Global-scale distribution of ozone in the remote troposphere from ATom and HIPPO airborne field missions.

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Abstract

Ozone is a key constituent of the troposphere where it drives photochemical processes, impacts air quality, and acts as a climate forcer. Large-scale in situ observations of ozone commensurate with the grid resolution of current Earth system models are necessary to validate model outputs and satellite retrievals. In this paper, we examine measurements from the Atmospheric Tomography (ATom, 4 deployments in 2016–2018) and the HIAPER Pole-to-Pole Observations (HIPPO; 5 deployments in 2009–2011) experiments, two global-scale airborne campaigns covering the Pacific (HIPPO and ATom) and Atlantic (ATom) basins.

ATom and HIPPO represent the first global-scale, vertically resolved measurements of O_3 distributions throughout the troposphere, with HIPPO sampling the Pacific basin and ATom sampling both the Pacific and Atlantic basins. Given the relatively limited temporal resolution of these two campaigns, we first compare ATom and HIPPO ozone data to longer-term observational records to establish the representativeness of our dataset. We show that these two airborne campaigns captured on average 53, 54, and 38 % of the ozone variability in the marine boundary layer, free troposphere, and upper troposphere/lower stratosphere (UTLS), respectively, at nine well-established ozonesonde sites. Additionally, ATom captured the most frequent ozone concentrations measured by regular commercial aircraft flights in the northern Atlantic UTLS. We then use the repeated vertical profiles carried out during these two campaigns to confirm and extend the existing knowledge ~~global-scale picture~~ of tropospheric ozone spatial and vertical distributions throughout the remote troposphere. We highlight a clear hemispheric gradient, with greater ozone in the northern hemisphere consistent with greater precursor emissions, on par with previous modeling and satellite studies. We also show that the ozone distribution below 8 km was similar in the extra-tropics of the Atlantic and Pacific basins likely due to zonal circulation patterns. However, twice as much ozone was found in the tropical Atlantic than in the tropical Pacific, due to well-documented dynamical patterns transporting continental air masses over the Atlantic. Finally, we show that the seasonal variability of tropospheric ozone over the Pacific and the Atlantic basins is driven by transported continental plumes and photochemistry, and the vertical distribution is driven by photochemistry and mixing with stratospheric air. This new dataset provides additional constraints for global climate and chemistry models to improve our understanding of both ozone production and loss processes in remote regions, as well as the influence of anthropogenic emissions on baseline ozone.

1. Introduction

Tropospheric ozone (O_3) plays a major role in local, regional, and global air quality and significantly influences Earth's radiative budget (IPCC, 2013; Shindell et al., 2012). In addition, O_3 drives tropospheric photochemical processes by controlling hydroxyl radical (OH) abundance, which subsequently controls the lifetime of other pollutants including volatile organic compounds (VOCs), greenhouse gases, and some stratospheric ozone-depleting substances (Crutzen, 1974; Levy, 1971). Sources of O_3 to the troposphere include downward transport from the stratosphere (Junge, 1962) and photochemical production from precursors such as carbon monoxide (CO), methane (CH_4), and VOCs in the presence of nitrogen oxides (NO_x) emitted by natural or anthropogenic sources (Monks et al., 2009). Tropospheric O_3 sinks include photo-dissociation, chemical reactions, and dry deposition. Owing to its relatively long lifetime (~23 days in the troposphere; Young et al., 2013), O_3 can be transported across intra-hemispheric scales. O_3 mixing ratios over a region thus depend not only on local and regional sources and sinks, but also on long-range transport. Further, the uneven density of O_3 monitoring locations around the globe leads to significant sampling gaps, especially near developing nations (Gaudel et al., 2018). The troposphere over the remote oceans is among the least-sampled regions, despite hosting 60–70 % of global tropospheric O_3 burden (Holmes et al., 2013).

Since the early 1980's, several aircraft campaigns have periodically addressed this paucity of remote observations, most notably under the umbrella of the Global Tropospheric Experiment (GTE), a major component of the National Aeronautics and Space Administration (NASA) Tropospheric Chemistry Program (https://eosweb.larc.nasa.gov/project/gte/gte_table). Airborne campaigns have targeted both the Pacific and Atlantic Oceans, providing novel characterization of O_3 sources, distribution, and photochemistry in the marine troposphere (Browell et al., 1996a; Davis et al., 1996; Jacob et al., 1996; Pan et al., 2015; Schultz et al., 1999; Singh et al., 1996c) and the low-O_3 tropical Pacific pool (Kley et al., 1996; Singh et al., 1996b), the pervasive role of continental outflow on O_3 production (Bey et al., 2001; Crawford et al., 1997; Heald et al., 2003; Kondo et al., 2004; Martin et al., 2002; Zhang et al., 2008), and the marked influence of African and South American biomass burning on O_3 production in the Southern Hemisphere (Browell et al., 1996b; Fenn et al., 1999; Mauzerall et al., 1998; Singh et al., 1996a; Thompson et al., 1996). Ozonesondes have been launched from remote sites for more than three decades in some places, and have provided additional constraints on the sources and

66 photochemical balance of tropospheric O_3 including a deep understanding of vertically-resolved
67 tropospheric O_3 climatology in select locations (Derwent et al., 2016; Diab et al., 2004; Jensen et
68 al., 2012; Liu et al., 2013; Logan, 1985; Logan and Kirchhoff, 1986; Newton et al., 2017;
69 Oltmans et al., 2001; Parrish et al., 2016; Sauvage et al., 2006; Thompson et al., 2012). Spatially-
70 resolved O_3 climatology has been provided by routine sampling by commercial aircraft, but has
71 mostly been limited to the upper troposphere or over continental regions (Clark et al., 2015;
72 Cohen et al., 2018; Logan et al., 2012; Petetin et al., 2016; Sauvage et al., 2006; Thouret et al.,
73 1998; Zbinden et al., 2013), and by satellite observations (Edwards et al., 2003; Fishman et al.,
74 1990, 1991; Hu et al., 2017; Thompson et al., 2017; Wespes et al., 2017; Ziemke et al., 2005,
75 2006, 2017), somewhat tempered by large uncertainties (Tarasick et al., 2019a). Recent overview
76 analyses depict the current understanding of global tropospheric O_3 sources, distribution, and
77 photochemical balance and underscore the insufficiency of observations in the remote free
78 troposphere (Cooper et al., 2014; Gaudel et al., 2018; Tarasick et al., 2019a) necessary to
79 improve the current representation of tropospheric O_3 in global chemical models (Young et al.,
80 2018). Spatial and temporal representativeness of O_3 observations is currently the biggest source
81 of uncertainty when inferring O_3 climatology in the free troposphere, even in regions where
82 observation are abundant but not ideally distributed (Lin et al., 2015a; Tarasick et al., 2019a).
83 The Atmospheric Tomography mission (ATom, https://espo.nasa.gov/atom) was a NASA
84 Earth Venture airborne field project to address the sparseness of atmospheric observations over
85 remote ocean regions by systematically sampling the troposphere over the Pacific and Atlantic
86 basins along a global-scale circuit (Fig. 1). ATom deployed an extensive payload on the NASA
87 DC-8 aircraft, measuring a wide range of chemical, microphysical, and meteorological
88 parameters in repeated vertical profiles from 0.2 km to over 13 km altitude, from the Arctic to
89 the Antarctic over the Pacific and Atlantic Oceans, in four separate seasons from 2016 to 2018.
90 One of the main goals of ATom was to develop an observation-based climatology of the
91 composition of the remote atmosphere using airborne in situ measurements from global-scale
92 sampling flights. ATom built on a previous study, the HIAPER Pole-to-Pole Observations
93 mission (HIPPO, https://www.eol.ucar.edu/field_projects/hippo). The goal of HIPPO was to
94 measure atmospheric distributions of important greenhouse gases and reactive species over the
95 Pacific Ocean, from the surface to the tropopause, five times during different seasons from 2009
96 to 2011. Together, ATom and HIPPO provide recent and comprehensive information about the

altitudinal, latitudinal, and seasonal composition of the remote troposphere over the Pacific, and over the Atlantic for ATom. In addition, ATom and HIPPO sampling strategies were designed to deliver an objective climatology of key species to enable modelling of air parcel reactivity of the remote troposphere (Prather et al., 2017).

Here we use existing ozonesonde and commercial aircraft observations of O_3 at selected locations along the ATom and HIPPO circuits to provide a climatological context for the altitudinal, latitudinal, and seasonal distributions of O_3 derived from the systematic airborne in situ "snapshots". Long-term O_3 observations are obtained from decades of ozonesonde vertical profiles (e.g., Oltmans et al., 2013; Thompson et al., 2017) and from ~60,000 flights using the In-service Aircraft for a Global Observing System (IAGOS) infrastructure (Petzold et al., 2015; http://www.iagos.org). Ozonesondes have typically been launched weekly for two decades or more, depending on the site, and have sampled a wide range of air masses across the globe, from O_3-poor remote surface locations to the O_3-rich stratosphere. IAGOS commercial aircraft have provided daily measurements in the upper troposphere and lower stratosphere (UTLS) for the past 25 years, especially over the northern midlatitudes between America and Europe. Combined, the ozonesonde and IAGOS datasets offer robust measurement-based climatologies that quantify the full expected range of atmospheric O_3 variability with altitude and season. The in-situ data from temporally-limited intensive field studies can be placed in context by comparing them with long-term ozonesonde and commercial aircraft monitoring data. We show that ATom and HIPPO measurements capture the spatial and, in some cases, the temporal dependence of O_3 in the remote atmosphere. Then, we use the geographically extensive ATom and HIPPO vertical profile data to establish a more complete measurement-based benchmark for O_3 abundance and distribution in the remote marine atmosphere.

2. Measurements

2.1 ATom

The four ATom circuits occurred in July–August 2016 (ATom-1), January–February 2017 (ATom-2), September–October 2017 (ATom-3), and April–May 2018 (ATom-4), thus spanning all four seasons in both hemispheres over a two-year timeframe (Table S1). The mission in total consisted of 48 science flights and 548 vertical profiles distributed nearly equally along the global circuit. All four deployments completed roughly the same loop, starting and

ending in Palmdale, California, USA (Fig. 1). A notable addition during ATom-3 and -4 were out-and-back flights from Punta Arenas, Chile to sample the Antarctic troposphere and UTLS.

O_3 was measured using the National Oceanic and Atmospheric Administration (NOAA) nitrogen oxides and ozone (NO_yO_3) instrument. The O_3 channel of the NO_yO_3 instrument is based on the gas-phase chemiluminescence (CL) detection of ambient O_3 with pure NO added as a reagent gas (Ridley et al., 1992; Stedman et al., 1972). Ambient air is continuously sampled from a pressure-building ducted aircraft inlet into the NO_yO_3 instrument at a typical flow rate of 1025.0 ± 0.2 standard cubic centimeters per minute (sccm) in flight. Pure NO reagent gas flow delivered at 3.450 ± 0.006 sccm is mixed with sampled air in a pressure (8.00 ± 0.08 Torr) and temperature (24.96 ± 0.01 °C) controlled reaction vessel. NO-induced CL is detected with a dry-ice-cooled, red-sensitive photomultiplier tube and the amplified digitized signal recorded using an 80 MHz counter; pulse coincidence corrections at high count rates were applied, but are negligible for the data presented in this work. The instrument sensitivity for measuring O_3 under these conditions is 3150 ± 80 counts per second per part per billion by volume (ppbv) averaged over the entire ATom circuit. CL detector calibrations were routinely performed both on the ground and during flight by standard addition of O_3 produced by irradiating ultrapure air with 185 nm UV light and independently measured using UV optical absorption at 254 nm. All O_3 measurements were taken at a temporal resolution of 10 Hz, averaged to 1 Hz, and corrected for the dependence of instrument sensitivity on ambient water vapor content (Ridley et al., 1992). Under these conditions the total estimated 1 Hz uncertainty at sea level is ± (0.015 ppbv + 2 %).

A commercial dual-beam photometer (2B Technologies model 211) based on UV optical absorption at 254 nm also measured O_3 on ATom, with an estimated uncertainty of ± (1.5 ppbv + 1 %) at a 2-second sampling resolution. Comparison of the 2B absorption instrument O_3 data to the NO_yO_3 CL instrument O_3 data agreed to within combined instrumental uncertainties, lending additional confidence to the NO_yO_3 CL instrument calibration. For the ATom project we use NO_yO_3 instrument O_3 data in the following analyses.

