



- 1 Consistency evaluation of tropospheric ozone from ozonesonde and
- 2 IAGOS aircraft observations: vertical distribution, ozonesonde types
- 3 and station-airport distance
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15 **Abstract:** The vertical distribution of tropospheric O₃ from ozonesondes is compared with that from

16 In-service Aircraft for a Global Observing System (IAGOS) measurements at 23 pairs of sites

17 between about 30°S and 55°N, from 1995 to 2021. Profiles of tropospheric O<sub>3</sub> from IAGOS aircraft

are in generally good agreement with ozonesonde observations, for Electrochemical concentration

cells (ECC), Brewer-Mast, and Carbon-Iodine sensors, with average biases of 7.03 ppb, 6.28 ppb,

and -4.48 ppb, and correlation coefficients (R) of 0.72, 0.86, and 0.82, respectively. Agreement

between the aircraft and Indian-sonde observations is poor, with an average bias of 24.07 ppb and

R of 0.41. The  $O_3$  concentration observed by ECC sondes is on average higher by 5-10% than that

23 observed by IAGOS aircraft, and the relative bias increases modestly with altitude. For other sonde

24 types, there are some seasonal and altitude variations in the relative bias with respect to IAGOS

- 25 measurements, but these appear to be caused by local differences.
- 26 The distance between station and airport within 4° has little effect on the comparison results. For
- 27 the ECC ozonesonde, the overall bias with respect to IAGOS measurements varies from 5.7 to 9.8
- 28 ppb, when the station pairs are grouped by station-airport distances of <1° (latitude and longitude),
- 29  $1-2^{\circ}$ , and  $2-4^{\circ}$ . Correlations for these groups are R=0.8, 0.9 and 0.7. These comparison results





30 provide important information for merging ozonesonde and IAGOS measurement datasets. They 31 can also be used to evaluate the relative biases of the different sonde types in the troposphere, using 32 the aircraft as a transfer standard. Key words: WOUDC; IAGOS; tropospheric O3; vertical distribution; ozonesonde; aircraft 33 34 1 Introduction 35 36 Ozone (O<sub>3</sub>) is a trace gas with small concentrations in the atmosphere (Ramanathan et al., 1985); it 37 is an important greenhouse gas in the atmosphere. In the planetary boundary layer, it is a major air 38 pollutant (Lefohn et al., 2018; Monks et al., 2015). It can endanger human health, damage 39 ecosystems, and affect climate change (Fu and Tai, 2015; Lefohn et al., 2018; Percy et al., 2003). 40 Therefore, it is of importance to study the temporal and spatial distribution and the factors and 41 mechanisms affecting the variation of tropospheric O<sub>3</sub> including near-surface O<sub>3</sub> (Logan, 1985; Ma 42 et al., 2020; Sharma et al., 2017; Young et al., 2018). 43 A large number of studies have been carried out on the spatiotemporal distribution, formation mechanism, and transport characteristics of ground O<sub>3</sub> (Li et al., 2020, 2021; Vingarzan, 2004; Wang 44 45 et al., 2017, 2023; Xu et al., 2021; Yu et al., 2021). However, due to the limitation of observations, 46 there are many unknowns on tropospheric ozone, especially the vertical distribution of tropospheric 47 O<sub>3</sub>. Satellites provide an effective platform for measuring O<sub>3</sub> globally. Satellite O<sub>3</sub> instruments, 48 including TES, GOME, GOME-2, SCIAMACHY, OMI, and TROPOMI, have been in operation for 49 decades (David et al., 2013; Ebojie et al., 2016; Hegarty et al., 2009; Hoogen et al., 1999; Hubert et 50 al., 2021; Miles et al., 2015). Although satellite observations can provide detailed temporally- and 51 horizontally-resolved maps of tropospheric O3 columns, in general satellite data lack vertical 52 resolution. While tropospheric differential absorption lidar can also provide vertical distribution 53 information for tropospheric O<sub>3</sub> (Keckhut et al., 2004; Yang et al., 2023), there are very few routinely 54 operating stations. The principal sources of vertically-resolved, trend-quality observations of tropospheric O<sub>3</sub> are 55 56 therefore balloon-borne ozonesondes, and IAGOS aircraft observations. The World Ozone and Ultraviolet Radiation Data Centre (WOUDC) and the In-service Aircraft for a Global Observing 57

System database (IAGOS) house the data from these two observation programs with the longest





59 duration and the most global stations, which are the most widely used for tropospheric O<sub>3</sub> studies 60 (Gaudel et al., 2020; Liao et al., 2021; Tarasick et al., 2019; Wang et al., 2022). These two datasets are used to study the distribution, variability and trends of tropospheric O3, and its sources and 61 62 transport, as well as satellite and model validation (Hu et al., 2017; Gaudel et al., 2018; 2020; Wang 63 et al., 2022; Zhang et al., 2008). The first phase of the Tropospheric Ozone Assessment Report 64 (TOAR-I), initiated in 2014, utilized available surface, ozonesonde, aircraft, and satellite observations to assess tropospheric O<sub>3</sub> trends from 1970 to 2014 (Schultz et al., 2017). Hu et al. 65 (2017) found that the largest bias in a chemical transport model, GEOS-Chem, with respect to 66 67 ozonesondes and IAGOS observations, is in high northern latitudes in winter-spring, where the simulated ozone is 10-20 ppb lower. Wang et al. (2022) examined observed tropospheric O<sub>3</sub> trends, 68 69 their attributions, and radiative impacts from 1995 to 2017, using aircraft observations from IAGOS, 70 ozonesondes, and a multi-decadal GEOS-Chem chemical model simulation, and found increases in 71 tropospheric ozone (950 - 250 hPa) of  $2.7 \pm 1.7$  ppbv per decade from IAGOS observations in the 72 Northern Hemisphere and at 19 of 27 global ozonesonde sites averaging  $1.9 \pm 1.7$  ppbv per decade. 73 There are also a number of comparative studies on these two datasets (Zbinden et al., 2013; Staufer 74 et al., 2013, 2014; Tanimoto et al., 2015; Tarasick et al., 2019). Staufer et al. (2013, 2014) used 75 trajectory calculations to match air parcels sampled by both sondes and aircraft. Zbinden et al. (2013) 76 compared coincidences (±24 hours) at three site pairs, while Tanimoto et al. (2015) examined 77 simultaneous observations (±3 hours for sonde versus aircraft) at several site pairs less than 100 km 78 apart. In general, these studies show small (6% or less) negative biases of aircraft measurements 79 against ECC sondes. Tarasick et al. (2019) compared trajectory-mapped averages over 20-70 N of 80 ozonesonde and MOZAIC/IAGOS profiles and concluded that over 1994-2012 ozonesonde 81 measurements were about  $5 \pm 1\%$  higher in the lower troposphere and  $8 \pm 1\%$  higher in the upper 82 troposphere. 83 In this study, we attempt to make the most comprehensive evaluation to date of the relative biases 84 of IAGOS and sonde profiles, using as many station pairs as possible. We identify 23 suitable pairs 85 of sites in the WOUDC and IAGOS datasets from 1995 to 2021, compare the average vertical 86 distribution of tropospheric O<sub>3</sub> shown by ozonesonde and aircraft measurements, and analyze their 87 differences by ozonesonde type and by station-airport distance.





2 Data and methods

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# 2.1 MOZAIC-IAGOS observations

90 91 The MOZAIC (Measurements of OZone and water vapor on Airbus In-service airCraft) program, 92 initiated in 1994 and incorporated into the IAGOS (In-service Aircraft for a Global Observing 93 System; www.iagos.org) program since 2011, takes advantage of commercial aircraft to provide 94 worldwide in-situ measurements of several trace gases (e.g., O<sub>3</sub> and CO) and meteorological 95 variables (e.g., water vapor) throughout the troposphere and the lower stratosphere (Marenco et al., 96 1998; Petzold et al., 2015; Nédélec et al., 2015). O<sub>3</sub> measurements are performed using a dual-beam 97 UV-absorption monitor (time resolution of 4 seconds) with an instrumental uncertainty of ±2 ppbv+2% (Thouret et al., 1998; Blot et al., 2021). It should be noted that this is only the instrumental 98 99 uncertainty, and does not include sampling uncertainties (possible losses) caused by the inlet line and the compressor before the UV-photometric measurements are made. Loss of ozone on the inlet 100 101 pump was an issue in earlier aircraft ozone sampling programs (Brunner et al., 2001; Dias-Lalcaca et al., 1998; Schnadt Poberaj et al., 2007), but Thouret et al. (1998) found it negligible for 102 103 MOZAIC/IAGOS. 104 More details on the new IAGOS instrumentation can be found in Nédélec et al. (2015). The 105 continuity of the dataset between the MOZAIC and IAGOS programs has been demonstrated based 106 on their 2-year overlap (2011~2012) (Nédélec et al., 2015). Blot et al. (2021) performed an 107 evaluation of the internal consistency of the O<sub>3</sub> and CO measurements since 1994, which confirmed 108 the instrumental uncertainty of ±2 ppbv. Moreover they found no drift in the bias amongst the 109 different instrument units (six O<sub>3</sub> and six CO IAGOS-MOZAIC instruments, nine IAGOS-Core 110 Package1 and the two instruments used in the IAGOS-CARIBIC aircraft). 2.2 WOUDC ozonesonde observations 111 The World Ozone and Ultraviolet Radiation Data Centre (WOUDC) is part of the Global 112 113 Atmosphere Watch (GAW) program of the World Meteorological Organization

