P-31 NOAA ESRL Global Monitoring Annual Conference 2015, May 19–20 2015, Boulder Colorado.

Methyl chloride as a tracer of tropical tropospheric air in the lowermost stratosphere inferred from IAGOS-CARIBIC passenger aircraft measurements

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ABSTRACT

Methyl chloride (CH_3Cl) is a predominantly natural trace gas which provides ozone-depleting chlorine to the stratosphere. Nitrous oxide (N_2O) is the third most important greenhouse gas that also plays a dominant role in stratospheric ozone depletion. In this study, we present variations of N_2O and CH_3Cl in the lower stratosphere (LMS) observed by the IAGOS-CARIBIC passenger aircraft observatory. N_2O undergoes clear seasonal variations in the LMS with a minimum in spring, the seasonal amplitude being pronounced going deeper in the LMS. Significant correlations between N_2O and CH_3Cl are found in the LMS from winter to early summer due to mixing between LMS and upper tropospheric air. This correlation disappears in late summer to autumn. Using the CH_3Cl-N_2O correlation slope, we estimate the stratospheric lifetime of CH_3Cl to be 35±7 years. We also examine the partitioning of stratospheric air, tropical tropospheric air and extra-tropical tropospheric air in the LMS based on a mass balance approach using N_2O and CH_3Cl. This analysis clearly indicates efficient inflow of tropical tropospheric air into the LMS in summer and demonstrates the usefulness of CH_3Cl as a unique tracer of tropical tropospheric air.

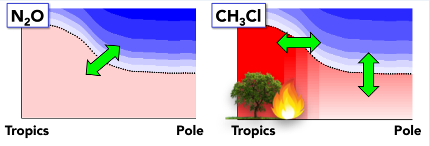
1. The IAGOS-CARIBIC flying observatory and CH_3Cl & N_2O data

- IAGOS-CARIBIC uses a 1.6-t air freight container with a variety of instruments onboard a Lufthansa Airbus A340-600 aircraft and has conducted almost monthly observation flights to various destinations from Germany (http://caribic-atmospheric.com).
- Two types of whole air samplers collect 116 samples in the UT/LMS (9–12 km altitudes) during each series of consecutive flights since May 2005.
- CH_3Cl mixing ratio is measured using GC-MS at UEA since 2005 and using GC-FID at MPIC since 2008 (Umezawa et al. 2014). The dataset is adjusted to the NOAA CH_3Cl scale (Montzka et al. 2011).
- N_2O mixing ratio is measured using GC-ECD at MPIC (Schuck et al. 2009) on the NOAA 2006 scale (Hall et al. 2007).
- In this study, N_2O data are expressed as ΔN_2O, deviations from the long-term trend observed at MLO (data provided by NOAA/GMD) (Umezawa et al. 2014).

Lufthansa Airbus A340-600 and the CARIBIC container

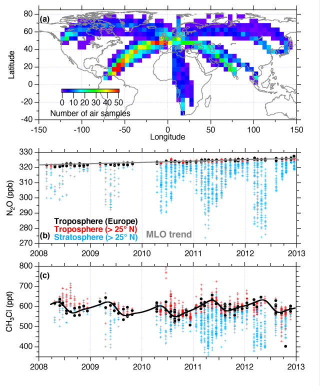
2. Background and concept

— CH_3Cl as a potentially useful tracer of tropical air in the LMS



A schematic view of N_2O and CH_3Cl distributions in the troposphere and the stratosphere. N_2O is almost uniform below the tropopause, shows a clear gradient across the tropopause and decreases going deeper in the troposphere. CH_3Cl decreases similarly in the stratosphere, but characteristic is the latitudinal distribution peaking in the tropical troposphere. Arrows indicate exchange of air that may be detectable in each gas.

3. Time series of CARIBIC N_2O and CH_3Cl data

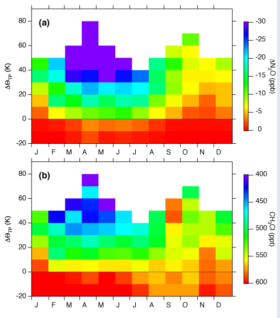


Number of air samples collected by CARIBIC for the period 2008–2012 in 5° grids.

Hereafter CARIBIC N_2O data are expressed as $\Delta N_2O = N_2O_{CARIBIC} - (N_2O\ trend)_{MLO}$

Both N_2O and CH_3Cl shows curtain-like seasonal pattern in the LMS, while the tropospheric variations are different.

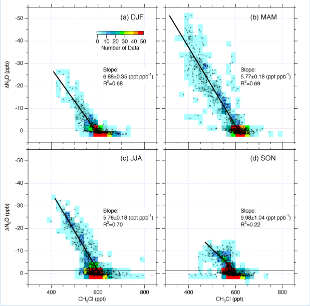
4. Seasonal and vertical variations of N_2O and CH_3Cl in the LMS



- N_2O undergoes a seasonal variation with a minimum in spring in the LMS and the amplitude increase going deeper in the stratosphere.
- CH_3Cl shows a seasonal variation similar in phase to that of N_2O in the LMS, while the seasonal cycle in the UT is different (a minimum in late summer).
- Vertical gradients of ΔN_2O and CH_3Cl vary with season: largest in spring and smallest in autumn.

