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Interpreting multi-year datasets of volatile organic compounds derived from MIPAS

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Contents

- **The aim of this presentation is to highlight a variety of novel MIPAS measurements of volatile organic compounds in the upper troposphere and lowermost stratosphere**
  - **What have we observed?**
  - **What have we learnt?**

1. **Monitoring long-term changes**
   - **Global ethane**
2. **Short-term stratospheric perturbations due to fire events**
   - **Australian wildfire event, "Black Saturday"**
3. **Seasonality of key VOCs in relation to tropospheric chemistry**
   - **Peroxyacetyl nitrate, comparison to models and other satellites**

Species	Source	Importance
PAN	secondary production including from fossil fuels, biomass burning	Principal tropospheric reservoir species for nitrogen oxide radicals, remote production of tropospheric ozone
HCOOH	vegetation	contributes to acidity in precipitation
C2H2	biomass burning	role in glyoxal formation
C2H6	fossil fuels, biomass burning, biofuel	precursor to tropospheric ozone, reacts with OH

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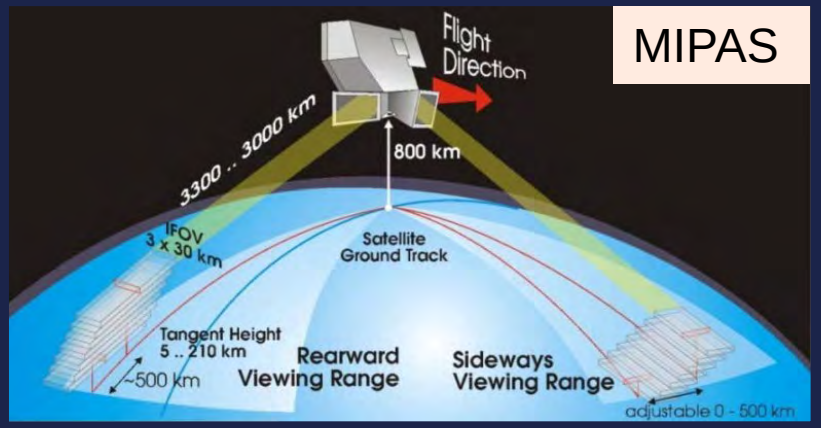
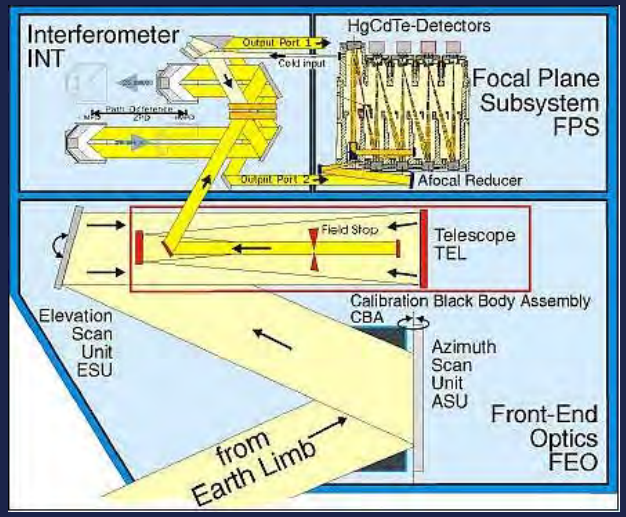
MIPAS instrument on Envisat

MIPAS specifics	
Platform	ENVISAT
Instrument type	**Fourier Transform Spectrometer**
Mass	320 kg
Spectral range	**4.15 to 14.2 microns in 5 spectral bands (685-2410 cm^{-1})**
Spectral resolution	0.025cm^{-1} (March 2002-March 2004) 0.0625 cm^{-1} (August 2004-April 2012)
Vertical range	**6 to 68 km**
Vertical resolution	3 km from 6 to 42 km, 5 km from 42 to 52 km, 8 km from 52 to 68 km
Species measured	**L1b emission spectra - Potential to observe many gases** **L2 vertical profiles- operational products: H_2O, O_3,CH_4, N_2O, NO_2, HNO_3**



MIPAS limb-observation geometry design (image credit: ESA)



Schematic view of MIPAS optical design (image credit: ESA)

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Multi-year volatile organic compound (VOC) products from MIPAS

- Produced using a version of the University of Oxford MORSE retrieval scheme designed for MIPAS
  - Used to cross-compare to operational ESA products (PT, H_2O, O_3)
- A dedicated MIPAS dataset of major volatile organic compounds is now available from the University of Leicester (V1.0)
  - PAN, HCOOH, C_2H_2, C_2H_6 (2007-2012)
  - Netcdf(4) format (but not yet CF compliant…)
- Plan to upload data to CEDA before August 2014 (www.ceda.ac.uk).
- Files contain retrieved vmr, pressure, cloud-index (test of cloudiness), retrieval error, averaging kernels, covariance matrices.
- Retrieval of VOCs also requires retrieval of interfering species and other parameters
  - Pressure/temperature, H_2O, O_3, HNO_3, $ClONO_2$, CCl_4, CH_4, N_2O, HCFC-22

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Monitoring long-term changes in atmospheric ethane (1)

