



Supplement of

Airborne measurements of oxygen concentration from the surface to the lower stratosphere and pole to pole

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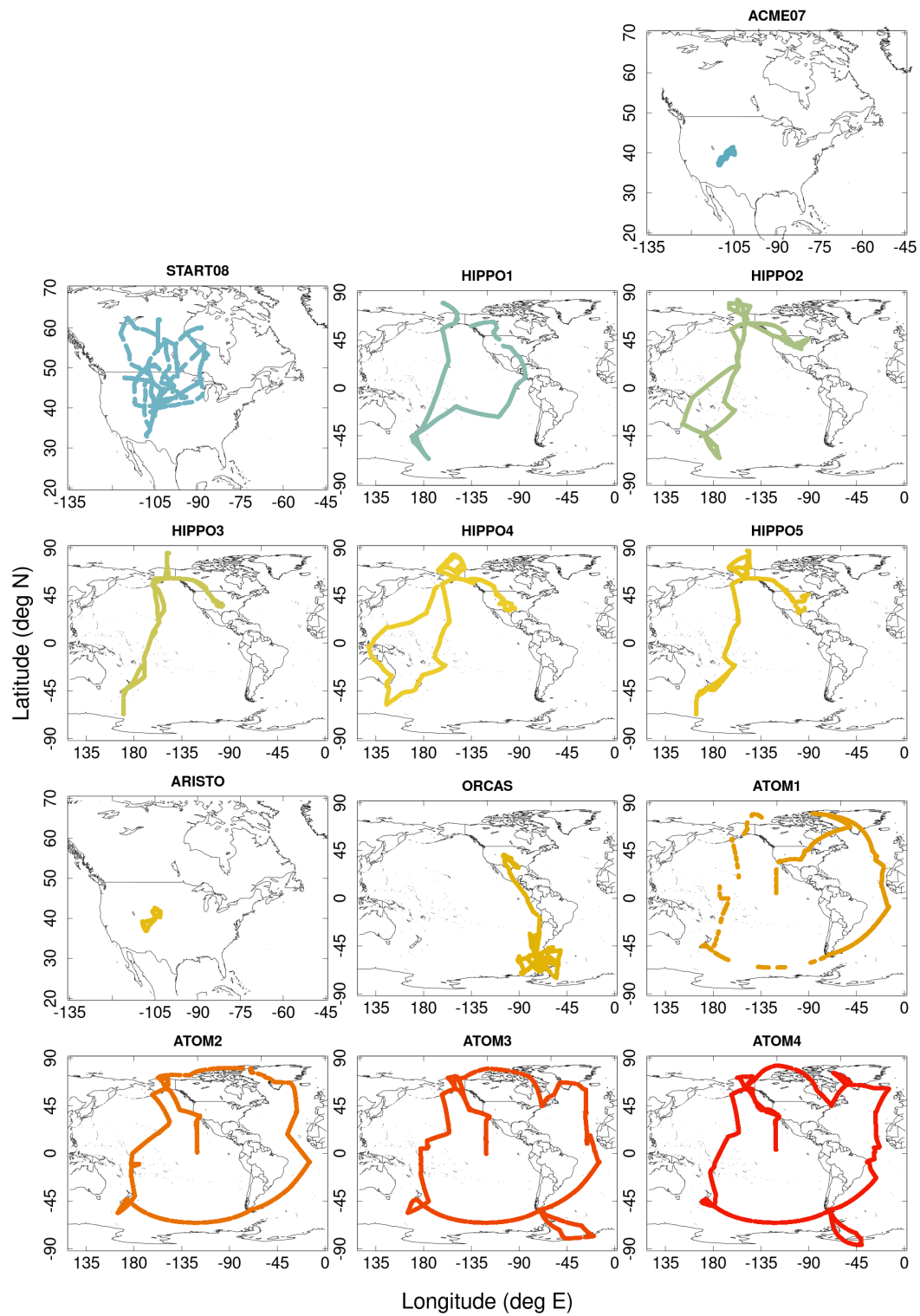


Figure S1. Locations of AO2 (this page) and Medusa (next page) sampling. COBRAtest-1999, IDEAS-1, IDEAS-2, and ACME-04 are not shown.

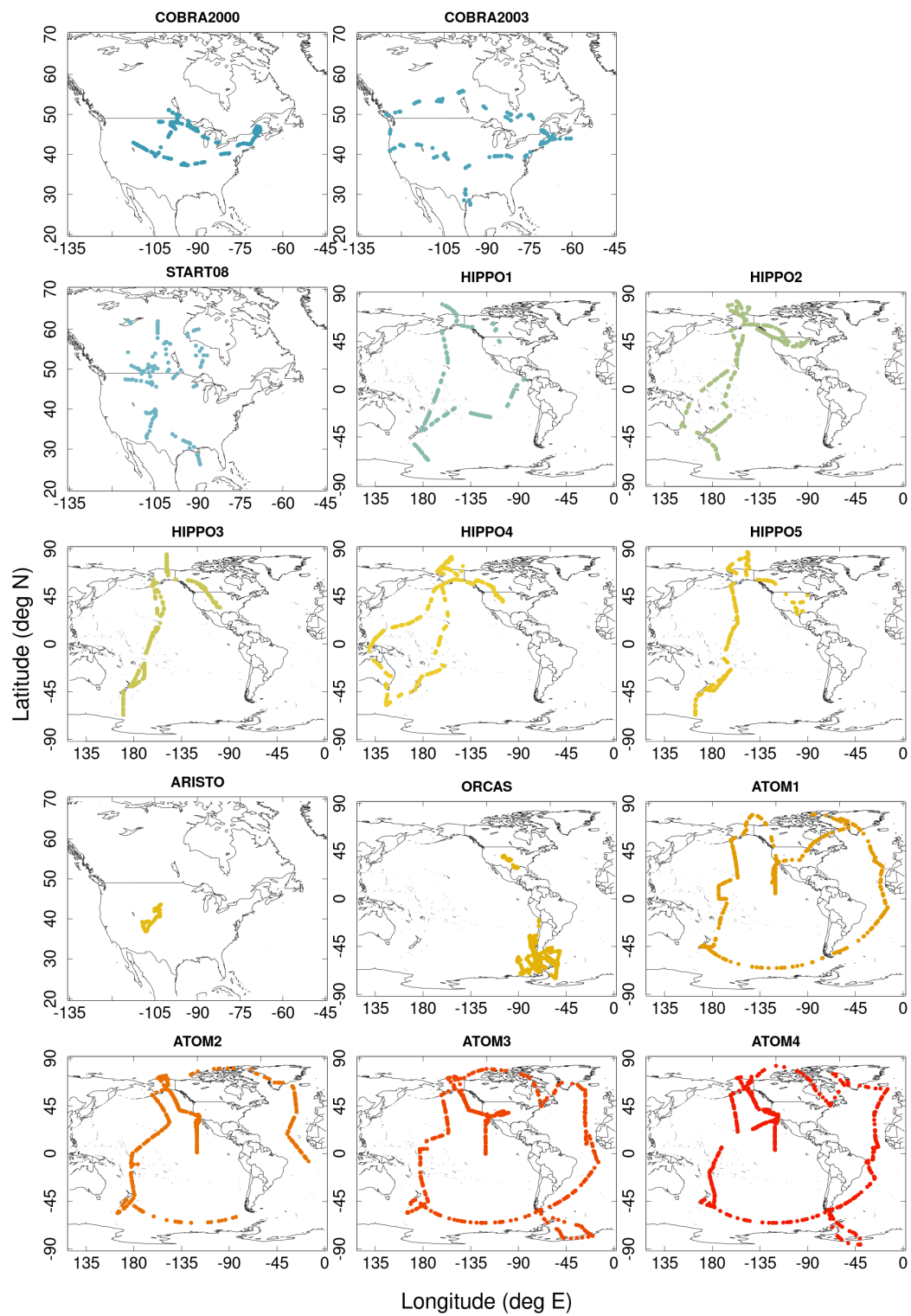


Figure S1. Continued.

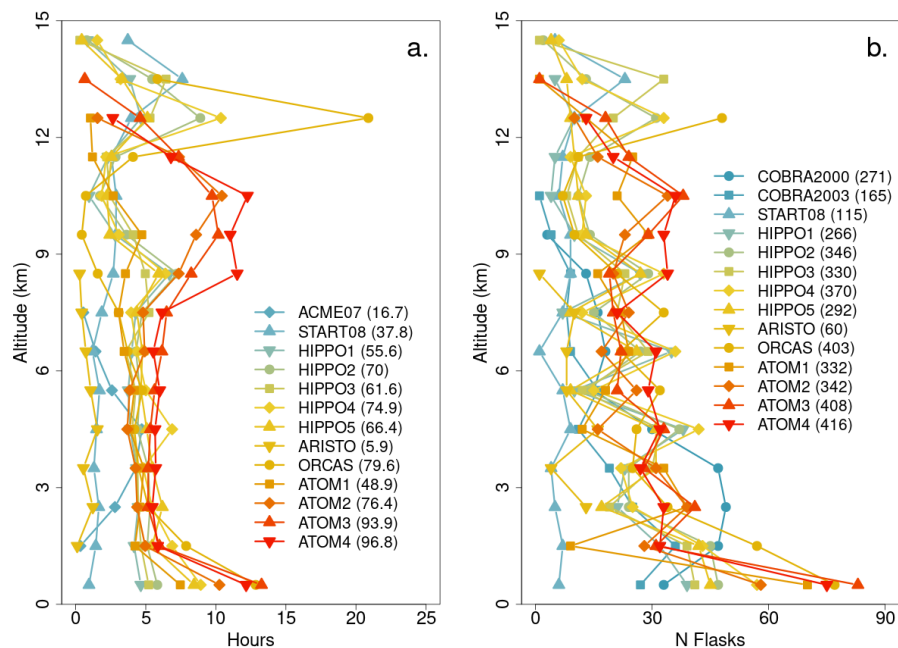


Figure S2. Vertical distributions of AO2 (a) and Medusa (b) sampling. The vertical distributions show the number of hours of AO2 data or number of flask samples per 1000 m altitude bin. Values in parentheses give total hours (a) or flasks (b) for each campaign. COBRAtest-1999, IDEAS-1, IDEAS-2, and ACME-04 are not shown.

