, 2021). Recent evaluations of ozonesonde data have established the quality of the global ECC network. Measurements of total column ozone (TCO) from 60 global stations average within ±2% agreement with total ozone from 4 uvtype satellites since 2005 (Stauffer et al., 2022). About half the SHADOZ stations exhibit a ~3-5% dropoff in stratospheric ozone (Stauffer et al., 2020) that is not completely understood (Nakano and Morofuji, 2023; Smit et al., 2024). Accordingly, our study only uses ozone data below ~50 hPa, defining the lowermost stratospheric (LMS) ozone as bounded by 15-20 km.

For the update to T21, that was based on 1998-2019 SHADOZ v06 ozonesonde data (https://doi.org/10.57721/SHADOZ-V06), the same records for 8 equatorial sites, located between 5.8N and 14S (color-coded in Fig. 1b; italicized in Table 1) are used with four additional years (2020-2023) of ozone and P-T-U (pressure-temperature-humidity) profiles. These 8 stations have at least 14 years of data between 1998 and 2023, although several have multi-year gaps (Figs. 2, 3 Fig. S1). For more reliable statistics three of the "stations" or "sites" as they are referred to (Fig. 1b), are defined by combining profiles from pairs of launch locations abbreviated as follows: SC-Para for San Cristóbal-Paramaribo (dark blue dots in Fig. 1b); Nat-Asc for Natal-Ascension (red dots in Fig. 1b); KL-Java (cyan dots in Fig. 1b) for Kuala Lumpur-Watukosek (Table 2). T21 (see Supplementary Material) describes multiple tests that were conducted to verify that these combinations are statistically justified. Annual cycles in absolute column amounts (Fig. 2) and anomalies for the pairs were well-correlated. In T21 (Supplementary Material) total tropospheric columns integrated from sondes (TrCO<sub>satellite</sub> from OMI/MLS data over the period 2005-2019. It is important to note that the 8 well-correlated sites are within 15 degrees latitude of the equator. The correlation falls to r²=0.50 when comparisons are made between sondes and

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satellite columns for the 4 subtropical SHADOZ stations. FT ozone at those locations are seasonally\_dependent mixtures of tropical and extra-tropical air masses, with latitudes (Table 4) spanning Hanoi (+21.0) to Irene (-25.9S).

We have also analyzed trends of tropospheric ozone column and free tropospheric ozone at individual SHADOZ stations using a QR model, following column definitions and guidelines for the TOAR II/HEGIFTOM (Harmonization and Evaluation of Ground-based Instruments for Free Tropospheric Ozone Measurements) project analysis (Chang et al., 2023). The tropospheric ozone column in HEGIFTOM trends analysis is defined as surface to 300 hPa; the FT is defined as a layer between 300 and 700 hPa and the results are given as ppbv O<sub>3</sub>/decade change and %/decade. The QR trends for 13 SHADOZ sites from 2000 to 2022 are summarized in **Table 4**; a subset of them appear in an evaluation of ground-based global ozone trends in HEGIFTOM-1.

# 2.1.2 SHADOZ and IAGOS-SHADOZ blended profiles. LMS and FT ozone,

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The MLR trend analyses (results in **Table 2 and 3**) use SHADOZ profile measurements in several ways. First, the trends are computed using monthly-averaged ozone mixing ratios at 100-m intervals from the surface to 20 km, as described in T21. Second, most results are illustrated as ozone column amounts (in,DU) for two FT segments, 5-10 km and 10-15 km, and for the LMS, Trends for ozone and P-T-U,data below 5 km are determined for completeness but are not tabulated because station sampling times and local pollution can vary, giving artifact biases among the individual sites (Thompson et al., 2014). We use 15-20 km for the LMS for two reasons. This is where several studies identified wave activity associated with convection and ENSO-La Niña oscillations (Lee at al., 2010; Thompson et al., 2011; Randel and Thompson, 2011; T21). Second Randel et al. (2007) identified a distinct ozone annual cycle driven by the Brewer-Dobson circulation.

A third way of using SHADOZ profiles in the MLR analysis is in a blend with IAGOS aircraft profile measurements within a lower FT pressure-defined region ("FTp" = 300-700 hPa, HEGIFTOM-1).

Calculations in the FTp segment are designed to add more samples within the SHADOZ-labeled combination sites (compare profile numbers in Tables 2 and 3) and to augment regional trends in Van Malderen et al. (2024b) where no results are reported for the equatorial Americas, Atlantic Ocean or African continent. In defining regions for merging SHADOZ and IAGOS observations, we follow locations presented by Tsivlidou et al. (2023). Profiles from the SHADOZ Quito station (2014-2023) and two IAGOS airports (Bogotá and Caracas) are added to the SHADOZ SC-Para profiles to define the equatorial Americas for determining trends within the FTp (Table 3). Also, for the SHADOZ-IAGOS calculations, sonde profiles from the Natal-Ascension pair are combined with 13 airports in west Africa (Table 3) to determine trends for a region designated "Atlantic+West Africa," as shown in Fig. 1b (color-coded circles) and the second column of Table 3. Nairobi is combined with IAGOS Addis Ababa profiles. The

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FTp-designated Equatorial SE Asia consists of KL-Java profiles from SHADOZ combined with IAGOS

landing and takeoff <u>data</u> from Kuala Lumpur and Singapore. Time-series of ozone column amounts (in DU and as anomalies) for SHADOZ stations and airports for these 4 "regional" sites appear in **Fig. 3**. The coordinates of individual SHADOZ stations used in the blended dataset (italicized) with IAGOS airports appear in **Table 1**. Calculations with FTp retain Samoa as a single station.

### 2.1.3 OMI/MLS satellite and sonde total ozone columns

Trends computed with MLR for <u>sonde-derived</u> total tropospheric ozone columns (TrCO) are based on integrating ozone mixing ratios from the surface to the thermal lapse-rate tropopause derived from the radiosondes that accompany each ozonesonde launch. The standard WMO definition of tropopause is used. For the <u>5</u> equatorial sites in our analyses, the tropopause is typically between 16 and 17 km. Our TrCO<sub>sonde</sub> columns and trends are compared to TrCO<sub>satellite</sub>, the troposphere ozone columns estimated from the OMI/MLS residual as described by Ziemke et al. (2019; updated in the TOAR II <u>paper by</u> Gaudel et al., 2024). These newest OMI/MLR TrCO estimates have been corrected for a ~1%/decade upward drift in OMI over the past two decades (Gaudel et al., 2024; SI material). The OMI/MLS product is available starting in October 2004. We use monthly average TrCO for both sondes and OMI/MLS between January 2005 and December 2023. These are identical to the data used in the Gaudel et al. (2024) TOAR II analyses of tropical ozone.