(https://woudc.org/data/explore.php). The WOUDC is operated by Environment and Climate

Change Canada. WOUDC ozonesonde data have been evaluated in a number of WMO-sponsored

international field intercomparisons (Attmannspacher and Dütsch, 1970, 1981; Kerr et al, 1994) and





(Smit et al., 2007, 2024; Thompson et al., 2019). In the global ozonesonde network, while different 118 ozonesonde types were common in the past, more than 95% of current sounding stations use 119 120 electrochemical concentration cells (ECC). ECC ozonesondes have a precision of 3-5% (1-σ) while 121 the precision of other sonde types is somewhat poorer, at about 5-10% for Brewer-Mast and the 122 Japanese KC (Carbon-Iodine) sonde, and somewhat larger for the Indian-sonde (Kerr et al., 1994; 123 Smit et al., 2007). Biases with respect to UV reference spectrometers have been estimated for ECC 124 sondes at 1-5% in the troposphere (Smit et al., 2021; Tarasick et al., 2019, 2021). 125 2.3 Data processing 126 The two datasets were first screened for airport-sonde station pairs within a latitude separation of 127 <4° and a longitude separation of <4°. Many sonde stations have observational records that do not 128 overlap with the IAGOS period (1994-present). In addition, the IAGOS dataset has large gaps at 129 many airports, because the frequency of visits to airports by aircraft that take part in IAGOS depends 130 on commercial airlines' operating constraints. In total, 23 station pairs (Fig. 1) were identified with 131 a separation of less than 4° in both latitude and longitude, and coincident observations over at least 132 nine months. The majority of the 23 ozonesonde site records are ECC (17), while four are Indian-133 sonde, one Brewer-Mast, and one Carbon-Iodine (the Japanese KC sonde). These stations were 134 divided into 3 groups according to the distance (D) between the ozonesonde station and the airport: 135 D<1°, 1°<D<2°, and 2°<D<4°. Specific information on the comparison stations is shown in Table 136 1. 137 The observation times of the ozonesonde and aircraft are generally not the same. Ozonesondes are 138 typically launched once a week, although a few stations have more frequent launches. The aircraft 139 records generally contain more frequent observations, but observation times vary. For the selected 140 23 stations, we calculated the mean ozone vertical profiles at 1km resolution (the first layer is from 141 the surface to 1 km above sea level) for each month during the observational period for the two 142 datasets. A minimum of four aircraft profiles were required to estimate a monthly mean profile; because ozonesonde launches are typically only a few times per month, no minimum was required 143 144 to estimate a monthly mean profile. Only data with monthly means in both datasets were included 145 for further analysis. Comparisons between the two datasets were made by ozonesonde type and by

more recently in laboratory simulation chamber experiments using a standard reference photometer





station-airport distance.

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#### 3. Results and discussion

### 3.1 Comparison of the vertical profiles of tropospheric O<sub>3</sub> from four types of ozonesondes and

#### aircraft observations

Previous intercomparisons of sondes launched on the same balloon (Attmannspacher and Dütsch, 1970, 1981; Beekmann et al, 1994, 1995; Deshler et al., 2008; Hilsenrath et al., 1986; Kerr et al, 1994; Smit et al., 2007) have shown that sondes of different types respond somewhat differently to the same ozone profile; that is, they have relative biases, that vary with altitude. Fig. 2 therefore compares the mean vertical profiles of tropospheric O<sub>3</sub> from ozonesonde and aircraft measurements, separated by ozonesonde type. Both O<sub>3</sub> concentrations and absolute differences between ozonesonde and aircraft increase with altitude, especially above 9 km. Average tropospheric O<sub>3</sub> profiles observed by ECC, Brewer-Mast, and Carbon-Iodine sondes are in good agreement with aircraft measurements, with biases of 7.03 ppb, 6.28 ppb and -4.48 ppb, respectively, and correlation coefficients (R) of 0.72, 0.86 and 0.82, respectively (Fig. 3a-3c). The Indian-sonde average shows a linear increase with altitude, while the aircraft measurements indicate an ozone decrease with altitude above 8 km (Fig. 2b). This behaviour is most clearly related to the comparisons of stations 2°-4° apart in spring (Fig. S8). The agreement between the Indian-sonde and aircraft observations is poor, with a bias of 24.07 ppb, and R of only 0.41 (Fig. 3d). The RMSE of O<sub>3</sub> observed with the four types of ozonesondes (ECC, Brewer-Mast, Carbon-Iodine and Indian-sonde) and the aircraft is small, at 49.15 ppb, 42.08 ppb, 33.55 ppb and 45.90 ppb, respectively. Fig. 2 shows that the mean differences between ozonesonde and aircraft measurements vary significantly with altitude. This can also be observed clearly from the relative differences (RD), expressed as (O<sub>3-ozonesonde</sub> - O<sub>3-aircraft</sub>)/ O<sub>3-aircraft</sub> × 100% (Fig. 4). O<sub>3</sub> concentrations from ECC measurements are higher than those from aircraft measurements in all altitudes except at the surface. Mean O<sub>3</sub> concentrations reported by Brewer-Mast sondes are lower than those from IAGOS below 7 km, but higher between 7 and 12 km. O<sub>3</sub> concentrations reported by Carbon-Iodine sondes are higher than those observed from aircrafts below 2 km, but significantly lower above 8 km. In relative

terms, the bias between ECC sonde and aircraft measurements has only a very modest variation





175 with altitude, except near the ground. The mean relative bias for Brewer-Mast measurements is at 176 an absolute maximum of -19 % near the ground, but increases slowly above 3 km, and is positive above 7 km, reaching more than +10 % at 10~11 km. The relative bias for Carbon-Iodine 177 178 measurements is about 8% below 2 km, is quite small from 2 - 8 km, and becomes large and negative 179 above ~8 km. 180 The Indian-sonde observations show much larger mean differences from the aircraft measurements. 181 Biases are everywhere positive, and as high as nearly 60% or 30 ppb, with much higher uncertainty 182 (standard errors) at each altitude as well (Fig. 2b, Fig. 4). 183 These results are broadly consistent with those from JOSIE 1996 (Smit et al. 1996; Smit and Kley, 1998; Thompson et al., 2019), and with the northern hemisphere average result from Tarasick et al. 184 (2019). (Their Figure 20b; note that it is largely based on ECC sondes, and the scale is inverted 185 186 (IAGOS-sondes) from the sense we use here.) 187 It should be noted that these comparisons only give an average relative bias between sondes and 188 IAGOS. The true value of the ozone profile remains unknown, as do the absolute biases of sondes 189 and IAGOS. 190 191 3.2 Seasonal dependence of relative biases 192 Fig. 5 compares mean profiles observed by ECC ozonesondes and IAGOS, separated by season. 193 There are modest seasonal differences in the relative bias profiles, with somewhat larger average 194 biases in winter and spring, but average biases are all positive (ECC sondes higher) and at all levels 195 the average seasonal biases are not statistically different at the 95% confidence level. 196 The modest seasonal differences that are apparent in Fig. 5 and in Figs. S1-S3 are likely due to the 197 modest sample size (for ECC sondes) and small sample sizes (for other types). The actual 198 coincidence in time for profiles can range from less than one day to about 1-3 weeks, depending on the number of ozonesonde and aircraft O<sub>3</sub> profiles collected within each month-bin. This means the 199 200 larger the atmospheric variability of O3 is, the larger the real differences between ozonesonde and 201 aircraft O<sub>3</sub> can become, particularly when the number of profiles within a month-bin are small. In 202 addition, there are errors due to variations in the aircraft take-off and landing trajectories and the 203 balloon rise rate, the geographical location of the observation stations (and any associated