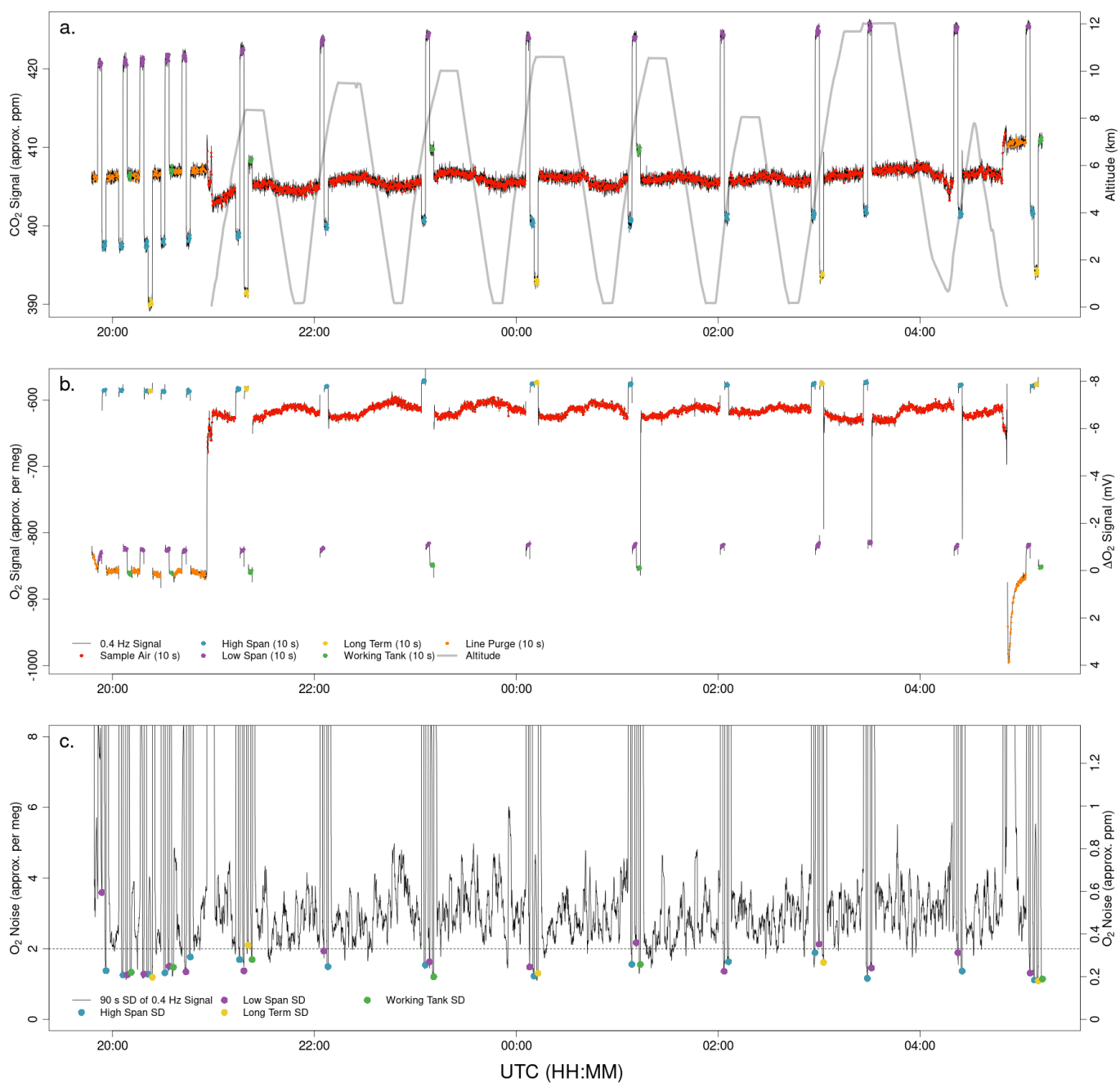


Figure S3. Same as Fig. 5 but for AO2 CO₂ and O₂ signals from ATom-4 research flight 5, from Nadi, Fiji to Christchurch, New Zealand, and including a third panel showing O₂ signal noise. The legend in (b) also applies to (a). In (c), the black line shows the running 90 s standard deviation (of forty 2.3 s points) and the symbols show the standard deviation of points within the 90 s calibration intervals. In (c), the left y axis shows values in per meg $\delta(O_2/N_2)$ and the right y axis shows ppm in O₂ mole fraction.

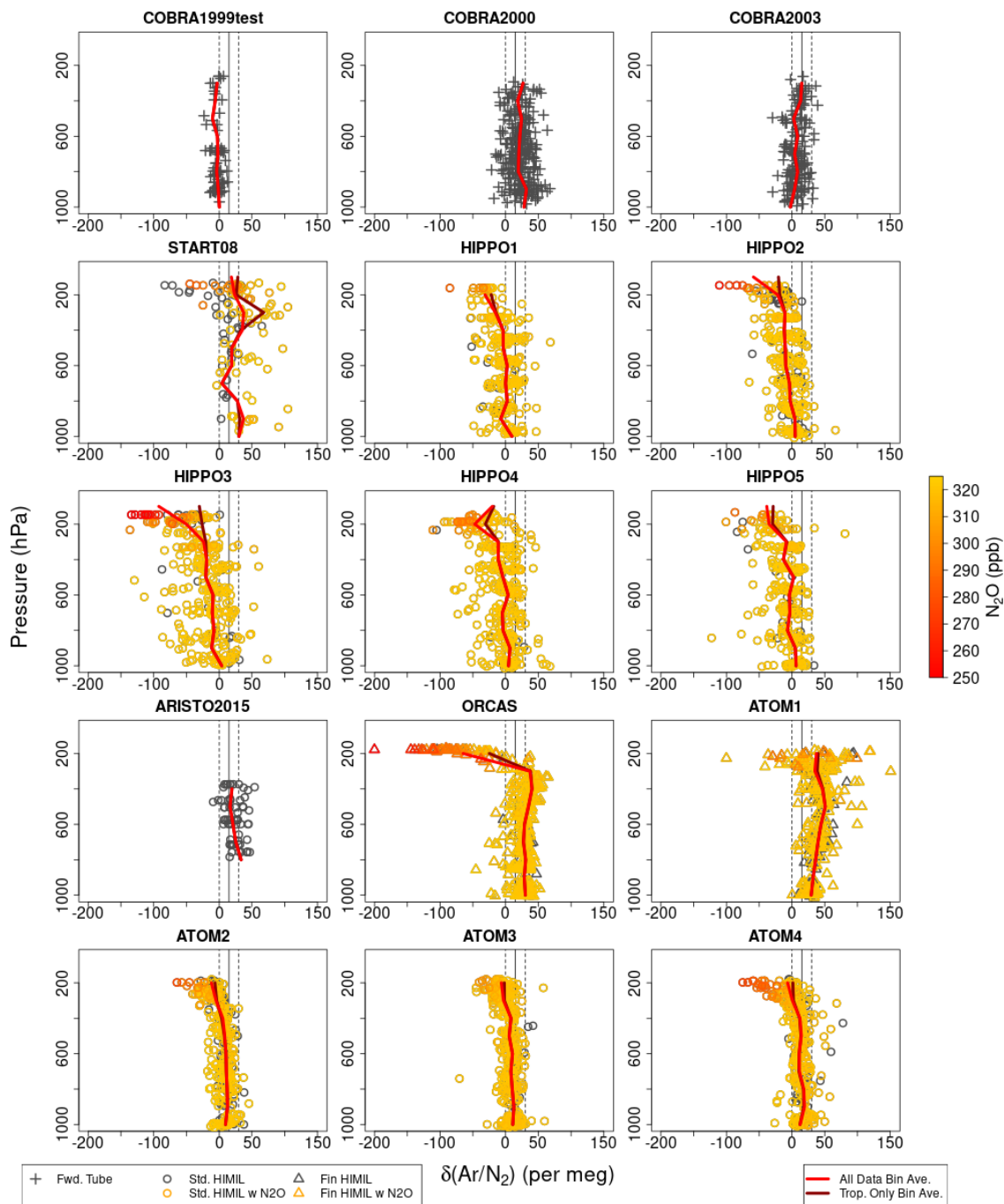


Figure S4. Measurements of $\delta(\text{Ar}/\text{N}_2)$ on Medusa flasks for each campaign plotted versus pressure. Symbol shapes distinguish the different Medusa inlet types. Colored symbols indicate N_2O concentrations detrended to a reference year of 2009 ($\text{N}_2\text{O}_{2009}$) as measured by the Harvard QCLS instrument for all campaigns except ATom-1 which used N_2O from the NOAA PANTHER instrument. Black points are for samples where N_2O measurements are not available. The COBRA and ARISTO-2015 campaigns did not include N_2O measurements. Red lines show averages for 100 hPa bins. Dark red lines show bin averages for data determined to be tropospheric based on $\text{N}_2\text{O}_{2009} < 318$.