### 2.2 Trend analyses

# 2.2.1 Multiple Linear Regression (MLR) model

As in T21 and S24, the Goddard MLR model (original version Stolarski et al., 1991, updated in Ziemke *et al.*, 2019) is used for analysis of monthly mean ozone amounts. The MLR model includes terms for annual and semi-annual cycles and oscillations prevalent in the tropics: QBO, MEI (Multivariate ENSO Index, v2) and IOD DMI (Indian Ocean Dipole Moment Index; only for KL-Java):

 $O_3(t) = A(t) + B(t) + C(t)MEI(t) + D(t)QBO1(t) + E(t)QBO2(t) + F(t)IOD(t) + \varepsilon(t)$ 

where t is month. The coefficients are as follows: A through F include a constant and periodic components with 12, 6, 4, and 3 month cycles, where A represents the mean monthly seasonal cycle and B represents the month-dependent linear trend. When annual trends are reported, the B term includes only the 12-month component to generate a single trend value over the period of computation. The model includes data from the MEIv2 (https://www.esrl.noaa.gov/psd/enso/mei/), the two leading QBO EOFs from Singapore monthly mean zonal radiosonde winds at 10, 15, 20, 30, 40, 50, and 70 hPa levels, and IOD DMI (https://psl.noaa.gov/gcos\_wgsp/Timeseries/Data/dmi.had.long.data). The  $\epsilon(t)$  is the residual, i.e., the difference between the best-fit model and the raw data. T21 noted that the monthly ozone data and MLR model fits for the mid FT (5-10 km) and LMS layers are well-correlated. For the LMS, for example, the correlation coefficients are r = 0.83-0.90 (Fig. S7 in T21). The IOD DMI term is included for KL-Java, the only station where the IOD impact on the ozone trend is reliably detected.

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The 95% confidence intervals and p-values for each term in the MLR model as presented here are determined using a moving-block bootstrap technique (10,000 resamples) in order to account for autocorrelation in the ozone time series (Wilks, 1997). The model is applied to ozone anomalies in all cases in order to minimize biases that might arise from intersite ozone differences between pairs for the combined stations: SC-Para, Nat-Asc, KL-Java (Table 2). In other words, we calculate ozone anomalies from the individual station's monthly climatology for all profiles before combining the pairs into monthly means and computing the MLR ozone trends. Anomalies are also analyzed for the Nairobi and Samoa station data, although this would be no different than computing MLR trends on the actual ozone timeseries themselves. The MLR model was separately applied to the monthly mean ozone profile anomalies at 100 m resolution, and the monthly mean partial column ozone anomaly amounts from 5-10 km, 10-15 km, and 15-20 km. The MLR model was also applied to the monthly mean tropopause height (TH) anomaly at each station, defined as the 380 K potential temperature surface (e.g., Wargan et al., 2018). Because TH and LMS ozone trends turn out to be strongly correlated (T21), the MLR analysis was also performed for the ozone column amount anomalies referenced to the tropopause. In that case LMS ozone trends refer to changes in the 5 km above the tropopause with the FT extending from the tropopause to 5km below the tropopause (Section 3, Table 2). Finally, the MLR model was applied to total tropospheric column amounts from the sondes ( $TrCO_{sonde}$ ) and corresponding  $TrCO_{satellite}$  from OMI/MLS (surface to Tp in Table 2).

Note that recent ozone trends studies and the TOAR II guidelines (Chang et al., 2020; Cooper et al., 2020; Chang et al., 2023) have discouraged the use of nomenclature associated with statistical significance, whereas the Figures and Tables presented here refer to trends using terminology of 95% confidence intervals (equivalent to p-value < 0.05), the most reliable results in **Section 3** (bold in **Tables 2, 3** and 4) are explicitly stated as based on p-values < 0.05,

Several studies of tropospheric ozone observations have noted a persistence of COVID-19 perturbations on post-2019 trends after 2019 (Ziemke et al., 2022; <a href="HEGIFTOM-1">HEGIFTOM-1</a>; Van Malderen et al., 2024b). A comparison of the extended SHADOZ mean ozone trends (1998-2023) relative to those from T21 (covering 1998 to 2019), both summarized in **Table 2**, represents the impact of COVID-19 in the deep tropics. Likewise, SHADOZ was initiated at the end of the powerful 1997-1998 ENSO. Accordingly, we applied MLR to the same 5 sites for 2000-2023 to evaluate any artifacts relative to the 1998 to 2023 trends. Those results also appear in **Table 2**.

# 2.2 Quantile Regression (QR) model

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Whereas MLR has been the standard tool for analyzing global total and stratospheric ozone trends, the latter often, with satellite data where zonal means can be used, the TOAR II project has recommended using QR, as better suited for trends in the troposphere where, for example, urban concentrations can vary by factors of 3-4, Because it is a percentile-based method (Koenker, 2005), the heterogeneously