204 meteorological differences) and any systematic difference in standard observational times. Table 2 indicates that in all four seasons ECC data correlate well with aircraft observations, with R 205 206 ranging from 0.71 to 0.76, but with larger average biases in winter and spring, as noted. It is not 207 clear if these seasonal average differences in bias are significant, as the uncertainty ranges on the 208 seasonal averages (lower plot of Fig. 5) overlap. 209 The vertical distribution of tropospheric O<sub>3</sub> observed by Brewer-Mast and IAGOS aircraft in the 210 four seasons is similar (Fig. S1). Differences are also similar, except above 7 km, where the 211 uncertainties are larger, and in general the uncertainty ranges on the seasonal difference averages 212 overlap. Since these comparisons come from only one station pair, some of the differences may be attributable to local differences in topography and meteorology. Table 2 shows that correlations for 213 214 the ensemble of Brewer-Mast stations are higher than those for ECC stations. Like the ECC sondes, 215 average biases are all positive, but this is determined by the biases above 7 km (Fig. 4); unlike the 216 ECCs, biases are negative in the lowest 3 km. 217 The vertical distribution of tropospheric O<sub>3</sub> concentrations observed by Carbon-Iodine sondes and 218 IAGOS aircraft in the four seasons are similar, except in summer when the tropopause is high (Fig. 219 S2). The difference plots are fairly similar, except in the lowest 3 km, where differences become 220 quite large in summer. Like the previous comparison for Brewer-Mast sondes, these comparisons 221 come from only one station pair, and so the large differences in the boundary layer in summer are 222 likely due to local ozone production sampled by the sonde but not the aircraft. Likely for this reason, 223 the consistency between Carbon-Iodine and aircraft observations is poor in summer, with R being 224 only 0.46 (Table 2). For the other three seasons it is fairly good. 225 The tropospheric O3 observed by Indian-sondes displays a consistently high bias relative to IAGOS 226 in all seasons, and the seasonal difference plots are quite similar, except in the lowest 3 km in winter 227 (Fig. S3). This different behavior in winter is likely due to local ozone production sampled by the 228 aircraft but not the sonde. Temperature inversions are common in the winter in northern India and trap local pollution. The very low values registered by the aircraft near the surface in summer also 229 230 suggest local effects, in this case titration by NOx. 231 Table 2 indicates poor consistency between Indian-sonde and aircraft observations in all four 232 seasons, with R in winter only 0.18.





233 3.3 Dependence of relative biases on station-airport distances 234 A major concern with comparing IAGOS and ozonesonde observations is that the stations and 235 airports are not generally co-located, and even where they are close, the flight paths taken by balloon 236 and aircraft are quite different. Fig. 6 compares the average vertical distribution of tropospheric O<sub>3</sub> 237 observed at different station-airport distances by ECC sondes and IAGOS aircraft. Note that we 238 continue to separate sonde station data by type --- only ECC data are used here. Sonde-aircraft pairs 239 have been grouped by station-airport distance (Table 1). The differences in average bias vary only 240 very modestly between the different station-airport distance categories, and those differences are 241 not statistically different at the 95% confidence level (Fig. 6d). This, partially owing presumably to the use of mean monthly averages, is encouraging, as this provides further evidence that the average 242 bias we have derived is an artifact strictly of instrument differences. 243 244 Table 3 indicates that the bias variation between ECC and aircraft observations at different station-245 airport distances is small, ranging from 5.7 ppb to 9.8 ppb. Correlations for these groupings are also 246 fairly similar, at R = 0.8, 0.9 and 0.7. 247 Compared with ECC sondes, the consistency between the Indian-sonde and aircraft observations is 248 poor at all station-airport distances, with much larger biases, and poor correlations, with R = 0.2 to 249 0.4. Nevertheless, Fig. S4 shows that the profiles of average differences are quite similar for station-250 airport distances  $< 1^{\circ}$ , and distances of  $2^{\circ} \sim 4^{\circ}$  (Fig. S4c). 251 Fig. 7 and Figs. S5-S7 examine possible seasonal variation in the differences at different station-252 airport distances, for ECC sondes. The mean differences for the different station-airport distance 253 categories are larger than for the annual averages (Fig. 6), but in general those differences are not statistically different at the 95% confidence level (Figs. 7d and S5d-S7d). 254 255 256 3.4 Comparison of ozonesonde relative biases under operational conditions using IAGOS 257 observations as a transfer standard The foregoing discussion demonstrates that, consistent with previous work, there is a fairly constant 258 259 relative bias between IAGOS and sondes, with considerable dependence on sonde type, as expected 260 from previous sonde intercomparisons like JOSIE 1996. Although uncertainties are sizeable due to 261 the relatively sparse nature of the available data, we find consistent differences at all sites, with little





262 dependence on season or on station-airport separation, and little regional dependence (not shown). 263 Notwithstanding this overall sonde-IAGOS bias, we can use these station-airport comparisons to 264 derive relative biases of the different sonde types in use in the global network. 265 This does not assume that the aircraft data are unbiased. The true value of the ozone profile (or even 266 its average) remains unknown, as do the absolute biases of sondes and IAGOS. It does assume: 267 1. That the measurement errors are random and normally distributed; 268 2. That there is one, constant bias for each measurement type (that is, if, for example, the Indian 269 sonde has changed over the period of comparison, or the IAGOS instruments have different biases, 270 there would be additional error that is not included in our uncertainty estimate); 271 3. That the measurement biases are not dependent on the geographic location or other variability of 272 the ozone profile. This does not assume that the average ozone profile is the same, just that the 273 instruments respond in the same way. 274 With these assumptions we can use the results of Fig. 2 to estimate the relative biases of each 275 sonde type to each other. The uncertainty of the comparisons will be the quadratic sum of the 276 uncertainties of the two IAGOS-sonde comparisons. The results are shown in Table 4. This 277 intercomparison of the different sonde types has an important advantage: it compares ozonesonde 278 relative biases under operational conditions, as it compares the data that are actually in databases 279 like the WOUDC. It also fills a gap, as the last WMO international intercomparison involving all four sonde types was JOSIE 1996. These results are broadly consistent with those from JOSIE 1996 280 281 (Smit and Kley, 1998; their Table 8 and Fig. 11). 282 In fact, the types of ozonesonde have changed during long-term observations at some stations (e.g. 283 Uccle and Payerne). De Backer et al. (1998) showed that with the use of an appropriate correction 284 procedure, accounting for the loss of pump efficiency with decreasing pressure and temperature, it 285 is possible to reduce the mean difference between ozone profiles obtained with both types of sondes 286 below 3%, which is statistically insignificant over nearly the whole operational altitude range (from 287 the ground to 32 km). Stübi et al. (2008) also found that the O<sub>3</sub> difference between the Brewer-Mast 288 and the ECC ozonesonde data shows good agreement between the two sonde types, and the profile 289 of the O<sub>3</sub> difference is limited to ±5% (±0.3 mPa) from the ground to 32 km. The results for Brewer-290 Mast sondes in Table 4 should also be applicable to the older Payerne and Uccle records, and are





292 2002; 2016). 293 The results in Table 4 will be quite valuable for addressing the problem of relative biases when 294 merging ozonesonde data into global climatologies (e.g. McPeters et al., 2007; McPeters and Labow, 295 2012; Bodeker et al., 2013; Liu et al., 2013; Hassler et al., 2018;). 296 4 Conclusions 297 The vertical distribution of tropospheric O3 observed by ozonesondes and IAGOS aircraft sensors are compared at 23 pairs of sites between ~30°S and 55°N from 1995 to 2021. Overall, ECC, 298 299 Brewer-Mast, and Carbon-Iodine sondes agree reasonably well with aircraft observations, with 300 average biases of 7.03 ppb, 6.28 ppb, and -4.48 ppb, and correlation coefficients of 0.72, 0.86, and 301 0.82, respectively. The agreement between the aircraft and Indian-sonde observations is poor, with 302 an average bias of 24.07 ppb and R of 0.41. 303 Notwithstanding this general agreement, all sonde types show significant average biases with 304 respect to IAGOS. The O<sub>3</sub> concentration observed by ECC sondes is on average higher by 5-10% 305 than that observed by IAGOS aircraft, and the relative bias increases modestly with altitude. 306 Seasonal variations in the relative bias are not in general statistically significant at the 95% 307 confidence level. The distance between station and airport within 4° also has little effect on the 308 comparison results. When the ECC station pairs are grouped by station-airport distances of <1° 309 (latitude and longitude), 1-2°, and 2-4°, biases with respect to IAGOS measurements vary from 5.7 310 to 9.8 ppb, and correlations from 0.7 to 0.9. 311 Thus, the observed average relative bias between sondes and IAGOS found in this study, also noted 312 by previous authors (Zbinden et al., 2013; Staufer et al., 2013, 2014; Tanimoto et al., 2015; Tarasick 313 et al., 2019), is a robust result. Possible reasons for the difference include: side reactions that cause 314 sondes to produce excess iodine (Saltzman and Gilbert, 1959), and/or loss of ozone on the inlet pump that could cause IAGOS monitors to read low at pressures below 800 hPa. The latter was an 315 316 issue in earlier aircraft ozone sampling programs (Schnadt Poberaj et al., 2007; Dias-Lalcaca et al., 317 1998; Brunner et al., 2001), but Thouret et al. (1998) found it negligible for MOZAIC/IAGOS. 318 This result implies that care must be taken when merging ozonesonde and IAGOS measurement 319 datasets. While the aircraft and sonde measurements are often complementary, filling in important

generally consistent with these results and with those for the older Canadian records (Tarasick et al.,