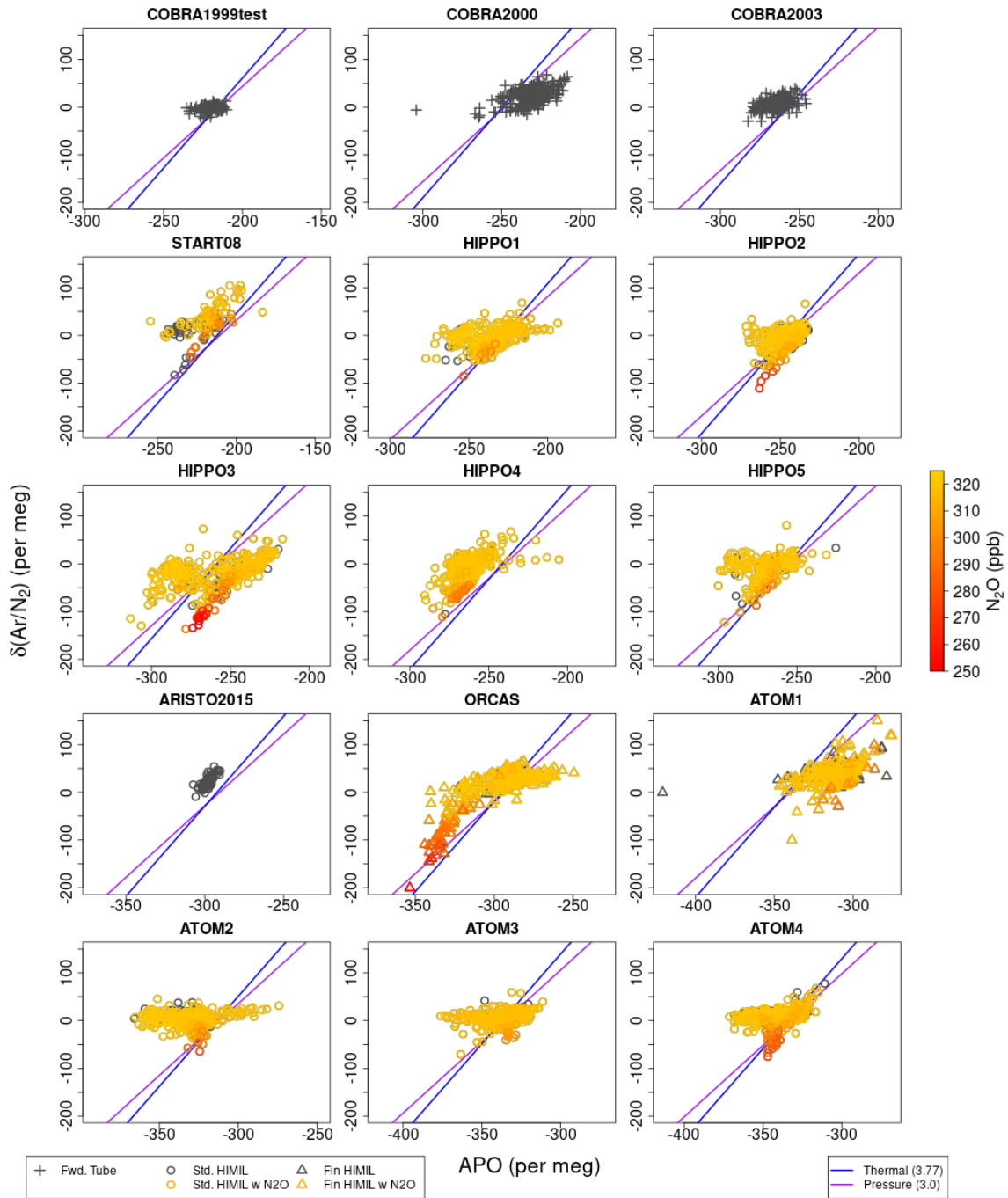


Figure S5. Measurements of Medusa flask $\delta(\text{Ar}/\text{N}_2)$ for each campaign plotted versus Medusa flask APO. Symbol shapes distinguish the different Medusa inlet types. Colored symbols indicate N_2O concentrations detrended to a reference year of 2009 as measured by the Harvard QCLS instrument for all campaigns except ATom-1 which used N_2O from the NOAA PANTHER instrument. The COBRA and ARISTO-2015 campaigns did not include N_2O measurements. Blue and magenta lines indicate expected slopes for thermal or pressure gradient fractionation at 1 atm. Variations along these lines could indicate fractionation or natural covariations between Ar and O_2 .

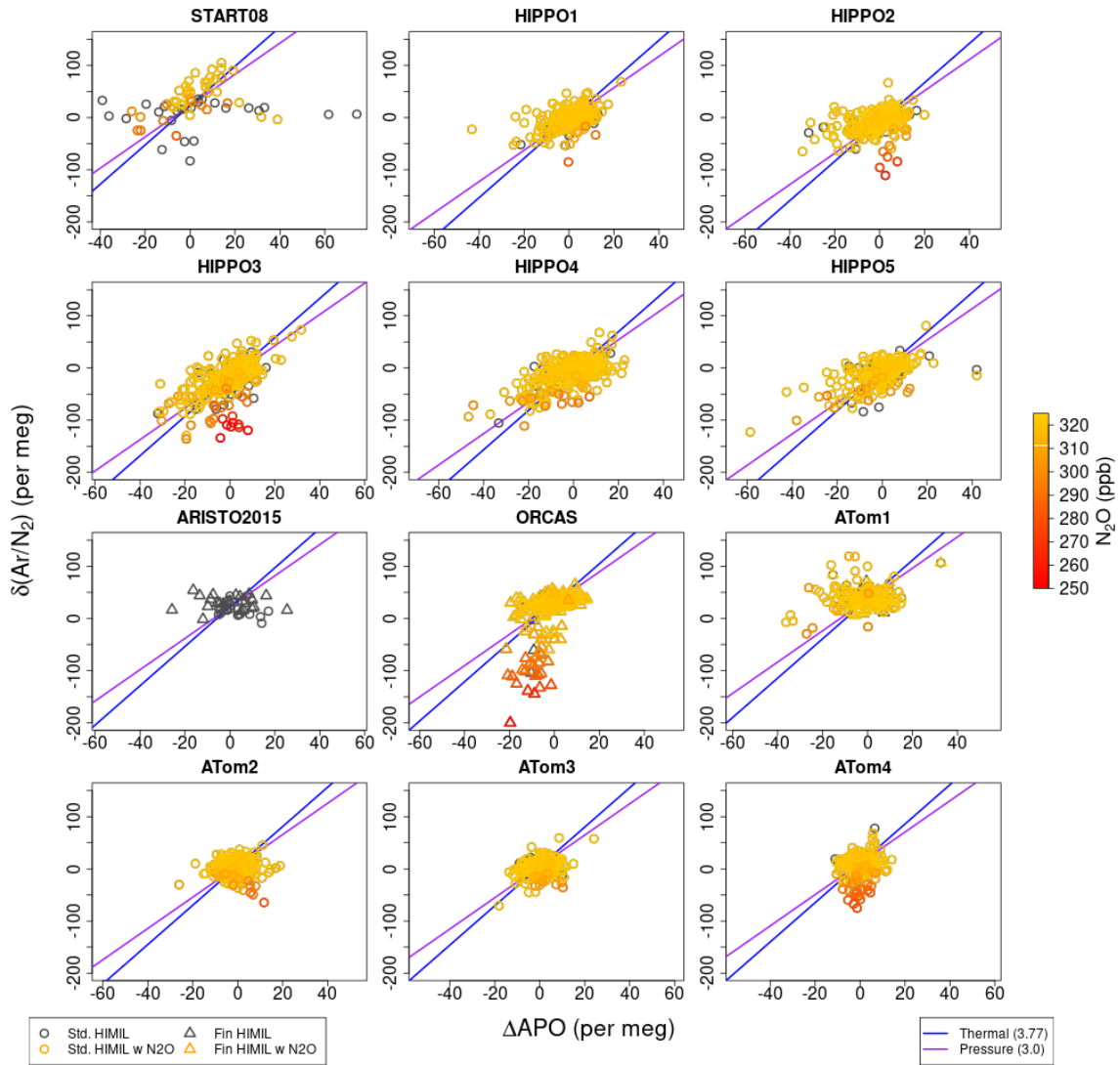


Figure S6. Measurements of Medusa flask $\delta(\text{Ar}/\text{N}_2)$ for each campaign plotted versus the Medusa minus AO2 APO difference. These APO differences are calculated using uncorrected Medusa values to highlight potential flask sampling fractionation, and have been normalized by subtracting the mean difference for each flight to avoid flight-to-flight AO2 variations. Symbol shapes distinguish the different Medusa inlet types. Colored symbols indicate N_2O concentrations detrended to a reference year of 2009 as measured by the Harvard QCLS instrument for all campaigns except ATom-1 which used N_2O from the NOAA PANTHER instrument. The ARISTO-2015 campaign did not include N_2O measurements. Blue and magenta lines indicate expected slopes for thermal or pressure gradient fractionation in Medusa samples at 1 atm. Variations along these lines suggest fractionation of Medusa samples. Variations parallel with the y axis could be fractionation affecting AO2 and Medusa identically, though this is less likely based on other evidence.

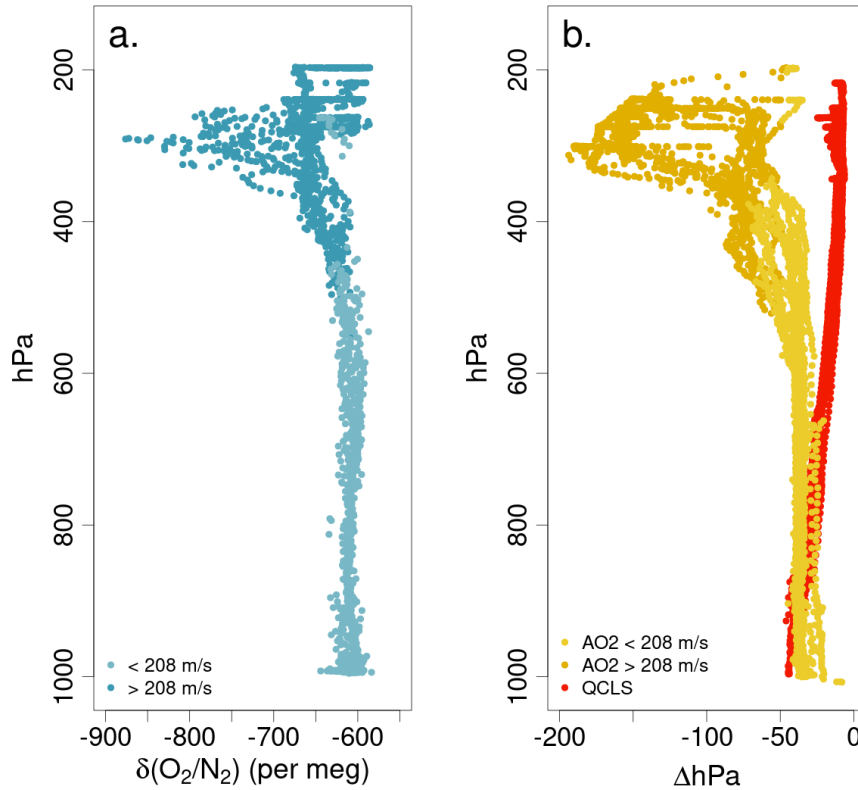


Figure S7. Evidence of inlet fractionation with the use of a 7.9 mm OD aft facing fin HIMIL inlet at fuselage station 770 on the left side of the DC-8 during the first half of ATom-1. a) Multiple vertical profiles of $\delta(\text{O}_2/\text{N}_2)$ measured by AO2 on July 29, 2016 on research flight 1 between California and the Eastern Equatorial Pacific. b) Multiple vertical profiles of the dynamic pressure deficit relative to static pressure (yellow) from the same inlet as (a) measured on Aug., 15, 2016 on research flight 7 between Punta Arenas, Chile and Ascension Island. The plumbing arrangement only allowed pressure measurement from an inlet while not in use for sampling, hence data from different flights are shown. For comparison, (b) also shows the dynamic pressure deficit for a typical flight measured from an identical fin HIMIL in use by the Harvard QCLS instrument at fuselage station 450.