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distributed changes of trends can be estimated, as shown, for example, in Gaudel et al. (2020). To date 580 581 the TOAR II HEGIFTOM trends studies for observations at individual sites (HEGIFTOM-1) and regionally Deleted: Van Malderen et al., 2024a 582 organized data (HEGIFTOM-2) have been studied with the QR approach. In those studies and for the 13 Deleted: Van Malderen et al., 2024b individual SHADOZ time-series (Table 4) QR has been applied to the median change of the trends, 583 Deleted: / S1 Formatted: Not Highlight which is equivalent to the least absolute deviation estimator (i.e. aiming to minimize mean absolute 584 deviation for residuals; Chang et al., 2021). The rationale is that compared to least-squares criterion, a 585 586 median-based approach is more robust when extreme values or outliers are present. Median trends are 587 estimated based on the following multivariate linear model: Observations[t] =  $a0 + a1*sin(Month*2\pi/12) + a2*cos(Month*2\pi/12)$ 588 589  $+ a3*sin(Month*2\pi/6) + a4*cos(Month*2\pi/6) + b*t + c*ENSO[t] + N[t],$ Eq. 1 590 where harmonic functions are used to represent the seasonality, a0 is the intercept, b is the trend value, c is the regression coefficient for ENSO, and N[t] represents the residuals. Autocorrelation is accounted 591 592 for by using the moving block bootstrap algorithm, and the implementation details are provided in the TOAR statistical guidelines (Chang et al., 2023). In the individual site analyses of HEGIFTOM 593 594 observations (HEGIFTOM-1), where all individual ozone records (L1) and monthly means (L3) have Deleted: Van Malderen et al., 2024a both been analyzed, annually averaged trends usually turned out to be the same within uncertainties. 595 2.2.3 Trend Sensitivity Studies 596 Three sensitivity studies were conducted, related to (1) sampling frequency; (2) the complementarity 597 Moved (insertion) [2] 598 of MLR and OR methods; (3) duration of time-series. Deleted: , sample numbers and Deleted: were conducted. 599 A number of TOAR-related studies (Chang et al., 2020; Chang et al., 2021; Chang et al., 2023) have 600 emphasized links between ozone time-series sampling characteristics, i.e., frequency of profile 601 measurements and/or temporal gaps, and trend uncertainty. Gaudel et al. (2024), for example, show 602 uncertainty (as 2-sigma) in median tropospheric ozone profiles; the inference is that 6-15 monthly 603 samples are required for meaningful FT trends (Fig. 1-2 in Supplemental Material, Gaudel et al., 2004). 604 For the first sensitivity test we examined trends dependence on sample size by comparing the annual 605 trends computed with MLR for the lower-mid-FT ozone segment (5-10 km) to the trends from the 606 combined SHADOZ-IAGOS merged monthly mean SHADOZ profiles (L3, 700-300 hPa) for 1998-2023. A 607 comparison of Tables 2 and 3 indicate that for the equatorial Americas and Atlantic regions, the sample 608 numbers are increased more than a factor of 2; the other two sites have enhancements of 1.3 and 1.5. 609 The results in DU/decade and %/decade, appear in Table 3. Moved up [2]: Three sensitivity studies related to 610 In the second sensitivity study, three calculations were made using the data from all individual methods, sample numbers and duration of time-series 611 SHADOZ stations as in HEGIFTOM-1, not only the 5-site equatorial profiles (Table 2), The individual were conducted. For the first Deleted: For the first 612 station results appear in Table 4. The trend (1998-2023) in mean tropospheric column (mixing ratio, **Deleted:** investigation 613 averaged from surface to the tropopause for each sample) was computed. The QR method was applied **Deleted:**, as in HEGIFTOM-1 to all profiles at each station (L1, level 1 data) and to the same set of profiles but monthly averaged for 614 Deleted:

each site (L3, level 3 data). The L1 sample numbers ranged from 326 (Watukosek) to 1142 (Hilo); the L1/L3 sample size ratios ranged from 2.2 to 4.3 with only 3 of 13 stations having a ratio below 3.2. The second calculation compared QR and MLR trends applied to the same L3 data set. The third calculation was a computation of trends at each station for the FT column amount used in HEGIFTOM-1 (700-300 hPa. comparable to our 5-10 km segment), HEGIFTOM-1 pointed out the complementarity of applying both MLR and QR to the 23-yr time-series of mean ozone column amounts. The advantage of MLR is a graphical display of monthly trends that indicate important ways ozone interacts with seasonally varying dynamics, as in S24 or Millet et al. (2025). QR distinguishes trends among different segments of the distribution, an advantage for stations where tropospheric ozone segments are highly variable. In this calculation the augmented FT ozone data (?L1) were used for the four regions: equatorial Americas: Atlantic+West Africa; East Africa; equatorial southeast Asia. With QR the median trends (50 %-ile) are computed as well as trends for the 5%-ile, 25%-ile, 75%-ile, 95%-ile, v. The third sensitivity test investigates the degree to which trends and uncertainties depend on the length of sampling. This issue arises because the most frequently used satellite estimates of tropospheric ozone begin after 2003 compared to SHADOZ (1998-) and IAGOS (1994-). The Boynard et al. (2025) study uses IASI products for 2008-2023, comparing only 7 of ~40 potential ozonesonde stations for evaluation. We used our HEGIFTOM-1 results: tropospheric columns computed for surface to 300 hPa based on 9 SHADOZ stations, with L1 data using OR for this exercise. The latter trends are for 2000-2022 and the trends are nearly the same as the individual trends in Table 4. To eliminate COVID-19 impacts, we selected a 12-year period for comparison, calculating trends for the same columns over 2008-2019. The results appear in Table 5.

3 Results and Discussion

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### 3.1 Monthly and seasonal ozone climatology at 5 SHADOZ sites

Figure 4 displays the 5-site monthly ozone climatology based on SHADOZ monthly averaged data from the surface to 20 km. Regional differences in vertical structure within the FT are pronounced. For example, the contours representing the 60-90 ppbv range (yellow to red colors) never appear in mid-FT ozone over KL-Java or Samoa (Figs. 4d,e). Conversely, FT ozone values  $\leq$  30 ppbv (darkest blue shades) observed over KL-Java and Samoa in the middle FT never appear over the other 3 stations: equatorial Americas (SC-Para, Fig. 4a), Nat-Asc or Nairobi (Figs. 4b,c). These contrasts may reflect regional differences in ascending vs. descending nodes of the Walker circulation. The latter feature is partly responsible for the tropospheric zonal wave-one (Thompson et al., 2003) that refers to a mean TrCO over the south tropical Atlantic Ocean that is sometimes twice as large as over the western Pacific. There is less regional variability in LMS ozone. At all stations (Fig. 4) above  $\sim$ 16 km the colors and contours are nearly uniform over the year. Mixing ratio contours of 100 ppbv and 200 ppbv may appear as a thick

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white line. The 100 ppbv level is sometimes referred to an ozonopause; typically it is within 1-2 km of the thermal lapse-rate tropopause.

### 3.2 FT and LMS ozone annual cycle (1998-2023)

The annual cycle of ozone at the two FT layers and for LMS ozone appear as anomalies in Fig. 5. FT ozone seasonality (Figs. 5a,b) is less uniform than for LMS ozone (Fig. 5c) and tropopause height (TH, Fig. 5d). Randel et al. (2007) showed that the near-uniform LMS ozone seasonality in the equatorial zone is due to the Brewer-Dobson circulation. The more varied FT ozone cycles in Figs. 5a,b are due to a range of different dynamical and chemical influences across the stations. As expected, the annual cycles for the pressure- and regionally defined FTp ozone (Fig. 6 in %) resemble those for the corresponding SHADOZ sites in the lower (5-10 km) FT layer in Fig. 5a; the magnitudes are similar as well although Figs. 5 and 6 are illustrated with different scales. In both cases it is seen that there are two seasonal maxima and minima for KL-Java (Fig. 5a) and equatorial SE Asia (Fig. 6a). The early year minima are associated with intense convective activity (T21, S24) that repeats in August at the onset of the Asian monsoon. KL and Watukosek are also affected by seasonal fire activity at the latter end of the rainy seasons. These features were described in detail in Stauffer et al. (2018) using Self-Organizing Map clusters and proxies for convection and fires.