320 spatial gaps that would otherwise exist if only one type were used, the records are not typically over 321 the same period, and so merging can introduce spurious jumps if relative biases are not taken into 322 account. 323 The importance of ozone in the troposphere as an air pollutant and a greenhouse gas, and therefore 324 of accurate measurements of its temporal and spatial distribution implies that it will be important to 325 resolve the causes of this bias, and so further research involving more direct comparisons of IAGOS 326 instrumentation and ozonesondes, e.g. in the WCCOS chamber, are strongly recommended. 327 These results are also useful to evaluate the relative biases of the different sonde types in the 328 troposphere, using the aircraft as a transfer standard. This intercomparison of the different sonde 329 types has the advantage that it compares ozonesonde relative biases under operational conditions; that is, the data that are actually in databases like the WOUDC. These results will be invaluable for 330 331 addressing relative biases when merging ozonesonde data into global climatologies (e.g. Bodeker et al., 2013; Hassler et al., 2018; Liu et al., 2013; McPeters et al., 2007; McPeters and Labow, 2012). 332 333 334 Competing interests. The contact authors have declared that none of the authors has any competing 335 interests. 336 Data availability. The global ozone sounding data were acquired from the World Ozone and 337 Ultraviolet Radiation Data Center (http://www.woudc.org) operated by Environment Canada. The 338 IAGOS data are created with support from the European Commission, national agencies in Germany 339 (BMBF), France (MESR), and the UK (NERC), and the IAGOS member institutions 340 (http://www.iagos.org/partners). 341 Author contributions. HW: Data curation, Methodology, Validation, Visualization, Writing -342 original draft preparation, Writing - review & editing, Funding acquisition. LS: Methodology, 343 Investigation, Writing - original draft. DWT: Data curation, Resources, Conceptualization, Supervision, Writing - original draft preparation, Writing - review & editing. JL: Data curation, 344 345 Resources, Methodology, Conceptualization, Supervision, Writing - original draft preparation, 346 Writing - review & editing, Funding acquisition. TZ: Funding acquisition, Writing - review & 347 editing. HGJS and RVM: Writing - review & editing.





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378	electrochemical sondes, a ground based lidar and an airborne UV-photometer. Atmospheric
379	Environment, 29(9), 1027-1042.
380	Bernhard, G.H., Bais, A.F., Aucamp, P.J., Klekociuk, A.R., Liley, J.B., McKenzie, R.L., 2023.
381	Stratospheric ozone, UV radiation, and climate interactions. Photochemical & Photobiological
382	Sciences, 1-53.
383	Blot, R., Nedelec, P., Boulanger, D., Wolff, P., Sauvage, B., Cousin, JM., Athier, G., Zahn, A.,
384	Obersteiner, F., Scharffe, D., Petetin, H., Bennouna, Y., Clark, H., and Thouret, V., 2021.
385	Internal consistency of the IAGOS ozone and carbon monoxide measurements for the last 25
386	years, Atmos. Meas. Tech., 14, 3935-3951, https://doi.org/10.5194/amt-14-3935-2021.
387	Bodeker, G. E., Hassler, B., Young, P. J., and Portmann, R. W., 2013. A vertically resolved, global,
388	gap-free ozone database for assessing or constraining global climate model simulations, Earth
389	Syst. Sci. Data, 5, 31-43, https://doi.org/10.5194/essd-5-31-2013.
390	Brunner, D., J. Staehelin, D. Jeker, H. Wernli, and U. Schumann, 2001. Nitrogen oxides and ozone
391	in the tropopause region of the Northern Hemisphere: Measurements from commercial aircraft
392	in 1995/96 and 1997, J. Geophys. Res., 106, 27673-27699.
393	Callis, L.B., Boughner, R.E., Natarajan, M., Lambeth, J.D., Baker, D.N. and Blake, J.B., 1991.
394	Ozone depletion in the high latitude lower stratosphere: 1979-1990. Journal of Geophysical
395	Research: Atmospheres, 96(D2), 2921-2937.
396	$David, L.M.\ and\ Nair, P.R., 2013.\ Tropospheric\ column\ O_3\ and\ NO_2\ over\ the\ Indian\ region\ observed$
397	by Ozone Monitoring Instrument (OMI): Seasonal changes and long-term trends. Atmospheric
398	Environment, 65, 25-39.
399	De Backer, H., De Muer, D. and De Sadelaer, G., 1998. Comparison of ozone profiles obtained with
400	Brewer-Mast and Z-ECC sensors during simultaneous ascents. Journal of Geophysical
401	Research: Atmospheres, 103(D16), 19641-19648.
402	Deshler, T., Mercer, J.L., Smit, H.G., Stubi, R., Levrat, G., Johnson, B.J., Oltmans, S.J., Kivi, R.,
403	Thompson, A.M., Witte, J., Davies, J., 2008. Atmospheric comparison of electrochemical cell
404	ozonesondes from different manufacturers, and with different cathode solution strengths: The
405	Balloon Experiment on Standards for Ozonesondes. Journal of Geophysical Research:
406	Atmospheres, 113(D4), D04307, doi:10.1029/2007JD008975.





407	Dias-Lalcaca, P., Brunner, D., Imfeld, W., Moser, W. and Staehelin, J., 1998. An automated system
408	for the measurement of nitrogen oxides and ozone concentrations from a passenger aircraft:
409	Instrumentation and first results of the NOXAR project. Environmental Science & Technology,
410	32(20), 3228-3236.
411	Ebojie, F., Burrows, J.P., Gebhardt, C., Ladstätter-Weißenmayer, A., Von Savigny, C., Rozanov, A.,
412	Weber, M. and Bovensmann, H., 2016. Global tropospheric ozone variations from 2003 to 2011
413	as seen by SCIAMACHY. Atmospheric Chemistry and Physics, 16(2), 417-436.
414	Fu, Y. and Tai, A.P.K., 2015. Impact of climate and land cover changes on tropospheric ozone air
415	quality and public health in East Asia between 1980 and 2010. Atmospheric Chemistry and
416	Physics, 15(17), 10093-10106.
417	García, O.E., Sanromá, E., Schneider, M., Hase, F., León-Luis, S.F., Blumenstock, T., Sepúlveda,
418	E., Redondas, A., Carreño, V., Torres, C. and Prats, N., 2022. Improved ozone monitoring by
419	ground-based FTIR spectrometry. Atmospheric Measurement Techniques, 15(8), 2557-2577.
420	García, O.E., Schneider, M., Sepúlveda, E., Hase, F., Blumenstock, T., Cuevas, E., Ramos, R., Gross,
421	J., Barthlott, S., Röhling, A.N. and Sanromá, E., 2021. Twenty years of ground-based NDACC
422	FTIR spectrometry at Izaña Observatory-overview and long-term comparison to other
423	techniques. Atmospheric Chemistry and Physics, 21(20), 15519-15554.
424	Gaudel, A., Cooper, O.R., Ancellet, G., Barret, B., Boynard, A., Burrows, J.P., Clerbaux, C., Coheur,
425	P.F., Cuesta, J., Cuevas, E. and Doniki, S., 2018. Tropospheric Ozone Assessment Report:
426	Present-day distribution and trends of tropospheric ozone relevant to climate and global
427	atmospheric chemistry model evaluation. Elementa: science of the anthropocene, 6.
428	Gaudel, A., Cooper, O.R., Chang, K.L., Bourgeois, I., Ziemke, J.R., Strode, S.A., Oman, L.D.,
429	Sellitto, P., Nédélec, P., Blot, R. and Thouret, V., 2020. Aircraft observations since the 1990s
430	reveal increases of tropospheric ozone at multiple locations across the Northern
431	Hemisphere. Science Advances, 6(34), eaba8272.
432	Gebhardt, C., Rozanov, A., Hommel, R., Weber, M., Bovensmann, H., Burrows, J.P., Degenstein,
433	D., Froidevaux, L. and Thompson, A.M., 2014. Stratospheric ozone trends and variability as
434	seen by SCIAMACHY from 2002 to 2012. Atmospheric Chemistry and Physics, 14(2), 831-
435	846.