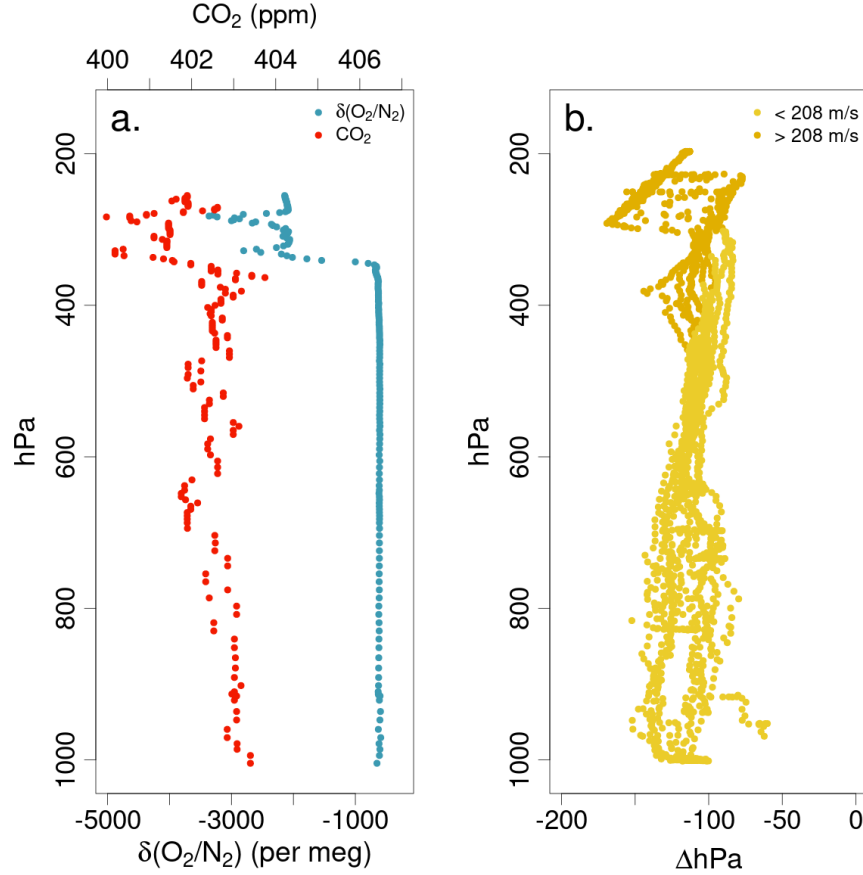


Figure S8. Evidence for exceptional fractionation of $\delta(\text{O}_2/\text{N}_2)$ and CO_2 during a test of a side facing 3.2 mm OD inlet during ATom-1. a) A single ascending vertical profile of $\delta(\text{O}_2/\text{N}_2)$ and CO_2 measured by AO2 on Aug. 8, 2016 on research flight 5, between American Samoa and Christchurch, New Zealand. b) Multiple vertical profiles of the dynamic pressure deficit relative to static pressure from the same inlet as (a) measured on the same flight but after switching AO2 to sample from a different inlet, so not including the same profile as (a).

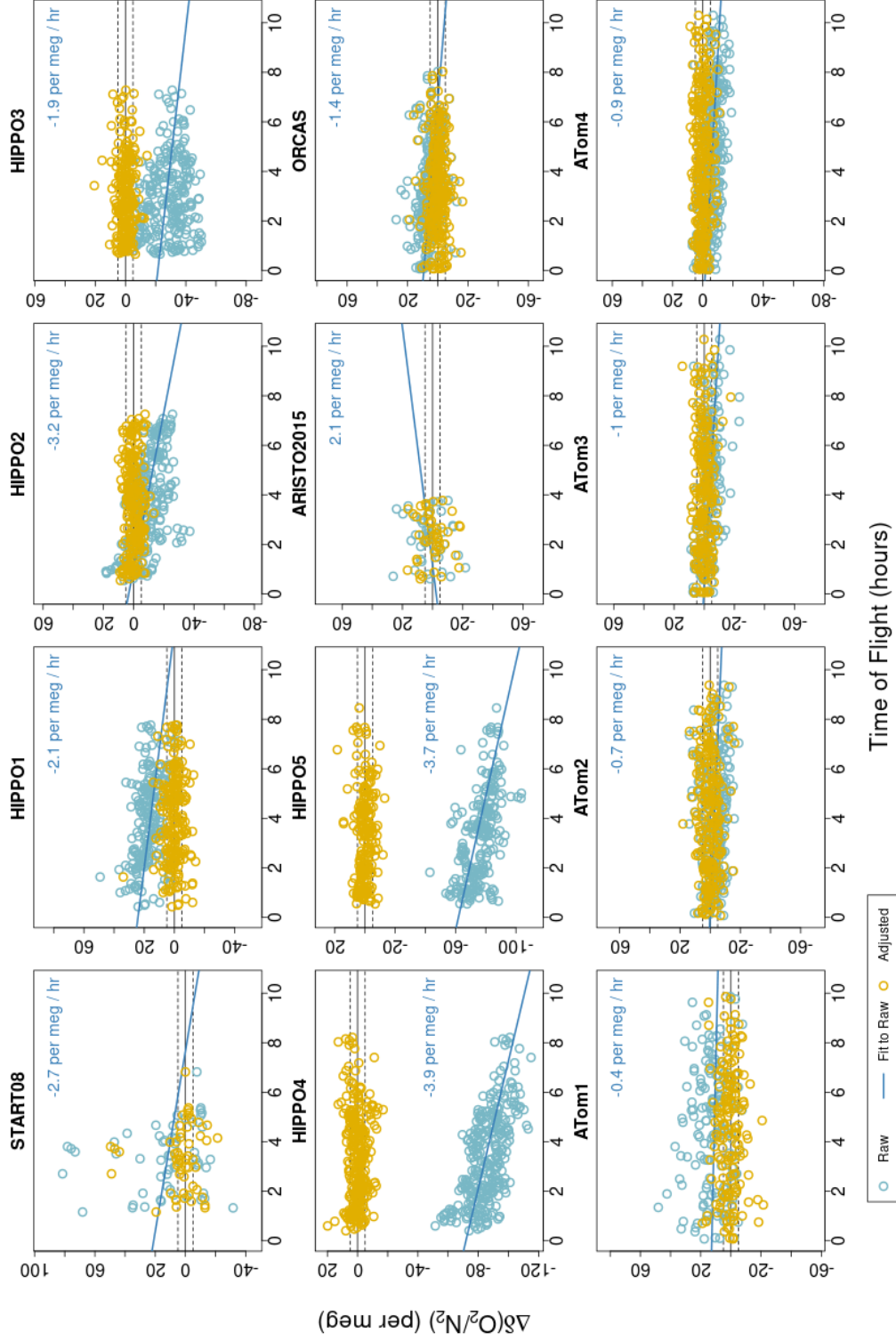


Figure S9. AO2 $\delta(\text{O}_2/\text{N}_2)$ minus Medusa $\delta(\text{O}_2/\text{N}_2)^*$ differences for each campaign plotted versus hours since takeoff. Blue symbols show differences between raw unadjusted AO2 $\delta(\text{O}_2/\text{N}_2)$ measurements and Medusa $\delta(\text{O}_2/\text{N}_2)^*$ measurements. Yellow symbols show differences between the AO2 measurements adjusted by a linear time-of-flight trend plus mean offset for each flight, except for the second half of ATom-1, which was adjusted by a pressure dependent fit. Lines and values in each panel give the slope of the campaign mean unadjusted time-of-flight dependency (Table S3).

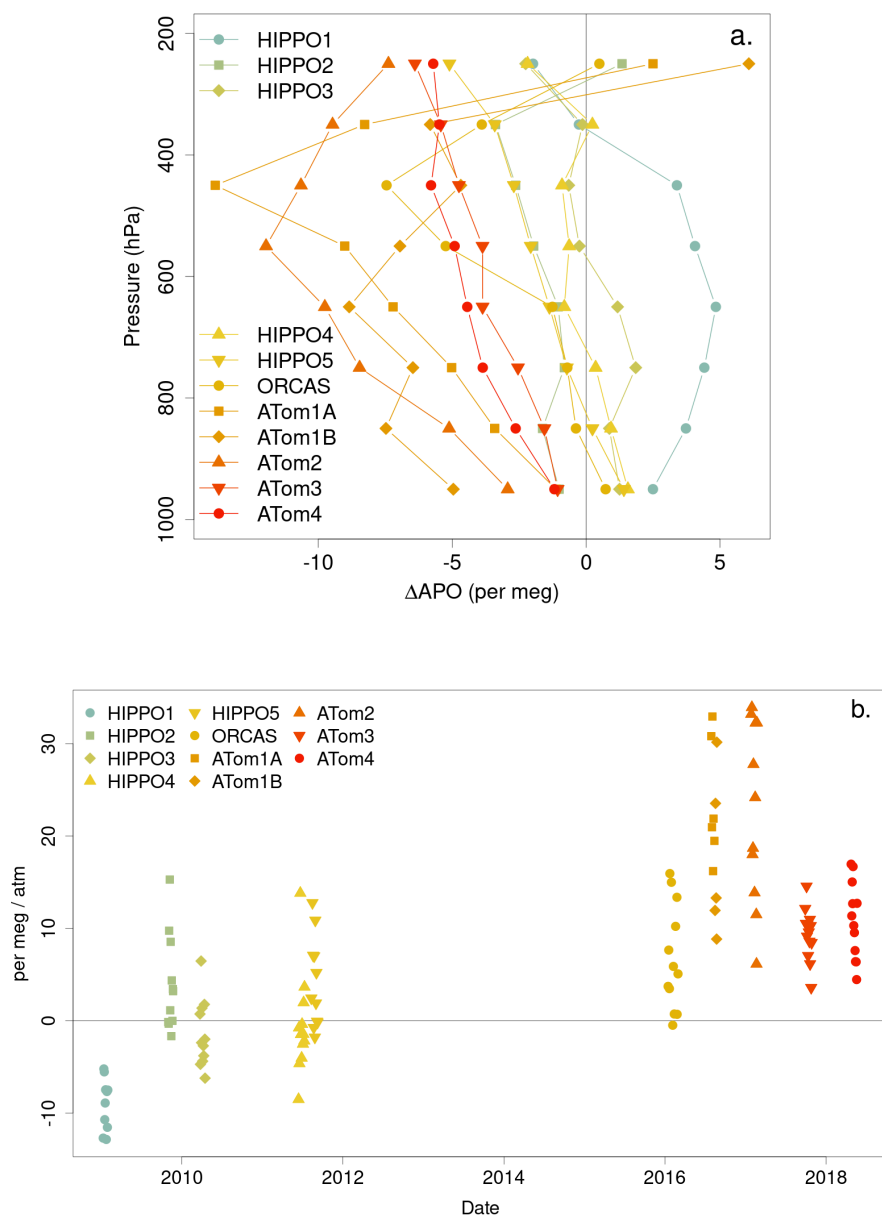


Figure S10. a) For each campaign, the average difference in AO2 APO measurements between ascending and descending profiles, plotted versus ambient pressure. Differences were calculated by first separating the observations by profile and sign of vertical speed, then subtracting an interpolated profile from the preceding and following profiles. These differences, sign adjusted to correspond to ascending minus descending, were then averaged into 100 hPa bins by flight and then for the entire campaign. b) Apparent ascending minus descending dependency of AO2 APO measurements by individual flight, shown as the slope of a linear fit to the differences calculated as for (a) versus pressure between 400 hPa and the surface.

