Bromoform and Tetrachloroethene in the Tropical Boundary Layer of the Maritime Continent

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Danum GAW inland site


Darvel Bay from Kunak

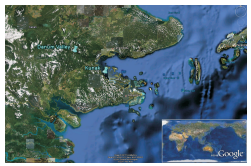
1) Overview

- Halogen source gases are important in stratospheric and tropospheric chemistry processes[1]. Sources are natural (marine and terrestrial) and anthropogenic.
- The west Pacific is an important source region for halocarbons (sea surface temperature and primary productivity are high) yet few observations have been made.
- The maritime continent is an important region for large scale convection, potentially lifting halocarbons rapidly from the surface to the tropopause.
- We compare observations of bromoform ($CHBr_3$) and tetrachloroethene (C_2Cl_4) at a coastal and an inland site, visited in June/July 2008. Then we show how two different models can reproduce some of the observed features of the data[2].

2) Instrument and measurement sites

- Our GC-ECDs measure a range of halocarbons[3] including short-lived compounds of marine origin: $CHBr_3$, $CHCl_3$, $CHCl_2Br/CH_2Br_2$, $CHBr_2Cl$ and CH_2CII.
- Typical precision is ± 5-10 % (1 s.d.) with calibration against a NOAA-ESRL standard.
- Sample frequency is 15-45 minutes and depends on mode of operation and column length.


photo: John Hofman / Nathan Pitt



- Danum Valley GAW site (5.0°N, 117.8°E) is ~400 m above sea level and ~35 km inland.
- Kunak (4.70°N, 118.24°E) is a small town on the coast in Darvel Bay.

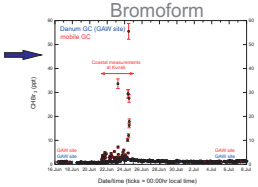
3) The models

- Numerical Atmospheric dispersion Modelling Environment (NAME):
  - Lagrangian dispersion model developed by the UK Met Office
  - abstract particles move through model using wind fields from UKMO unified model
  - also uses random walk turbulence scheme
  - use it backwards in time (e.g. air mass origin) or forwards in time (e.g. plume dispersion)
- p-TOMCAT running at high horizontal resolution (0.5° x 0.5°):
  - tracer transport based on 6 hourly meteorological fields (ECMWF)
  - bromoform emissions:
    - on uniform latitude bands over tropical/extra tropical oceans
    - also with tropical coastline emissions
  - bromoform loss by OH and photolysis using pre-calculated time-varying 3-D fields

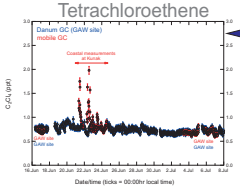
4) The measurements

- Instruments show nearly identical readings at Danum.
- $CHBr_3$ level at Danum is low (1-2 ppt) but at Kunak the baseline is 2-5 ppt and with very high spikes (max ~60 ppt).
- When the mobile GC returns to Danum it still shows an excellent agreement with the GAW site GC.

Bromoform

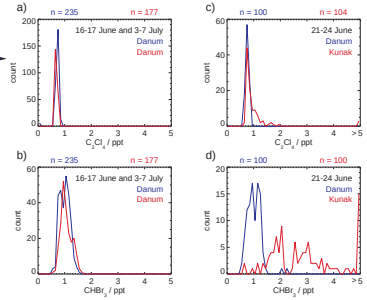


Tetrachloroethene



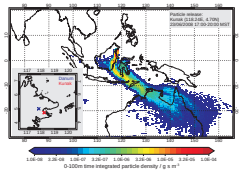
- The C_2Cl_4 baseline is very similar at both sites (and for both instruments).
- Spikes in C_2Cl_4 at Kunak are likely due to proximity of the town (this is a predominantly man-made tracer).
- C_2Cl_4 does not correlate with $CHBr_3$, they have different sources.

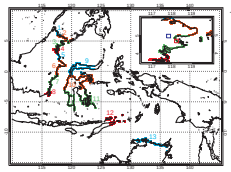
- Probability density functions (pdfs) show excellent instrument agreement at Danum for C_2Cl_4 and $CHBr_3$ (a & b).
- At Kunak the C_2Cl_4 baseline matches that at Danum and the polluted events are seen as a tail of higher values (c).
- $CHBr_3$ at Kunak shows a much broader spectrum, it's difficult to deduce a baseline (d). Notice the tail of very high values >5 ppt.



5) Modelling and interpretation

- What can NAME tell us? When run backwards it tells us that the large scale flow is south-easterly passing over potentially rich emission regions.
- But this figure cannot explain why $CHBr_3$ at Kunak is higher than at Danum.

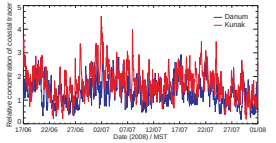




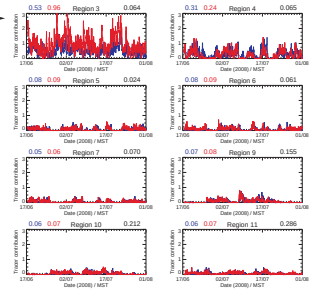
- So let's run NAME forwards in time. From each of these 13 coastal regions we release a tracer which has an e-folding time of 15 days (similar to $CHBr_3$). Emissions per unit area are constant but the regions do not have the same areas. $CHBr_3$ also has an open ocean source, here we focus on the coastal source.
- We can sum the individual tracers arriving at each location (Danum and Kunak) to get the total tracer concentration there.

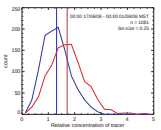
5) Modelling and interpretation (continued)

- As with the observations, the tracer results at Danum (mean 1.31 ppt) are lower than at Kunak (mean 1.72 ppt).
- Modelled $CHBr_3$ at Kunak also shows higher maxima than at Danum and the variability is also higher at Kunak.



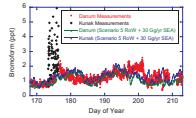
- The tracer concentration at Kunak is dominated by region 3. Danum is also heavily influenced by this region, though at Kunak the contribution is ~2x more than at Danum.
- Region 4 is slightly more important for Danum than for Kunak.
- All the other regions only have a very minor influence on the total concentration at the sites. Regions 1 & 2 (not shown) were almost always downwind of the sites.



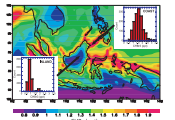


- The model pdfs are broader than for the observations and at Kunak they are shifted to higher $CHBr_3$ values compared to Danum. The model does not reproduce the very high levels actually recorded at Kunak.

- Can p-TOMCAT reproduce the observations? We use a 'best fit' emission scenario including South East Asia coasts, open ocean and 'rest of world'. The model captures the baseline values at Danum well and also shows a gradient between the coast and inland. Due to the 60 km model resolution, the very high Kunak $CHBr_3$ values are not captured.



- Surface monthly mean $CHBr_3$ mixing ratios from p-TOMCAT do capture a clear difference between the centre of the island of Borneo and the coast (locations a few grid boxes apart). Here the model appears to indicate the presence of the Borneo vortex.



6) Conclusions

- Shows the potential of these instruments for long-term observations at a range of sites and for key halocarbons of marine and anthropogenic origin.
- NAME works well with the observations. It confirms that air arriving at the sites passes over potentially strong emission regions and it captures several features of the observations.
- p-TOMCAT is able to reproduce the baseline observations but only by reducing the emission strengths in the South East Asia region from earlier estimates.

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