Global seasonal distributions of HCN and acetonitrile

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Atmospheric Tomography (ATom) mission

NASA's Earth Venture suborbital (EVS-2) ATom dataset provides a unique opportunity to investigate the global distributions of the biomass burning (BB) tracers HCN and CH_3CN during four different seasons spanning 2016 through 2018. Here we assess the distributions of these BB tracers in the remote troposphere during the first two ATom deployments, and compare to CAM-chem, a global chemical model, using two different BB emissions inventories.

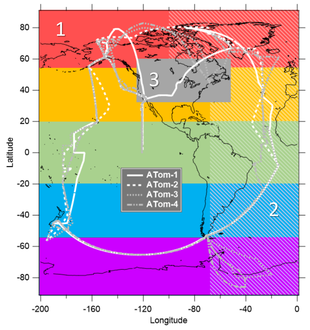


Figure 1. ATom-1 to -4 NASA DC-8 flight tracks and study regions.

Study Regions:

1. **PACIFIC/WEST ARCTIC**: solid regions, 5 lat. bands
2. **ATLANTIC/EAST ARCTIC**: hashed regions, 5 lat. bands
3. **CONUS**: 23°N to 60°N, 125° to 54°W

Gas-phase biomass burning tracer observations

During the four ATom deployments, biomass burning trace gases including HCN, CH_3CN, and CO were measured by several *in situ* 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
CIT-CIMS	Chemical Ionization Mass Spectrometer	Paul Wennberg (CalTech)	HNO_3, H_2O_2, CH_3OOH, MHP, HCN, PAA, PNA, SO_2
NOAA Picarro	Picarro Inc. wavelength-scanned cavity ring down spectroscopy	Kathryn McKain (NOAA)	CO_2, CH_4, CO
QCLS	Quantum Cascade Laser System	Bruce Daube (Harvard)	CO_2, CO, CH_4, N_2O

