Table S1 Characteristics of the different O_3 measurements used in this study.

	Timeframe	Spatial coverage (in this study)	Altitude range (in this study)	O_3 measurement technique	O_3 measurement Uncertainty
ATom	2016–2018	86°S–80°N, 195°W–14°W	0.2–12 km	Chemiluminescence	0.015 ppbv + 2%
HIPPO	2009–2011	55°S–87°N, 200°W–140°W	0.2–12 km	UV absorption	5 % + 1 ppbv
IAGOS	1994–2017	0–80°N, 100°W–10°W	8–12 km	UV absorption	2 ppbv + 2 %
Ozonesondes	Site specific	Site specific	0–12 km	ECC sondes (KI)	5–10 %

Table S2 Characteristics of the ozonesonde launching sites from which data were used in this study.

	Location	Timeframe (in this study)	Sampling frequency	Operating network
Ascension Island	7.98°S, 14.42°W	1998–2010, 2016–2018	~ weekly	SHADOZ/NASA-Goddard
Eureka	79.98°N, 85.95°W	1991–2018	~ weekly	Environment and Climate Change Canada
Hilo	19.40°N, 155.0°W	1990–2018	~ weekly	SHADOZ/NOAA-GMD
Lauder	45.04°S, 169.68°W	1987–2018	~ weekly	NIWA
Marambio	64.23°S, 56.62°W	1988–2018	~ weekly to biweekly	SMNA/FMI
Pago-Pago	14.23°S, 170.56°W	1995–2018	~ weekly	SHADOZ/NOAA-GMD
Suva	18.13°S, 112.65°E	1997–2018 (2006 & 2014 missing)	~ biweekly	SHADOZ/NOAA-GMD
Trinidad Head	41.06°N, 124.15°W	1997–2018	~ weekly	NOAA GMD
Ushuaia	54.85°S, 68.31°W	2008–2012, 2012–2018 (July–December)	~ weekly	SMNA/FMI

Table S3 Sensitivity analysis of the 1 km-vertically-binned S_{score} from 0–12 km between ATom and ozonesonde O_3 distributions at Ascension Island in August. The first variable indicates the distance from the ozonesonde launching site within which ATom data were used to define O_3 distributions. The second variable corresponds to the size of the bin used to compare O_3 distributions. The third variable is the length of the ozonesonde record to which ATom is compared. The first row corresponds to the variables used in this study. In the following rows, the changing variable is indicated in bold.

S_{score} / Variables	Range	Average
500 km, 5 ppbv, full record	21–61	42
250 km, 5 ppbv, full record	21–68	36
1000 km, 5 ppbv, full record	40–66	50
500 km, **2 ppbv**, full record	15–53	34
500 km, **10 ppbv**, full record	32–66	48
500 km, 5 ppbv, **2 years**	15–62	40
500 km, 5 ppbv, **8 years**	20–63	42