Data from two CO measurements were combined in this analysis. The Harvard quantum cascade laser spectrometer (QCLS) instrument used a pulsed quantum cascade laser tuned at ~2160 cm^{-1} to measure the absorption of CO through an astigmatic multi-pass sample cell with 76 m path length and detection using a liquid-nitrogen-cooled HgCdTe detector (Santoni et al., 2014). In-flight calibrations were conducted with gases traceable to the NOAA World

159 Meteorological Organization (WMO) X2014A scale, and the QCLS observations have an
160 accuracy and precision of 3.5 and 0.15 ppb for 1 Hz data, respectively. CO was also measured by
161 the NOAA cavity ring-down spectrometer (CRDS, Picarro, Inc., model G2401-m) in the 1.57
162 μm region with a total uncertainty of 5.0 ppbv for 1 Hz data (Karion et al., 2013). The NOAA
163 Picarro was also calibrated to the NOAA CO-X2014A scale. The combined CO data (CO-X)
164 used here corresponds to the QCLS data, with the Picarro measurement used to fill calibration
165 gaps in the QCLS time series.
166 Water (H_2O) vapor was measured using the NASA Langley Diode Laser Hygrometer
167 (DLH), an open-path infrared absorption spectrometer that uses a laser locked to a water vapor
168 absorption feature at ~1.395 μm. Raw data are processed at the instrument's native ~100 Hz
169 acquisition rate and averaged to 1 Hz with an overall measurement accuracy within 5 %.
170
171 2.2 HIPPO
172 The HIPPO mission consisted of five seasonal deployments over the Pacific basin
173 between 2009 and 2011, from the North Pole to the coastal waters of Antarctica (Wofsy, 2011).
174 HIPPO deployments consisted of two transects, southbound and northbound, and occurred in
175 January 2009 (HIPPO-1), October–November 2009 (HIPPO-2), March–April 2010 (HIPPO-3),
176 June–July 2011 (HIPPO-4) and August–September 2011 (HIPPO-5). The platform used was the
177 NSF Gulfstream V (GV) aircraft. More details can be found in Table S1.
178 A NOAA custom-built dual-beam photometer based on UV optical absorption at 254 nm
179 was used to measure O_3 (Proffitt and McLaughlin, 1983). The uncertainty of the 1 Hz O_3 data is
180 estimated to be ± (1 ppbv + 5 %) for 1 Hz data. A commercial dual-beam O_3 photometer (2B
181 Technologies model 205) based on UV optical absorption at 254 nm was also included in the
182 HIPPO payload. Comparison of the 2B O_3 data to the NOAA O_3 data showed general agreement
183 within combined instrument uncertainties on level flight legs. For the HIPPO project we use
184 NOAA O_3 data in the following analyses.
185 Data from two CO measurements were combined in this analysis. The QCLS instrument
186 was the same instrument as used during ATom and described in section 2.1. CO was also
187 measured by an Aero-Laser AL5002 instrument using vacuum UV resonance fluorescence (in
188 the 170–200 nm range) instrument with an uncertainty of ± (2 ppbv + 3 %) at a 2-second

189 sampling resolution. The combined CO data (CO-X) used here corresponds to the QCLS data,
190 with the Aero-Laser measurement used to fill calibration gaps in the QCLS time series.
191
192 2.3 IAGOS
193 IAGOS is a European Research Infrastructure that provides airborne in situ chemical,
194 aerosol, and meteorological measurements using commercial aircraft (Petzold et al., 2015). The
195 IAGOS Research Infrastructure includes data from both the CARIBIC (Civil Aircraft for the
196 Regular Investigation of the atmosphere Based on an Instrument Container; Brenninkmeijer et
197 al., 2007) and MOZAIC (Measurements of OZone and water vapor by Airbus In-service
198 airCraft; Marenco et al., 1998) programs, providing measurements from ~60,000 flights since
199 1994. We note the relative lack of IAGOS data over the Pacific compared to the Atlantic (shorter
200 temporal record, lower flight frequency, and much fewer flights with concomitant O_3 and CO
201 measurements), and therefore limited the comparison to the Atlantic. Because commercial
202 aircraft cruise altitudes over the ocean are predominantly between 9 and 12 km, the comparison
203 between ATom and IAGOS is further limited to the UTLS (Fig. 1). More details are shown in
204 Table S1.
205 Identical dual-beam UV absorption photometers measured O_3 aboard the IAGOS flights.
206 An instrument comparison demonstrated that the photometers (standard model 49, Thermo
207 Scientific, modified for aircraft use) showed good consistency in measuring O_3 following an
208 inter-comparison experiment (Nédélec et al., 2015). The associated uncertainty is ± (2 ppbv + 2
209 %) at a 4-second sampling resolution (Thouret et al., 1998).
210 CO measurements were made using infra-red absorption photometers (standard model 48
211 Trace Level, Thermo Scientific, modified for aircraft use) with an uncertainty of ± (5 ppbv + 5
212 %) at a 30-second sampling resolution (Nédélec et al., 2003, 2015).
213
214 2.4 Ozonesondes
215 Ozonesondes have measured the vertical distribution of O_3 in the atmosphere for decades,
216 and provide some of the longest tropospheric records that are commonly used to determine
217 regional O_3 trends (Gaudel et al., 2018; Leonard et al., 2017; Oltmans et al., 2001; Tarasick et
218 al., 2019a; Thompson et al., 2017). Ozonesonde launching sites are operated by the NOAA
219 ESRL Global Monitoring Division (GMD), NASA Goddard's Southern Hemisphere Additional

OZonesondes (SHADOZ) program, the New Zealand National Institute of Water & Atmospheric Research (NIWA), the National Meteorological Center of Argentina (SNMA) in collaboration with the Finnish Meteorological Institute (FMI), or Environment and Climate Change Canada. A more detailed description of each ozonesonde site and corresponding dataset can be found in Tables S1 and S2. All sites use electrochemical concentration cell (ECC) ozonesondes that rely on the potassium iodide electrochemical detection of O_3, and which provide a vertical resolution of about 100 m (Komhyr, 1969). The associated uncertainty is usually ± (5–10 %) (Tarasick et al., 2019a; Thompson et al., 2019; Witte et al., 2018).

2.5 Data analysis

In this analysis, ATom flight tracks were divided into the Atlantic and Pacific basins, and further subdivided into five regions within those basins: tropics, and northern and southern middle- and high-latitudes. Vertical profiles presented graphically in this paper show O_3 median values and the 25^{th} to 75^{th} percentile range within the 0–12 km tropospheric column sampled by the DC-8 aircraft. These medians were obtained by averaging with equal weight the individual profiles within each region over 1 km altitude bins.

HIPPO flight tracks are illustrated in Figure 1. The flight segments used for comparison with ATom were binned into the same Pacific latitude and longitude bands as for ATom. HIPPO vertical profile data are derived using the same methodology as for ATom.

All IAGOS flight tracks over the northern and tropical Atlantic are represented in Figure 1 in green. The latitude bands used to parse IAGOS data are consistent with the ones used for ATom. The longitude bands are 50° W to 20° W in the tropics, 50° W to 10° W in the northern midlatitudes, and 110° W to 10° W in the northern high-latitudes. Variations of the longitude band widths do not significantly affect the O_3 distributions measured by IAGOS. Data from all flights from 1994 to 2017 were included in the IAGOS dataset considered here, and were then divided into two altitude bins (8–10 km and 10–12 km) in order to better understand the influence of different O_3 sources (e.g., anthropogenic, stratospheric) on these two layers of the atmosphere.

We compare the ozonesonde measurements to ATom and HIPPO aircraft data sampled within 500 km of each ozonesonde launching site, since we expect a robust correlation in the free troposphere within this distance (Liu et al., 2009). We used the surface coordinates of the

ozonesonde sites because the in-flight coordinates of ozonesondes are not available at all sites. For comparison with ozonesonde long term records, we consider three regions of the atmosphere: boundary layer (0–2 km), free troposphere (2–8 km), and UTLS (8–12 km). For each layer, we compared monthly O_3 distributions from ozonesondes with the corresponding seasonal O_3 distributions from aircraft measurements using the skill score (S_{score}) metric (Perkins et al., 2007). The S_{score} is calculated by summing the minimum probability of two normalized distributions at each bin center, and therefore measures the overlapping area between two probability distribution functions. If the distributions are identical, the skill score will equal 100 % (see Fig. S1 for further examples). Note the S_{score} is positively correlated with the size of the bin used to compare distributions. Here we chose a bin size of 5 ppbv, which is larger than the combined precision of ATom, HIPPO, and IAGOS measurements, but small enough to separate distinct air masses and their influence on O_3 distribution. Variables such as the distance to each ozonesonde launching site (500 km in this study), the bin size of the O_3 distributions (5 ppbv in this study), and the length of each ozonesonde record (full length in this study) can shift the vertically-averaged S_{score} value by up to 8 % (Table S3). We therefore treat this 8 % as a rough estimate of the precision of the S_{score} values presented here.

All three techniques (chemiluminescence, UV absorption, and ECC) used to measure O_3 for the datasets analyzed in this work have been shown to provide directly-comparable accurate measurements with well-defined uncertainties (Tarasick et al., 2019a).

2.6 Back trajectory analysis

Analysis of back trajectories for air masses sampled during airborne missions is useful to examine the air mass source regions and causes for O_3 variability over the Pacific and Atlantic Oceans. We calculated ten-day back trajectories using the Traj3D model (Bowman, 1993; Bowman and Carrie, 2002) and National Centers for Environmental Prediction (NCEP) global forecast system (GFS) meteorology. Trajectories were initialized each minute along all of the ATom flight tracks.

3. Comparison of ATom and HIPPO O_3 distributions to longer-term observational records

281 Here we use existing ozonesonde and IAGOS observations of O_3 at selected locations
282 along the ATom and HIPPO circuits to provide a climatological context for O_3 distributions
283 derived from the systematic airborne in situ "snapshots". We quantify how much of O_3
284 variability, occurring on timescales ranging from hours to decades, was captured by the
285 temporally-limited HIPPO and ATom missions.
286
287 3.1. Comparison to ozonesondes
288 ATom and HIPPO explored the fidelity with which airborne missions represent O_3
289 climatology in the remote troposphere. Here, we show that aircraft-measured median O_3 follows
290 the seasonal ozonesonde-measured median O_3 cycle at most of the sites studied here, and at
291 almost all altitudes – with a few exceptions (Figs. 2 and 3). Figure 2 plots the monthly median O_3
292 measurements from the tropical ozonesonde sites in three altitude bins, along with the median
293 values obtained from HIPPO and ATom measurements. Figure 3 plots the same for the
294 extratropical sites. Figure 4 correlates the median O_3 measured by aircraft in Figures 2 and 3
295 with those measured by ozonesondes. At the Eureka site, the winter and spring ATom
296 deployments recorded a significantly lower median O_3 compared to the corresponding
297 ozonesonde monthly median O_3 in the 0–2 km range (Fig. 3). Eureka is frequently subject to
298 springtime O_3 depletion events at the surface due to atmospheric bromine chemistry, which is
299 well recorded by the ozonesonde record (Fig. 3; Tarasick and Bottenheim, 2002). Sampling
300 during O_3 depletion events significantly lowered the ATom winter and springtime O_3
301 distributions near this site. In the 2–8 km range, there is a very good seasonal agreement between
302 ATom/HIPPO and the ozonesondes (Fig. 4b). Most seasonal differences are found above 8 km
303 (e.g., ATom in February at Trinidad Head and in May at Eureka; Fig. 3) and can be linked to the
304 occurrence – or absence – of stratospheric air sampling during ATom and HIPPO. However, it is
305 straightforward to remove stratospheric airmasses from airborne data using filters based on
306 meteorology (potential vorticity) or composition (H_2O/O_3) (e.g., Cohen et al., 2018). In the
307 absence of stratospheric air mixing (< 8 km in Fig. 4), ATom/HIPPO successfully capture a large
308 fraction of O_3 climatology everywhere (Figs. 4b and 4c).
309
310 Figures 5 and 6 show vertical profiles of O_3 distributions by season at each ozonesonde
311 site, along with comparisons to HIPPO and ATom vertical profiles. Our analysis reveals that O_3

distributions derived from the ATom and HIPPO seasonal "snapshots" capture 30–71 % of the 1 km-vertically binned O_3 distribution established by long-term ozonesonde climatologies. For the nine ozonesonde sites considered here, ATom and HIPPO captured on average 53 %, 54 %, and 38 % of the O_3 distribution in the 0–2 km, 2–8 km, and 8–12 km altitude bins, respectively.