3.3 FT ozone trends: regional and seasonal variability

# 3.3.1 Trends for 1998-2023

In **Fig.** 7 the trends in %/decade computed with MLR at 100-m intervals, for 1998 to 2023, are displayed (update of Fig. 6 in T21 for the 1998-2019 trends). Changes in the ozone column amounts for 1998-2023 computed from the model (DU/decade and %/decade) for the two FT layers (5-10 km, 10-15 km) appear in **Fig.** 8. A summary of values for the two layers (and for LMS ozone) appears in **Table 2**. The percentage values in **Fig.** 7 and **Table 2** are the result of dividing the MLR B(t) term by the A(t) annual cycle of ozone term (Section 2.2.1). The MLR-calculated A(t) annual cycle derived from monthly mean ozone profiles (i.e., no anomaly calculation) is used to convert the B(t) trend in ppmv/decade (profiles) or DU/decade (partial columns) to %/decade. Ozone trends for both percent/decade and DU/decade are given in **Table 2**. Shades of red (blue) in **Fig.** 7 represent ozone increases (decreases); cyan hatching denotes trends with p-values < 0.05. The annual mean trends in **Table 2** are computed by taking the average of the 12 monthly trends in DU and dividing by the mean seasonal ozone in DU to yield the annual percentage trend.

For 3 of 5 stations in **Figs. 8a** and **c**, there is a pattern of ozone increase at both FT layers in January to April. Percentage-wise the greatest increases are at KL-Java and Nairobi,  $\sim (10-15)\%/\text{decade}$  in March and April. However, SC-Para and Samoa at 5-10 km (**Fig. 8a**) exhibit almost no trend at any time of year; at 10-15 km SC-Para and Nairobi show losses up to 10%/decade in February and (5-10)%/decade

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**Deleted:** The corresponding column trends changes in the FTp layer appear in **Fig. 9** with tabulated trends in **Table 3.** Note that for both **Figs. 8** and **9**, the annual trend for each station at the right of the figure is essentially zero.? *Say more* 

losses in August and September. However, Table 2 displays no trend on an annual basis for SC-Para and Nairobi. Inspection of Fig. 7 suggests small FT trends at Nat-Asc; Table 2 displays a +3.4%/decade increase in the 10-15 km layer from 1998-2023. The total column, integrated to the tropopause, TrCO, over Nat-Asc, has increased (1.9±1.8)%/decade, p<0.05. There are no other annually averaged trends in the FT layers but TrCO for KL-Java (KL-Watukosek in **Table 2**), also increased, (2.6±2.3)%/decade.

### \_\_3.3.2 FT ozone trends sensitivity to COVID-19 and 1997-1998 ENSO

A comparison of the **Table 2** columns for 1998-2023 relative to those for 1998-2019 (the latter is from T21) reveals little. Only the 10-15 km layer at Nat-Asc has entries with p<0.05 for both periods. The extra 4 years reduced the positive trend slightly. This is consistent with studies that found lingering COVID-related declines in sondes and satellites (Ziemke et al., 2022; HEGIFTOM-1). In Table 2 columns for trends for 2000-2023 can be compared to those for 1998-2023. There is little information in the 2000-2023 column, i.e., no trends anywhere except for the TrCO for KL-Java, an area that was wellstudied with satellite and some sonde measurements for the period affected by the large ENSO, amplified by the Indian Ocean Dipole pattern (Thompson et al., 2001). After August 1997, as a result of exceptionally high fire activity, ozone increased greatly. That could have meant a smaller change between ozone levels from 1998 through 2023 which would be consistent with a larger, more robust trend for 2000-2023 (4.6%/decade for KL-Java) compared to T21, 2.6%/decade (both p<0.05). cf to QR

more than twice the number of profiles as those in the other lower FT layer, Fig. 8a. The trends are similar to those in the 5-10 km layer in both magnitude and confidence level (uncertainty). Table 3 shows no trends (p<0.05) at any location. This was unexpected given that Chang et al. (2023) suggest that the uncertainty should decline with more samples and a positive trend might be amplified. For example, adding west African IAGOS data to Natal and Ascension increased the number of profiles by a factor of > 2.5 (compare Tables 2 and 3). Further analysis is needed, including with the QR approach. ??? to sensitivity section below T

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# 3.4 LMS ozone trends and mean vertical trend over 5 SHADOZ sites\_

TrOC and HEGIF KL trend

In T21 (Figs. 10 and 11) trends in the LMS (nominally 15-20 km) showed 5-10%/decade decreases for Nat-Asc, KL-Java and SC-Para between July and October. For the same months those locations exhibited a tropopause increase ~100 m/decade, suggesting that the seasonal ozone increase is an artifact of a changing tropopause. In other words, if the TH increased more air with relatively lower ozone would be located in the 15-20 km layer. We tested this hypothesis by recomputing ozone column changes referenced to the TH for 1998-2019, i.e., evaluating trends in a 5-km thick layer above the TH. The result was that the apparent loss of LMS ozone from July to September or October disappeared. The same analyses performed with LMS ozone and TH for the 1998-2023 period (Fig. 2) are the same as for 1998-2019 (T21). Whatever the cause(s) of ozone loss in the LMS, it is a feature clearly captured by SHADOZ data as seen

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760 in annually averaged ozone trends derived from the analyses displayed in Fig. 10. At 18 km the

composite trend from the 8 SHADOZ stations analyzed with MLR is (-4+3)%/decade. The mean trend

from  $\sim$ 13 to 3 km is zero, albeit with a  $\pm 2\sigma$  (95%)  $\pm 3$ %/decade. Only below  $\sim$ 2km is the mean ozone

trend clearly positive. Most of that increase originates from near-surface pollution over equatorial SE

764 Asia (Fig. 6 in S24).

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### 3.5 Sensitivity tests. Trends method. FT sample numbers. Length of time-series

