QOS2016-223: Convective influence on the CH_3I distribution in the Tropical Tropopause Layer in the Pacific Ocean

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1. Introduction and Goals

- Convection and chemistry control the composition of the tropical troposphere as well as the air that enters lower stratosphere.
- Understanding these processes involves modelling interactions between ocean biology, atmospheric composition and climate change.
- UK Met Office Numerical Atmospheric-dispersion Modelling Environment (NAME) model is used to assess spatial and temporal variability in transport of very short-lived halogenated organic substances (VSLS): methyl iodide, CH3I.
- The convective influence on the CH3I in the TTL is investigated quantitatively using NAME.
- NAME results are compared with airborne tracer measurements taken during NASA ATTREX in 2013 & 2014 made in conjunction with NCAR CONTRAST and NERC CAST campaigns.

2. CAST-CONTRAST-ATTREX

- CAST aimed to characterise distribution and emission sources of VSLS in the tropical lower troposphere. CONTRAST and ATTREX aircraft flew at higher altitudes to sample the mid/upper-troposphere and lower stratosphere to study convective outflow and its impact on UTLS composition.
- NAME: (i) tool for flight planning and flight coordination (pre- and in-campaign); and (ii) post-campaign analysis of tracer data .

Research Campaign	Research Aircraft	Time and location	Altitude range [km]	Research Flights	AWAS/GWAS samples
NASA **ATTREX II**	Global Hawk	Jan-Mar 2013 Dryden	14-19	6	388
NASA **ATTREX III**		Jan-Mar 2014 Guam		8	670
NCAR **CONTRAST**	Gulfstream V	Jan-Feb 2014 Guam	0-12	16	715
NERC **CAST**	BAe-146		0-6	24	695

4. NAME modelling activities for ATTREX – RESULTS

- ATTREX III 2014, Research Flight 03, 16-17/02/2014

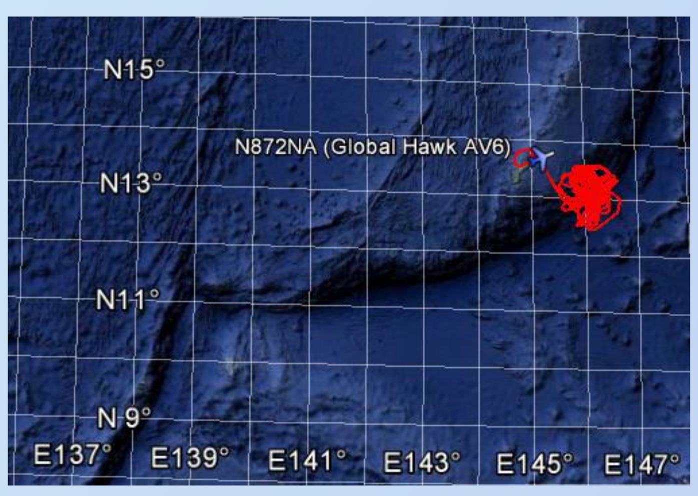


Figure 2. The ATTREX III RF03 flight path.

- the Global Hawk made 20 TTL profiles at the single point location (east off Guam, the US).
- Unique sampling of TTL over 18 hours.
- CH3I observations: high degree of variability in the TTL.
- Highest at 14-15 km, decreasing with altitude.

