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Transport of Gas-Phase Anthropogenic VOCs to the Remote Troposphere during the NASA ATom Mission

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Atmospheric Tomography (ATom) Mission

Primary questions: What are chemical processes that control the short-lived climate forcing agents CH_4, O_3, and BC in the atmosphere? How is the chemical reactivity of the atmosphere on a global scale affected by anthropogenic emissions? How can we improve chemistry-climate modeling of these processes?

Study Regions:

1. PACIFIC: < 70°W
2. ATLANTIC: > 70°W
3. ARCTIC: > 60°N, separated at 100°W
4. CONUS: 23°N to 60°N, 125° to 54°W

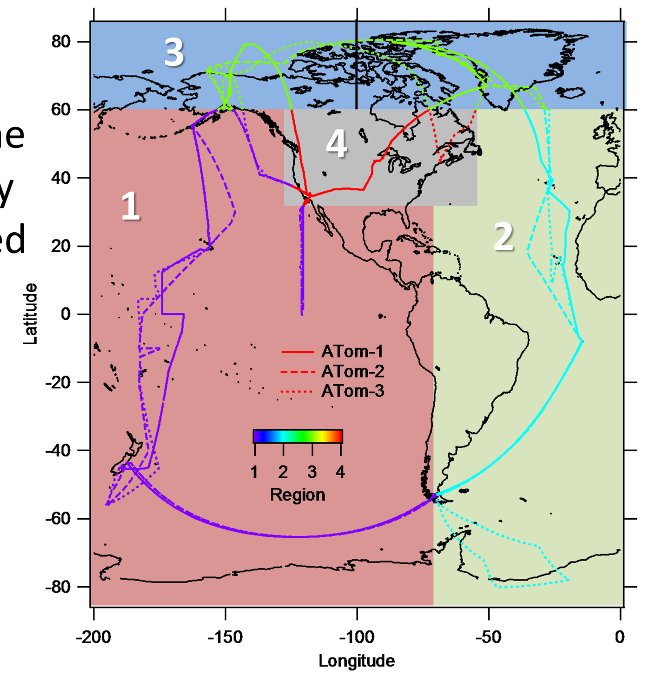


Figure 1. ATom-1, -2, and -3 NASA DC-8 flight tracks and study regions.

Trace Gas Measurements

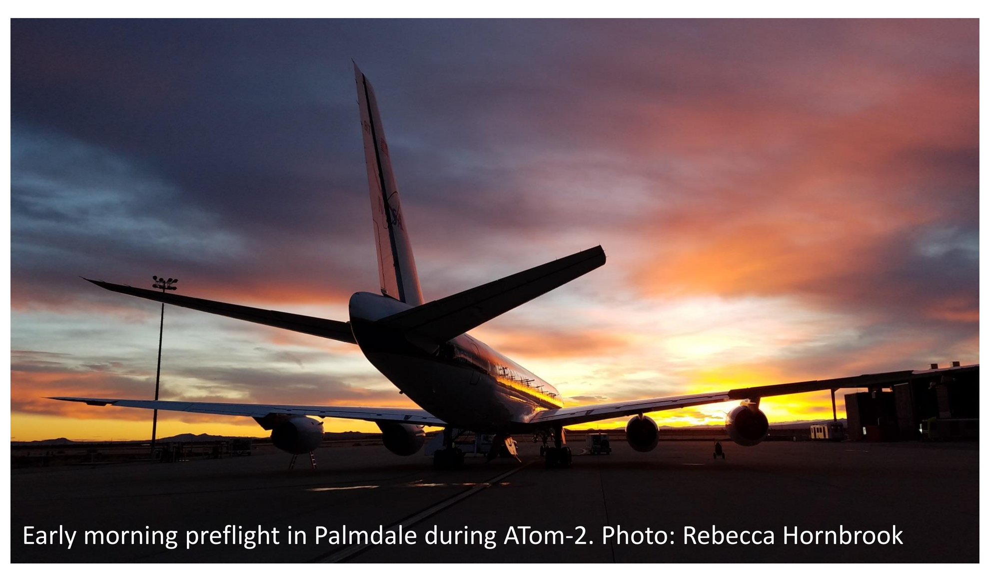
For the four ATom deployments, VOCs (Volatile Organic Compounds) and other trace gases are being measured by a large payload of instruments.

Table 1. Selected instruments in the ATom NASA DC-8 payload.