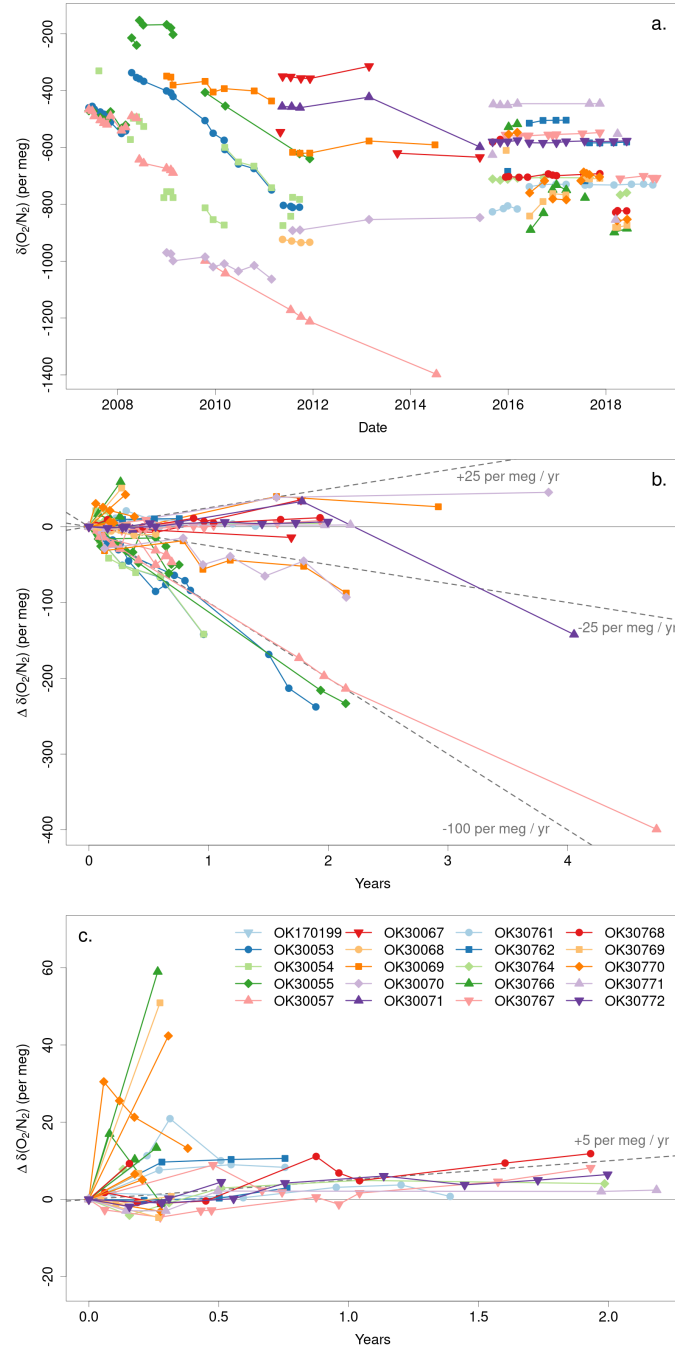


Figure S11. Laboratory measurements of AO2 cylinders, (a) all measurements on the Scripps O₂ Program scale, propagated as described in the main text (b) all measurements relative to the initial measurement for that cylinder and fill, and (c) same as (b) but only for measurements since July 2015. Each point represents the average of multiple overnight runs within a period of 2 weeks. The legend in (c) lists cylinder IDs for all plots. In (a), new fills of the same cylinder are distinguished by gaps in the lines.

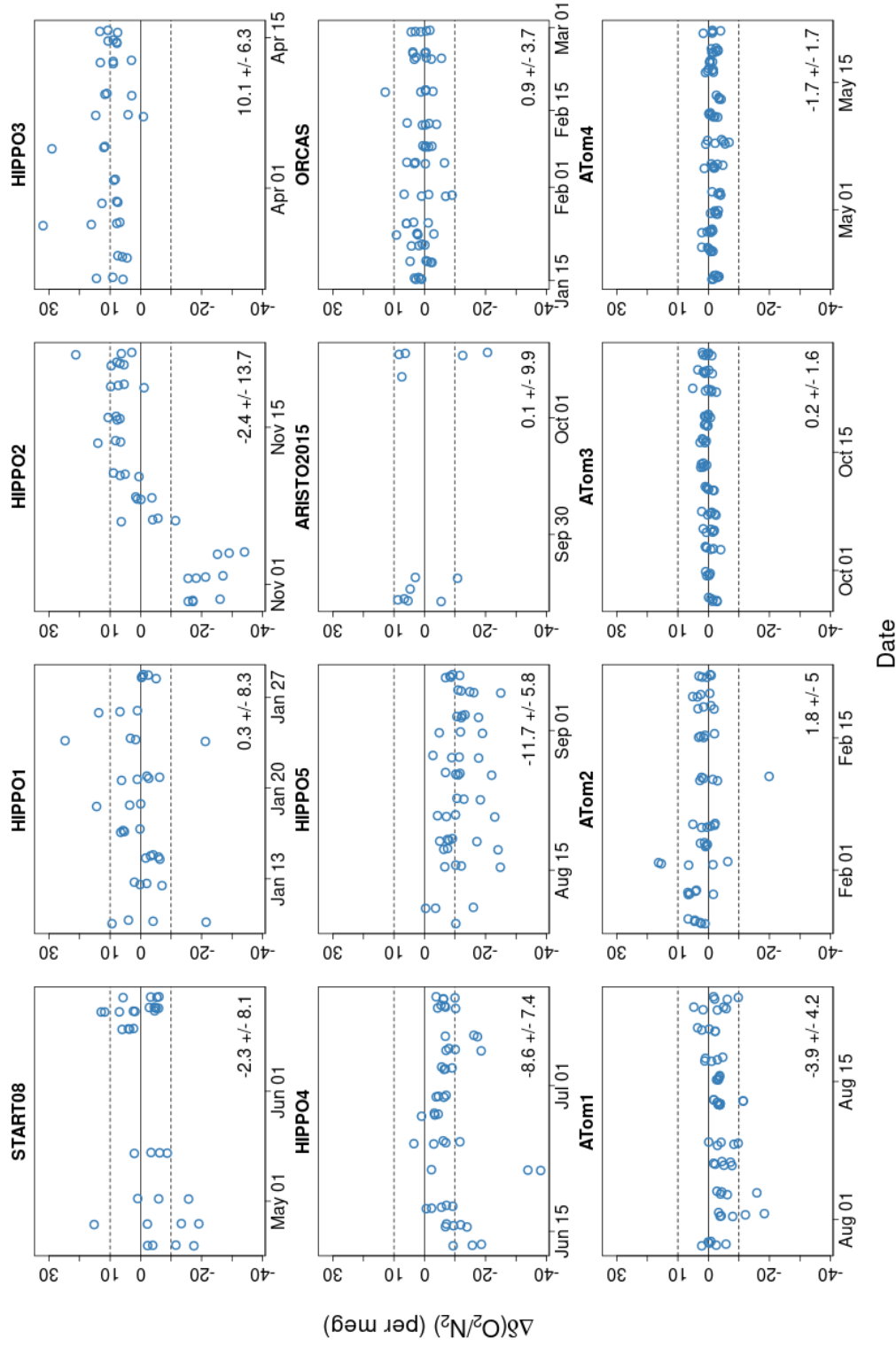


Figure S12. Differences between airborne determinations of the long term reference cylinder by AO2 and the laboratory assigned value for this cylinder. AO2 measures this cylinder approximately every 150 min in flight. The values in each panel give the average offset and standard deviation for that campaign.

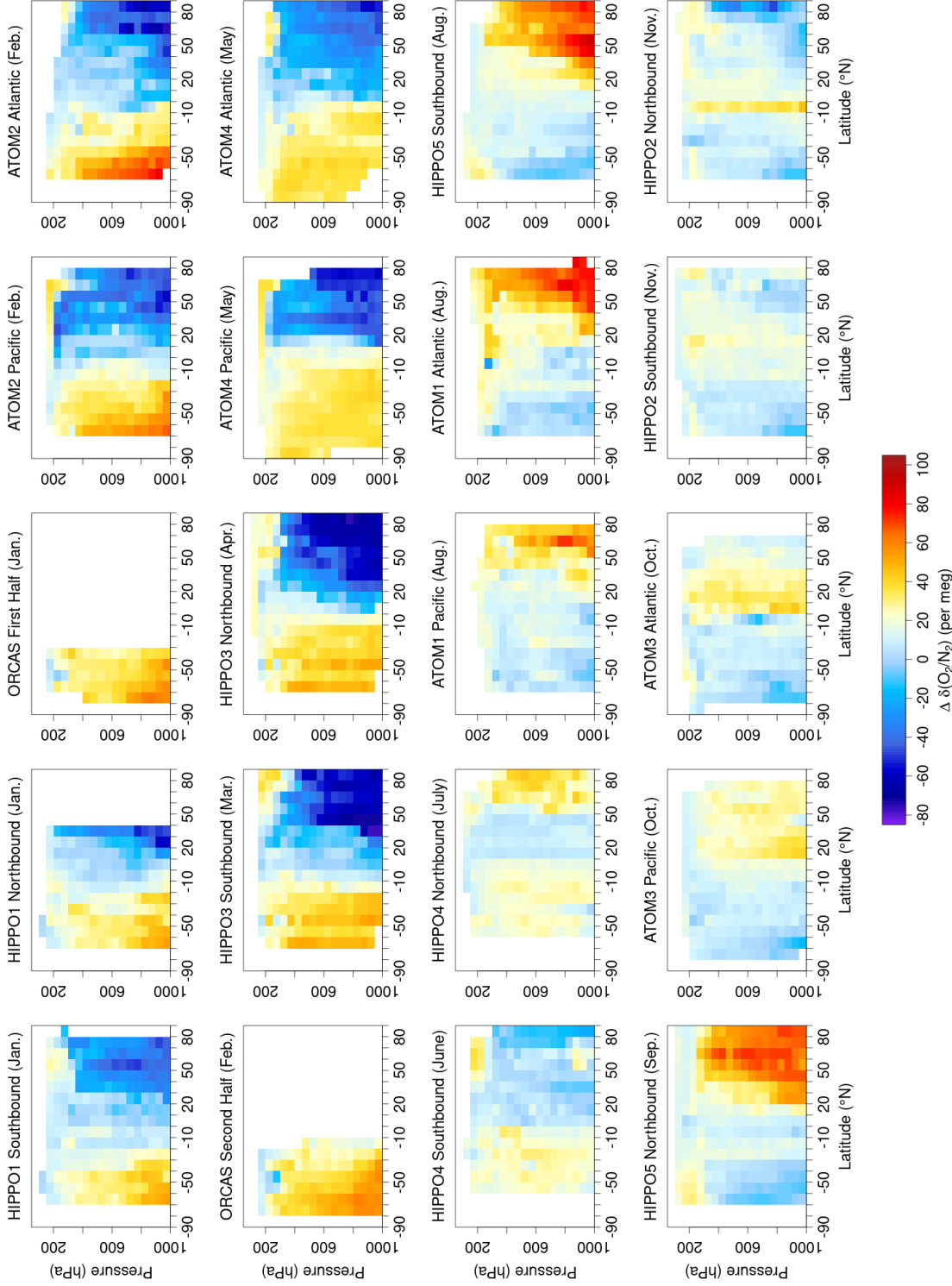


Figure S13. AO2 and Medusa detrended $\delta(\text{O}_2/\text{N}_2)$ altitude latitude cross-sections for all HIPPO, ORCAS, and ATom campaigns. Plotted as for Fig. 11, except the flight tracks and individual Medusa points are not shown for clarity, the binning is by 10 degrees latitude and 50 hPa, and the values have been detrended by removing a long-term deseasonalized trend fit to Scripps O₂ Program measurements from Mauna Loa. Plots are arranged in seasonal order. Each campaign is divided in half. For HIPPO the southernmost flight is included in both halves, and for ORCAS research flight 8 is included in both halves.