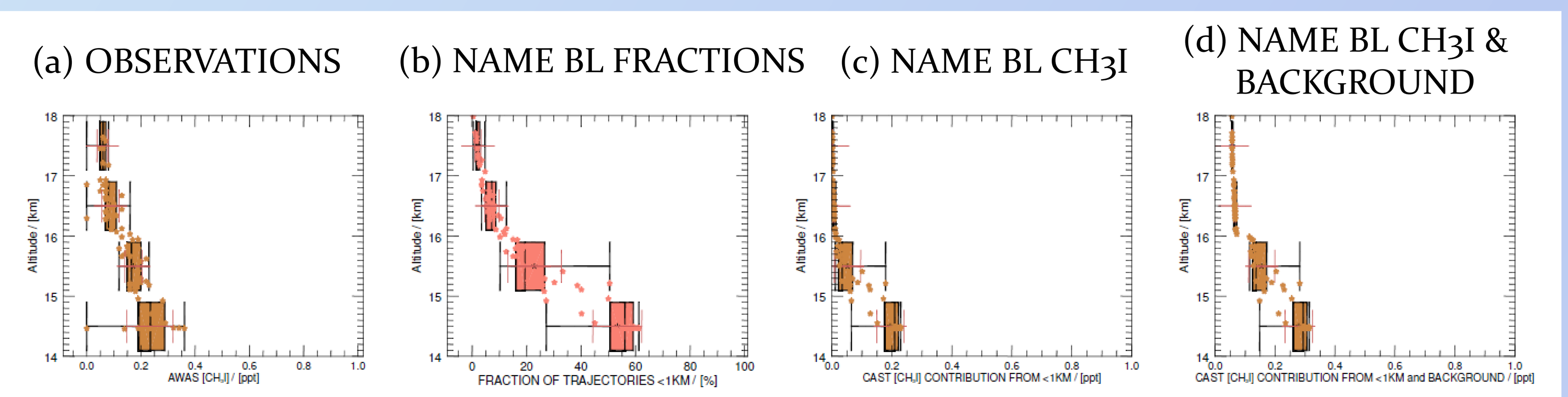


Figure 3. The ATTREX III RF03 TTL 1-km profiles for (a) CH3I AWAS observations, (b) the 1 km fractions of trajectories, (c) boundary layer CH3I contribution from the 1 km fractions, and (d) the sum of the boundary layer and the background CH3I concentration estimates.

- The calculated fraction is highest for 14-15 km (Fig. 3(b)), indicating significant contribution of 'fresh' air masses recently lofted from the boundary layer.
- The fractions drop off with altitude indicating considerably less convective influence.
- The calculated boundary layer CH3I contribution (Fig. 3(c)) from the 1 km fractions shows highest values for the 14-15 km, dropping off with altitude (indicative of higher fractions bringing higher CH3I concentrations aloft).
- At 14-15 km, the modelled contribution of boundary layer CH3I explains most of the observed AWAS concentrations. At higher altitudes, the measurements are higher than the boundary layer estimates.
- A true comparison with observations also requires an estimate of the background TTL CH3I concentration.
- The background is estimated in 2 ways, both using NAME calculations to identify AWAS samples in all 2014 flights which had small convective influence:
  (a) Boundary layer fraction values less than 5%;
  (b) Lowest 10% of boundary layer fractions. This had to be used in 2014 as no data available for (a).
- The sum of the NAME boundary layer and the background CH3I estimates (Fig. 3(d)) are in good agreement with the AWAS observations.
- The background component may be an overestimate since it is hard to identify samples with no convective influence. This is especially true at the lower altitudes since the ATTREX 2014 flights were close to regions of strong convection.

References

1. Meneguz E., Filus M., Harris N., et al., (2016), *Improved parametrization scheme to represent tropospheric moist convection in the atmospheric dispersion model NAME*, JAMC (submitted).
2. Navarro M., Harris N., Filus M., et al., (2015), *Airborne measurements of organic bromine compounds in the Pacific Tropical Tropopause Layer*, PNAS (doi: 10.1073.pnas.1514631112).
3. Harris N., Filus M., et al., (2016), *Co-ordinated Airborne Studies in the Tropics (CAST)*, BAMS (doi: 10.1175.bams-d-14-00290.1).
4. Ashfold M., Harris N., et al., (2012), *Transport of short-lived species into the Tropical Tropopause Layer*, ACP (doi: 10.5194.acp-12-6309-2012).