Instrument	Instrument Description	PI	Species Measured
TOGA	Trace Organic Gas Analyzer; gas chromatography/mass spectrometry	Eric Apel (NCAR)	NMHCs, OVOCs, Halocarbons, CH_3CN, HCN, DMS
WAS	Whole Air Sampler, canister sampling and laboratory analysis.	Don Blake (UCI)	NMHCs, Halocarbons, Alkyl Nitrates, OCS, DMS, CS_2
$NOyO_3$	NOAA Nitrogen Oxides and Ozone, chemiluminescence	Tom Ryerson (NOAA)	NO, NO_2, NOy, O_3
PANTHER/ UCATS	PAN and Trace Hydrohalocarbon ExpeRiment/UAS Chromatograph for Atmospheric Trace Species	Jim Elkins (NOAA)	PANTHER: $(CH_3)_2CO$, PAN, H_2, CH_4, CO, N_2O, SF_6, $CFCl_3$, CF_2Cl_2, Halon-1211 UCATS: N_2O, SF_6, CH_4, CO, O_3
NOAA Picarro	NOAA Picarro, wavelength-scanned cavity ring down spectroscopy	Kathryn McKain (NOAA)	CO_2, CH_4, CO
PFP	Programmable Flask Package glass flask automated whole air sampler and laboratory analysis.	Steve Montzka (NOAA)	N_2O, SF_6, H_2, CS_2, OCS, CO_2, CH_4, CO, CFCs, HCFCs, HFCs, Solvents, Methyl Halides, Hydrocarbons, Perfluorocarbons
QCLS	Quantum Cascade Laser System	Bruce Daube (Harvard)	CO_2, CO, CH_4, N_2O

Global Model

- **CESM1**: Community Earth System Model, version1
- **CAM-chem** (Community Atmospheric Model Version 5.4 with comprehensive tropospheric and stratospheric chemistry) 0.9° x 1.25° horizontal resolution
- **MOZART TS1** chemical mechanism
- **HTAP2** 2010 anthropogenic emissions used for every year
- **FINN** (Fire INventory from NCAR) daily fire emissions
- **MEGAN** (Model of Emissions of Gases and Aerosols from Nature) biogenic emissions



Early morning preflight in Palmdale during ATom-2. Photo: Rebecca Hornbrook

MMS and TOGA on DC-8
Photo: Paul Bui

VOCs during ATom-1 (Aug 2016) and ATom-2 (Feb 2017)

The ATom mission is providing global seasonal cross-sections of a large suite of gases in the remote regions of the troposphere. These allow for seasonally-based analyses of transport and chemistry impacting regions far removed from emission sources, providing opportunity for evaluation and improvement of our emissions inventories and photochemical modeling capabilities.

Figure 2 contrasts NH Summer and NH Winter measurements of selected primary anthropogenic VOCs (n-butane, benzene, CH_2Cl_2, MTBE; methyl t-butyl ether), and OVOCs both directly emitted and photochemically produced (MEK, acetone). North-south, east-west, and seasonal differences exist in most VOC observations due to a variety of factors including primary emission region, OH lifetime, and seasonal changes in photochemical production and loss.