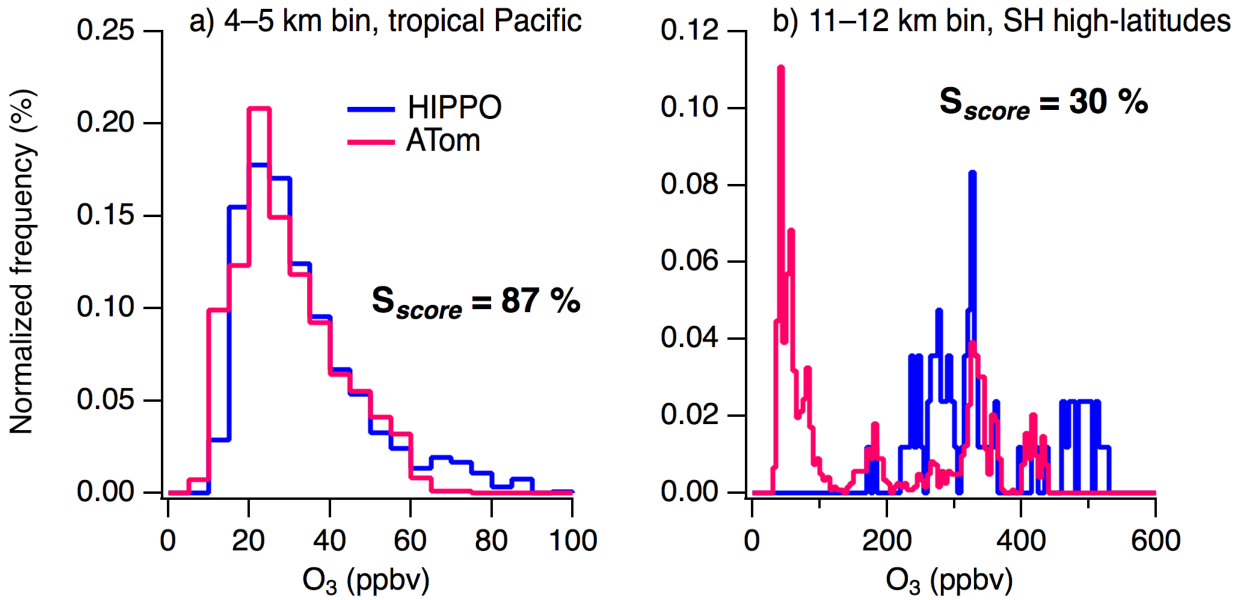


Figure S1 Two illustrations of the Skill Score (S_{score}), a metric used in this study to quantify the overlap of O_3 distributions. Here are shown in a) a high S_{score} between HIPPO (blue) and ATom (pink) from 4 to 5 km in the tropical Pacific, and b) a low S_{score} between HIPPO (blue) and ATom (pink) from 11 to 12 km in the high-latitudes of the Southern Hemisphere.