Larger differences between ATom/HIPPO and the ozonesonde records in the UTLS (8–12 km) can be ascribed to O_3 variability from stratospheric–tropospheric exchanges, which are not always captured by the ATom and HIPPO missions. This increased O_3 variability in the UTLS is well-described by the long term ozonesonde records at Lauder, Trinidad Head, Eureka, Ushuaia, and Marambio (Figs. 3 and 6). In these middle- and high-latitude locations in both hemispheres, O_3 variability is especially pronounced during winter and spring, time periods favorable to more frequent stratospheric air mixing (Greenslade et al., 2017; Lin et al., 2015b; Tarasick et al., 2019b). Furthermore, the probability of sampling stratospheric air masses at ATom and HIPPO ceiling altitude (12–14 km) increases with latitude, resulting in a lower S_{score} between the ATom/HIPPO and ozonesonde datasets at the extra-tropical sites than at the tropical sites (Figs. S2a and S2b).

In the boundary layer (0–2 km) of the remote troposphere, O_3 variability is predominantly impacted by loss mechanisms. Ozonesonde records show instances of O_3 mixing ratios lower than 10 ppbv throughout the year in the boundary layer at the nine sites studied here (Figs. 2 and 3). The lowest O_3 mixing ratios are a result of (a) photochemical destruction over the oceans in the tropics (Monks et al., 1998, 2000; Thompson et al., 1993), (b) O_3-destroying halogen emissions in polar regions in springtime (e.g., Fan and Jacob, 1992), and (c) transport of O_3-poor oceanic air over the midlatitude sites (e.g., Neuman et al., 2012).

ATom and HIPPO best describe the O_3 distribution in the free troposphere (2–8 km; Figs. S2a and S2b). This suggests that airborne campaigns can capture global baseline O_3 values, along with the long-range transport of O_3 pollution plumes often lofted to this altitude range and responsible for O_3 variability.

While ATom consisted of one transect per ocean per season, HIPPO covered the Pacific twice per seasonal deployment (southbound and northbound). The 1 km-binned S_{score} is on average higher when two combined seasonal HIPPO flights (southbound and northbound) were available to compare to ozonesonde records, as opposed to when comparing O_3 profiles from

individual HIPPO transects with ozonesonde records (Fig. S2c). In addition, two seasonal flights during HIPPO reduced the occurrence of low S_{score} values. This S_{score} decrease from flying only one Pacific transect only during ATom was traded for the increase of vertical profiles over the Atlantic Basin, which were not sampled during HIPPO. Future airborne missions with multiple seasonal vertical profiles over large-scale regions would be ideal to better depict the full range of tropospheric O_3 variability.

3.2. Comparison to IAGOS

IAGOS O_3 and CO observations in the northern Atlantic UTLS provide a measurement-based climatology at commercial aircraft cruise altitudes for comparison to ATom. Simultaneous measurements of O_3 and CO are of particular interest because CO provides a long-lived tracer of continental emissions, which helps to differentiate O_3 sources (Cohen et al., 2018). We note that while IAGOS measurements encompass hundreds of seasonal flights (depending on the region), ATom sampled within each latitude band and season on one or two flights only (Fig. 1). Thus, variability in the UT that occurred on timescales longer than a day were not captured by ATom. Consequently, it is not surprising to see that ATom systematically under-sampled tropospheric O_3 (and CO) variability compared to IAGOS at all latitudes in the northern Atlantic (Figs. 7 and 8). ATom captured on average 40 % of the O_3 variability measured by IAGOS in the Atlantic UTLS (Fig. 7), on par with the S_{score} of 38 % obtained when comparing ATom and HIPPO to ozonesonde data (see section 3.1).

In the middle- and high-latitudes, the shapes of the O_3 vs. CO scatterplots from IAGOS data demonstrate that distinct sources contribute to O_3 levels in the UTLS (Figs. 8a and 8b; Gaudel et al., 2015). The high O_3 (>150 ppbv) – low CO (<100 ppbv) range corresponds to intrusions of stratospheric air, which were mostly sampled in the spring season during ATom, supporting previous observations of increased stratospheric air mixing during this season (Lin et al., 2015b; Tarasick et al., 2019b). The low O_3 (<50 ppbv) – low CO (<100 ppbv) range corresponds to the tropospheric baseline air, whereas the intermediate O_3 (50–120 ppbv) – high CO (>100 ppbv) range generally represents the influence of air masses transported from continental regions. During ATom, high O_3 and low CO in the middle- and high-latitude UTLS were typical of stratospheric and baseline tropospheric air mixing.

374

375 O_3 measured during IAGOS rarely exceeds 150 ppbv in the northern tropical Atlantic
376 UTLS (Fig. 8c). This is expected because the tropical tropopause is typically situated between 13
377 and 17 km altitude and IAGOS flights typically cruise below 12 km. Therefore, instances of
378 stratospheric intrusions at IAGOS flight altitudes are limited. O_3 measured during ATom in the
379 tropical Atlantic above 8 km was generally positively correlated with CO, showing the
380 contribution of tropospheric O_3 production from continental sources reaching high altitudes.
381 Given this variability, the ATom data do not capture the extrema of UTLS O_3 variability in the
382 IAGOS measurements (Figs. 7 and 8). However, the most frequently measured O_3 and CO
383 values from ATom overlap with the most frequently measured O_3 and CO values from IAGOS
384 (contours in Fig. 8), suggesting that ATom captured the mode of the O_3 and CO distributions
385 from IAGOS in the northern Atlantic UTLS.
386

387 **4. O_3 distributions in the remote troposphere from ATom and HIPPO**

388 We have established the fidelity of ATom and HIPPO O_3 data by comparison to
389 measurement-based climatologies of tropospheric O_3 from well-established ozonesonde and
390 commercial aircraft monitoring programs. In the following sections we exploit the systematic
391 nature of the ATom and HIPPO vertical profiles to provide a global-scale picture of tropospheric
392 O_3 distributions in the remote atmosphere. Figure 9 presents the altitudinal, latitudinal, and
393 seasonal distribution of tropospheric O_3 during ATom and HIPPO. Higher O_3 was measured
394 during ATom & HIPPO in the Northern Hemisphere (NH) than in the Southern Hemisphere
395 (SH), both in the Pacific and in the Atlantic. This distribution gradient has previously been
396 shown by global O_3 mapping from modeling, satellite, and ozonesonde analyses (e.g., Hu et al.,
397 2017; Liu et al., 2013). This finding holds true throughout the tropospheric column from 0 to 8
398 km, both in the middle- and high-latitudes (Fig. S3). In the midlatitudes below 8 km, median O_3
399 ranged between 25 and 45 ppbv in the SH, and between 35 and 65 ppbv in the NH. In the high
400 latitudes below 8 km, median O_3 ranged between 30 and 45 ppbv in the SH, and between 40 and
401 75 ppbv in the NH. Notable features in the global O_3 distribution are discussed in more detail in
402 the following sections. Figure 10 presents the vertically-resolved distribution of tropospheric O_3
403 from 0–12 km for the Atlantic (ATom in green) and for the pacific (ATom in pink, HIPPO in
404 blue). S_{score} values resulting from the comparison of HIPPO and ATom Pacific distributions are

405 shown with blue diamonds, and from the comparison of ATom Atlantic and Pacific distributions
406 with pink squares. Figure 11 is derived from Figure 10 and gives the S_{score} values against altitude
407 in the first panel, as well as the relative difference of median O_3 from 0 to 8 km in the second
408 panel.
409
410 4.1. Tropics
411 **Vertical distribution.** O_3 is at a minimum in the tropical marine boundary layer (MBL),
412 especially over the Pacific (Fig. 10a). The lowest measured O_3 in this region was 5.4 ppbv in
413 May during ATom, and 3.5 ppbv in January during HIPPO. The tropical MBL is a net O_3 sink
414 owing to very slow O_3 production rates – NO levels averaged 22 ± 12 pptv in the Pacific and
415 Atlantic MBL during ATom – and rapid photochemical destruction rates of O_3 in a sunny, humid
416 environment (Kley et al., 1996; Parrish et al., 2016; Thompson et al., 1993). Deep stratospheric
417 intrusions into the Pacific MBL were not observed in ATom or HIPPO, in contrast to reports
418 from previous studies (e.g., Cooper et al., 2005; Nath et al., 2016). In the tropics, marine
419 convection within the intertropical convergence zone (ITCZ) is associated with relatively low O_3
420 values throughout the tropospheric column, with median O_3 mixing ratios less than 25 ppbv
421 below 4 km altitude in the tropical Pacific (Fig. 10a; Oltmans et al., 2001). The relative
422 difference between ATom Atlantic and Pacific median O_3 in the tropics below 8 km is
423 consistently higher than a factor of 1.5, with an average S_{score} of 43 % (Figs. 10a and 11b). We
424 ascribe this difference to O_3 production from biomass burning (BB) emissions in the continental
425 regions surrounding the tropical Atlantic; back trajectories from the ATom flight tracks show the
426 tropical Atlantic is strongly affected by transport from BB source regions in both Africa and
427 South America (Fig. S4; Jensen et al., 2012; Sauvage et al., 2006; Stauffer et al., 2018;
428 Thompson et al., 2000). In addition, the positive correlation of O_3 enhancements with black
429 carbon (Katich et al., 2018) and reactive nitrogen species (Thompson et al., personal
430 communication) also indicate BB influence. Although ATom and HIPPO data show evidence for
431 extensive and widespread BB influence on O_3 in the Pacific as well, O_3 mixing ratios are
432 consistently more elevated throughout the tropospheric column in the Atlantic. One reason is
433 closer proximity of the mid-ocean Atlantic flight tracks to O_3 precursor source regions. These
434 findings confirm studies that previously highlighted the impact of African BB emissions on O_3
435 production in the tropical Atlantic (e.g., Andreae et al., 1994; Fishman et al., 1996; Jourdain et

al., 2007; Williams et al., 2010). Lightning NO_x also play a role in the buildup of O_3 over the tropical Atlantic at certain times of year (Moxim and Levy, 2000; Pickering et al., 1996).

Seasonality. The seasonal variation of vertical profiles of O_3 in the tropics is lower throughout the column compared to the extra-tropics (Fig. 12), in part due to less stratospheric influence at the highest tropical altitudes. The remoteness of the tropical Pacific flight paths from continental pollution sources also drives the lower seasonal variability here compared to the tropical Atlantic, where BB influence peaks in June–August and October–November, characterized by high O_3 (> 75 ppbv) and high CO (>100 ppbv) (Fig. 13f), significantly increases the O_3 vertical distribution compared to the other seasons (Figs. 12c, 12h, and 12m). Finally, photochemistry, which regulates O_3 net balance in the troposphere, is less seasonally variable in the tropics than in the extra-tropics, where the photolysis frequency of O_3 ($j(O_3)$) and photochemical production of O_3 fluctuate annually with solar zenith angle.

O_3 minima and maxima. Coincident O_3 and CO enhancements were observed in the tropical Atlantic for each ATom circuit (Figs. 9 and 13f), suggesting a year-round influence of continental emissions and distinctive dynamics in this region (Krishnamurti et al., 1996; Thompson et al., 1996). In the tropical Pacific, the April–May period stands out due to an O_3 and CO enhancement episode during HIPPO (Fig. 9) that was attributed to the transport of anthropogenic and BB emissions from southeast Asia (Shen et al., 2014).