436 Hassler, B., Kremser, S., Bodeker, G. E., Lewis, J., Nesbit, K., Davis, S. M., Chipperfield, M. P., 437 Dhomse, S. S., and Dameris, M., 2018, An updated version of a gap-free monthly mean zonal mean ozone database. Earth System Science Data, 10, 1473-1490. 438 Hegarty, J., Mao, H. and Talbot, R., 2009. Synoptic influences on springtime tropospheric O 3 and 439 440 CO over the North American export region observed by TES. Atmospheric Chemistry and 441 Physics, 9(11), 3755-3776. Hilsenrath, E., Attmannspacher, W., Bass, A., Evans, W., Hagemeyer, R., Barnes, R., Komhyr, W., 442 Mauersberger, K., Mentall, J., Proffitt, M., Robbins, D., 1986. Results from the balloon ozone 443 444 intercomparison campaign (BOIC). Journal of Geophysical Research: Atmospheres, 91(D12), 13137-13152. 445 Hoogen, R., Rozanov, V.V. and Burrows, J.P., 1999. Ozone profiles from GOME satellite data: 446 447 Algorithm description and first validation. Journal of Geophysical Research: 448 Atmospheres, 104(D7), 8263-8280. 449 Hu, L., Jacob, D.J., Liu, X., Zhang, Y., Zhang, L., Kim, P.S., Sulprizio, M.P. and Yantosca, R.M., 450 2017. Global budget of tropospheric ozone: Evaluating recent model advances with satellite 451 (OMI), aircraft (IAGOS), and ozonesonde observations. Atmospheric Environment, 167, 323-452 334. 453 Hubert, D., Heue, K.P., Lambert, J.C., Verhoelst, T., Allaart, M., Compernolle, S., Cullis, P.D., Dehn, 454 A., Félix, C., Johnson, B.J. and Keppens, A., 2021. TROPOMI tropospheric ozone column data: 455 geophysical assessment and comparison to ozonesondes, GOME-2B and OMI. Atmospheric 456 Measurement Techniques, 14(12), 7405-7433. 457 Keckhut, P., McDermid, S., Swart, D., McGee, T., Godin-Beekmann, S., Adriani, A., Barnes, J., 458 Baray, J.L., Bencherif, H., Claude, H. and di Sarra, A.G., 2004. Review of ozone and 459 temperature lidar validations performed within the framework of the Network for the Detection 460 of Stratospheric Change. Journal of Environmental Monitoring, 6(9), 721-733. 461 Kerr, J.B., Fast, H., McElroy, C.T., Oltmans, S.J., Lathrop, J.A., Kyro, E., Paukkunen, A., Claude, 462 H., Köhler, U., Sreedharan, C.R., Takao, T., 1994. The 1991 WMO international ozonesonde 463 intercomparison at Vanscoy, Canada. Atmosphere-Ocean, 32(4), 685-716. 464 Lefohn, A.S., Malley, C.S., Smith, L., Wells, B., Hazucha, M., Simon, H., Naik, V., Mills, G.,





- 465 Schultz, M.G., Paoletti, E. and De Marco, A., 2018. Tropospheric ozone assessment report:
- 466 Global ozone metrics for climate change, human health, and crop/ecosystem
- research. Elementa: Science of the Anthropocene, 6.
- 468 Li, K., Jacob, D.J., Liao, H., Qiu, Y., Shen, L., Zhai, S., Bates, K.H., Sulprizio, M.P., Song, S., Lu,
- 469 X. and Zhang, Q., 2021. Ozone pollution in the North China Plain spreading into the late-
- winter haze season. Proceedings of the National Academy of Sciences, 118(10), e2015797118.
- 471 Li, K., Jacob, D.J., Shen, L., Lu, X., De Smedt, I. and Liao, H., 2020. Increases in surface ozone
- 472 pollution in China from 2013 to 2019: anthropogenic and meteorological
- influences. Atmospheric Chemistry and Physics, 20(19), 11423-11433.
- 474 Liao, Z., Ling, Z., Gao, M., Sun, J., Zhao, W., Ma, P., Quan, J. and Fan, S., 2021. Tropospheric
- 475 ozone variability over Hong Kong based on recent 20 years (2000-2019) ozonesonde
- observation. Journal of Geophysical Research: Atmospheres, 126(3), e2020JD033054.
- 477 Liu, G., Liu, J., Tarasick, D.W., Fioletov, V.E., Jin, J.J., Moeini, O., Liu, X., Sioris, C.E. and Osman,
- 478 M., 2013. A global tropospheric ozone climatology from trajectory-mapped ozone soundings.
- 479 Atmospheric Chemistry and Physics, 13(21), 10659-10675.
- 480 Logan, J.A., 1985. Tropospheric ozone: Seasonal behavior, trends, and anthropogenic
- 481 influence. Journal of Geophysical Research: Atmospheres, 90(D6), 10463-10482.
- 482 Ma, Y., Ma, B., Jiao, H., Zhang, Y., Xin, J. and Yu, Z., 2020. An analysis of the effects of weather
- and air pollution on tropospheric ozone using a generalized additive model in Western China:
- 484 Lanzhou, Gansu. Atmospheric Environment, 224, 117342.
- 485 McPeters, R.D., Labow, G.J. and Logan, J.A., 2007. Ozone climatological profiles for satellite
- 486 retrieval algorithms. Journal of Geophysical Research: Atmospheres, 112(D5), D05308.
- 487 doi:10.1029/2005JD006823.
- 488 McPeters, R.D. and Labow, G.J., 2012. Climatology 2011: An MLS and sonde derived ozone
- climatology for satellite retrieval algorithms. Journal of Geophysical Research: Atmospheres,
- 490 117(D10), D10303. doi:10.1029/2011JD017006.
- 491 Miles, G.M., Siddans, R., Kerridge, B.J., Latter, B.G. and Richards, N.A.D., 2015. Tropospheric
- 492 ozone and ozone profiles retrieved from GOME-2 and their validation. Atmospheric
- 493 Measurement Techniques, 8(1), 385-398.





494 Meng, K., Zhao, T., Xu, X., Zhang, Z., Bai, Y., Hu, Y., Zhao, Y., Zhang, X. and Xin, Y., 2022. 495 Influence of stratosphere-to-troposphere transport on summertime surface O3 changes in North China Plain in 2019. Atmospheric research, 276, 106271. 496 497 Monks, P.S., Archibald, A.T., Colette, A., Cooper, O., Coyle, M., Derwent, R., Fowler, D., Granier, 498 C., Law, K.S., Mills, G.E. and Stevenson, D.S., 2015. Tropospheric ozone and its precursors 499 from the urban to the global scale from air quality to short-lived climate forcer. Atmospheric 500 Chemistry and Physics, 15(15), 8889-8973. 501 Percy, K.E., Legge, A.H. and Krupa, S.V., 2003. Tropospheric ozone: a continuing threat to global 502 forests? Developments in Environmental Science, 3, 85-118. Perlwitz, J., Pawson, S., Fogt, R.L., Nielsen, J.E. and Neff, W.D., 2008. Impact of stratospheric 503 ozone hole recovery on Antarctic climate. Geophysical Research Letters, 35(8). 504 505 Ramanathan, V., Cicerone, R.J., Singh, H.B. and Kiehl, J.T., 1985. Trace gas trends and their 506 potential role in climate change. Journal of Geophysical Research: Atmospheres, 90(D3), 507 5547-5566. 508 Saltzman, B.E. and Gilbert, N., 1959. Iodometric microdetermination of organic oxidants and ozone. 509 Resolution of mixtures by kinetic colorimetry. Analytical Chemistry, 31(11), 1914-1920. 510 Schnadt Poberaj, C., Staehelin, J., Brunner, D., Thouret, V. and Mohnen, V., 2007. A UT/LS ozone 511 climatology of the nineteen seventies deduced from the GASP aircraft measurement program. 512 Atmospheric Chemistry and Physics, 7(22), 5917-5936. 513 Schultz, M.G., Schröder, S., Lyapina, O., Cooper, O.R., Galbally, I., Petropavlovskikh, I., Von 514 Schneidemesser, E., Tanimoto, H., Elshorbany, Y., Naja, M. and Seguel, R.J., 2017. 515 Tropospheric Ozone Assessment Report: Database and metrics data of global surface ozone 516 observations. Elementa: Science of the Anthropocene, 5. 517 Sharma, S., Sharma, P. and Khare, M., 2017. Photo-chemical transport modelling of tropospheric 518 ozone: A review. Atmospheric Environment, 159, 34-54. 519 Smit, H.G.J., Sträter, W., Helten, M., Kley, D., Ciupa, D., Claude, H.J., Köhler, U., Hoegger, B., 520 Levrat, G., Johnson, B., Oltmans, S.J., Kerr, J.B., Tarasick, D.W., Davies, J., Shitamichi, M., Srivastav, S.K. and Vialle, C., 1996. JOSIE: The 1996 WMO international intercomparison of 521 522 ozonesondes under quasi-flight conditions in the environmental chamber at Jülich, Proc.