Global chemistry-climate model

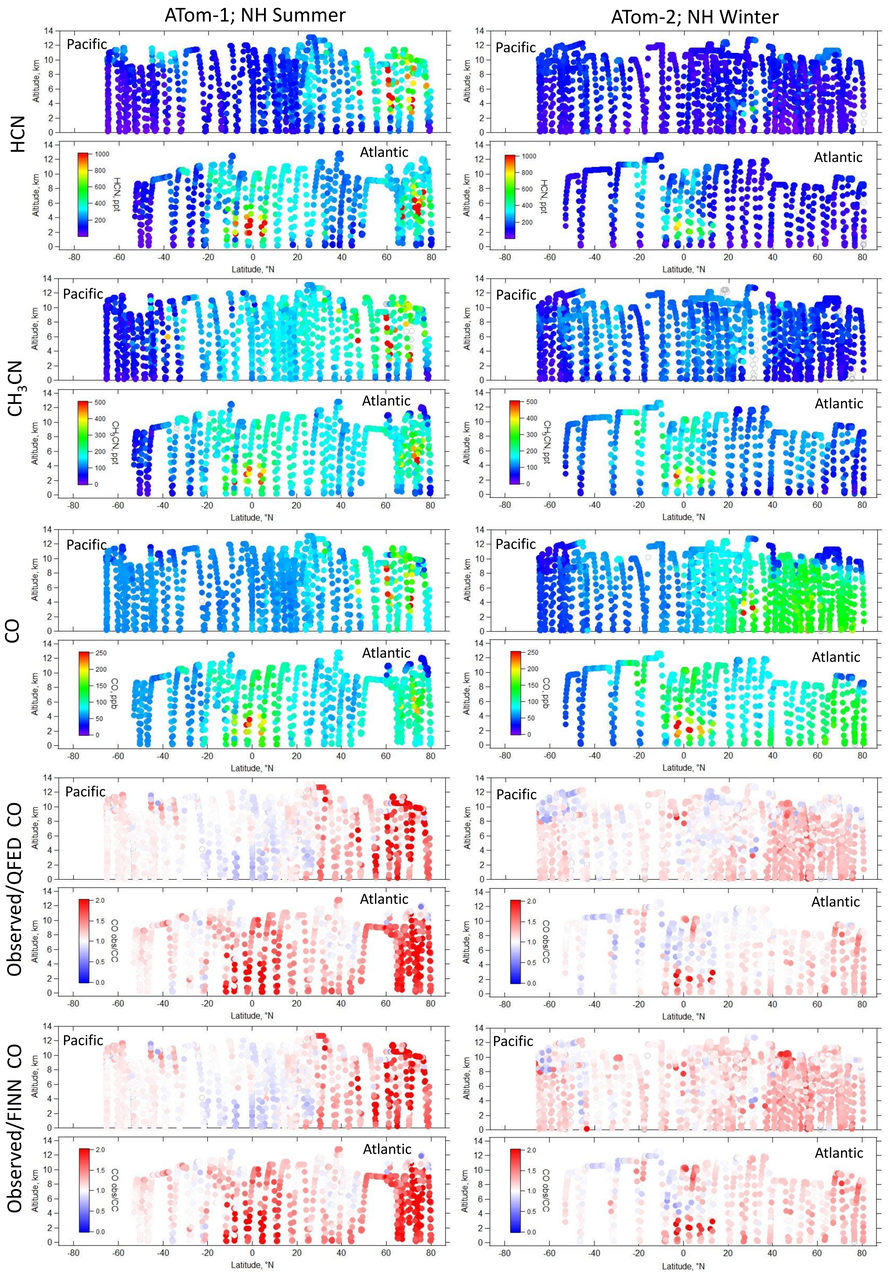
- **CESM2**: Community Earth System Model, Version 2.1, exp003
- **CAM-chem** (Community Atmospheric Model Version 6.0 with comprehensive tropospheric and stratospheric chemistry) 0.9° x 1.25° horizontal resolution
- **Meteorology** nudged to MERRA2 meteorological reanalysis
- **CMIP6** 2014 anthropogenic emissions used for every year
- **MEGAN** (Model of Emissions of Gases and Aerosols from Nature) biogenic emissions; calculated online
- **Biomass Burning emissions inventories:**
  - **FINN** (Fire INventory from NCAR) daily fire emissions (Wiedinmyer et al., Geosci.Model Dev., 2011)
  - **QFED** (Quick Fire Emissions Dataset) non-native emissions created using FINN emission factors and QFED CO emissions (NASA GMAO)


Thick biomass burning layer during ATom-1 near ASI, Photo: Rebecca Hornbrook


MMS and TOGA on DC-8 Photo: Paul Bui

Biomass burning gas-phase tracers in ATom-1 and ATom-2

Figure 2. NH Summer (August 2016) and NH Winter (February 2017) TOGA-merge observations of BB tracers HCN (average of CIT-CIMS and TOGA), Acetonitrile (CH_3CN; TOGA), and CO (average of QCLS and Picarro). Stronger BB emissions influenced the tropical Atlantic and the Arctic in August 2016. In the QFED and FINN model simulations, the CO measurement/model comparison are reasonable in most regions except for the NH Summer Arctic, where the models underestimate the observed CO; FINN failed to capture the enhanced CO in the tropical Atlantic during ATom-1.