3. NAME modelling approach for ATTREX

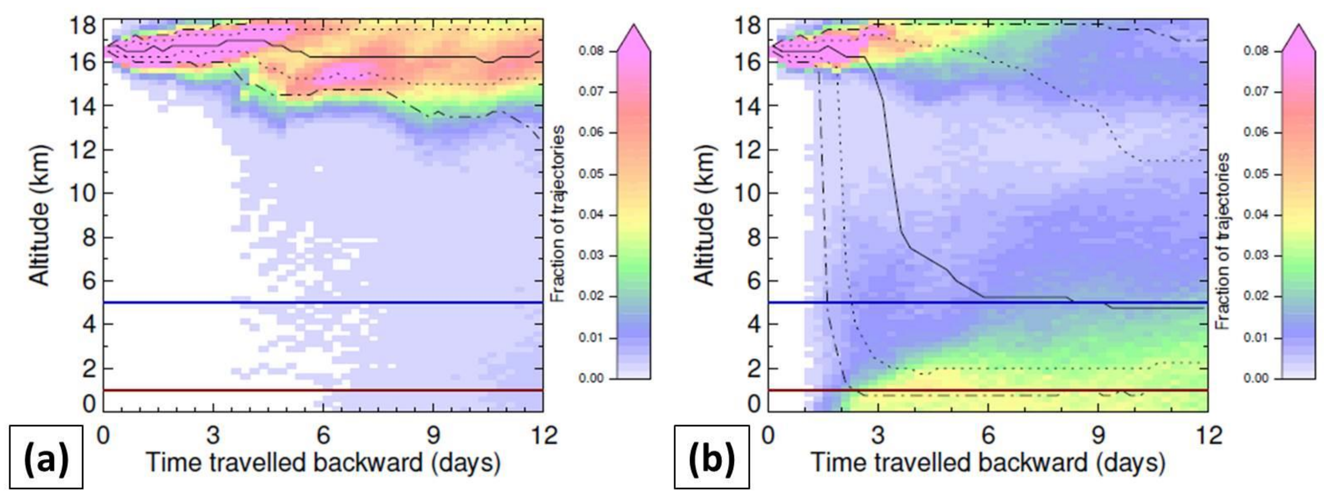
- NAME is a Lagrangian Particle Dispersion model developed by the UK Met Office
- We use the Unified Model global wind fields at 25km resolution (*17km from 2016)
- 12-day back trajectories with new convection scheme[1]
- 15,000 particles released from each AWAS sampling location along all ATTREX Global Hawk flight tracks
- Calculate fractions of trajectories which crossed below 1 km (and 5 km, not shown) in previous 12 days, as a quantitative measure of the boundary layer air mass contribution to those samples (Fig. 1)
- Assign initial CH3I concentrations (from the CAST observations) to trajectories < 1 km and calculate the CH3I contribution from the boundary layer, allowing for the decay of CH3I in each trajectory since it was last < 1 km:

The 1 km Contribution; Fraction of trajectories in time interval

$$[X]_{0-1\_to\_TTL} = [X_0]_{0-1km} \times \sum Fraction_t \times e^{(t/\tau)}$$