Deep convection in the tropics brings O_3-poor (<15 ppbv) air to the upper troposphere (Kley et al., 1996; Pan et al., 2015; Solomon et al., 2005). However, the spatial extent of these events remains poorly constrained. Results from ATom and HIPPO suggest that deep convection can loft O_3-poor air at least up to 12 km (the altitude ceiling of this study) in the tropical Pacific, and occurred more frequently between January and May (Figs. 12c and h). During the rest of the year, O_3-poor air was typically confined below 4 km. Conversely, O_3-poor air is confined to the first 2 km in the tropical Atlantic (Fig. S5). Meteorological analysis of tropical ozonesondes shows that subsidence of higher-O_3 air aloft over the Atlantic is one reason O_3-poor air is found only in the boundary layer (Thompson et al., 2000, 2012).

4.2. Middle- and high-latitudes

Vertical distribution. In the middle- and high-latitudes, tropospheric O_3 was generally at a minimum in the MBL and increased with altitude. Above 8 km, increasing O_3 with altitude

(Figs. 10b–e) and its persistent anticorrelation with CO (Fig. 13) points to stratospheric air sampling as the cause for higher O_3 variability in the extra-tropical UTLS, especially at high latitudes where the tropopause is lower and wave breaking of the polar jet streams can lead to stratospheric intrusions. As a result, the S_{score} decrease above 8 km, summarized in Figure 11a, is ascribed to variability in the influence of stratospheric air. ATom detected little change in the O_3 distribution over the Pacific Ocean since HIPPO, with a S_{score} averaging 74 % in the 0–8 km range. The relative difference between median O_3 values from HIPPO and ATom in the Pacific is generally lower than 20 % (Fig. 11b). Similarly, the relative difference between median O_3 mixing ratios between ATom Atlantic and Pacific below 8 km is consistently lower than 20 %, with an average S_{score} of 75 % between (Fig. 11b). The southern high-latitudes are the only region where the S_{score} below 8 km occasionally fell below 60 % (Fig. 10e). However, a lower S_{score} was expected there as the Atlantic vertical profile is based on only two seasonal flights to Antarctica, whereas there were four seasonal flights in the Pacific. Additionally, HIPPO was less spatially extensive – resulting in fewer data points – in this latitude bin compared to ATom (Fig. 1), which could explain the low S_{score} values when comparing the two missions (Fig. 10e). Nevertheless, the similar O_3 distribution in the extra-tropical free troposphere above the two oceans is consistent with an O_3 lifetime sufficiently long for rapid zonal transport to smooth out variations in baseline O_3 distribution in the remote troposphere, across a relatively wide range of longitudes (Figs. 10b–e). The comparison of O_3 seasonal cycles at remote ozonesonde launching sites of the northern midlatitudes yields similar results and further supports this conclusion (Logan, 1985; Parrish et al., 2020). Studies of the spatial representativeness of tropospheric O_3 monitoring networks have concluded that tropospheric O_3 distributions varied significantly with longitude, especially in the northern middle- and high-latitudes over continents (Liu et al., 2013; Tilmes et al., 2012). ATom findings stem from O_3 measurements predominantly over the oceans, possibly yielding a different picture of O_3 longitudinal distribution away from regional precursor emissions.

Seasonality. The extra-tropical vertical profiles of O_3 vary seasonally during ATom and HIPPO. The summer season in the middle- and high-latitudes was remarkable over both oceans and hemispheres for the steep O_3 gradients in the tropospheric column (Fig. 12 in black). In the MBL, median O_3 was consistently under 25 ppbv in the summer, whereas O_3 was over 25 ppbv in other seasons. Low O_3 in the MBL in summer reflects the enhanced O_3 photochemical

498 destruction in this NO_x-limited region. Photochemical destruction decreases in dry air in the
499 upper troposphere, thus leading to the steep O_3 gradients observed here. The summer O_3
500 minimum was especially apparent in the high latitudes of the southern Pacific during ATom and
501 extended well above the MBL into the free troposphere (Fig. 12 in black).
502 O_3 mixing ratios were highest in the tropospheric column during springtime in both
503 hemispheres, and over both oceans (Fig. 12 in gold). A notable exception occurred during
504 springtime in the high latitudes of the NH, where several O_3 depletion events were sampled in
505 the lower legs of the Arctic transit. During these events, O_3 mixing ratios lower than 10 ppbv
506 were measured, resulting in a lower 25th percentile of O_3 distribution at the lowest altitude
507 compared to the other seasons (Fig. 12e in gold). A tropospheric O_3 springtime maximum has
508 often been reported in the NH (e.g., Monks, 2000) when meteorology favors efficient transport
509 of O_3 and precursors from continental air from North America and Eurasia (Owen et al., 2006;
510 Zhang et al., 2017, 2008). Another contributing factor is the increased frequency of stratospheric
511 air mixing in spring that significantly contributes to higher O_3 levels (Lin et al., 2015b; Tarasick
512 et al., 2019b). Further, the tropospheric O_3 springtime maximum in the SH is often attributed to
513 BB emissions reaching a peak ((Fishman et al., 1991; Gaudel et al., 2018), but stratospheric air
514 mixing also occurs (Diab et al., 1996, 2004; Greenslade et al., 2017). Here, the O_3/CO
515 relationship in spring shows that the enhanced stratospheric mixing with tropospheric air during
516 this season, both in the northern and southern middle- and high-latitudes, contributes to the
517 increase in column O_3 (Fig. 13).
518 Fall and winter seasons shared similar features in the middle- and high-latitudes: no
519 strong O_3 gradient was measured in the free troposphere, and O_3 values varied over similar
520 ranges – about 40 ppbv in the NH and about 30 ppbv in the SH – during the two seasons (Fig. 12
521 in red and blue).
522 **O_3 enhancements.** The linear increase of O_3 with CO >100 ppbv highlights the
523 contribution of natural and anthropogenic pollution plumes lofted from continental areas into the
524 remote troposphere.
525 In the NH, these events occur almost year-round (Figs. 13b–c and 13g–h). Higher CO
526 enhancements in the Pacific (Figs. 13g–h) than in the Atlantic (Figs. 13b–c) have been observed
527 before and attributed to sampling bias (Clark et al., 2015). Here, our findings suggest a year-
528 round influence of continental emissions on the Pacific atmosphere despite its remoteness.

Modeled back trajectories show that most air masses sampled in the NH during ATom were influenced by long-range transport of continental emissions from Asia, Africa, and North America (Fig. S6). Previous studies have shown anthropogenic and BB emission outflow from Asia significantly contributed to O_3 pollution events measured over the northern Pacific or in California (e.g., Heald et al., 2003; Jaffe et al., 2004; Lin et al., 2017). Intercontinental transport of anthropogenic emissions from Europe can also contribute to the Asian outflow of anthropogenic pollution (e.g., Bey et al., 2001; Liu et al., 2002; Newell and Evans, 2000). Finally, O_3 enhancements in the northern Atlantic were frequently observed and attributed to midlatitude anthropogenic and boreal forest fire emissions (e.g., Honrath et al., 2004; Martín et al., 2006; Trickl et al., 2003).

In the SH, polluted air is encountered more often in spring and summer over the Atlantic, but springtime CO is greater than in other seasons over the Pacific (Figs. 13d–e and 13i–j). During spring, median O_3 above 50 ppbv was measured throughout the free troposphere in the southern midlatitudes (Fig. 12). Several air masses intercepted during these flights originated from regions that were intensively burning at the time, notably equatorial and southern Africa, Australia, and southern South America, contributing to the observed enhanced O_3 and CO (Fig. S4).

5. Conclusion

We present tropospheric O_3 distributions measured over remote regions of the Pacific and Atlantic Oceans during two airborne chemical sampling projects: the four deployments of ATom (2016–2018) and the five deployments of HIPPO (2009–2011). The data highlight several regional- and large-scale features of O_3 distributions, and provide ~~valuable new~~ insight into current O_3 distributions in remote regions. The main findings are as follows:

- ATom and HIPPO provide a unique perspective on vertically-resolved global baseline O_3 distributions over the Pacific and Atlantic basins, and expand upon spatially-limited O_3 climatologies from long-term datasets to highlight large-scale features necessary for model output and satellite retrieval validation.
- ATom and HIPPO O_3 data are consistent – where they overlap – with measurement-based climatologies of tropospheric O_3 from well-established ozonesonde and commercial aircraft monitoring programs. ATom and HIPPO seasonal median O_3 showed

high correlation ($R^2 > 0.7$) with corresponding seasonal median O_3 from ozonesondes, giving confidence in the accurate depiction of the emerging global O_3 climatology by these diverse research activities. ATom and HIPPO captured 30–71 % of O_3 variability measured by ozonesondes launched in the vicinity of the aircraft flight tracks, and had the same mode of the O_3 distribution as determined by IAGOS in the northern Atlantic UTLS.

- Higher O_3 loading in the NH compared to the SH is consistent with the heterogeneous distribution of O_3 precursor emissions around the globe, mostly concentrated in the NH, a result consistent with previous modeling studies and satellite observations. ATom Atlantic vs. Pacific comparison reveals a similar O_3 distribution in the free troposphere up to ~8 km in the middle- and high-latitudes, but not in the tropics. Similar O_3 distributions across latitude bands have been suggested in the past, but these studies were limited to the northern midlatitudes. Conversely, other observation-based studies indicated significant O_3 longitudinal gradients. Here, our findings are consistent with zonal transport smoothing the baseline O_3 distribution longitudinally from the Pacific to the Atlantic. In the tropics, median O_3 mixing ratios are about twice as high in the Atlantic than in the Pacific, due to a well-documented mixture of dynamical patterns interacting with the transport of continental air masses.
- A comparison of seasonal O_3 vertical profiles did not reveal a marked seasonality in the tropics, but instead highlighted the influence of specific events, most notably BB emissions from Africa and South America, which have been extensively documented in the literature. In the extra-tropics, the summer season was characterized by a steeper tropospheric O_3 gradient driven by very low O_3 abundance in the MBL. Fall and winter seasons generally led to near-constant O_3 mixing ratios from the surface to the upper troposphere, while the highest O_3 abundance was recorded during the spring season when more frequent and intense stratospheric intrusions and transport of air masses from continental regions occur. ATom and HIPPO provide the first airborne in situ vertically-resolved O_3 climatology covering both the Atlantic and Pacific Oceans in the NH and in the SH. They confirm and extend the current understanding of O_3 variability in the remote troposphere, built over several decades by airborne campaigns, monitoring networks, and satellite observations.

- Overall, this paper highlights the value of the ATom and HIPPO datasets, which cover spatial scales commensurate with the grid resolution of current Earth system models, and further useful as a priori estimates for improved retrievals of tropospheric O_3 from satellite remote sensing platforms. In addition, ATom and HIPPO in situ measurements help to establish the quantitative legacy of global pollution transport and chemistry by through the evaluation of key, covarying species – in this case O_3 and CO. ATom and HIPPO datasets should be critical for improving the scientific community's understanding of O_3 production and loss processes, and the influence of anthropogenic emissions on baseline O_3 in remote regions. They provide a timely addition to the Tropospheric Ozone Assessment Report (TOAR) effort to characterize the global-scale O_3 distribution, and address some of the measurement gaps identified therein.