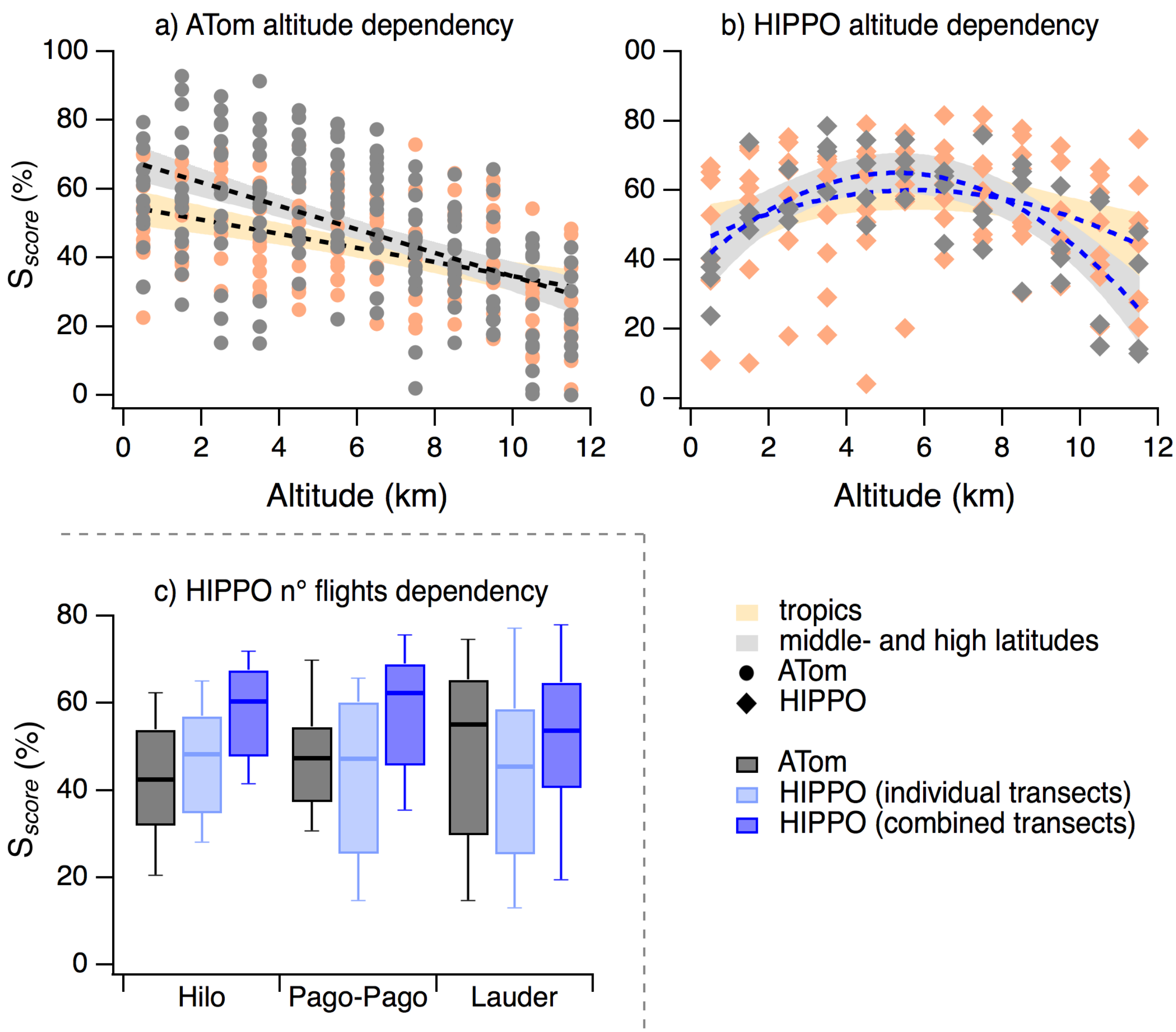