Initial Tracer Concentration; Atmospheric Lifetime

- Compare NAME model results with ATTREX AWAS measurements of CH3I



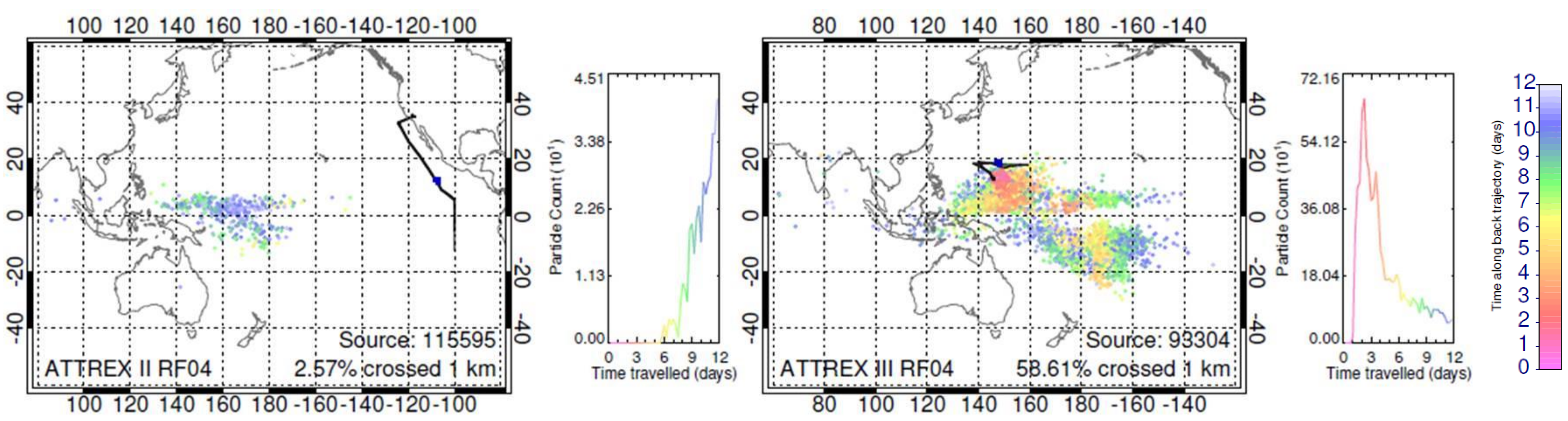


Figure 1. Trajectory information derived for one AWAS sample in 2013 (a, SRC115595, ATTREX II RF04, Alt: 16.54km - left) and one in 2014 (b, SRC93304, ATTREX III RF04, Alt: 16.58km - right).

The top row shows the altitude dependence of the particle density over the previous 12 days; the bottom row displays the 1 km fractions of trajectories and the locations where they first crossed 1 km. The location of the AWAS sample is marked as a blue square, and the adjacent histogram shows the 1km trajectory crossing time distribution.

5. NAME modelling activities for ATTREX – RESULTS

- All ATTREX III 2014 Research Flights

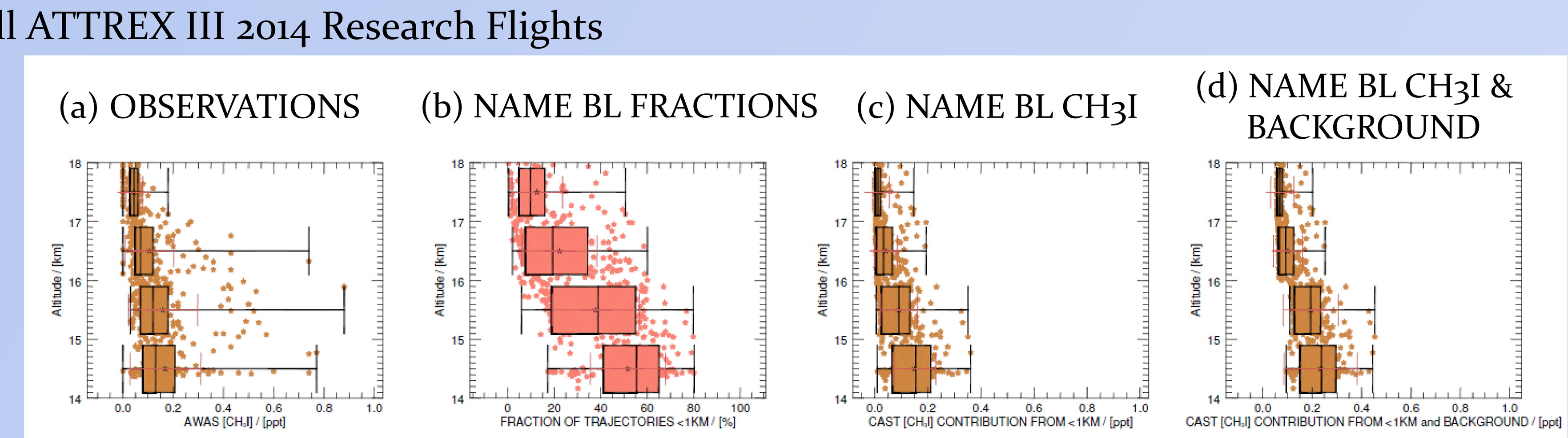


Figure 4. All the ATTREX III 2014 Research Flights: TTL 1-km profiles for (a) CH3I AWAS observations, (b) the 1 km fractions of trajectories, (c) boundary layer CH3I contribution from the 1 km fractions, and (d) the sum of the boundary layer and the background CH3I concentration estimates.

