1 Supplementary Text

- 2 Sea-air exchange calculations
- 3 To support the interpretation of our results, we calculate nominal equilibration times. For
- 4 estimates of bulk sea-air equilibration times for HVOCs, O₂, and CO₂, we assume a mixed layer
- 5 depth of 30 m, a temperature of 0° C, a salinity 35 PSU, and carbonate buffering according to eq.
- 6 8.3.10 in Sarmiento and Gruber (2006), and transfer velocities according to Nightingale et al.,
- 7 (2000). The Schmidt number (i.e. the ratio of the kinematic viscocity of a gas, divided by the
- 8 molecular diffusivity) for O_2 , CO_2 and CH_3Br were calculated according to Wanninkof (2014),
- 9 and the Schmidt numbers for CHBr₃ and CH₃I were calculated according to Quack and Wallace
- 10 (2003), and Moore and Groszko (1999), respectively. The results are provided in Sect. 2.5.
- 11 Comparisons of TOGA, WAS and PFP
- 12 Despite overall good agreement between co-located inflight AWAS, WAS, and PFP samples and
- 13 TOGA measurements, we observed notable discrepancies in several cases (e.g. Fig. S1b; Fig.
- 14 S2a-b). On ORCAS, we observed a non-linear relationship between inflight TOGA
- 15 measurements and co-located AWAS samples of CH₃I (Fig. S1b), driven by a few samples with
- 16 high mixing ratios. Close inspection of upwind and downwind flights over Region 2 with the
- 17 campaign's high mixing ratios of CH₃I indicated that TOGA measurements were consistent with
- 18 a modest flux of CH₃I from the ocean to the atmosphere. On ATom-2, TOGA measurements
- 19 agreed better with co-located PFP samples than with co-located WAS samples; and differences
- 20 on the sixth and seventh research flights (i.e. the data used here) were relatively small.
- 21 Nevertheless these differences motivated an instrument inter-comparison following the ATom
- 22 campaign between these instruments. Thus far, results of this inter-comparison show that TOGA
- and PFP measurements differ by < 25%.
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- 26 Supplementary Tables
- 27 Table S1. The TOGA-PFP instrument comparison was done by sampling a 50L SS pontoon,
- created at NCAR from a humidified dilution of the TOGA ATom standard. Data were analyzed
- and reported by Rebecca Hornbrook (NCAR, TOGA) and Steve Montzka (NOAA, PFP).

Pontoon Inter-	Concentration	TOGA (10/12/2018)	PFP (10/24/2018)
comparison	(dilution-based calc.)		
CHBr3	34	21.0 ± 0.1	26.6 ± 0.8
CHClBr2	26	19.9 ± 1.0	22.9 ±0.1
CH2Br2	52	47.7 ± 0.2	51.7 ± 2.0

33 Supplementary Figures

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Figure S1. Comparison between AWAS samples and TOGA measurements during ORCAS below 10 km, when these two shared over half their sampling period Points are colored by altitude. Dashed lines represent \pm 10% of the 1:1 line. Sample points below the DL are not included in this quantitative comparison.

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Figure S2. Comparison between WAS, PFP and TOGA measurements during ATom-2 below 10
km, when these instruments shared over half their sampling period. WAS measurements are
shown in larger circles, PFP measurements in smaller circles, and measurements from the
research flights six and seven used in this analysis are shown in color, while measurements on
other research flights in ATom-2 are shown in gray. Dashed lines represent ± 10% of the 1:1
line. Sample points below the DL are not shown.



57 Figure S3. Mixing ratios of CHBr₃, CH₂Br₂ and CH₃I vs. CO₂ in Region 1 (a-c) and Region 2 (d-

58 f). Type II major axis regression model (bivariate least squares regression) fits are shown for

combined ORCAS and ATom-2 data, using data below 7 km for $CHBr_3$, CH_2Br_2 , and below 2

- km for CH₃I. Fossil fuel and terrestrial CO2 signals were not subtracted prior to regressions.



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Figure S4. Consecutive samples in and out of dips into the MBL on two different days (a-c; Jan. 66 21,2016, and d-f; Jan. 30, 2016). For illustrative purposes, sampling locations are denoted by a black circle, 24-hour back trajectories are shown in red, and surface influences are shown with 67 68 black squares in each subpanel, overlying weekly composites of remotely sensed chl. a. Surface 69 influence is multiplied by the underlying chl a (or other) field and averaged for each sample to

70 yield a geostatistical surface influence.

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