### 3.5.1 Complementarity of Trends Methods

In Fig. 11 the median trends from 1998 to 2023 (50%-ile) for the FT ozone segments for the four regions with IAGOS-enhanced SHADOZ stations are displayed with the lowest and highest (5%, 95%, respectively), 25%-ile and 75%-ile quantiles. Trends of anomalies in DU (red denotes p< 0.10) are shown. The medians are statistically nearly the same, although as expected, the 2- $\sigma$  uncertainty bars are smaller with the QR method than with MLR. The MLR trends are higher in all cases except the equatorial Americas (Fig. 11a). In the latter case, the positive anomalies have increased significantly for the 5% and 25% quantiles over the 26-yr period with no change for the 50%, 75% or 95% quantiles. This signifies that the background (lowest-ozone) air has increasing ozone but the highest-ozone distribution has not changed. Over the Atlantic+West Africa (Fig. 11b) there are also small increases in the lowest part of the distribution but the median and higher %iles show no change. East Africa (Fig. 11c) shows marked increases in the lowest-ozone quantiles and the median but a significant decrease in the highest-ozone (more polluted) air. The opposite is true over equatorial southeast Asia (Fig. 11d). The most polluted air is increasing but the background (lowest ozone) FT segment has decreased

### 3.5.2 FT sample numbers

Figure 12 monthly ozone trends are based on a total of 1.8 times the number of profiles as those in the other lower FT layer, Fig. 8a and 8b. For the equatorial Americas (blue) twice as many profiles contribute to the trends in Fig. 12 than in Figs. 8a and 8b; for the Atlantic+West Africa includes 2.5 times more profiles than in Figs. 8a and 8b. In all four regions, the seasonality of the trends is nearly the same between the 5-10 km FT segment (Figs. 8a and 8b) and the corresponding SHADOZ+ IAGOS trend (Fig. 12). Furthermore, month by month, the trends are similar to those in the 5-10 km layer in both magnitude and confidence level (uncertainty). Table 3 shows no trends (p<0.05) at any location. The null trends are illustrated in the annual means at the right of each image in Figs. 8 and 12. This was unexpected given the Chang et al. (2023) and Gaudel et al. (2024) suggestions that the uncertainty should decline with more samples and positive trends might be amplified.

### 3.5.3 Length of time-series

**Table 5** displays a comparison of TrOC trends determined by QR for the HEGIFTOM-1 23-yr period, 2000-2022 for 10 individual SHADOZ stations, and for the same time-series only between 2008 and 2019. The uncertainties (expressed as  $+2-\sigma$ ) increased by factors 2-3 or more for 8 of 9 stations. These results are excerpted from a Comment on Boynard et al. (2025) that also compared the time-series changes for 17 mid-latitude ozonesonde stations. Similar uncertainty increases were noted for all 27 stations. Of that total, 9 stations exhibited ozone trend sign changes with the 12-year time-series

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although only 10 trends were statistically significant. Because so many of the HEGIFTOM-1 sonde trends are marginal, our results reinforce the need for multi-decade time-series.

# 3.6 Total tropospheric ozone trends, TrCO (1998-2023), from OMI/MLS and SHADOZ

Trends for the most recent version of OMI/MLS TrCO are based on monthly mean satellite data and determined with MLR over the period 2005 through 2023. Trends for total tropospheric column ozone (TrCO<sub>sonde</sub>) at the 5 SHADOZ sites for the same period appear in circles on the map in Fig. 13 where the stippling indicates no trend can be determined. For both OMI/MLS and the sondes (Fig. 7) shades of red indicate total column ozone increases; blue represents declining ozone over the period of analysis. The mean annual TrCO<sub>sonde</sub> trends appear in the two rightmost columns in **Table 2**. In **Fig. 13**.0MI/MLS shows trends > 1DU/decade (typically 2-9%/decade) only over equatorial SE Asia and parts of South America and the eastern Pacific at  $\sim$ 5N latitude. Circles indicate locations and trends for the individual SHADOZ stations. The latter display lower trends than OMI/MLS. On a month by month basis the sonde and OMI/MLS trends are compared in Fig. 14. In 3 cases the seasonality of TrCO trends from sonde and OMI/MLS are similar and the annually averaged OMI/MLS TrCO<sub>satellite</sub> trends are not different from zero (symbols at right of each image). However, the seasonality of the KL-Java monthly trends agree well with OMI/MLS; the satellite mean is +5%/decade, gray in Fig. 14d. The sonde SC-Para trend (Fig. 14a) is guite a bit lower early in the year than the OMI/MLS trends over San Cristóbal and Paramaribo that average +(2-3)%/decade. The Samoa sonde trend and OMI/MLS TrCO trends diverge most of the year; the satellite mean annual trend is close to +10%/decade whereas there is no trend for TrCO<sub>sonde</sub>.

# **4 Summary and Conclusions**

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We have presented a two-part evaluation of tropical ozone trends using a 26-yr record of ozonesonde (SHADOZ, 1998-2023) profiles with selected FT aircraft (IAGOS) ozone data and the most recent OMI/MLS estimates of tropospheric column ozone for 2005-2023. The next section summarizes the findings. It is followed by Section 4.2 which compares our trends to related TOAR II studies. Section 4.3 concludes with a consensus view of FT ozone trends and perspectives relevant to the overall TOAR-II climate and tropical assessments.

### 4.1 Summary of findings

The first part of the study updates trends in FT and LMS ozone for 5 stations, Nairobi, Samoa and three combination sites (San Cristóbal-Paramaribo, Natal-Ascension, Kuala Lumpur-Watukosek) extending the T21 trends, that covered 1998-2019, by 4 years. The new analysis added monthly averaged data from 2000 to 2023 to the Goddard MLR model with standard proxies. Trends in FT (5-10km, 10-15 km) and LMS (15-20 km) layers are illustrated with monthly means and annually averaged changes in DU/decade and %/decade. Trends determined for the period 2000-2023 assessed impacts of