523	Quadrennial Ozone Symposium 1996, l'Aquila, Italy, edited by R. D. Bojkov and G. Visconti,
524	pp. 971-974, Parco Sci. e Tecnol. d'Abruzzo, Italy, 1996.
525	Smit, H.G., Straeter, W., Johnson, B.J., Oltmans, S.J., Davies, J., Tarasick, D.W., Hoegger, B., Stubi,
526	R., Schmidlin, F.J., Northam, T., Thompson, A.M., 2007. Assessment of the performance of
527	ECC-ozonesondes under quasi-flight conditions in the environmental simulation chamber:
528	Insights from the Juelich Ozone Sonde Intercomparison Experiment (JOSIE). Journal of
529	Geophysical Research: Atmospheres, 112(D19), D19306, doi:10.1029/2006JD007308.
530	$Smit, H.G.J., 2014.\ Ozones ondes, in Encyclopedia of Atmospheric Sciences, Second Edition, edited$
531	by G.R. North, J.A. Pyle, and F. Zhang, Vol 1, pp. 372–378, Academic Press, London.
532	Smit, H. G. J., Poyraz, D., Van Malderen, R., Thompson, A. M., Tarasick, D. W., Stauffer, R. M.,
533	Johnson, B. J., and Kollonige, D. E., 2023. New insights from the Jülich Ozone Sonde
534	Intercomparison Experiment: calibration functions traceable to one ozone reference instrument.
535	Atmospheric Measurement Techniques, 17, 73-112, <a href="https://doi.org/10.5194/amt-17-73-2024">https://doi.org/10.5194/amt-17-73-2024</a> .
536	Smit, H.G.J., Thompson, A.M., and the ASOPOS 2.0 Panel, 2021. Ozonesonde Measurement
537	Principles and 1300 Best Operational Practices, WMO Global Atmosphere Watch Report
538	Series, No. 268, World Meteorological Organization, 1301 Geneva, [Available online at
539	https://library.wmo.int/doc_num.php?explnum_id=10884].
540	Solomon, S., 1999. Stratospheric ozone depletion: A review of concepts and history. Reviews of
541	Geophysics, 37(3), 275-316.
542	Staehelin, J., Harris, N.R., Appenzeller, C. and Eberhard, J., 2001. Ozone trends: A review. Reviews
543	of Geophysics, 39(2), 231-290.
544	$Staufer, J., Staehelin, J., St\"{u}bi, R., Peter, T., Tummon, F., and Thouret, V., 2013. \ Trajectory\ matching$
545	of ozonesondes and MOZAIC measurements in the UTLS - Part 1: Method description and
546	application at Payerne, Switzerland. Atmospheric Measurement Techniques, 6, 3393-3406.
547	doi:10.5194/amt-6-3393-2013.
548	Staufer, J., Staehelin, J., Stübi, R., Peter, T., Tummon, F., and Thouret, V., 2014. Trajectory matching
549	of ozonesondes and MOZAIC measurements in the UTLS - Part 2: Application to the global
550	ozonesonde network. Atmospheric Measurement Techniques, 7, 241-266. doi:10.5194/amt-7-
551	241-2014.





552	Stübi, R., Levrat, G., Hoegger, B., Viatte, P., Staehelin, J. and Schmidlin, F.J., 2008. In-flight						
553	comparison of Brewer-Mast and electrochemical concentration cell ozonesondes. Journal of						
554	Geophysical Research: Atmospheres, 113(D13). https://doi.org/10.1029/2007JD009091.						
555	Tanimoto, H., Zbinden, R.M., Thouret, V. and Nédélec, P., 2015. Consistency of tropospheric ozone						
556	observations made by different platforms and techniques in the global databases. Tellus B:						
557	Chemical and Physical Meteorology, 67(1), 27073.						
558	Tarasick, D. W., Davies, J., Anlauf, K., Watt, M., Steinbrecht, W., Claude, H. J., 2002. Laboratory						
559	investigations of the response of Brewer-Mast ozonesondes to tropospheric ozone. Journal of						
560	Geophysical Research: Atmospheres, 107(D16), ACH-14,						
561	https://doi.org/10.1029/2001JD001167.						
562	Tarasick, D. W., Davies, J., Smit, H. G., Oltmans, S. J., 2016. A re-evaluated Canadian ozonesonde						
563	record: measurements of the vertical distribution of ozone over Canada from 1966 to 2013.						
564	Atmospheric Measurement Techniques, 9(1), 195-214.						
565	Tarasick, D.W., Galbally, I., Cooper, O.R., Schultz, M.G., Ancellet, G., LeBlanc, T., Wallington, T.J.,						
566	Ziemke, J., Liu, X., Steinbacher, M., Stähelin, J., Vigouroux, C., Hannigan, J., García, O., Foret,						
567	G., Zanis, P., Weatherhead, E., Petropavlovskikh, I., Worden, H., Neu, J.L., Osman, M., Liu, J.,						
568	$Lin, M., Granados-Mu\~noz, M., Thompson, A.M., Oltmans, S.J., Cuesta, J., Dufour, G., Thouret, A.M., Cuesta, J., Dufour, G., Thouret, $						
569	V., Hassler, B., Thompson, A.M., and Trickl, T., 2019. TOAR- Observations: Tropospheric						
570	ozone from 1877 to 2016, observed levels, trends and uncertainties. Elementa: Science of the						
571	Anthropocene, 7(1), 39. DOI: <a href="http://doi.org/10.1525/elementa.376">http://doi.org/10.1525/elementa.376</a> .						
572	Tarasick, D.W., Smit, H.G., Thompson, A.M., Morris, G.A., Witte, J.C., Davies, J., Nakano, T., Van						
573	Malderen, R., Stauffer, R.M., Johnson, B.J., Stübi, R., Oltmans, S.J., and Vömel, H., 2021.						
574	Improving ECC ozonesonde data quality: Assessment of current methods and outstanding						
575	issues. Earth and Space Science, 8, e2019EA000914. <a href="https://doi.org/10.1029/2019EA000914">https://doi.org/10.1029/2019EA000914</a> .						
576	Thouret, V., Marenco, A., Logan, J.A., Nédélec, P. and Grouhel, C., 1998. Comparisons of ozone						
577	measurements from the MOZAIC airborne program and the ozone sounding network at eight						
578	locations. Journal of Geophysical Research: Atmospheres, 103(D19), 25695-25720. doi:						
579	10.1029/98JD02243.						
580	Thompson, A. M., Smit, H. G. J., Witte, J. C., Stauffer, R. M., Johnson, B. J., Morris, G., von der						