- Large flight-to-flight variability (Fig. 4(a)). As in RF03, the 1 km fractions drop off with altitude (Fig. 4(b)).
- Boundary layer CH3I contribution can explain most of the observations for the 14-15 and 15-16 km, but there are bigger differences above 16 km.
- The sum of the NAME boundary layer and the background CH3I estimates (Fig. 4(d)) show reasonable agreement with AWAS observations.
- Comparison of ATTREX II (2013) and ATTREX III (2014)

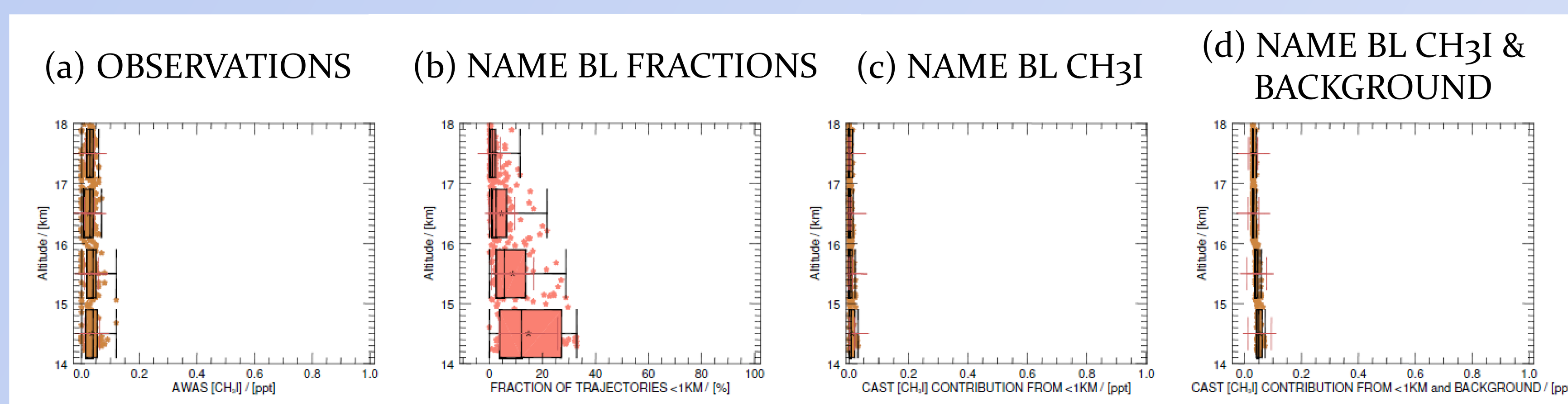


Figure 5. All the ATTREX II 2013 Research Flights: TTL 1-km profiles for (a) CH3I AWAS observations, (b) the 1 km fractions of trajectories, (c) boundary layer CH3I contribution from the 1 km fractions, and (d) the sum of the boundary layer and the background CH3I concentration estimates.

- CH3I observations, boundary layer fractions and related CH3I contributions are considerably lower than in 2014.
- Background component significantly lower (the 5% approach), close to the limit of detection[2].
- The sum of the boundary layer contribution and background CH3I estimates match the AWAS observations.
- ATTREX II flights were in East Pacific away from the main region of strong convection.
- Longer transport timescales were seen as horizontal transport more important in ATTREX 2013, with much less recent convective influence than in ATTREX 2014. More chemical removal of CH3I occurs before sampling, leading to lower CH3I concentrations.

6. Results and on-going work

- Approach developed to estimate relative contributions of air with different origins for comparison with observations, and used to study the influence of convective transport on the TTL.
- Higher CH3I concentrations observed in ATTREX 2014 are explained by much more recent convective uplift from the boundary layer than in ATTREX 2013. In ATTREX 2013, the longer transport times resulting from the horizontal transport in the TTL led to more photochemical destruction and lower CH3I concentrations.
- Good quantitative agreement between modelled estimates and AWAS observations in 2014 using the NAME new convection scheme[1].
- This approach is now being applied to study the budget of the longer-lived substances CHBr3 and CH2Br2, where an accurate assessment of both the background and boundary layer contribution is important.