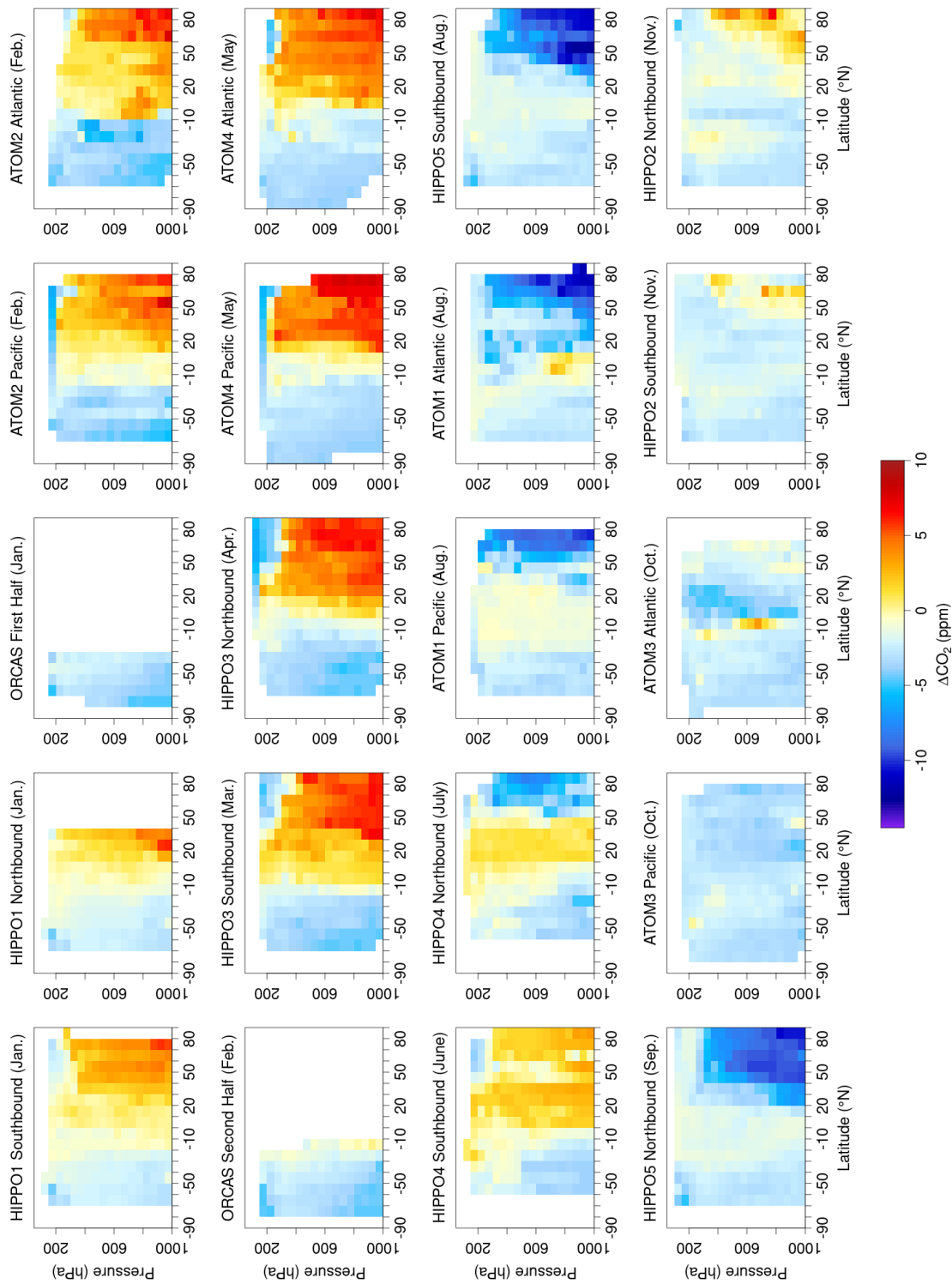


Figure S14. Same as Fig. S13 but for CO₂ as measured by AO2 and Medusa.

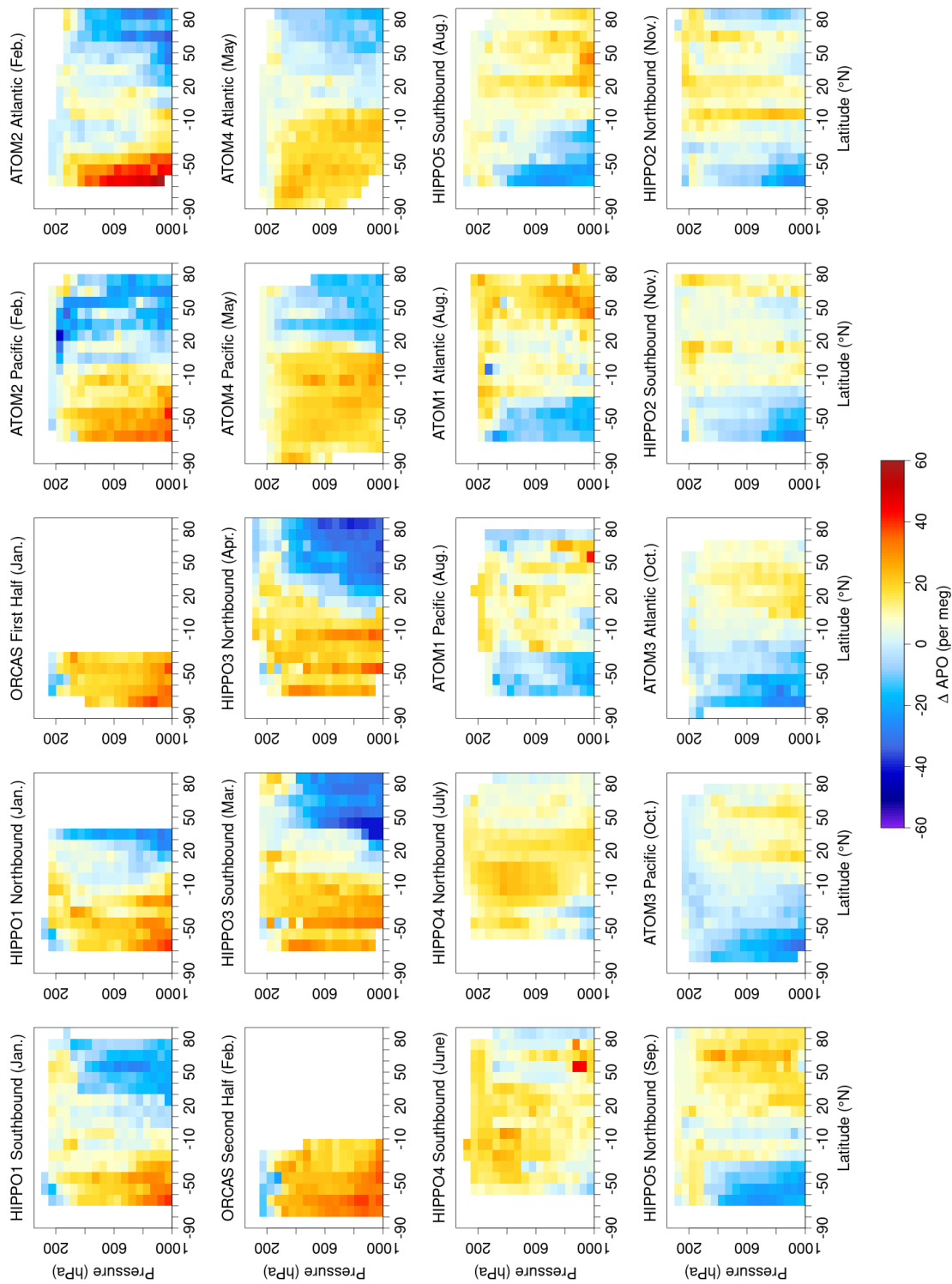


Figure S15. Same as Fig. S13 but for APO as measured by AO2 and Medusa.

Table S1. Vendors and part numbers for select AO2 and Medusa components

Component	Vendor and part number
AO2 tubing	Restek 29056
AO2 inlet and detector control valves	MKS 248A with #7 orifice
AO2 absolute pressure sensors	Honeywell FPA2BM2D5A6N
AO2 selection manifold valves	Numatics TM Series
AO2 calibration gas cylinders	Luxfer L45M
AO2 calibration gas cylinder seals	Technetics Helicoflex
AO2 calibration gas cylinder valves	Swagelok SS-0KM2-S2
AO2 regulators	Alphagaz 1001 / Scott Specialty Model 14
AO2 downstream pump	Vacuubrand MD Vario-SP
AO2 span and working tank control valves	MKS 248A with #6 orifice
AO2 mass flow meters	MKS M10MB23CP3BV
AO2 differential pressure sensors	MKS 223B-25038
AO2 changeover valves	ASCO 18801086
AO2 detector orifice	Beswick CJ-1010-008
AO2 bypass needle valve	Matheson HA-3
AO2 lamp	Ophos C3
AO2 lamp power supply	LCF A044
AO2 detector	Hamamatsu R1187
AO2 24-bit A/D boards	Micro/sys MPC 624
Inlet pumps	KNF N726, brushless DC
Cryotrop heat transfer fluid	3M Novec HFE-7500
CO ₂ sensors	LI-COR 840
Flame resistant insulation	Rohacell 110 S
Medusa pressure controllers	MKS 640
Medusa multiposition valves	VICI DLST16MWM and DL6UWM
Medusa downstream pump	KNF Type 814, brushless DC
Medusa individual flask lines	Eaton Synflex type 1300
Medusa flask o-ring grease	Apiezon type N

Table S2: AO2 and Medusa campaign history and instrument configuration details. Noted configuration changes persist for following campaigns, unless otherwise stated.