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We have presented a two-part assessment of tropical ozone trends using a 26-yr record of ozonesonde (SHADOZ, 1998-2023) profiles with selected FT aircraft (IAGOS) ozone data and the most recent OMI/MLS estimates of tropospheric column ozone for 2005-2023. ¶ The first part of the study updates trends in FT and LMS ozone for 5 stations, Nairobi, Samoa and three combination sites (San Cristóbal-Paramaribo, Natal-Ascension, Kuala Lumpur-Watukosek) extending the T21 trends, that covered 1998-2019, by 4 years. The new analysis added monthly averaged data from 2000 to 2023 to the Goddard MLR model with standard proxies. Trends in FT (5-10km, 10-15 km) and LMS (15-20 km) layers are illustrated with monthly means and annually averaged changes in DU/decade and%/decade. Trends determined for the period 2000-2023 assessed impacts of the strong 1997-1998 ENSO-La Niña on a possible anomalous starting point, Comparisons of trends for the monthly averaged Aura-derived OMI/MLS total tropospheric column ozone product (TrCO<sub>satellite</sub>) were made to those from monthly sonde-derived TrCO<sub>sonde</sub> for 8 equatorial SHADOZ stations. The principal results of the SHADOZ trend updates and comparisons are as follows: ¶ The overall characteristics of T21 trends in the FT and LMS are confirmed with 4 additional years of SHADOZ observations. From 1998 to 2023, regional and seasonal variability remains pronounced with FT ozone increases at 4 of 5 SHADOZ stations in thin layers ~(10-25)%/decade, mostly between January and May. The exception is at SC-Para there was a 5-10% decrease in between 10-15 km during 1998 to 2023 compared to 1998-2019 (T21). The greatest increases occur in multiple layers below 10 km over Nairobi and KL-Java and between 10-15 km over Samoa but these features do not translate into annually averaged trends (p<0.05) in the 5-10 km or 10-15 km segments except over Nat-Asc. Thus, adding 4 years of data to equatorial SHADOZ trends does not modify the T21 picture of little or no FT ozone change. Only when the total tropospheric column (TrCO<sub>sonde</sub>) trend is evaluated do Nat-Asc (1.9+1.8)%/decade and KL-Java (2.6+2.3)%/decade) exhibit the slightest trend (p<0.05). Examining the 5 station average in vertical form shows a null trend from ~3 to 17 km ( $\pm$ 2% within 2 $\sigma$  up to 7 km and ~ $\pm$ 3% from 7 to 17 km). The marginal overall mean increase +5%/decade below 3 km, is driven by KL-Java, possibly with a contribution from Nairobi. ¶ With the starting year delayed to 2000, the TrCO<sub>sonde</sub> KI the 1997-1998 ENSO on a possible anomalous starting point. Comparisons of trends for the monthly averaged Aura-derived OMI/MLS total tropospheric column ozone product (TrCO<sub>satellite</sub>) were made to those from monthly sonde-derived TrCO<sub>sonde</sub> for 8 equatorial SHADOZ stations. The principal results of the SHADOZ trend updates and comparisons are as follows:

- The overall characteristics of T21 trends in the FT and LMS are confirmed with 4 additional years of SHADOZ observations. From 1998 to 2023, regional and seasonal variability remains pronounced with FT ozone increasing in thin layers at 4 of 5 SHADOZ stations ~(5-20)%/decade, mostly between January and May. The exception is at SC-Para where there was a 5-10% ozone decrease between 10-15 km during 1998 to 2023 compared 5-10%/decade increases in 1998-2019 (T21). For 1998-2023, the greatest ozone increases occur in multiple layers below 10 km over Nairobi and KL-Java and between 10-15 km over Samoa. However, these features do not translate into annually averaged trends (p<0.05) in the 5-10 km or 10-15 km segments except over Nat-Asc, i.e., adding 4 years of data to equatorial SHADOZ data does not modify the T21 picture of little or no FT ozone change. Only when the total tropospheric column (TrCO<sub>sonde</sub>) trend is evaluated do Nat-Asc (1.9±1.8)%/decade and KL-Java (2.6±2.3)%/decade) exhibit the slightest trend (p<0.05). Examining the 5-station average in vertical form shows a null trend from ~3 to 17 km (0±2% within 2σ up to 7 km and ~0±3% from 7 to 17 km). The marginal overall mean increase, +5%/decade below 3 km, is primarily driven by KL-Java changes.
- With the starting year delayed to 2000, the TrCO<sub>sonde</sub> KL-Java trend (2000-2023) is almost twice as large as for 1998-2023, indicating an effect of the 1997-1998 ENSO on equatorial SE Asia. This is not surprising. Watukosek soundings (1997-1998) show ENSO-induced anomalously high ozone over Indonesia that was also captured by satellite tropospheric ozone estimates from TOMS (Thompson et al., 2001).
- The T21 LMS ozone and TH trends are also confirmed with 4 more years of data. For the layer 15-20 km, ozone losses ~5%/decade from June through October, on average, give an all-site average of -3%/decade at 17.5 km, a value similar to satellite averages (Godin-Beekmann et al., 2022). As in T21, re-determining the LMS trends for an ozone column 5 km above the tropopause from 1998 to 2023, causes the trend to disappear.
- Annually-averaged trends, 2005-2023, determined with MLR for OMI/MLS columns, TrCO<sub>satellite</sub>, over the 8 individual equatorial SHADOZ stations (members of the 5 combined sites) and TrCO<sub>sonde</sub> overlap within the uncertainties of each. Trends are close to zero at Nat-Asc, Nairobi and KL-Java. The OMI/MLS TrCO<sub>satellite</sub> trends are marginally positive at SC-Para, with monthly cycles diverging in the early part of the year at SC-Para. OMI/MLS trends do not capture large monthly seasonal variations seen in SHADOZ profiles, especially for negative trends. The large

positive trend from OMI/MLS over Samoa does not align with determinations of FT ozone from this or other studies (see below).

The second part of the investigation was motivated by statistical issues raised in related TOAR II trends analyses. The results of these analyses are summarized:

- Trends methods. The relative merits of trends computed with QR and MLR, previously demonstrated in HEGIFTOM-1, were reinforced with analysis of combined FT SHADOZ-IAGOS data. Although median trends are the same, QR uncertainties are smaller. MLR is superior for capturing seasonal influences but QR provides vital information on whether the background, low-ozone or high-ozone (polluted) populations are changing the most.
- Sampling frequency. The sensitivity of the 1998 to 2023 FT ozone trends to sample number was explored by using IAGOS profiles to increase the SHADOZ sample size for the equatorial stations by 80% overall, including a doubling over the equatorial Americas and Atlantic regions, then applying MLR. Median trends were nearly unchanged. No FT trends over the 4 regions (plus Samoa) are detected with p<0.05 although uncertainties, expressed in ppbv/decade, improved 30% over the equatorial Americas and by ~15% over 3 of the 4 other sites. These results indicate that current SHADOZ sampling with the 3 combined site records is sufficient in this radiatively-important region.

Length of trends. These were examined for the individual station  $TrCO_{sonde}$  trends by comparing trends for 1998 to 2023 with a 12-year trend (2008-2019), one of two scenarios investigated by Boynard et al. (2025). The uncertainties (as 2- $\sigma$  limits) increase by a factor of 2-3 for TrCO at 10 of 13 SHADOZ stations and some median trends change sign compared to the 26-year trends (**Table 4**). The next section shows that even 16-year IASI/Metop trends have an unreasonably low bias with respect to SHADOZ, IAGOS and OMI/MLS trends (**Table 6**), reinforcing a need for multidecade datasets where ozone changes are relatively small.