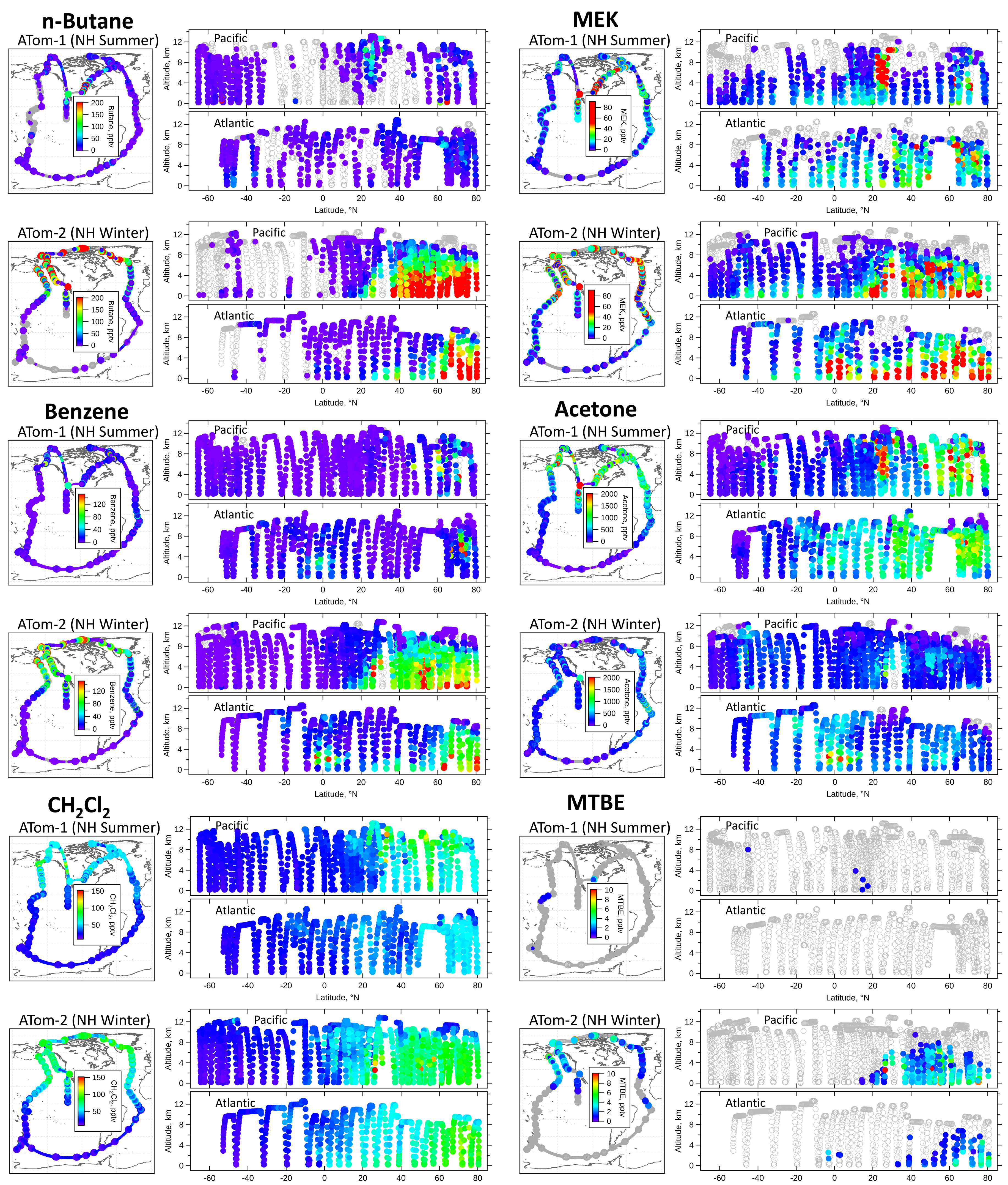


Figure 2. TOGA observations from ATom-1 and -2 of selected VOCs shown on a global map in which the largest points indicate the lowest altitude data, and by altitude-latitude separated into Pacific + western Arctic (top) and Atlantic + eastern Arctic (bottom) for each VOC for each deployment. Points in grey are below the detection limit.

Comparisons with the Global Model

CAM-chem model output along the flight tracks agree fairly well with observed O_3 and CH_2O (not shown). In Figure 3, the model compound BIGALK agrees very well with the observed sum of butanes in the remote troposphere where alkanes > C_4 are low. Modeled and observed benzene agree reasonably well in the NH, but the model predicts a large benzene signal in the tropical Atlantic region that was not observed. A significant summer to winter difference in acetone was observed in the NH mid-latitudes and Arctic region, presumably due to differences in photochemical production, but this was not predicted by the model. The model far overpredicts MEK in the NH during NH winter (not shown), but as the MOZART TS1 MEK is a composite of ketones produced from BIGALK oxidation, it is difficult to directly compare to observed MEK.