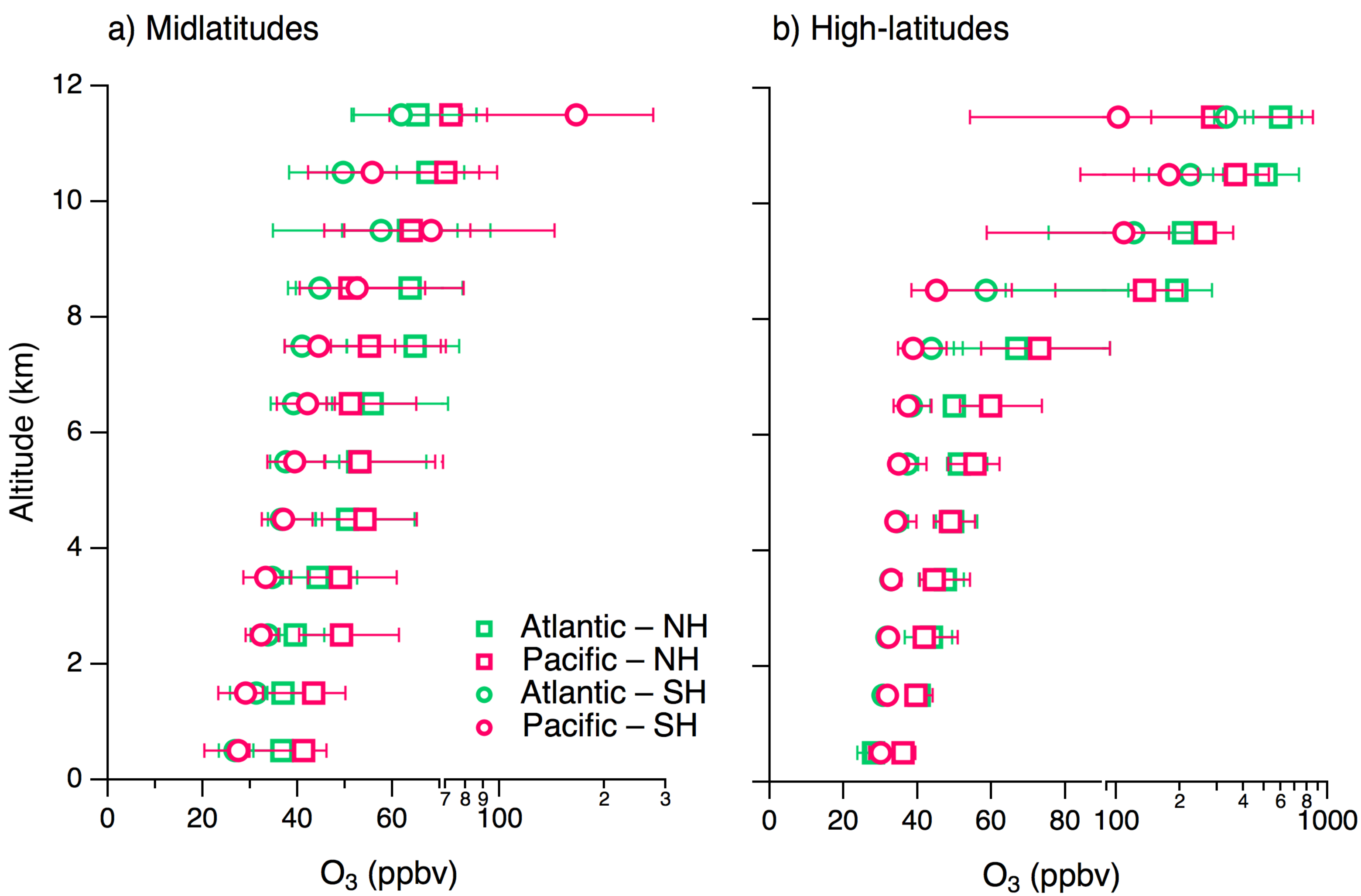