COBRA-1999test
Name: The CO ₂ Budget and Rectification Airborne Study, North America, 1999 test
Aircraft: University of North Dakota Citation II
Dates: Jun. 3-10, 1999
Flights: 6
Location: U.S.
Instrument: Medusa
Medusa configuration: Earlier version with open architecture, 16 flask capacity, no bypass line, no CO ₂ / H ₂ O sensor, and fewer diagnostic signals. Flask stopcocks closed at the end of the flight.
Inlet: 9.5 mm OD forward facing tube
Data availability: Work towards providing on a public repository is in progress. In the meantime, data are available by emailing the lead author.
COBRA-2000
Name: The CO ₂ Budget and Rectification Airborne Study, North America, 2000
Aircraft: University of North Dakota Citation II
Dates: Aug. 1-24, 2000
Research flights: 25
Location: U.S. and Canada
Instrument: Medusa
Medusa configuration: Same as for COBRA-1999test
Inlet: 9.5 mm OD forward facing tube
Data availability: Work towards providing on a public repository is in progress. In the meantime, data are available by emailing the lead author.
IDEAS-1
Name: Instrument Development and Education in Airborne Science, Phase 1
Aircraft: NSF/NCAR C-130
Dates: Apr. 5 - May 14, 2002
Flights: 8
Location: Broomfield, CO
Instruments: AO2 and Medusa

Configuration: Early prototype version of AO2. Medusa same as for COBRA-2000.

Inlets: 9.5 mm and 3.2 mm forward facing tube, and 9.5 mm aft facing tube

Data availability: Test data only

Website: <https://archive.eol.ucar.edu/raf/Projects/IDEAS-1>

IDEAS-2

Name: Instrument Development and Education in Airborne Science, Phase 2

Aircraft: NSF/NCAR C-130

Dates: Oct. 3 - Dec. 17, 2002

Flights: 8

Location: Broomfield, CO

Instrument: AO2

AO2 configuration: Early prototype version

Inlets: 9.5 mm and 3.2 mm forward facing tube

Data availability: Test data only

Website: <https://archive.eol.ucar.edu/raf/Projects/IDEAS-2>

COBRA-2003

Name: The CO₂ Budget and Rectification Airborne Study, North America, 2003

Aircraft: University of North Dakota Citation II

Dates: Apr. 25 - June 28, 2003

Research flights: 37

Location: U.S. and Canada

Instrument(s): Medusa

Medusa configuration: Same as for COBRA-2000

Inlet: 9.5 mm forward facing tube

Data availability: Work towards providing on a public repository is in progress. In the meantime, data are available by emailing the lead author.

ACME-04

Name: Airborne Carbon in the Mountains Experiment, 2004

Aircraft: NSF/NCAR C-130

Dates: May 14 - Aug. 2, 2004

Research flights: 16

Location: U.S. Rocky Mountains

Instrument(s): Medusa

Medusa configuration: Same as for COBRA-2003 but sampled smaller 100 ml flasks for U. Utah CO₂ isotopologue measurements.

Website: https://www.eol.ucar.edu/field_projects/acme

ACME-07

Name: Airborne Carbon in the Mountains Experiment, 2007

Aircraft: University of Wyoming King Air

Dates: May 3 - Aug. 9, 2007 (Jul. 27 - Aug. 9 for AO2)

Research flights: 18 (8 for AO2)

Location: U.S. Rocky Mountains

Instrument(s): AO2

AO2 configuration: First campaign for AO2 after initial build. Same as current configuration except: 1) no line purge cylinder, 2) included inlet fridge trap, 3) Synflex inlet tube, 4) pump outlet pressure control relative to gauge pressure and set to 8 psig, 5) 3.2 mm by 40 cm stainless steel U-tube second stage traps, 6) original inlet pump heads, 7) flow 140 sccm, 8) Li-840 downstream of second stage traps, and 9) 10 μ m screen between cell and downstream pressure transducer.

Inlet: 9.5 mm aft facing tube

Processing notes: Inlet pressure effect required empirical correction.

Data availability: AO2: <https://archive.eol.ucar.edu/homes/stephens/AO2>, Aircraft data: <http://flights.uwyo.edu/projects/acme07/data>

Website: <http://flights.uwyo.edu/projects/acme07>

START-08

Name: Stratosphere-Troposphere Analyses of Regional Transport, 2008

Aircraft: NSF/NCAR Gulfstream V

Dates: Apr. 18 - Jun. 27, 2008

Research flights: 18

Location: U.S. and Canada

Instrument(s): AO2 and Medusa

AO2 configuration: Same as ACME-07 but with 1) custom inlet pump heads and flow reduced to 115 sccm, 2) bypassed fridge trap before Research Flight 10, 3) moved inlet control valve to fuselage and replaced Synflex inlet tubing with 3.2 mm stainless steel before Research Flight 13, 4) changed to 9.5 mm ID inlet cryotrap before Research Flight 13, 5) 3.2 mm stainless steel second stage traps varied from 40-120 cm in length, and 6) absolute pressure control on pump outlet. Medusa configuration: First campaign after repackaging. Only sampled 16 flasks per flight. Flasks plumbed alternately in versus out the dip tube.

Inlets: AO2: HIMIL pylon on bottom of fuselage with internal 6.4 mm aft and forward facing tubes, primarily sampling from aft tube. Medusa: HIMIL pylon on top of fuselage with internal 6.4 mm aft facing tube.

Data availability: AO2: <https://doi.org/10.5065/D6DJ5CZ5>, Medusa flask data: <https://doi.org/10.26023/AWHD-19RV-GC14>, Medusa kernels: <https://doi.org/10.26023/CS11-P31Q-V713>, 1-Hz merge: <https://doi.org/10.5065/D61834TJ>

Websites: https://www.eol.ucar.edu/field_projects/start08, <https://www.acom.ucar.edu/start>

HIPPO-1

Name: HIAPER Pole-to-Pole Observations 1

Aircraft: NSF/NCAR Gulfstream V

Dates: Jan. 8 - Jan. 30, 2009

Research flights: 11

Location: Global, Pacific Basin

Instrument(s): AO2 and Medusa

Configuration: Medusa expanded to 32 flask capacity. AO2: Still using 9.5 mm ID inlet cryotrap and replaced inlet tubing with electropolished 2.2 mm ID stainless steel tubing.

Inlets: HIMIL pylons with internal 6.4 mm aft facing tubes, AO2 inlet now on top of fuselage.

Data availability: AO2: <https://doi.org/10.5065/D6J38QVV>, Medusa flask data: <https://doi.org/10.26023/J0VT-J67P-330R>, Medusa kernels: <https://doi.org/10.26023/4NM6-3MPG-WC14>, all HIPPO 10-sec merge: https://doi.org/10.3334/CDIAC/HIPPO_010, all HIPPO Medusa merge: https://doi.org/10.3334/CDIAC/HIPPO_014

Website: https://www.eol.ucar.edu/field_projects/hippo-1

HIPPO-2

Name: HIAPER Pole-to-Pole Observations 2

Aircraft: NSF/NCAR Gulfstream V

Dates: Oct. 31 - Nov. 22, 2009

Research flights: 11

Location: Global, Pacific Basin

Instrument(s): AO2 and Medusa

Configuration: Added vent tubes to HIMIL pylon struts. Moved Medusa inlet pressure controller to fuselage and replaced tubing from inlet to pump with 6.4 mm electropolished stainless steel. Added heater to Medusa trap inlet. All flasks plumbed to have flow out the dip tube.

Inlets: HIMIL pylons with internal 6.4 mm aft facing tubes

Data availability: AO2: <https://doi.org/10.5065/D65Q4TF0>, Medusa flask data: <https://doi.org/10.26023/30T9-FZ21-4G04>, Medusa kernels: <https://doi.org/10.26023/P4PE-KKYS-FZ07>, all HIPPO 10-sec merge: https://doi.org/10.3334/CDIAC/HIPPO_010, all HIPPO Medusa merge: https://doi.org/10.3334/CDIAC/HIPPO_014
Website: https://www.eol.ucar.edu/field_projects/hippo-2

HIPPO-3

Name: HIAPER Pole-to-Pole Observations 3

Aircraft: NSF/NCAR Gulfstream V

Dates: Mar. 24 - Apr. 16, 2010

Research flights: 11

Location: Global, Pacific Basin.

Instrument(s): AO2 and Medusa

AO2 configuration: First campaign using line purge cylinder. Re-implemented inlet fridge trap.

Inlets: HIMIL pylons with internal 6.4 mm aft facing tubes

Data availability: AO2: <https://doi.org/10.5065/D67H1GXJ>, Medusa flask data: <https://doi.org/10.26023/MYW6-DQQ6-PZ0R>, Medusa kernels: <https://doi.org/10.26023/GA02-K0FR-C10M>, all HIPPO 10-sec merge: https://doi.org/10.3334/CDIAC/HIPPO_010, all HIPPO Medusa merge: https://doi.org/10.3334/CDIAC/HIPPO_014
Website: https://www.eol.ucar.edu/field_projects/hippo-3

HIPPO-4

Name: HIAPER Pole-to-Pole Observations 4

Aircraft: NSF/NCAR Gulfstream V

Dates: June 14 - July 11, 2011

Research flights: 12

Location: Global, Pacific Basin.

Instrument(s): AO2 and Medusa

Configuration: AO2: Inlet fridge trap bypassed again. Replaced 3.2 mm electropolished tubing with 3.2 mm electropolished Sulfinert tubing. Medusa: flask stopcocks closed in flight soon after sampling.

Inlets: HIMIL pylons with internal 6.4 mm aft facing tubes

Data availability: AO2: <https://doi.org/10.5065/D679431D>, Medusa flask data: <https://doi.org/10.26023/XQW5-YHPP-XG0M>, Medusa kernels: <https://doi.org/10.26023/FF65-2RZM-ZB00>, all HIPPO 10-sec merge: https://doi.org/10.3334/CDIAC/HIPPO_010, all HIPPO Medusa merge: https://doi.org/10.3334/CDIAC/HIPPO_014
Website: https://www.eol.ucar.edu/field_projects/hippo-4

HIPPO-5

Name: HIAPER Pole-to-Pole Observations 5

Aircraft: NSF/NCAR Gulfstream V

Dates: Aug. 9 - Sep. 9, 2011

Research flights: 14

Location: Global, Pacific Basin.