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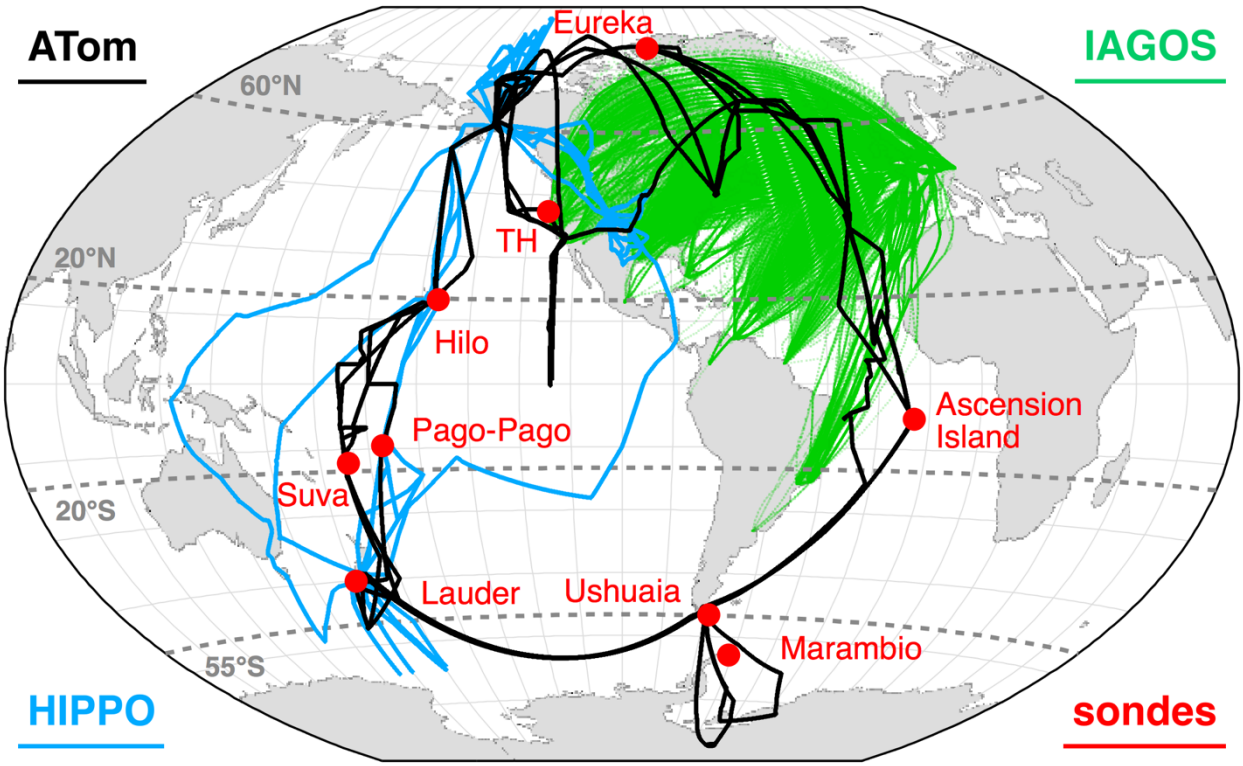

Figure 1 The location and flight tracks of all O_3 monitoring platforms used in this work are illustrated with different markers and colors. The ATom flight track is in black, the HIPPO flight track is in blue, IAGOS flight paths are in green, and the ozonesonde launching sites are indicated by the red markers. The dotted grey lines define the latitudinal bands over which individual ATom and HIPPO profiles were averaged to derive a regional O_3 distribution: the tropics (20° S – 20° N), the midlatitudes (55° S – 20° S; 20° N – 60° N), and the high-latitudes (90° S – 55° S; 60° N – 90° N). Only data from remote oceanic flight segments of ATom and HIPPO missions were used in this work.

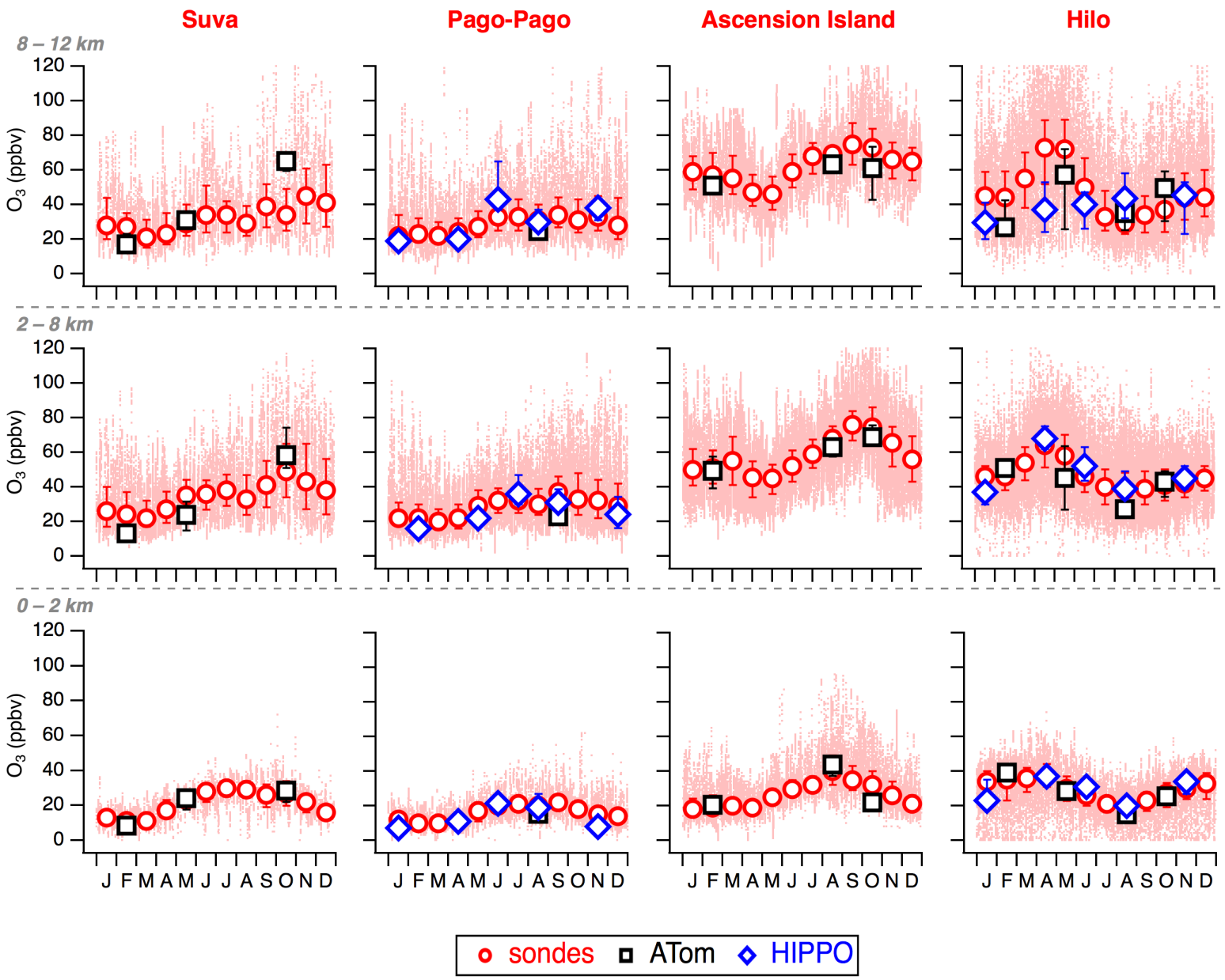
Suva
Pago-Pago
Ascension Island
Hilo
8 – 12 km
2 – 8 km
0 – 2 km
O3 (ppbv)
0
20
40
60
80
100
120
J F M A M J J A S O N D
sondes
ATom
HIPPO

Figure 2 Comparison of ATom (black squares) and HIPPO (blue diamonds) monthly median O_3 with ozonesonde (red circles) records from the four tropical sites. Markers indicate the median and the bars indicate the 25th and 75th percentiles. The three rows, from bottom to top, correspond to the boundary layer (0–2 km), the free troposphere (2–8 km), and the UTLS (8–12 km). The pink dots show every O_3 data point measured by ozonesondes for the timeframes indicated in Table S2.

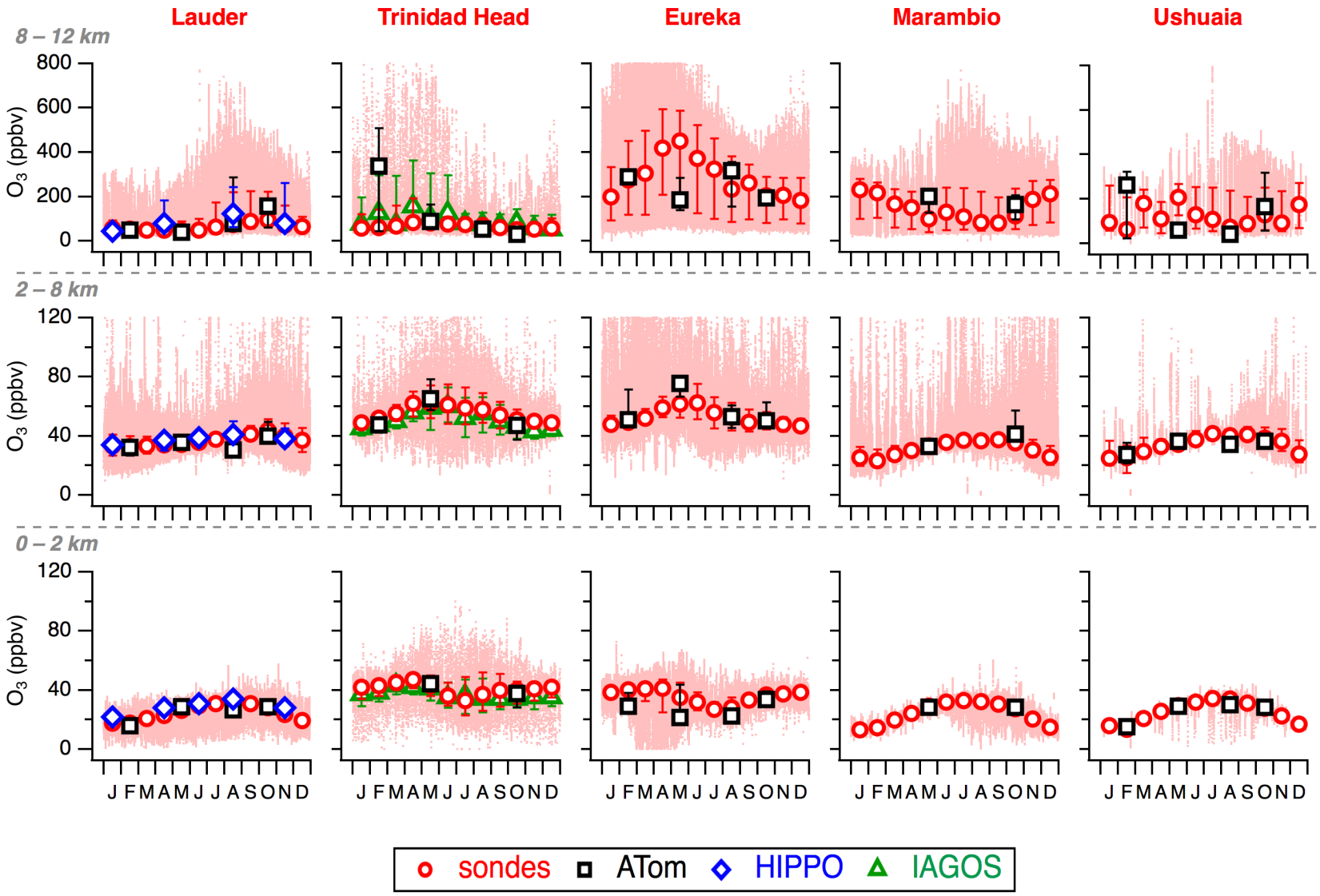
Lauder
Trinidad Head
Eureka
Marambio
Ushuaia
8 – 12 km
2 – 8 km
0 – 2 km
O3 (ppbv)
800
600
400
200
0
120
80
40
0
J F M A M J J A S O N D
sondes
ATom
HIPPO
IAGOS

Figure 3 Same as in Figure 2 but for ozonesonde launching sites located in the middle- and high-latitudes. O_3 data obtained from the IAGOS program (green triangles) during descents into San Francisco Bay-area airports were also added to the Trinidad Head site for comparison.