# 4.2 Comparison of this study to related TOAR-II investigations

How do our tropical tropospheric ozone trends compare to those in other studies that use SHADOZ and IAGOS profiles and/or satellite data? **Table 6** summarizes our results for the FT, UT segments and for TrCO. The FT ozone comparisons are made with Gaudel et al. (2024) and with results for two HEGIFTOM studies (HEGIFTOM-1; HEGIFTOM-2). For UT ozone, SHADOZ trends are compared to those derived from the lowest 3 layers of MLS (Froidevaux et al., 2025; see their Table 2). TrCO trends are taken from the 5 SHADOZ stations, OMI/MLS (this study) and IASI/Metop (Boynard et al., 2025). Note that the latter study only spans 2008-2023, much less than the SHADOZ data but close to the OMI/MLS period, 2005-2023.

The tropical trends study of Gaudel et al. (2024) groups SHADOZ and IAGOS profiles somewhat differently from this study and only extends through 2019 (period of trends are shown in the  $4^{th}$  column of **Table 6**). Our trends and those of Gaudel et al. (2024) for FT ozone and TrCO (**Table 6**) for the equatorial Americas are similar although some are insignificant. The equatorial Americas FT ozone trends range from (-0.01%/decade to -3.3%)/decade, For TrCO<sub>sonde</sub>, derived from profiles, the range is (-1.0 to -3.1)%/decade in between the satellite-based trends. They are (3.1±2.5)%/decade for OMI/MLS (2005-2023) and (-4.0±1.0)%/decade for IASI/Metop (2008-2023), an offset of 7.1%/decade for the median trends. Within given uncertainties, IAGOS, IAGOS-SHADOZ-combined and IASI/Metop agree: their trends all slightly negative.

For the Atlantic and western Africa regions, the profile-based comparisons differ in station-airport combinations among our study, Gaudel et al. (2024) and the two HEGIFTOM analyses. The FT and TrCO trends among the 4 studies fall in a relatively small range: (-1.3±2.8)%/decade (FT, Gaudel et al, 2024) to (1.9±1.8)%/decade (TrCO, this study). The larger TrCO from the Natal-Ascension combination appears to result from a higher positive trend in the UT (3.4±2.9)%/decade. Because the FT ozone trend of Gaudel et al. (2024) is negative and the Nat-Asc FT ozone trend is positive (Table 6), combining western African IAGOS profiles with the Natal and Ascension measurements, reduces the larger area trend compared to the sonde-only FT ozone trend. Table 6 shows that for all regions the MLS-derived UT ozone trend estimates fall between +3%/decade and +4%/decade (Froidevaux et al. 2025). Only over the Atlantic region do any of the SHADOZ UT ozone trends fall in this range. Overall, the Atlantic and western Africa FT ozone and TrCO<sub>sonde</sub> trends are nearly zero, in agreement with OMI/MLS (1.3±1.4)%/decade. As for the equatorial Americas, the 2008-2023 trends from IASI/Metop over the Atlantic and western Africa is much lower: (-4.9±2.0)%/decade. Compare to the OMI/MLS trend, the IASI/Metop median trend has a 6.2%/decade low bias. The picture for east Africa (both SHADOZ and IAGOS profiles over Nairobi) is similar to the Atlantic and western Africa. FT ozone, TrCO<sub>sonde</sub> and OMI/MLS TrCO trends are essentially null but IASI/Metop displays a (-3.6±1.0)%/decade trend.

There is more variability in trends among the ground-based studies for the equatorial SE Asia region than the equatorial Americas, Atlantic and Africa, most likely because different combinations of IAGOS profiles and SHADOZ data were used. Supplementing FT KL-Java SHADOZ profiles with IAGOS data (Tables 3 and 6) in our study, a 50% increase in sample size, did not change the trend appreciably: (1.0±2.6)%/decade vs (1.6±3.2)%/decade. The corresponding TrCO<sub>sonde</sub> over KL-Java increased (2.6±2.3)%/decade. S24 computed trends with MLR for KL-Java for 1998-2022: the TrCO<sub>sonde</sub> was (3.4±2.6)%/decade. Although the trend period only differs by one year, the smaller trend with the extra year in this study (to 2023) might reflect some COVID impact (columns 5 and 7 in Table 2). For OMI/MLS TrCO, determined from a mean of changes averaged over 5°x5° grid boxes for KL and Watukosek (Java), there was a (5.6±6.0)%/decade, 2005-2023. The latter change is nearly the same as

Gaudel et al. (2024) for both FT ozone and OMI/MLS changes over the same interval. There are several reasons for why Gaudel et al. (2024) FT ozone trends in **Table 6** are larger than our SHADOZ-IAGOS FT ozone trends. First, the fusing of SHADOZ and IAGOS profiles in Gaudel et al. (2024) may be more heavily weighted to polluted IAGOS segments than our merging. Second, reprocessed IAGOS profiles have not been rigorously compared to SHADOZ data up to this point; a new evaluation of IAGOS instrumentation in the World Calibration Center for Ozonesondes may facilitate consistent referencing to an absolute ozone standard in the future (Smit et al., 2024; Smit et al., 2025). Third, with the shorter trend period, especially ending in 2019, the data of Gaudel et al. (2024) would not be affected by lower, COVID-pertrubed ozone concentrations in 2020-2023 as some SHADOZ records were (**Table 2**).

The HEGIFTOM-1 FT ozone trend (2000-2022) for Kuala Lumpur is nearly identical to the KL-Java trend, 1998-2023: (2.2±3.0)%/decade. In HEGIFTOM-2 (based on SHADOZ KL and IAGOS from several SE Asia airports) FT ozone increases over a longer period (1995-2022) are (6.2±2.2)%/decade. Thus, in general, over SE Asia, as for the equatorial Americas, Atlantic and Africa, the GB and OMI/MLS trends are in reasonable agreement, given some differences in data selection and minor differences in the trend start and end dates: FT and TrCO ozone increases ~+(2-8)%/decade. Likewise, the IASI/Metop trend for TrCO, (0.0±1.4)%/decade over 2008-2023 (Boynard et al., 2025) is an outlier over SE Asia.

Nowhere are the satellite data as divergent from the FT sonde trends, near zero change from our study (1998-2023), Gaudel et al. (2024, for 2004-2019) and HEGIFTOM-1 (2000-2022), than at Samoa (Table 6). TrCO from the SHADOZ sondes has no significant change: (-1.4 ±4.8)%/decade, this study); (-3.1±5.4)%/decade (Gaudel et al. 2024). However, the OMI/MLS trend for TrCO is (9.1±8.3)%/decade, 2005-2023, and IASI/Metop is (-9.0±1.7)%/decade. The large disagreement in trends from sondes and both satellite instruments, the latter with median TrCO trends that are offset by 18% from one another, underscore the need for profile-based tropospheric ozone trends as an unbiased reference.