HCN and CH3CN comparisons to global chemistry model

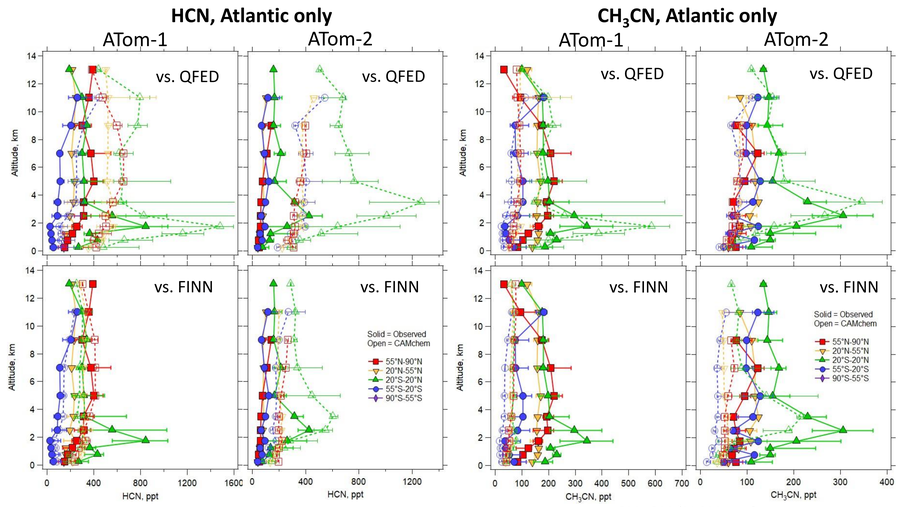


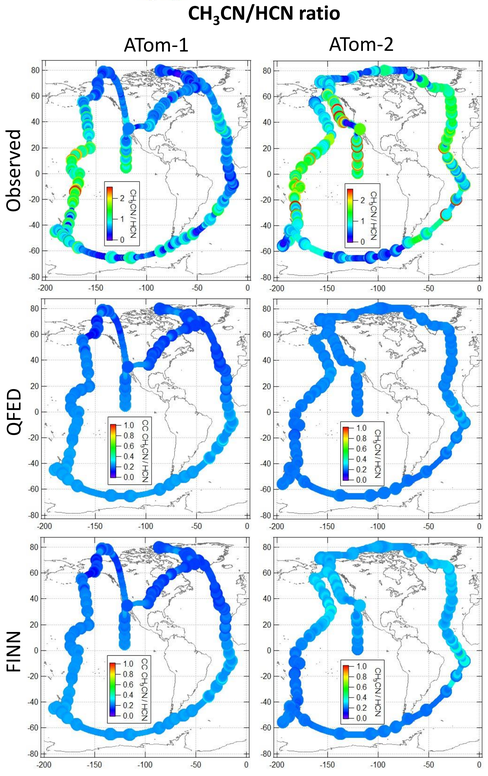
Figure 3. (above) Latitudinally and vertically binned observed and modeled Atlantic HCN and CH_3CN medians (and 25th and 75th percentiles) for ATom-1 and -2.

QFED overestimates HCN globally by approximately a factor of 2 during ATom-1 and more during ATom-2, but underestimates CH_3CN in ATom-1 except for in the tropical Atlantic (African BB).

FINN HCN is closer to observed globally, but also underestimates CH_3CN. FINN fails to capture the African BB during ATom-1, but represents it fairly well for both HCN and CH_3CN during ATom-2.

Figure 4. (right) Observed and modeled ratios of CH_3CN/HCN for ATom-1 and -2. (Note color scale range for observed ratio is 2.5x greater than modeled). Observed CH_3CN/HCN are 0.3-0.5 in regions with recent BB emissions, but much higher in remote regions, indicating differences in the loss rates of these species.

These differences are not captured by CAM-chem simulations with either QFED or FINN BB emissions inventories, indicating that the actual atmospheric loss mechanisms (reaction with OH and deposition to the surface) of CH_3CN and HCN are not as similar to and more nuanced than the model predicts.



Summary

- While both BB inventories capture the CO distribution fairly well, QFED overpredicts HCN and underpredicts CH_3CN, but predicts the African BB more reliably than FINN.
- CAM-chem predicts the ratio of CH_3CN/HCN to remain consistent globally, while the observations indicate loss mechanisms and differences in the losses that are not included in the model.