The Tropospheric Budget of PAN

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Introduction

Recently, the first global measurements of a number of important upper tropospheric organic species such as PAN have been retrieved from the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) instrument on board ENVISAT. Peroxyacetyl nitrate (PAN) is a key species in tropospheric chemistry. It acts as a reservoir for NOx, allowing it to be transported over large distances in the cold upper troposphere (UT). PAN therefore plays an important role in the long-range transport of pollution to remote clean areas. These new observations allow us, for the first time, to evaluate simulated global PAN distributions from the TOMCAT 3-D chemical transport model (CTM), which has recently been updated to include isoprene chemistry and new temperature dependent quantum yields for acetone photolysis. We are then able to use the model to investigate factors controlling the tropospheric budget of PAN.

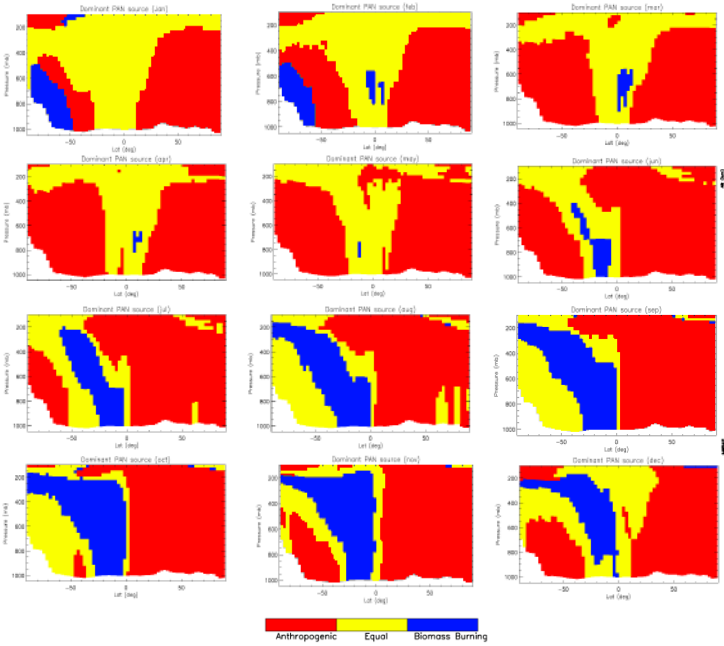
Updates to TOMCAT for this study

- Implementation of isoprene chemistry scheme
  - Based on Mainz condensed isoprene oxidation mechanism (Poschl et al. 2000).
  - Includes 16 species, 34 chemical reactions and 10 photolytic reactions. (=> ~50% increase of existing TOMCAT)
- Inclusion of new temperature dependent acetone photolysis quantum yield (Blitz et al. 2004) – Leads to ~50% increase in acetone lifetime
- Updated emissions based on IPCC, also inclusion of a monoterpene source for acetone

Investigating the Tropospheric PAN Budget

- Initial comparisons with MIPAS and aircraft suggest TOMCAT PAN is reasonable – so we may use the model to investigate the tropospheric PAN budget.
- Perform a series of model sensitivity runs:
  1. Base run for 2004, standard emissions and chemistry
  2. Switch off anthropogenic NOx emissions
  3. Switch off biomass burning NOx emissions
  4. Switch off lightning NOx
  5. Switch off biogenic emissions of isoprene
- Differences between runs will identify the factors controlling PAN in the troposphere

Zonal Mean NO_x Contribution to PAN



The TOMCAT CTM

Off-line 3-D chemical transport model

Vertical coordinate (σ-p, σ-θ). Variable resolution.

Horizontal winds and temperatures specified from analyses (e.g. ECMWF).

Vertical winds from analysed divergence *or* diagnosed heating rates (in stratosphere).

Advection: Prather [1986] second-order moments *or* semi-Lagrangian.

Physics: Tiedtke [1989] convection scheme.
Holtslag and Boville [1993] *or* Louis [1979] PBL schemes.

Chemistry:

Stratosphere: Ox, NOy, HOx, Cly, Bry, CHOx, source gases. Aerosols/PSCs..

Troposphere: Ox, NOy, HOx C1-C3. Wet/dry deposition. Emissions etc…
23 advected species (+ short lived),
83 chem reactions + 21 photolytic reactions
+ Mainz isoprene chemistry scheme (16 species + 44 reactions)

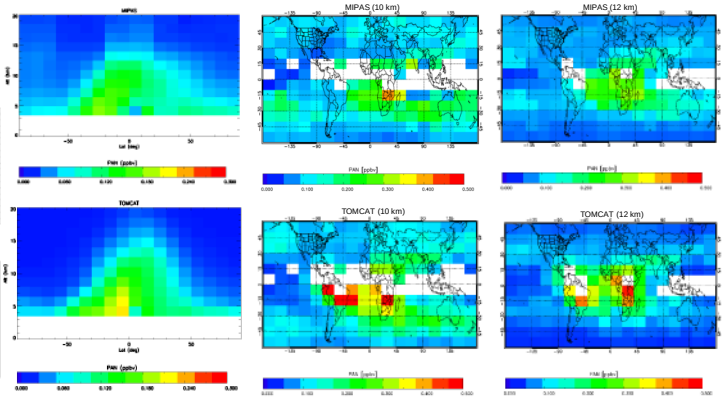
Aerosols:

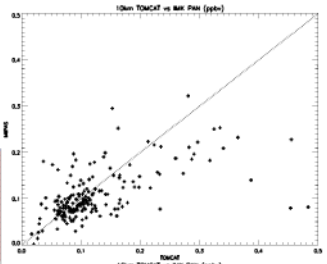
Troposphere: Sulphate, sea-salt, (SOA) (GLOMAP)

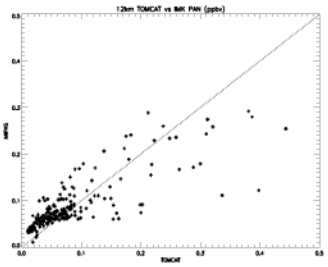
Stratosphere: Denitrification microphysical model (DLAPSE)

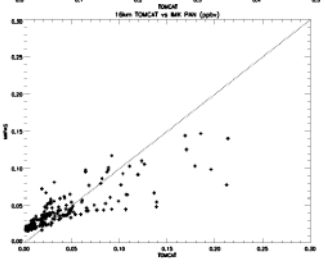
MIPAS-TOMCAT Comparison Strategy

- Retrievals of upper tropospheric PAN from MIPAS for October and November 2003 were provided by IMK.
- The TOMCAT fields are then sampled at MIPAS measurement times and locations and interpolated onto the MIPAS retrieval levels.
- Both datasets are then averaged over the whole period onto a 10 by 20 degree latitude/longitude grid.
- MIPAS and TOMCAT are then compared in the -60 to 60 degrees latitude range









MIPAS vs. TOMCAT correlation coefficients

Altitude	PAN	O3	HNO3
10 km	0.64	0.95	0.88
12 km	0.81	0.95	0.87
16 km	0.85	0.97	0.97

Initial Conclusions

- MIPAS and TOMCAT PAN, O_3 and HNO_3 show a high degree of correlation in the 10 – 16 km altitude range.
- TOMCAT PAN is generally higher than MIPAS but shows similar morphology.
- This gives confidence in using TOMCAT to explore the tropospheric PAN budget
- Initial sensitivity studies show that throughout most of the year Northern Hemispheric PAN is dominated by anthropogenic emissions, with boreal fires becoming equally important during the summer
- Southern Hemispheric PAN is dominated by biomass burning emissions in Austral spring with anthropogenic sources gaining importance in the autumn

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