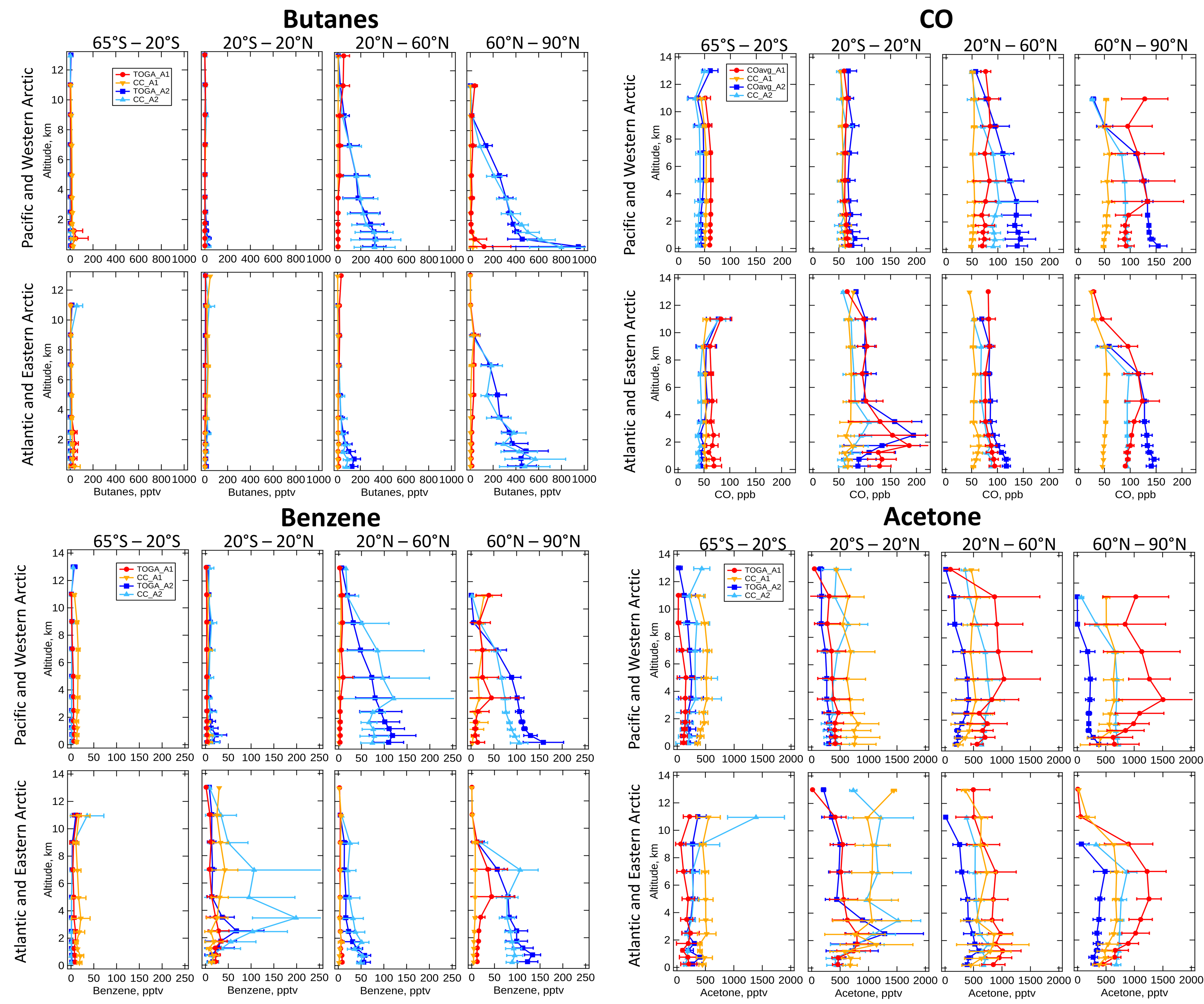
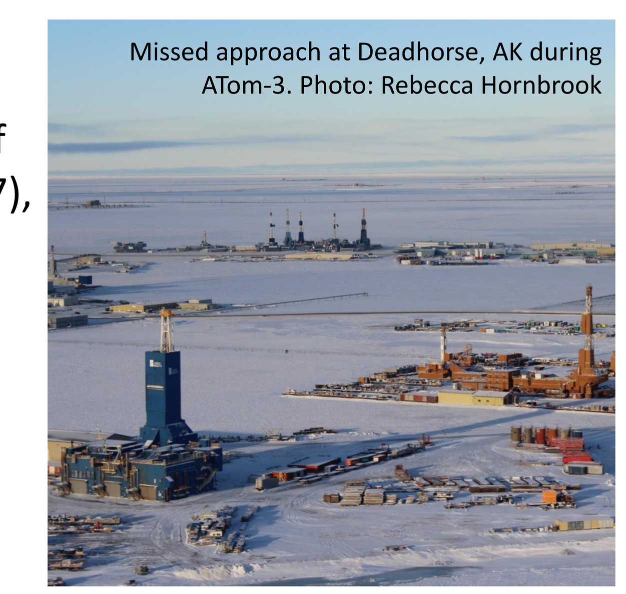


Figure 3. Measurements and model output for selected VOCs and CO, binned by altitude, latitude bands, and study region. "Butanes" is the sum of TOGA isobutane and n-butane and the C_{4+} alkane model compound BIGALK. CO_{avg} is the mean of the TOGA-merge NOAA CO and QCLS CO.

Summary

- The ATom mission has provided a comprehensive suite of observations in the remote troposphere that demonstrates seasonal and hemispheric variability in the atmospheric loading of trace gases. With two more deployments (May 2018 and Oct 2017), the resulting data set will be unprecedented.
- Comparison with a CAM-chem model simulation shows areas of good agreement while highlighting several areas where the emission inventories and chemistry need improvement. The addition of long-lived species (e.g., CH_2Cl_2), and the inclusion of speciated butanes and their oxidation products will allow assessment of both inventories and chemistry using this data set.



Missed approach at Deadhorse, AK during ATom-3. Photo: Rebecca Hornbrook