582 Hoang Anh, N. T., Corrales, E., Machinini, T., da Silva, F. R., Paiman, G., Thiong'o, K., Zainal, Z., Brothers, G. B., Wolff, K. R., Nakano, T., Stübi, R., Romanens, G., Coetzee, G. J. R., Diaz, 583 584 J. A., Mitro, S., Mohamad, M., and Ogino, S., 2019. Ozonesonde quality assurance: The 585 JOSIE-SHADOZ (2017) experience. Bull. Am. Meteorol. Soc., 100(1), 155-171. 586 Vingarzan, R., 2004. A review of surface ozone background levels and trends. Atmospheric 587 environment, 38(21), 3431-3442. Wang, H., Ke, Y., Tan, Y., Zhu, B., Zhao, T. and Yin, Y., 2023. Observational evidence for the dual 588 589 roles of BC in the megacity of eastern China: Enhanced O3 and decreased PM2. 5 590 pollution. Chemosphere, 327, 138548. 591 Wang, H., Lu, X., Jacob, D.J., Cooper, O.R., Chang, K.L., Li, K., Gao, M., Liu, Y., Sheng, B., Wu, K. 592 and Wu, T., 2022. Global tropospheric ozone trends, attributions, and radiative impacts in 1995-593 2017: an integrated analysis using aircraft (IAGOS) observations, ozonesonde, and multi-decadal 594 chemical model simulations. Atmospheric Chemistry and Physics, 22(20), 13753-13782. 595 Wang, T., Xue, L., Brimblecombe, P., Lam, Y.F., Li, L. and Zhang, L., 2017. Ozone pollution in 596 China: A review of concentrations, meteorological influences, chemical precursors, and 597 effects. Science of the Total Environment, 575, 1582-1596. 598 Williamson, C.E., Neale, P.J., Hylander, S., Rose, K.C., Figueroa, F.L., Robinson, S.A., Häder, D.P., 599 Wängberg, S.Å. and Worrest, R.C., 2019. The interactive effects of stratospheric ozone 600 depletion, UV radiation, and climate change on aquatic ecosystems. Photochemical & 601 Photobiological Sciences, 18(3), 717-746. Xu, J., Huang, X., Wang, N., Li, Y. and Ding, A., 2021. Understanding ozone pollution in the 602 603 Yangtze River Delta of eastern China from the perspective of diurnal cycles. Science of the 604 Total Environment, 752, 141928. Yang, T., Li, H., Wang, H., Sun, Y., Chen, X., Wang, F., Xu, L. and Wang, Z., 2023. Vertical aerosol 605 606 data assimilation technology and application based on satellite and ground lidar: A review and 607 outlook. Journal of Environmental Sciences, 123, 292-305. 608 Young, P.J., Naik, V., Fiore, A.M., Gaudel, A., Guo, J., Lin, M.Y., Neu, J.L., Parrish, D.D., Rieder, 609 H.E., Schnell, J.L. and Tilmes, S., 2018. Tropospheric Ozone Assessment Report: Assessment

Gathen, P., Van Malderen, R., Davies, J., Piters, A., Allaart, M., Posny, F., Kivi, R., Cullis, P.,

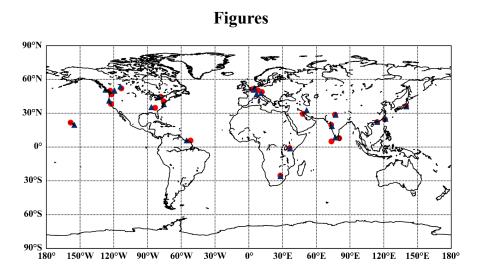




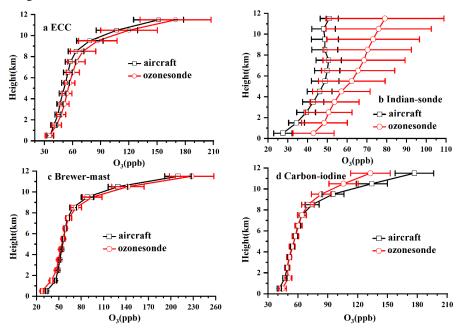
610	of global-scale model performance for global and regional ozone distributions, variability, and						
611	trends. Elementa: Science of the Anthropocene, 6.						
612	Yu, R., Lin, Y., Zou, J., Dan, Y. and Cheng, C., 2021. Review on atmospheric Ozone pollution in						
613	China: Formation, spatiotemporal distribution, precursors and affecting						
614	factors. Atmosphere, 12(12), p.1675.						
615	Zang, Z., J.J. Liu, D.W. Tarasick, O. Moeini, B. Jianchun, J. Zhang, R. Van Malderen, A.M. Thompson,						
616	H.G.J. Smit, R.M. Stauffer and B.J. Johnson (2024), An updated and improved Trajectory-mapped						
617	Ozonesonde dataset for the Stratosphere and Troposphere (TOST) from 1970-2021, to be submitted						
618	to ACP.						
619	Zbinden, R.M., Thouret, V., Ricaud, P., Carminati, F., Cammas, J.P. and Nédélec, P., 2013. Climatology						
620	of pure tropospheric profiles and column contents of ozone and carbon monoxide using MOZAIC						
621	in the mid-northern latitudes (24° N to 50° N) from 1994 to 2009. Atmospheric Chemistry and						
622	Physics, 13(24), 12363-12388, https://doi.org/10.5194/acp-13-12363-2013, 2013.						
623	Zhang, L., Jacob, D.J., Boersma, K.F., Jaffe, D.A., Olson, J.R., Bowman, K.W., Worden, J.R., Thompson, A.D., Thompson, A.D., College, A.D.,						
624	A.M., Avery, M.A., Cohen, R.C. and Dibb, J.E., 2008. Transpacific transport of ozone pollution and						
625	the effect of recent Asian emission increases on air quality in North America: an integrated analysis						
626	using satellite, aircraft, ozonesonde, and surface observations. Atmospheric Chemistry and						
627	Physics, 8(20), 6117-6136.						
628	Zhao, K., Huang, J., Wu, Y., Yuan, Z., Wang, Y., Li, Y., Ma, X., Liu, X., Ma, W., Wang, Y. and Zhang, Mang, Mang						
629	X., 2021. Impact of stratospheric intrusions on ozone enhancement in the lower troposphere						
630	and implication to air quality in Hong Kong and other South China regions. Journal of						
631	Geophysical Research: Atmospheres, 126(18), e2020JD033955.						







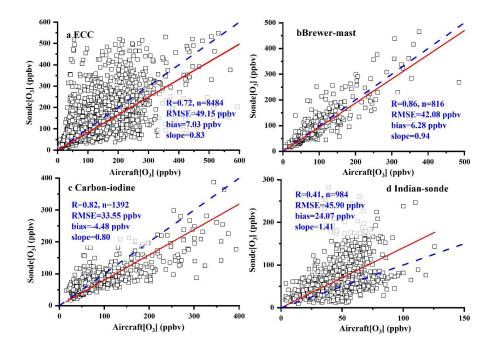
**Figure 1.** Map of 23 pairs of sites used in this study. Red circle markers are IAGOS sites, blue triangle markers are WOUDC sites.



**Figure 2.** Comparison of the vertical profiles of tropospheric O<sub>3</sub> observed between aircraft measurements and four types of ozonesondes, ECC, Indian-sonde, Brewer-mast, and Carbon-iodine. The error bar length is 4 times the standard error (SE) of the mean (equivalent to 95% confidence limits on the averages).



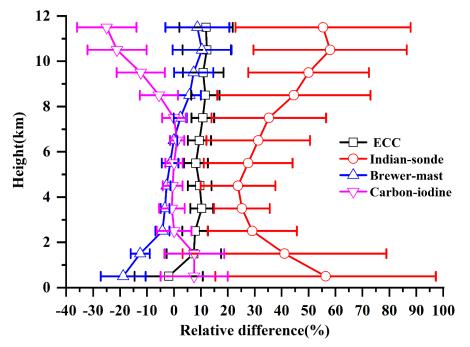




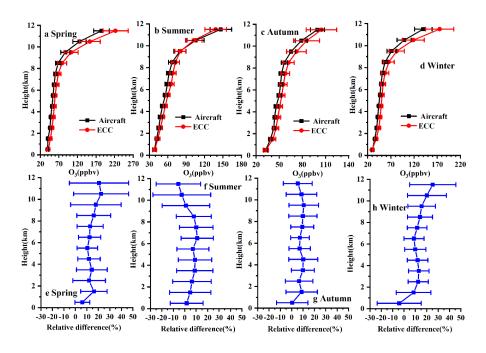
**Figure 3.** Correlation (R) of ozone mixing ratios between ozonesonde and aircraft measurements. The blue dashed line shows the 1:1 axis. Correlations are significant at the 99% level (p < 0.01). N denotes the number of data points, and RMSE is the root mean square error.







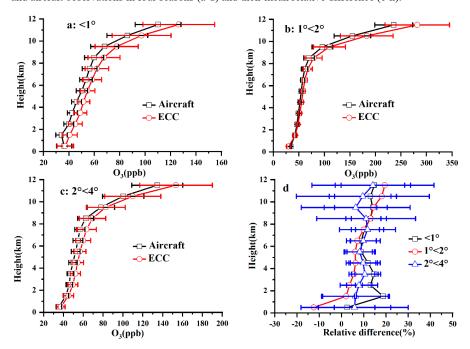
**Figure 4.** Mean relative difference (RD) between the ozonesonde  $O_3$  and aircraft  $O_3$  data. RD is calculated from  $(O_{3\text{-ozonesonde}} - O_{3\text{-aircraft}})/O_{3\text{-aircraft}} \times 100\%$ .