Seasonal variations of ΔN_2O and CH_3Cl at different potential temperature layers with respect to the thermal tropopause ($\Delta\Theta_{TP}$).

5. Scatterplots between N_2O and CH_3Cl



Scatterplots of CH_3Cl as a function of ΔN_2O for different seasons (DJF, MAM, JJA and SON). Geometric mean regression lines for the stratospheric data are shown. Grid colors indicate number of data. The horizontal line shows the N_2O-based tropopause.

6. Estimate of stratospheric lifetime of CH_3Cl

Stratospheric lifetime of two long-lived trace gases are related as follows (Plumb and Ko, 1992):

$$\frac{\tau_1}{\tau_2} \sim \frac{\sigma_1 / \sigma_2}{d\sigma_1 / d\sigma_2}$$

← Global average N_2O and CH_3Cl values

← Correlation slope in the LMS in spring

Given τ_{N2O} = 122±24 yr (Volk et al. 1997), we calculate the stratospheric lifetime of CH_3Cl to be 35±7 yr. This value is half of a satellite data based estimate of 69 yr (Brown et al. 2013) but in agreement with a model-based estimate of 30.4 yr (SPARC, 2013).

7. Partitioning LMS air into stratospheric and tropical/extra-tropical tropospheric air

- We utilize N_2O and CH_3Cl to partition air of different origins: (1) the stratospheric overworld, (2) the tropical upper troposphere and (3) the extra-tropical surface air.

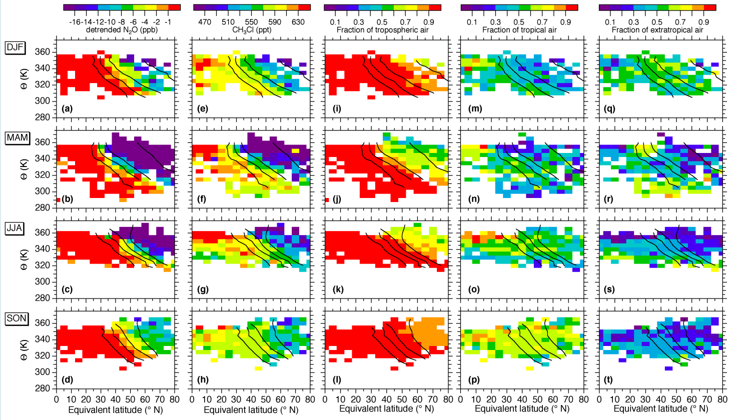
$$[N_2O]_{obs} = \alpha_{trop}[N_2O]_{trop} + \alpha_{strat}[N_2O]_{strat}$$

$$\alpha_{trop} + \alpha_{strat} = 1$$

$$[CH_3Cl]_{obs} = \alpha_{tropics}[CH_3Cl]_{tropics} + \alpha_{ex\text{-}tropics}[CH_3Cl]_{ex\text{-}tropics} + \alpha_{strat}[CH_3Cl]_{strat}$$

$$\alpha_{tropics} + \alpha_{ex\text{-}tropics} + \alpha_{strat} = 1$$

- The boundary values are given (1) from literature (Bönisch et al. 2009; Engel et al. 2002) for the stratospheric overworld, (2) from CARIBIC measurements (Umezawa et al. 2014) for the tropical upper troposphere and (3) from NOAA/GMD data at MHD (Montzka et al. 2011) for the extra-tropical surface air.
- The α_{trop} contours are in parallel to the PV isolines year round.
- The $\alpha_{tropics}$ contours also follow the PV isolines in spring, but in summer, a high tropical tongue extends across the tropopause, which is expanded in the entire LMS in autumn. Namely, high fraction of tropospheric air is dominated by flushing of the LMS by the tropical tropospheric air.



Potential temperature-equivalent latitude cross sections of (from left to right) N_2O, CH_3Cl, fraction of tropospheric air based on N_2O, fraction of tropical tropospheric air and extratropical air based on CH_3Cl for different seasons. Black lines PV isolines (PV = 2, 4, 6 and 8 PVU) indicating dynamical tropopause.

***Bönisch et al. (2009)**, Atmos. Chem. Phys., doi:10.5194/acp-9-5905-2009; **Brown et al. (2013)**, Atmos. Chem. Phys., doi:10.5194/acp-13-6921-2013; **Engel et al. (2002)**, J. Geophys. Res., doi:10.1029/2001JD000584; **Hall et al. (2007)**, J. Geophys. Res., doi:10.1029/2006JD007954; **Montzka et al. (2011)**, Science, doi:10.1126/science.1197640; **Plumb and Ko (1992)**, J. Geophys. Res., doi:10.1029/92JD00450; **Schuck et al. (2009)**, Atmos. Meas. Tech., doi:10.5194/amt-2-449-2009; **SPARC (2013)**, SPARC Report No. 6; **Umezawa et al. (2014)**, J. Geophys. Res. doi:10.1002/2013JD021396; **Volk et al. (1997)**, J. Geophys. Res., doi:10.1029/97JD02215.*

Acknowledgement *we are grateful to Lufthansa Airlines and Fraport for their support of the CARIBIC project and we thank to the CARIBIC team members. CARIBIC is part of IAGOS and we acknowledge funding from the German Ministry of Education and Research (BMBF). We also thank S. Montzka for NOAA CH_3Cl data, J. Elkins and B. Hall for NOAA N_2O data.*