Instrument(s): AO2 and Medusa

AO2 configuration: Changed from 2.5 to 3 min calibration periods, excluding first 2 min.

Inlets: HIMIL pylons with internal 6.4 mm aft facing tubes

Data availability: AO2: <https://doi.org/10.5065/D6WW7G0D>, Medusa flask data: <https://doi.org/10.26023/R8JN-Z3TG-2E0N>, Medusa kernels: <https://doi.org/10.26023/X9KY-CK34-VR10>, all HIPPO 10-sec merge: https://doi.org/10.3334/CDIAC/HIPPO_010, all HIPPO Medusa merge: https://doi.org/10.3334/CDIAC/HIPPO_014

Website: https://www.eol.ucar.edu/field_projects/hippo-5

ARISTO-2015

Name: Airborne Research Instrumentation Testing Opportunity 2015

Aircraft: NSF/NCAR C-130

Dates: Sep. 9 - Oct. 2, 2015

Flights: 5

Location: Broomfield, CO

Instrument(s): AO2 and Medusa

Configuration: AO2 and Medusa mounted to a vibration isolation plate for this campaign only. AO2: Secured loose wires inside lamp box before research flight 4. Removed sapphire window before research flight 5. Switched second stage traps to 60 cm coiled 3.2 mm electropolished stainless steel. Switched back to 1.6 cm ID inlet cryotrap.

Inlets: HIMIL pylon with internal 6.4 mm aft facing tube and 6.4 mm forward facing tube, and fin HIMIL with 7.9 mm aft facing tube.

Data availability: AO2: <https://doi.org/10.26023/PPR2-97CQ-N106>, Medusa flask data: <https://doi.org/10.26023/PV4Q-2XBJ-670C>, Medusa kernels: <https://doi.org/10.26023/2H6N-AZ1Y-N04>

Website: https://www.eol.ucar.edu/field_projects/aristo-2015

ORCAS

Name: O₂/N₂ Ratio and CO₂ Airborne Southern Ocean Study

Aircraft: NSF/NCAR Gulfstream V

Dates: Jan. 15 - Feb. 29, 2016

Research flights: 19

Location: Southern Ocean adjacent to South America and Antarctic Peninsula

Instrument(s): AO2 and Medusa

Configuration: AO2: Sapphire window moved to lamp side of detector cell. Started running line purge cylinder for 15 min on maintenance days to dry inlet tubing. Medusa: Implemented line purge cylinder for use purging flasks in preflight.

Inlets: fin HIMIL with 7.9 mm aft facing tube

Data availability: AO2: <https://doi.org/10.5065/D6N29VC6>, Medusa flask data: <https://doi.org/10.5065/D6H130FW>,

Medusa kernels: <https://doi.org/10.5065/D6MS3R6C>, 10-sec and Medusa merges: <https://doi.org/10.5065/D6SB445X>

Website: https://www.eol.ucar.edu/field_projects/orcas

ATom-1

Name: Airborne Tomography Mission 1

Aircraft: NASA DC-8

Dates: July 29 - Aug. 23, 2016

Research flights: 11

Location: Global, Pacific and Atlantic Basins.

Instrument(s): AO2 and Medusa

Configuration: Same as ORCAS but on NASA DC-8. Added inlet pressure sensor.

Inlets: fin HIMIL with 7.9 mm aft facing tube, 3.2 mm aft facing tube, 3.2 mm side facing tube, and 3.2 mm forward facing tube (see text)

Data availability: All ATom AO2: <https://doi.org/10.3334/ORNLDAAAC/1880>, All ATom Medusa flask data and kernels: <https://doi.org/10.3334/ORNLDAAAC/1881>, All ATom 10-sec and Medusa merges: <https://doi.org/10.3334/ORNLDAAAC/1581>

Website: <https://espo.nasa.gov/atom>

ATom-2

Name: Airborne Tomography Mission 2

Aircraft: NASA DC-8

Dates: Jan. 26 - Feb. 21, 2017

Research flights: 11

Location: Global, Pacific and Atlantic Basins.

Instrument(s): AO2 and Medusa

AO2 configuration: Sapphire window returned to detector side of cell.

Inlets: HIMIL solid-strut pylon with internal 6.4 mm (Medusa) and 3.2 mm (AO2) aft facing tubes

Data availability: All ATom AO2: <https://doi.org/10.3334/ORNLDAAAC/1880>, All ATom Medusa flask data and kernels: <https://doi.org/10.3334/ORNLDAAAC/1881>, All ATom 10-sec and Medusa merges: <https://doi.org/10.3334/ORNLDAAAC/1581>

Website: <https://espo.nasa.gov/atom>

ATom-3

Name: Airborne Tomography Mission 3

Aircraft: NASA DC-8

Dates: Sep. 28 - Oct. 27, 2017

Research flights: 13

Location: Global, Pacific and Atlantic Basins.

Instrument(s): AO2 and Medusa

AO2 configuration: Returned to 2.5 min calibration periods, excluding first 1.5 min. Removed 10 μ m screen between cell and downstream pressure transducer. Moved Li-840 upstream of second stage traps.

Inlets: HIMIL solid-strut pylon with internal 6.4 mm (Medusa) and 3.2 mm (AO2) aft facing tubes

Data availability: All ATom AO2: <https://doi.org/10.3334/ORNLDAAAC/1880>, All ATom Medusa flask data and kernels: <https://doi.org/10.3334/ORNLDAAAC/1881>, All ATom 10-sec and Medusa merges: <https://doi.org/10.3334/ORNLDAAAC/1581>

Website: <https://espo.nasa.gov/atom>

ATom-4

Name: Airborne Tomography Mission 4

Aircraft: NASA DC-8

Dates: Apr. 24 - May 21, 2018

Research flights: 13

Location: Global, Pacific and Atlantic Basins.

Instrument(s): AO2 and Medusa

AO2 configuration: Same as ATom-3

Inlets: HIMIL solid-strut pylon with internal 6.4 mm (Medusa) and 3.2 mm (AO2) aft facing tubes

Data availability: All ATom AO2: <https://doi.org/10.3334/ORNLDAAAC/1880>, All ATom Medusa flask data and kernels: <https://doi.org/10.3334/ORNLDAAAC/1881>, All ATom 10-sec and Medusa merges: <https://doi.org/10.3334/ORNLDAAAC/1581>
Website: <https://espo.nasa.gov/atom>

Table S3. Campaign mean corrections and offsets (see main text for details)

Parameter	S08 ¹	H1	H2	H3	H4	H5	ORC	A1a	A1b	A2	A3	A4
Medusa $\delta(\text{Ar}/\text{N}_2) > 800 \text{ hPa}$ (per meg)	37.9	3.1	3.5	-6.7	1.5	1.9	28.9	29.1	29.2	10.9	10.7	15.9
Scripps O ₂ Program global station $\delta(\text{Ar}/\text{N}_2)$ (per meg)	13.2	12.9	12.6	12.9	13.7	13.7	15.5	15.4	15.4	15.6	15.9	15.4
Medusa correction from $\delta(\text{O}_2/\text{N}_2)$ to $\delta(\text{O}_2/\text{N}_2)^*$ (per meg $\pm 1 \sigma$)	-6.5 ± 7.0	5.0 ± 5.3	5.8 ± 4.8	8.8 ± 7.8	6.2 ± 6.7	6.1 ± 5.9	-1.4 ± 6.8	-5.4 ± 5.2	-7.4 ± 6.2	2.6 ± 3.5	2.5 ± 3.4	1.0 ± 3.6
Medusa minus Scripps O ₂ Program station APO* (per meg $\pm 1 \sigma$, n)	–	4.5 $\pm 3.5, 3$	3.4 $\pm 6.1, 9$	4.5 $\pm 7.4, 9$	0.9 $\pm 12, 9$	-4.9 $\pm 9, 12$	1.8 $\pm 7.4, 9$	-2.9 $\pm 14, 7$	5.3 $\pm 0.1, 3$	-0.3 $\pm 4.0, 9$	1.7 $\pm 5.0, 5$	-2.2 $\pm 5, 13$
AO2 ascent-minus-descent adjustment pressure dependency ² (per meg atm ⁻¹)	–	5.1	-1.9	1.2	0.4	-0.7	-2.3	-7.7	-9.0	-10.1	-3.6	-4.7
Raw ³ AO2 $\delta(\text{O}_2/\text{N}_2)$ minus Medusa $\delta(\text{O}_2/\text{N}_2)^*$ (per meg $\pm 1 \sigma$)	11.6 ± 25.8	16.1 ± 8.3	-8.9 ± 9.9	-28.3 ± 11.7	-86.0 ± 11.4	-76.7 ± 9.3	3.8 ± 5.7	6.3 ± 8.5	15.2 ± 10.7	-2.8 ± 6.4	-3.2 ± 5.4	-5.8 ± 5.1
AO2 time-of-flight correction to match Medusa $\delta(\text{O}_2/\text{N}_2)^*$ (per meg hour ⁻¹)	-4.8	-2.1	-3.2	-2.0	-3.9	-3.7	2.1	-1.2	–	-0.7	-1.1	-1.0
AO2 pressure correction to match Medusa $\delta(\text{O}_2/\text{N}_2)^*$ (per meg atm ⁻¹)	–	–	–	–	–	–	–	–	34	–	–	–
Corrected AO2 $\delta(\text{O}_2/\text{N}_2)$ minus Medusa $\delta(\text{O}_2/\text{N}_2)^*$ (per meg $\pm 1 \sigma$)	1.6 ± 14.7	0.0 ± 5.7	0.0 ± 4.1	0.0 ± 4.2	-0.1 ± 5.1	0.0 ± 4.2	0.0 ± 4.5	0.1 ± 4.8	1.2 ± 7.9	0.0 ± 5.3	0.0 ± 4.2	0.0 ± 3.6
AO2 long-term surveillance cylinder $\delta(\text{O}_2/\text{N}_2)$ results (per meg $\pm 1 \sigma$)	-2.3 ± 8.1	0.3 ± 8.3	-2.4 ± 13.7	10.1 ± 6.3	-8.6 ± 7.4	-11.7 ± 5.8	0.1 ± 9.9	0.9 ± 3.7	-3.9 ± 4.2	1.8 ± 5.0	0.2 ± 1.6	-1.7 ± 1.7

¹Campaign names abbreviated as START-08 (S08), HIPPO 1–5 (H1–H5), ORCAS (ORC), ATom-1A (A1A), and ATom 2–4 (A2–A4)

²From fit to campaign mean values > 400 hPa

³After ascent-minus-descent adjustment but before Medusa-based time-of-flight or pressure corrections