**Figure 5.** The mean difference in vertical profiles of the tropospheric O<sub>3</sub> between ECC ozonesonde and aircraft observations in four seasons (a-d) and their mean relative difference (e-h).



**Figure 6.** The annual mean vertical profiles of tropospheric  $O_3$  between ECC ozonesonde and aircraft observations at station-pair distances (D) of D<1° (a), 1°< D <2° (b), and 2°< D <4°. The relative differences for the three categories are shown in (d).





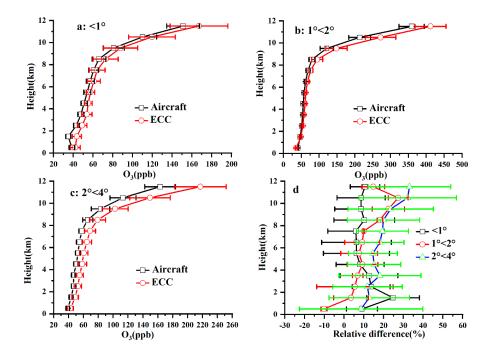


Figure 7. The seasonal mean vertical profiles of tropospheric  $O_3$  in spring between ECC ozonesonde and aircraft observations at station-pair distances (D) of D<1° (a), 1°< D<2° (b), and 2°< D<4°. The relative differences for the three categories are shown in (d).





**Tables** 

Table 1. Summary of the station information, including station's name, geolocation, the number of profiles, observational period, and the station-pair distance used in

r e	MOZAIC-IAGOS	·IAGOS			ĸ	WOUDC					
Station name	Lon	Lat	No. profiles	Station name	Lon	Lat	No. profiles	Type	No. valid data months	observation period	station- airport distance
Toronto	-78.50	44.58	321	Egbert	-79.78	44.23	181	ECC	33	2004-2008	
Dusseldorf	4.96	51.82	412	De Bilt	5.18	52.10	333	ECC	63	1995-2013	
Munich	11.63	48.84	2136	Hohenpeissenberg	11.01	47.80	1032	Brewer-mast	29	1996-2006	
Johannesburg	28.07	25.32	199	Irene	28.22	25.91	135	ECC	26	1998-2003	
Nairobi	36.33	-0.94	114	Nairobi	36.75	-1.30	42	ECC	10	1997-1998	7
Mumbai	73.27	19.70	122	Pune	73.85	18.53	99	Indian-sonde	35	1996-2003	S.T.V
Delhi	76.65	28.73	342	New Delhi	77.18	28.63	88	Indian-sonde	50	1995-2016	
Hongkong	114.11	22.10	123	King's Park	114.17	22.31	115	ECC	25	2000-2005	
Taipei	121.08	24.59	2115	Taipei	121.48	25.02	58	ECC	31	2014-2018	
Tokyo	139.73	36.33	1342	Tateno (Tsukuba)	140.13	36.05	655	Carbon- iodine	116	1995-2006	
Calgary	113.25	52.03	170	Edmonton	-114.10	53.55	112	ECC	17	2009-2011	1°~2°
Brussels	3.24	51.21	2412	Uccle	4.36	50.80	736	ECC	55	1997-2009	
Honolulu	158.33	21.66	169	Hilo (HI)	155.07	19.58	107	ECC	16	2015-2017	2°~4°

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2003-2015	1999-2001	2003-2009	1999-2006	1994-2014	2002-2013	2002-2020	2001-2004	1997-2000	1998-2000
89	10	45	10	80	6	204	17	24	11
ECC	ECC	ECC	ECC	ECC	ECC	ECC	ECC	Indian-sonde	Indian-sonde
594	53	317	85	616	64	2673	34	45	37
50.69	41.05	49.97	35.28	37.94	5.81	46.81	32.48	8.48	8.48
- 127.38	124.15	- 119.38	-86.58	-75.46	-55.21	6.94	51.43	76.95	76.95
Kelowna	Trinidad Head (CA)	Kelowna	Huntsville (AL)	Wallops Island (VA)	Paramaribo	Payerne	Esfahan	Trivandrum	Trivandrum
595	34	385	34	610	200	12742	105	92	31
49.95	38.30	46.76	34.78	40.52	5.75	50.16	29.52	5.00	7.79
- 123.14	122.50	122.06	-83.28	-75.59	-51.78	8.30	48.01	73.49	80.41
Vancouver	San- Francisco	Portland	Atlanta	Washington	Cayenne	Frankfurt	Kuwait-City	Male	Colombo

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# 1 Table 2. Bias, correlation coefficient (R), and RMSE for four types of ozonesonde and aircraft

## 2 observations in four seasons.

Туре	Season	Bias (O <sub>3-ozonesonde</sub> - O <sub>3-aircraft</sub> ) (ppb)	R	RMSE (ppb)
	Spring	17.34	0.76	65.52
ECC	Summer	1.96	0.76	40.15
ECC	Autumn	1.75	0.71	34.47
	Winter	7.61	0.71	51.74
	Spring	10.22	0.94	43.51
D	Summer	2.99	0.83	48.79
Brewer-mast	Autumn	6.53	0.79	29.40
	Winter	6.11	0.88	45.45
	Spring	-9.19	0.84	38.34
Carbon-iodine	Summer	3.83	0.46	29.31
Carbon-iodine	Autumn	2.33	0.68	15.10
	Winter	-16.68	0.88	44.72
	Spring	19.64	0.44	44.30
Indian-sonde	Summer	19.58	0.57	37.44
indian-sonde	Autumn	20.38	0.45	37.30
	Winter	40.07	0.18	64.99

3

4 Table 3. Bias, correlation coefficient(R) and RMSE for ECC and Indian-sonde ozonesonde and

5 aircraft observations at different station-airport distances.

Tree	Station-pair	Diag (O O ) (such)		DMCE (1.)	
Туре	distance	Bias (O <sub>3-ozonesonde</sub> - O <sub>3-aircraft</sub> ) (ppb)	R	RMSE (ppb)	
	<1°	9.78	0.78	47.46	
ECC	1°~2°	8.91	0.90	40.73	
	2°~4°	5.65	0.67	51.00	
Indian-sonde	<1°	26.71	0.37	49.54	
indian-sonde	2°~4°	15.35	0.24	30.86	





**Table 4.** Comparison of the sondes of each type to IAGOS. (average  $\pm$  2 times the standard error (SE)) Indian-sonde/ECC is (Indian-sonde/IAGOS)/(ECC/IAGOS), Brewer-mast/ECC is (Brewer-mast/IAGOS)/(ECC/IAGOS), Carbon-iodine/ECC is (Carbon-iodine/IAGOS)/(ECC/IAGOS)

Altitude(km)	Indian-	Brewer-	Carbon-	ECC/ IAGOS
Attitude(kiii)	sonde/ECC	mast/ECC	iodine/ECC	ECC/ IAGOS
0~1	$1.59 \pm 1.74$	$0.83 \pm 0.96$	$1.10 \pm 1.36$	$0.98 \pm 1.28$
1~2	$1.31 \pm 1.83$	$0.81 \pm 0.90$	$1.00\pm1.05$	$1.07 \pm 1.58$
2~3	$1.20 \pm 1.62$	$0.89 \pm 0.97$	$0.93 \pm 0.85$	$1.08\pm\!1.54$
3~4	$1.14 \pm 1.57$	$0.88 \pm 0.94$	$0.90 \pm 0.87$	$1.10\pm\!1.48$
4~5	$1.13 \pm 1.61$	$0.89 \pm 1.02$	$0.91 \pm 0.99$	$1.10\pm\!1.44$
5~6	$1.18\pm\!1.76$	$0.91 \pm 1.05$	$0.92 \pm 1.04$	$1.08\pm\!1.37$
6~7	$1.20 \pm 1.89$	$0.91~{\pm}1.00$	$0.92 \pm 0.82$	$1.09 \pm 1.54$
7~8	$1.22 \pm 1.92$	$0.92 \pm 0.94$	$0.90 \pm 0.64$	$1.11 \pm 1.69$
8~9	$1.29 \pm 2.09$	$0.95 \pm 0.99$	$0.85 \pm 0.55$	$1.12 \pm 1.61$
9~10	$1.35 \pm 2.35$	$0.97 \pm 1.09$	$0.79 \pm 0.62$	$1.11 \pm 1.46$
10~11	$1.41 \pm 3.26$	$0.98 \pm 1.21$	$0.70 \pm 0.68$	$1.12 \pm 1.37$
11~12	$1.39 \pm 4.61$	$0.97 \pm 1.19$	$0.67 \pm 0.72$	$1.12 \pm 1.42$