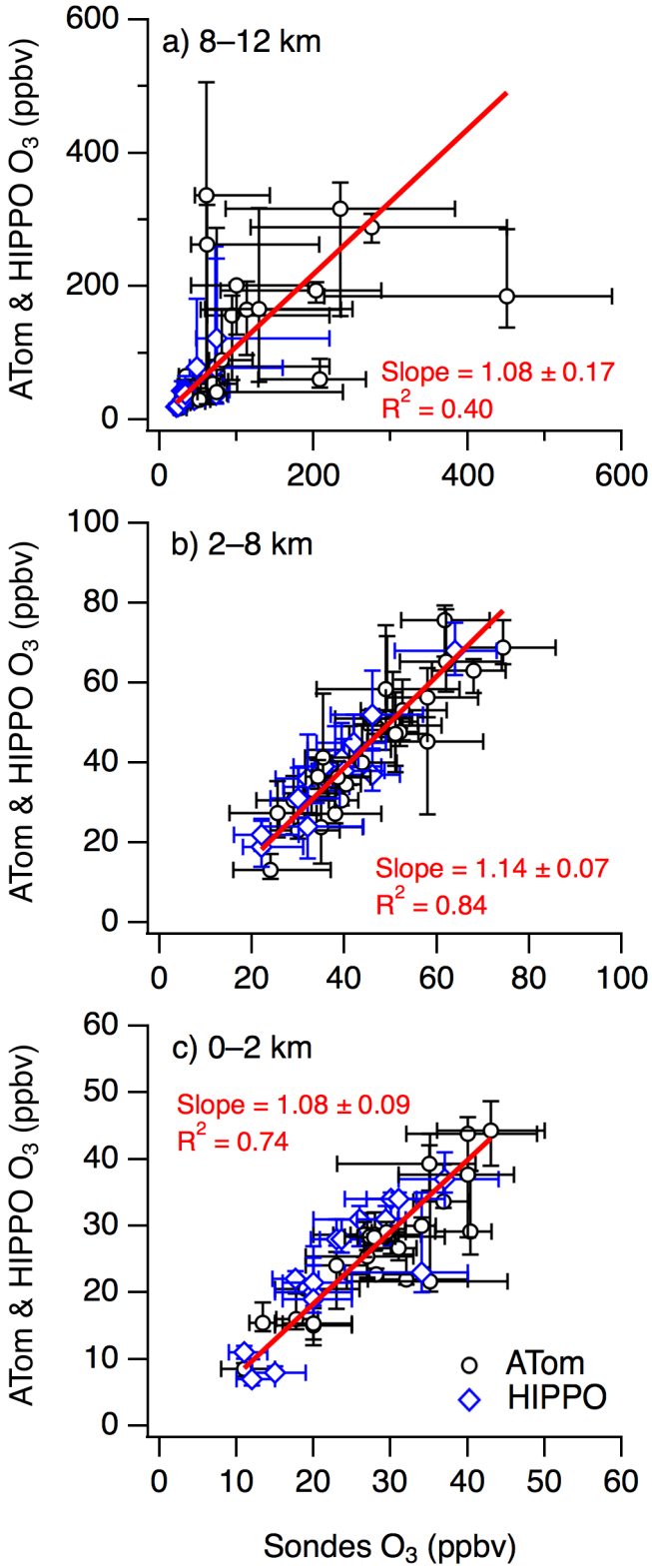

Figure 4 ATom (black circles) and HIPPO (blue diamonds) combined monthly median O_3 vs. monthly median O_3 from ozonesondes at the nine sites considered in this study. The three panels

indicate the correlations for a) the UTLS (8–12 km), b) the free troposphere (2–8 km), and c) the boundary layer (0–2 km). The orthogonal regression fits are two-sided but not weighted.

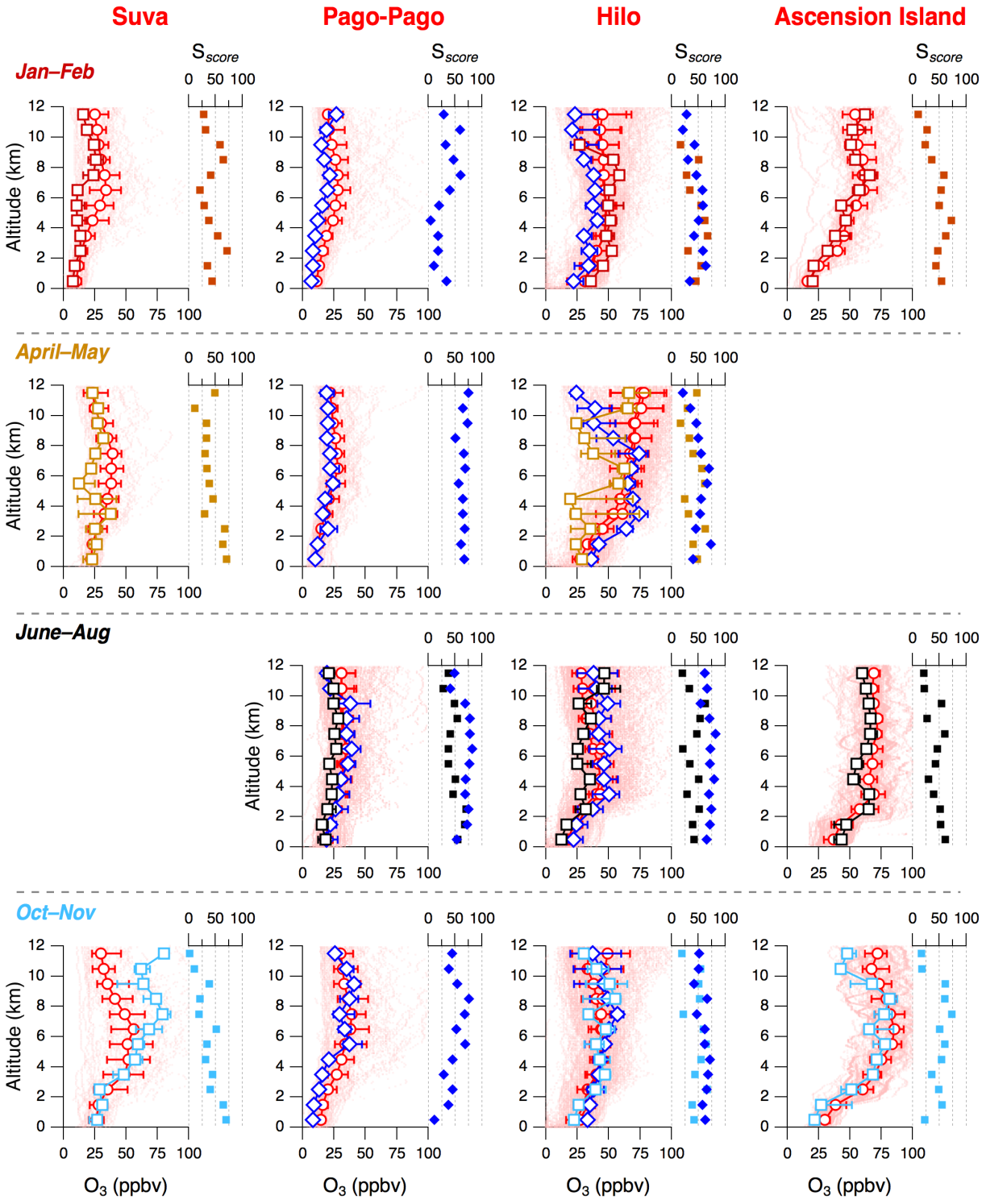

Figure 5 Seasonal comparison of 1 km-vertically-binned ATom (colored squares) and HIPPO (blue diamonds) median O_3 with ozonesonde (red circles) records at four sites in the tropics (Suva in Fiji, Pago-Pago in American Samoa, Hilo in Hawaii, and Ascension Island). Markers indicate the median and the bars are the 25th and 75th percentiles. The S_{score} is a metric of how well ATom

and HIPPO 1 km-binned O_3 probability distribution functions (PDFs) overlap with the corresponding 1 km-binned O_3 PDFs from ozonesondes. The S_{score} shown with squares compares ATom with ozonesondes, and the S_{score} shown with blue diamonds compares HIPPO with ozonesondes. The pink dots show every O_3 data point measured by ozonesondes for the timeframes indicated in Table S2.

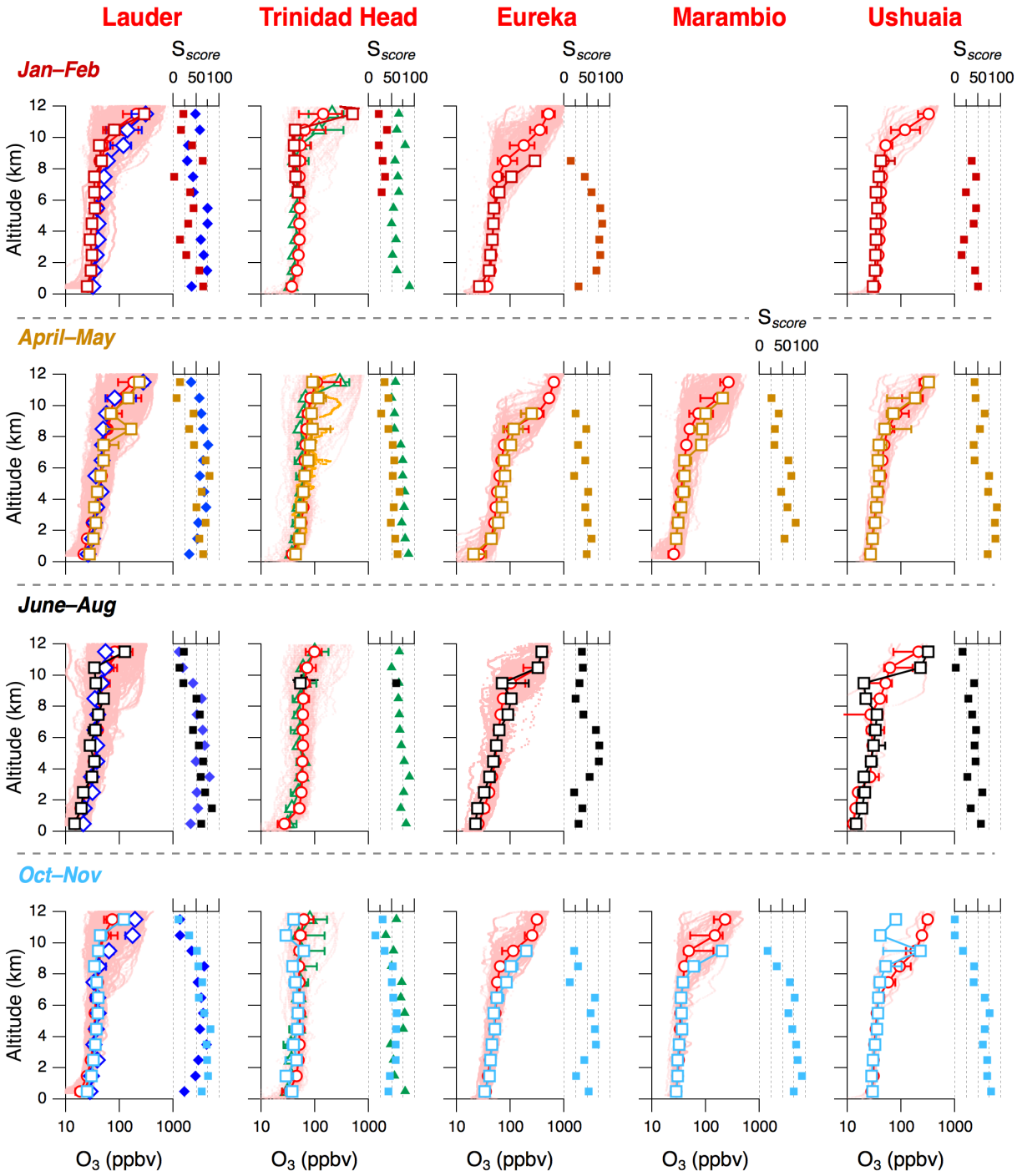

Figure 6 Same as in Figure 5 but for ozonesonde launching sites located in middle- and high-latitudes (Lauder in New Zealand, Trinidad Head in the USA, Eureka in Canada, Ushuaia in Argentina, and Marambio in Antarctica). O_3 data obtained from the IAGOS program (green triangles) during descents into San Francisco Bay-area nearby airports were also added to the Trinidad Head site for comparison.