Figure S2 S_{score} correlation with altitude for a) ATom (circles) vs. ozonesonde comparison, and b) HIPPO (diamonds) vs. ozonesonde comparison. The comparison is split between the tropics (orange) and the extra-tropics (grey) to show the increased dependency with altitude of the S_{score} in the extra-tropics. In c) is shown the distribution of the S_{score} values between ATom and ozonesondes (in black), HIPPO (comparing the northbound and southbound transects individually) and ozonesondes (in light blue), and HIPPO (comparing the combined northbound and southbound transects) and ozonesondes (in dark blue).



a) Midlatitudes
b) High-latitudes
Altitude (km)
12
10
8
6
4
2
0
0
20
40
60
7
8
9
100
2
3
80
4
6
1000
O3 (ppbv)
O3 (ppbv)
Atlantic – NH
Pacific – NH
Atlantic – SH
Pacific – SH

Figure S3 Inter-hemispheric comparison of tropospheric O_3 distribution in a) the mid latitudes and b) the high latitudes, both in the Pacific (pink) and in the Atlantic (green). The NH is delineated by the squares, and the SH is delineated by the circles. The x-axes start with linear scales then switch to a logarithmic scale in both panels.

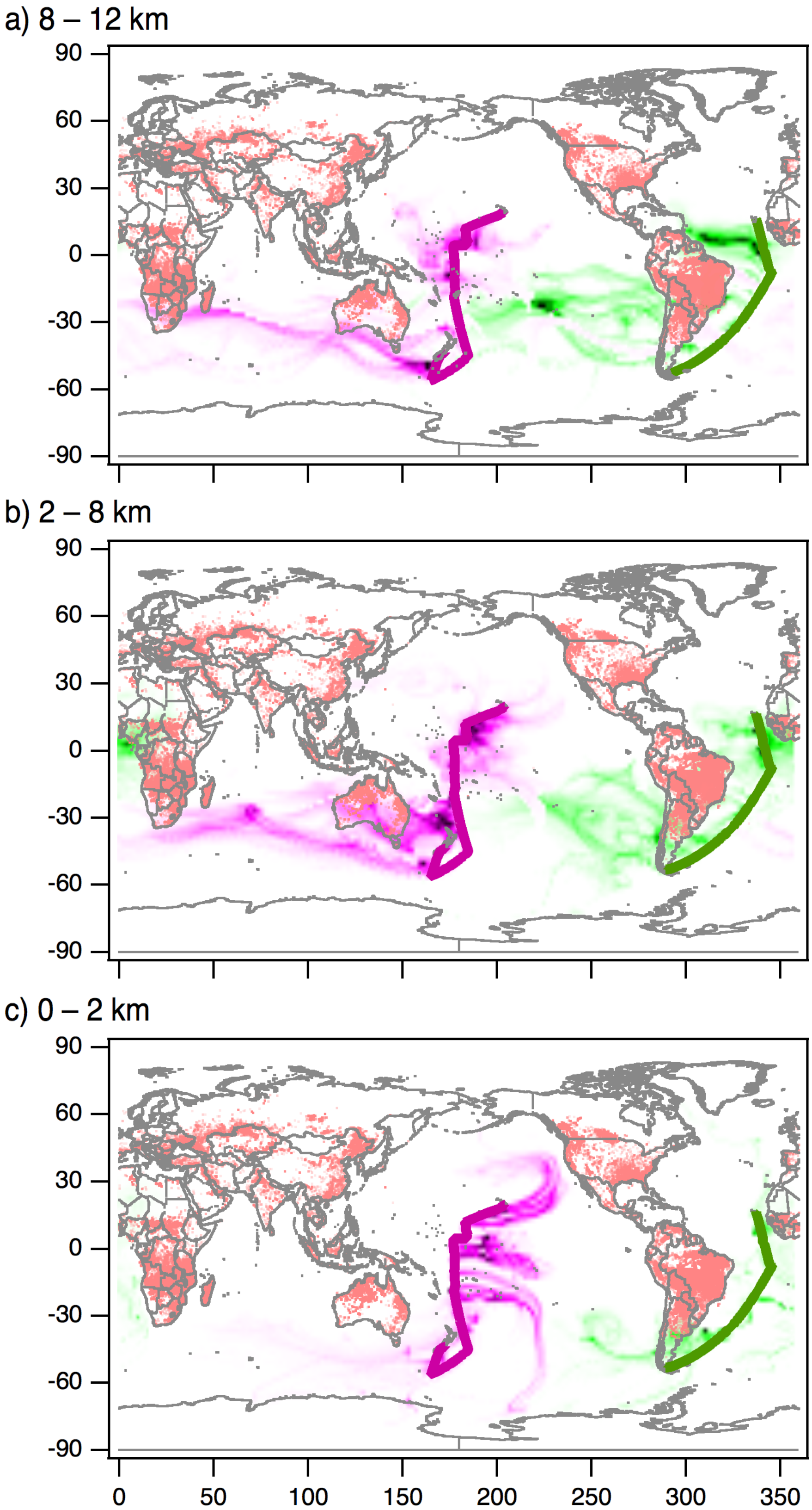


Figure S4 Modeled 10 days back trajectories for ATom in October (ATom-3) for different altitude levels corresponding to a) 0 – 2 km, b) 2 – 8 km, and c) 8 – 12 km. The red dots on the map are fire counts in October 2017 from MODIS.