### 4.3 Implications of this study for TOAR II and related assessments

How do our findings apply to an overall TOAR II assessment for tropical ozone? First, there is consensus among the four GB studies: this study, Gaudel et al (2024) and the two HEGIFTOM articles (HEGIFTOM-1; HEGIFTOM-2). These results provide well-characterized trends in FT ozone for input to climate models and a reference for evaluating TrCO, total tropospheric column ozone trends from evolving tropospheric ozone satellite products. In short, over the past 20-25 years, except for equatorial SE Asia, tropical FT and total ozone trends are negligible to within  $\pm$ (1-2)%/decade. For SE Asia both FT and TrCO have increased from  $\sim$ (2-8)%/decade. Detailed analyses of sonde profiles by S24 suggest that the annually averaged FT ozone trends are (2-3)%/decade. The (5-7)%/decade increases are seasonal

in FT ozone. However, for TrCO it is the large ozone increases in the boundary layer found throughout the year that are dominating the total tropospheric column change.

Second, as far as methods of computing trends, we have shown both the similarity of median trends from OR and MLR as relative advantages of each with SHADOZ and combined SHADOZ-IAGOS profile data. HEGIFTOM-1 reached similar conclusions about OR and MLR trends for 34 ozonesonde stations, including 10 from SHADOZ. The complementarity of QR and MLR for trends attribution was also highlighted in HEGIFTOM-1. The HEGIFTOM papers (HEGIFTOM-1, HEGIFTOM-2), with a multiinstrument perspective, show variable trends among 55 global stations, including over western Europe and North America where both positive and negative tropospheric column trends occur within few hundred km. Overall, except for southeast Asia, FT ozone trends from HEGIFTOM are small to moderate and not distinguishable from zero at a number of tropical and extratropical locations. Third, both HEGIFTOM-1 and this study investigated the matter of sample density in ground-based data, the former by cutting sample size roughly in half for TrCO and our SHADOZ-IAGOS analyses for FT ozone, roughly doubling it. In both studies trend changes determined with MLR were small (a few tenths of a ppbv/decade) and included both increases and decreases (well-illustrated in Fig. S6 in HEGIFTOM-1). Uncertainty changes ranged from 15-30%, usually improving with more sampling. We conclude that arguments for excluding ground-based trends from TOAR II on the basis of sample size have little merit, particularly given ongoing uncertainties and limited record lengths of many satellite products. Fourth, by reducing the trend period from 26 years to 12 years, we quantified the degradation of too-short trends for the relatively small changes that apply to much of global FT ozone.

The length of trends is only one issue for ozone products derived from satellites that have operated less than two decades. Trends based on the lowermost levels of ozone from MLS (Froidevaux et al., 2025) were largely the same at all SHADOZ stations, (3-4%/decade for the UT) whereas the sondederived trends are geographically variable. Only one of 5 tropical regions we analyzed, Atlantic and western Africa, displayed a UT ozone trend in this range. The discrepancies between GB-based trends is not surprising because MLS does not capture observed variability in UT ozone concentrations, up to a factor of 2 between the Atlantic and western Indian Ocean, that is detected by the sondes or uv sensors (Thompson et al., 2003; Thompson et al., 2017). We compared our profile-based trends to two typical satellite products: IASI/Metop and the well-characterized OMI/MLS. With the uncertainty of the satellite trends, OMI/MLS TrCO trends agreed with TrCO<sub>sonde</sub> trends in 4 of 5 regions. Where sonde-derived trends were near-zero, IASI/Metop trends sometimes agreed well but for equatorial SE Asia the IASI/Metop zero trend clearly underestimated trends relative to those derived from SHADOZ, SHADOZ-IAGOS combined profiles and OMI/MLS. At Samoa IASI/Metop (-9%/decade) and OMI/MLS (+9%/decade) were both in error, displaying much greater disagreement with the sonde-based trend

1157 than anywhere else. There is no easy explanation, but the differing wavelengths, vertical sensitivities, 158 and respective biases in the satellite products likely all play a role. 159 The S24 TOAR-II study is a reminder that the near-zero annually averaged FT ozone trends over most of the tropics (Figs. 8 and 12) in the tropics may mask strong seasonal trends (T21). S24 linked the 160 161 strong February-April increase in FT ozone over KL-Java (1998 to 2022) to declining convection with 4 162 proxies, e.g., outgoing long-wave radiation, velocity potential at 200 hPa. A role for changing dynamics 163 must be considered in tropical tropospheric ozone increases; increasing ozone precursor emissions are 164 apparently not the only driver. In a similar manner, decreases observed in LMS ozone appear to be related to tropopause height changes during the 1998 to 2023 period. 165 In summary, our updated results for SHADOZ stations remain the most reliable reference for trends 166 167 throughout the tropical troposphere and LMS. We provide a definitive standard for evaluating monthly trends and regional variability of satellite-based products and related models being used for tropical 168 ozone trends. Together with the more extensive HEGIFTOM and IAGOS coverage, our SHADOZ analyses 169 170 lead to a conclusion that the most reliable trends information for TOAR II is based on re-processed GB 171 measurements. However, coverage of the ground-based instruments is uneven and space-based 172 observations are needed for truly global trends. In order for satellite products to mature and for 173 differences among them to be understood, independent ozone data collection, particularly from ozonesondes referenced to an absolute standard are required. As our investigations with the high-174 175 resolution SHADOZ profiles have demonstrated, sonde data will remain the gold standard for deriving trends in the FT and LMS, the two most critical regions where ozone interacts with the climat system. 176 177

**Data Availability** 

180 All datasets used in this study are openly and publicly accessible. 181 V06 SHADOZ data are available at https://doi.org/10.57721/SHADOZ-V06 (NASA Goddard Space Flight Center 182 (GSFC) SHADOZ Team, 2019). 183

IAGOS and SHADOZ HEGIFTOM data are available at: https://hegiftom.meteo.be 184

Trends in table (\*.csv) format are available at: https://tropo.gsfc.nasa.gov/shadoz/SHADOZ PubsList OMI/MLS data are available at https://acd-ext.gsfc.nasa.gov/Data services/cloud slice/new data.html.

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Competing Interests. All authors declare that we have no competing interests.

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