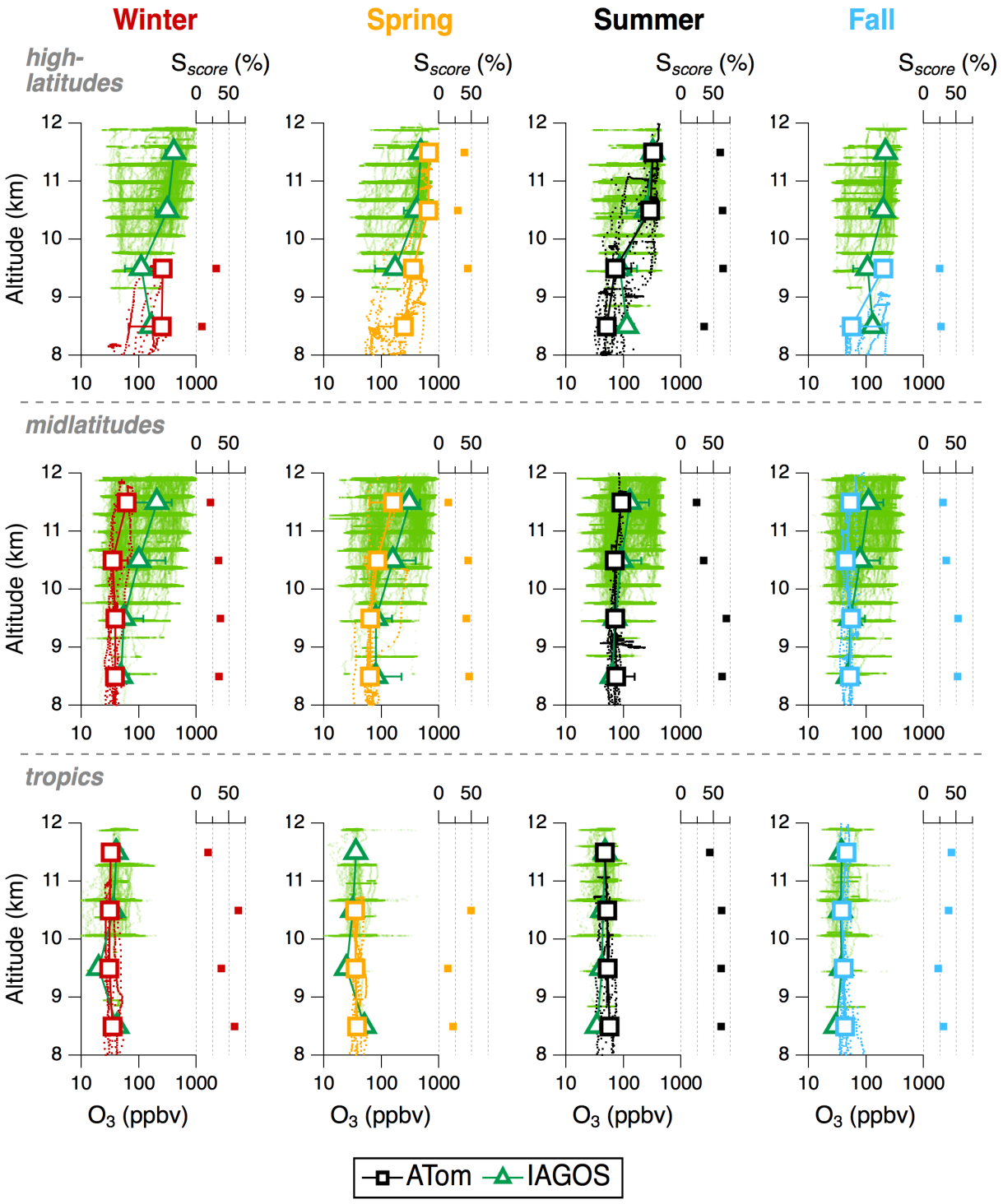

Figure 7 Seasonal comparison of 1 km-binned ATom (colored squares) median O_3 with IAGOS (green triangles) in the northern Atlantic UTLS. Markers indicate the median and the bars are the 25th and 75th percentiles. The three different rows indicate the latitudinal bands. The four columns

indicate the seasons. The green dots show every O_3 data point measured by IAGOS flights for the timeframe indicated in Table S1.

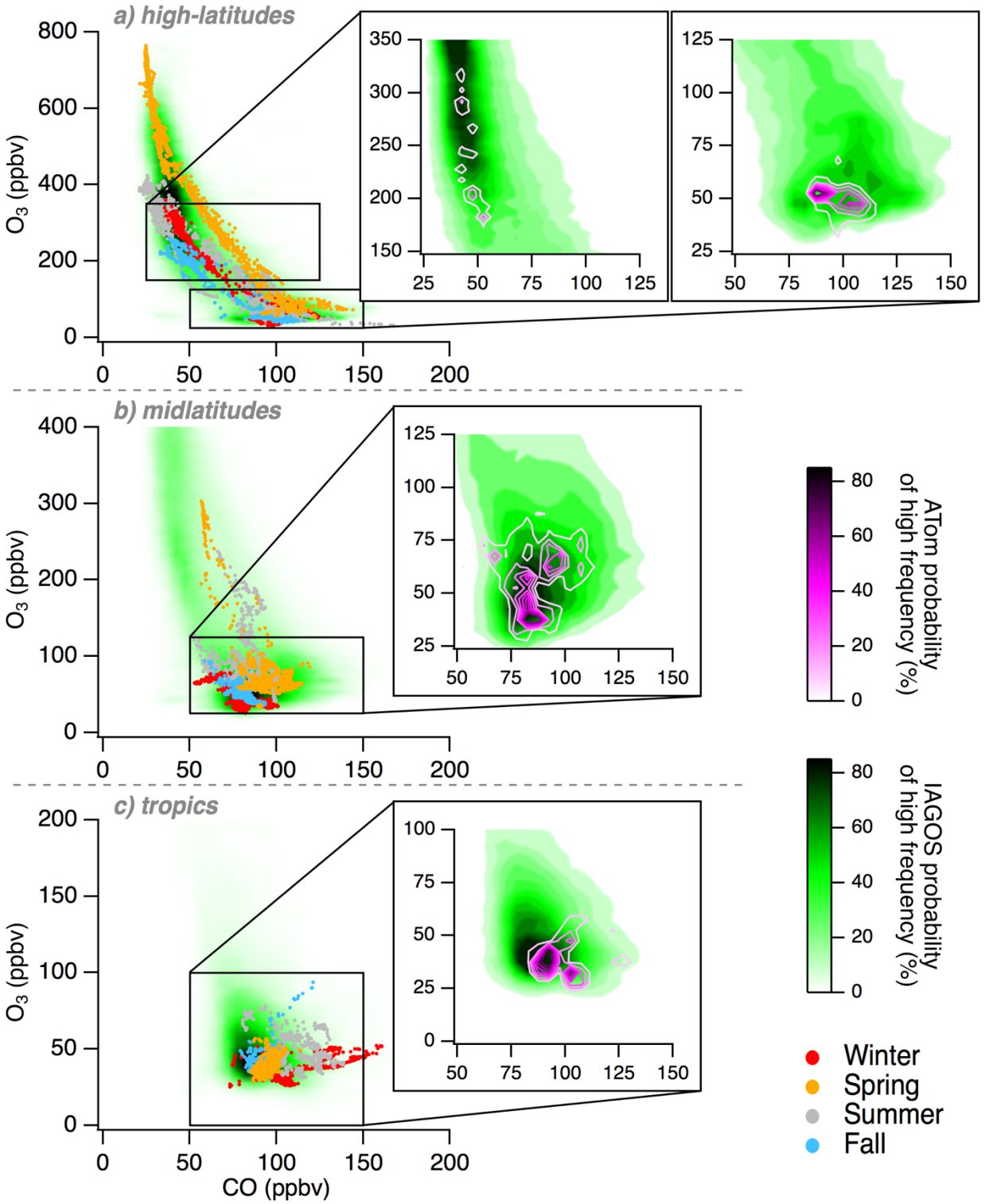

Figure 8 IAGOS and ATom seasonal O_3 vs. CO scatterplots, with insets showing the most frequent O_3 values measured during IAGOS and ATom. ATom seasonal deployments are

colored according to the legend. The frequency gradient of O_3 counts is illustrated by the color scales (green for IAGOS, magenta for ATom). ATom measurements have been combined for the frequency gradients shown in the insets. The probability of high frequency refers to the probability of finding frequently measured O_3 values within the contour boundaries

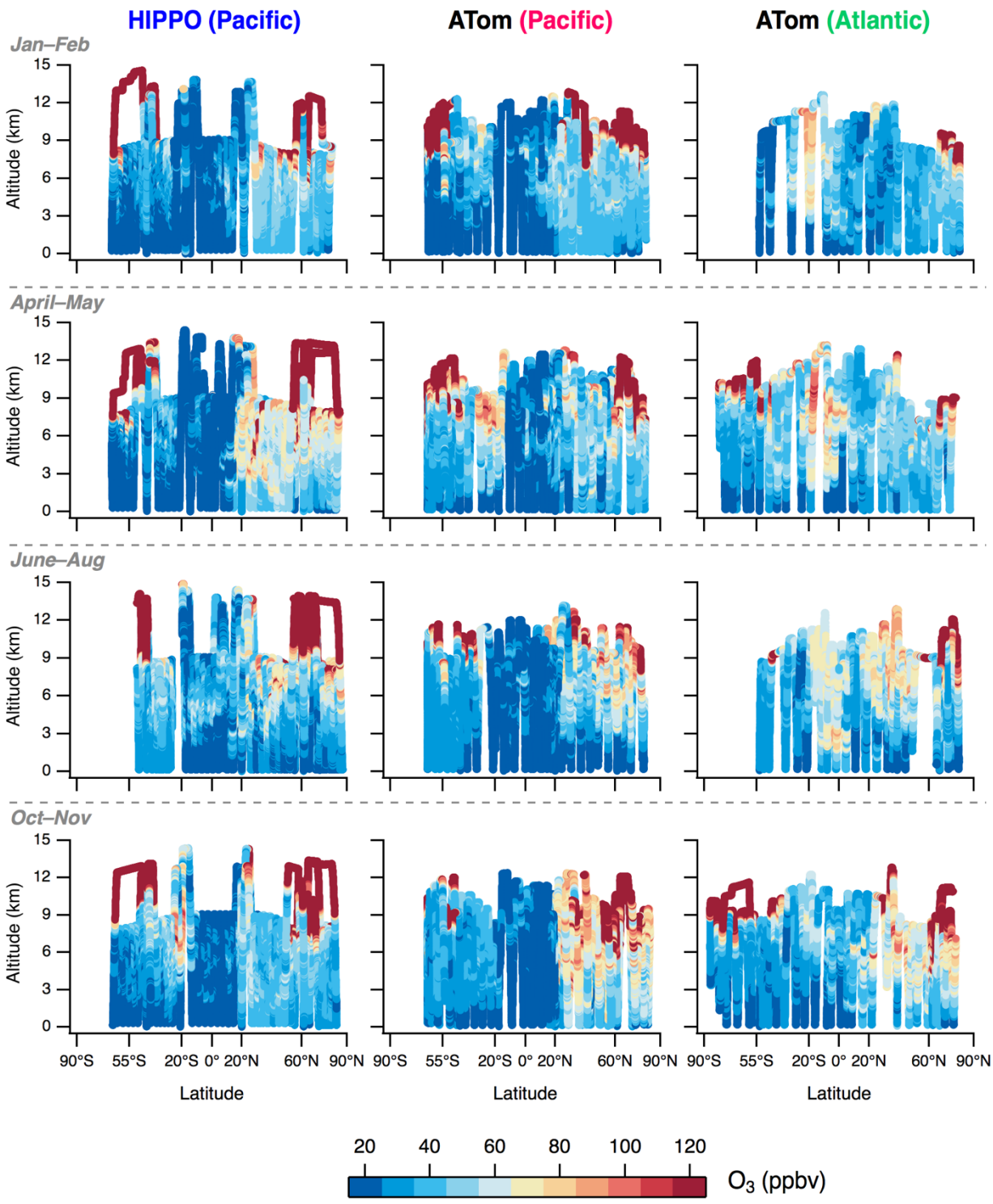

Figure 9 Global-scale distribution of tropospheric O_3 for each ATom and HIPPO seasonal deployment. The rows separate the seasonal deployments, while the columns indicate the mission and the ocean basin. The O_3 color-scale ranges from 20 to 120 ppbv, and all values outside of this

range are shown with the same extremum color (red for values > 120 ppbv, blue for values < 20 ppbv). HIPPO deployments in June and August were combined together.