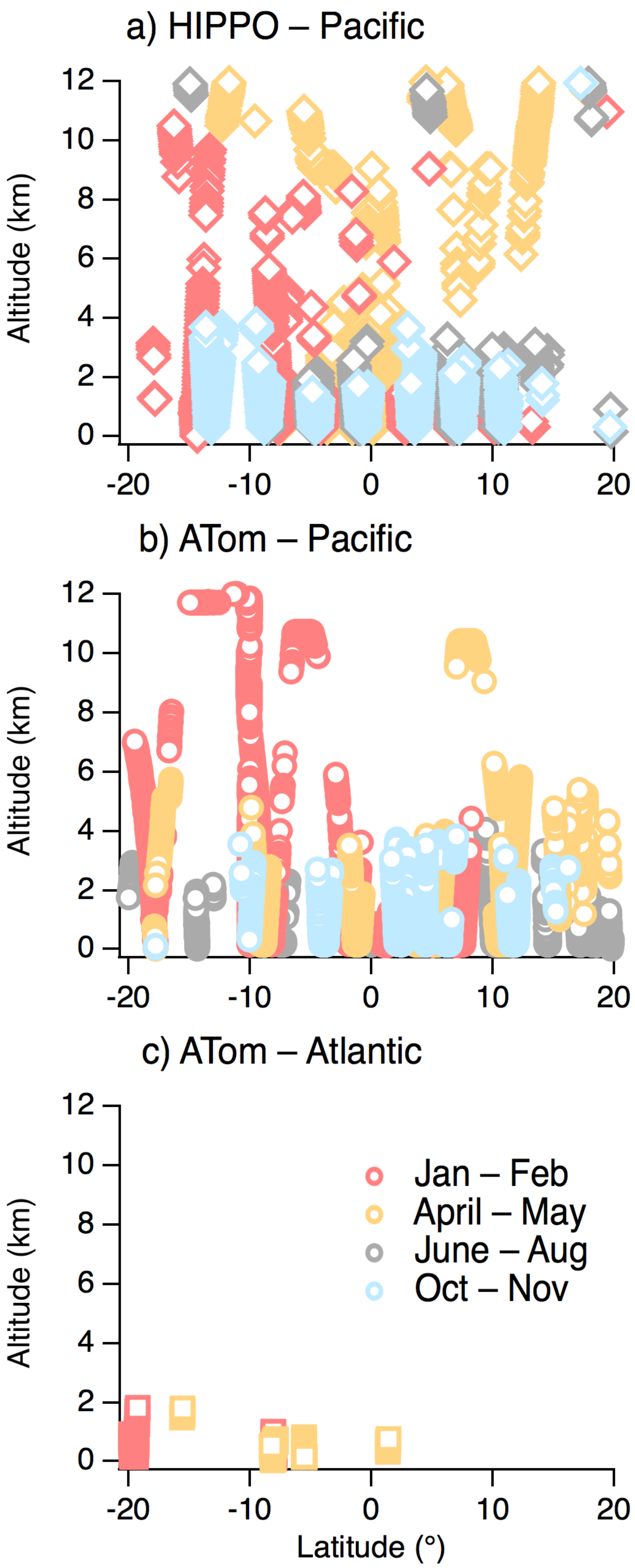
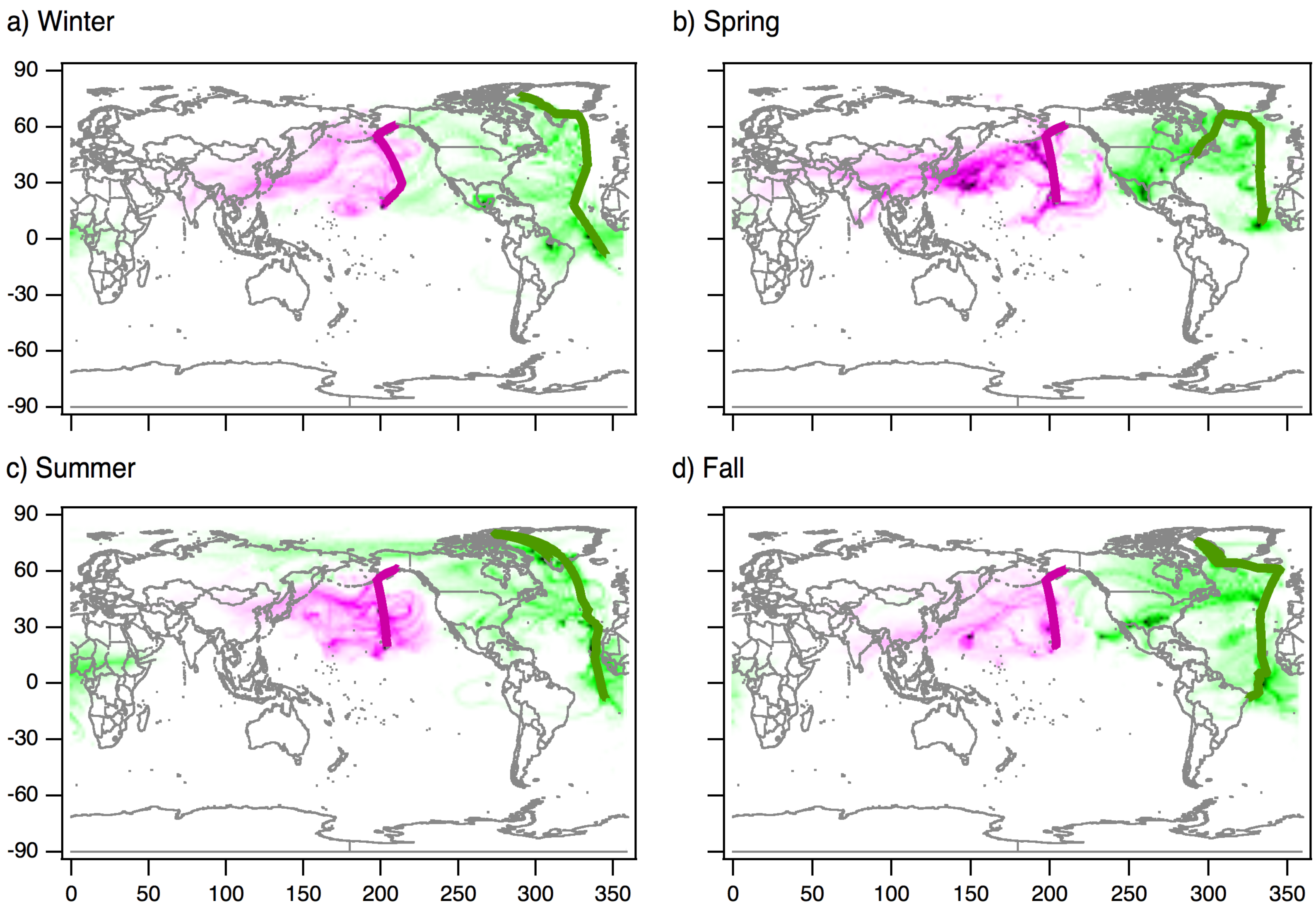


Figure S5 Occurrences of low O_3 (< 15 ppbv) in the tropical oceans during a) HIPPO in diamonds, b) ATom (Pacific) in circles, and c) ATom (Atlantic) in squares. The seasonal deployments are delineated by the colors, as shown in the legend.



a) Winter
b) Spring
c) Summer
d) Fall
90
60
30
0
-30
-60
-90
0
50
100
150
200
250
300
350

Figure S6 Modeled 10 days back trajectories for ATom 1–4 corresponding to a) boreal winter, b) boreal spring, c) boreal summer, and d) boreal fall.