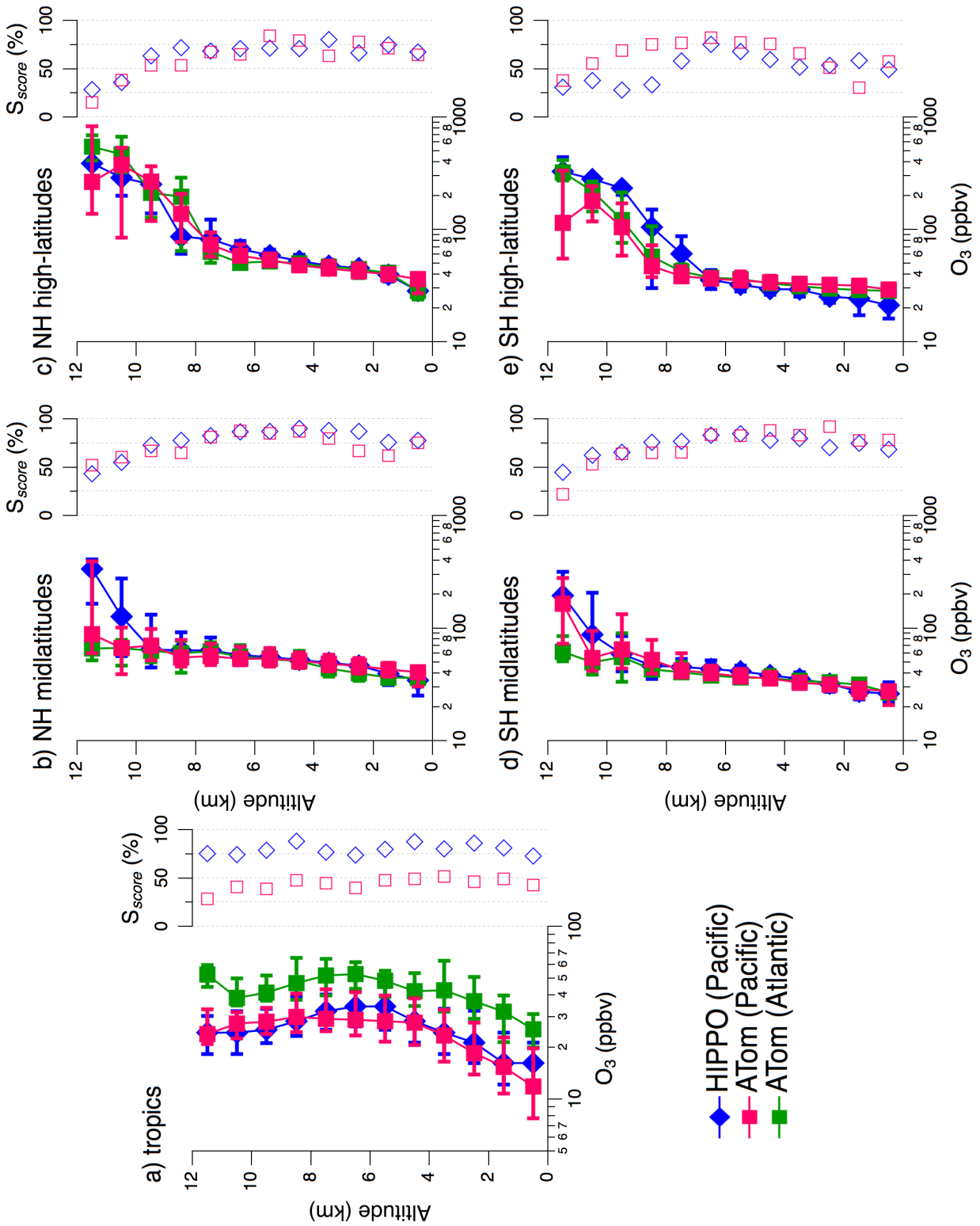

Figure 10 Vertically-resolved O_3 distributions from 0–12 km are plotted for the Atlantic (ATom in green) and for the Pacific (ATom in pink, HIPPO in blue). The five broad latitude regions correspond to the data parsing illustrated by Fig. 1. Markers indicate median O_3, and bars are the

25th and 75th percentiles, per 1 km altitude bin. Note the log scale on the x-axis. S_{score} values resulting from the comparison of HIPPO and ATom Pacific distributions are shown with blue diamonds, and from the comparison of ATom Atlantic and Pacific distributions with pink squares.

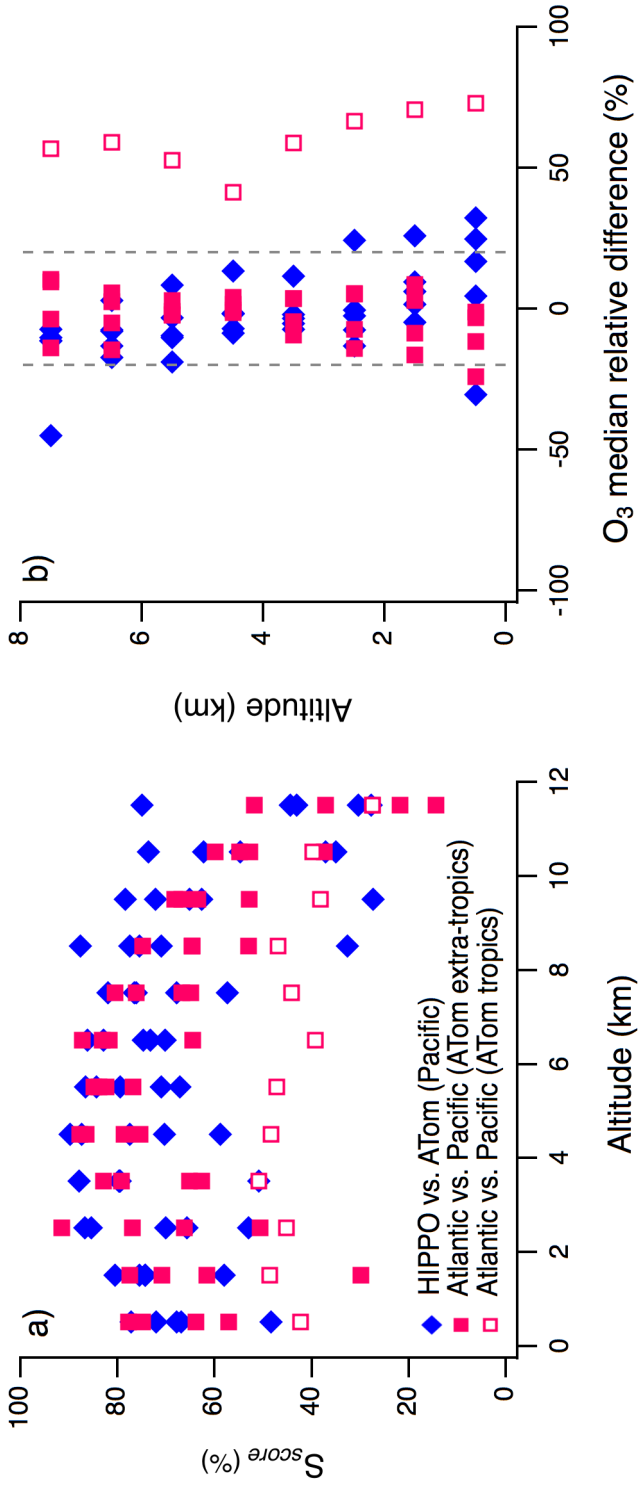

Figure 11 All S_{score} values from Fig. 10 are shown in panel a) and plotted against altitude. The HIPPO and ATom comparison in the Pacific basin is shown with blue diamonds, and a comparison of the Atlantic and Pacific basins during ATom is shown with filled pink squares for the extra-

tropics and open pink squares for the tropics. The relative difference of median O_3 from 0 to 8 km given in Fig. 10 is shown in panel b), with the same color and marker code as in panel a). The dotted grey lines indicate a relative difference of 20 %.

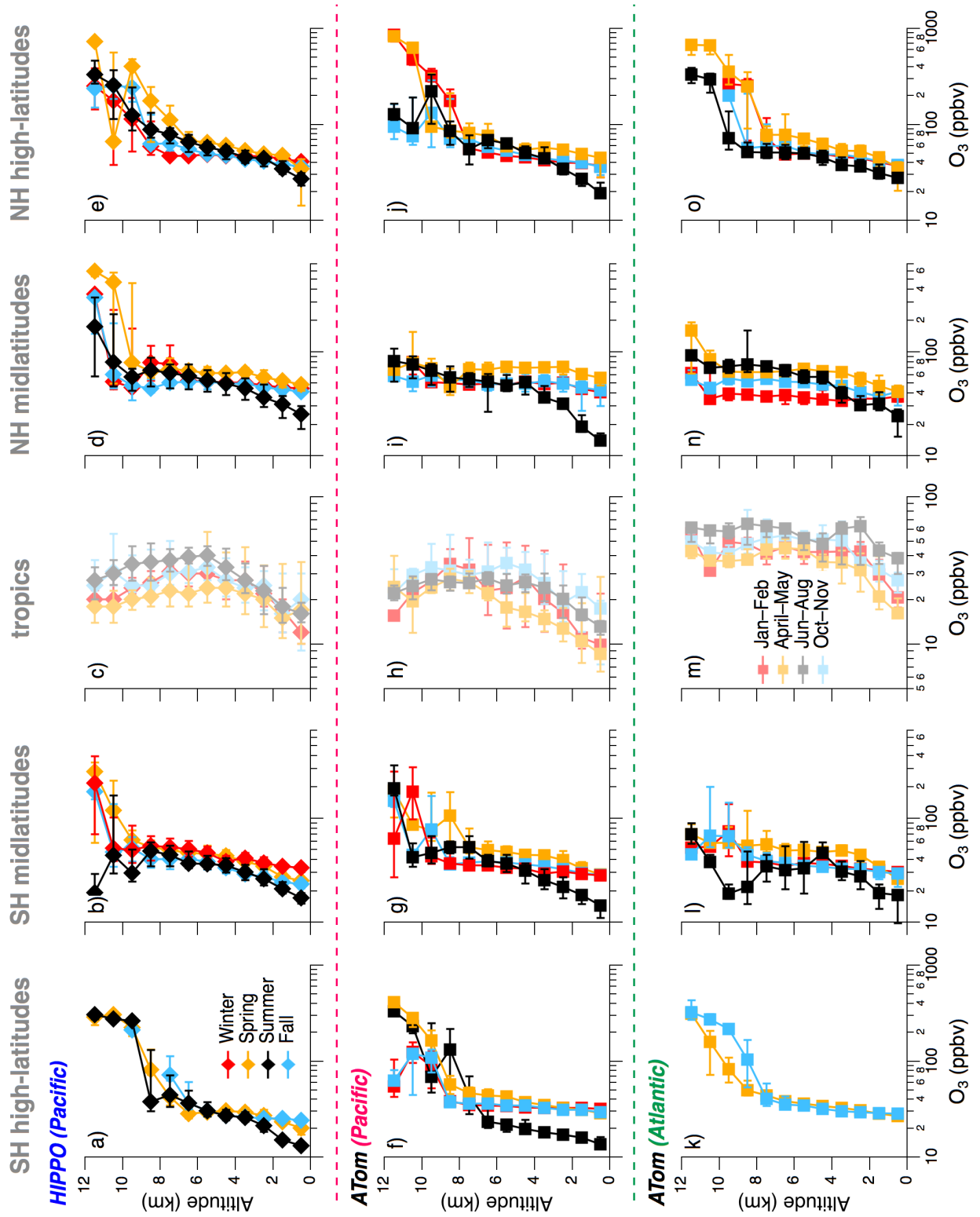

Figure 12 Seasonal variability of regional O_3 distribution in the Pacific (HIPPO in the first and ATom in the second row) and in the Atlantic (ATom in the third row). The colors designate the

local seasons with red as winter, gold as spring, black as summer, and blue as fall (corresponding months are indicated for the tropics, with lighter colors). The markers and associated bars correspond to the median, 25^{th} and 75^{th} percentiles, respectively, of O_3 distribution in every 1 km altitude bin. Note the logarithmic scale on the x-axes in all panels, and the changing scale with latitudinal bin.

Figure 13 O_3 vs. CO plots using combined ATom and HIPPO data. Each panel denotes a different latitudinal band in each basin. Seasonal deployments are colored according to the legend. Note the logarithmic scale on the y-axes in all panels, and the changing scale with latitudinal bin