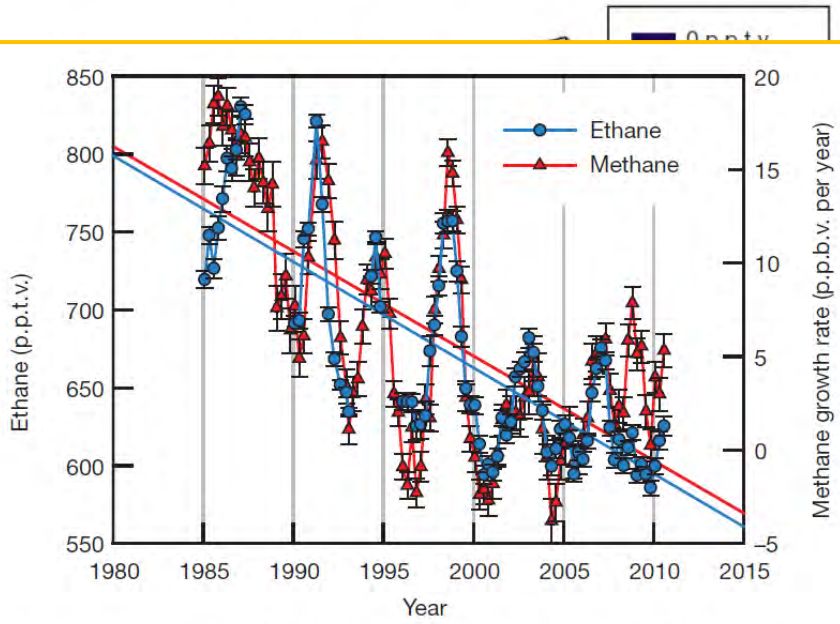

Figure 4 | Running global averages of ethane mixing ratios and methane growth rate. The methane measurements, which were co-sampled with ethane, are also from the UCI global monitoring network and are described in ref. 10. Solid lines are linear fits to the ethane (blue circles) and methane (red triangles) data using a least-squares regression. Each global average is based on 55–75 individual mixing ratios. The averaging procedure and uncertainty calculations are described in the Supplementary Information.

- After methane, ethane is the most abundant hydrocarbon in the remote atmosphere
- Precursor to tropospheric ozone and influences the atmosphere's oxidative capacity through its reaction with the hydroxyl radical (OH)
- Recent studies, i.e. Simpson et al., 2012 in Nature, report a long-term decline of global atmospheric ethane concentrations and its implications for methane
  - Ethane emission rates decreased globally from 14.3 to 11.3 teragrams (21%) between 1984 and 2010 (-6.8±0.6 ppt/yr)
  - Most likely due to decreased venting and flaring from oil fields

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Monitoring long-term changes in atmospheric ethane (2)

- Ethane's major emission sources are shared with methane
  - Papers have disagreed on whether reduced fossil fuel or microbial emissions caused methane's atmospheric growth to slow at the early part of this century.
  - Reduced fossil fuel emissions account for at least 10-21 teragrams per year (30-70 %) of the decrease in methane's slowing atmospheric growth rate since the mid-1980s

- All data in Simpson et al. study taken from **surface sites only**, across the Pacific Ocean
  - What is happening in the upper troposphere and lower stratosphere?
  - What is happening globally?
  - This is where **MIPAS can fill in the gaps**.

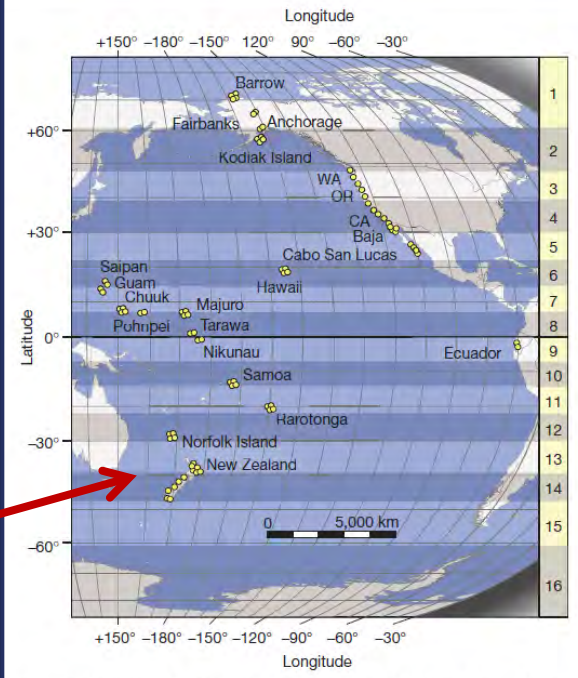

Figure 1 | Individual air sampling locations for the UCI global trace gas monitoring network. Each air sample is collected for one minute into a conditioned, evacuated two-litre stainless steel canister, typically at a site (yellow circles) on the coast when the wind is blowing from the ocean. Air samples are usually collected over a 3-week period each season (March, June, September and December), though samples were collected less frequently in 1991 and 1992 (April, August and December) because of political events at the time. We have also collected biweekly air samples on Norfolk Island (29° S) since June 2001. Boundaries that separate the Earth's surface into 16 latitudinal bands, each with an equal volume of air, are shown on the right (see Supplementary Information). The global representativeness of our Pacific-based measurements, both spatial and temporal, is discussed in the Supplementary Information. WA, Washington; OR, Oregon; CA, California.

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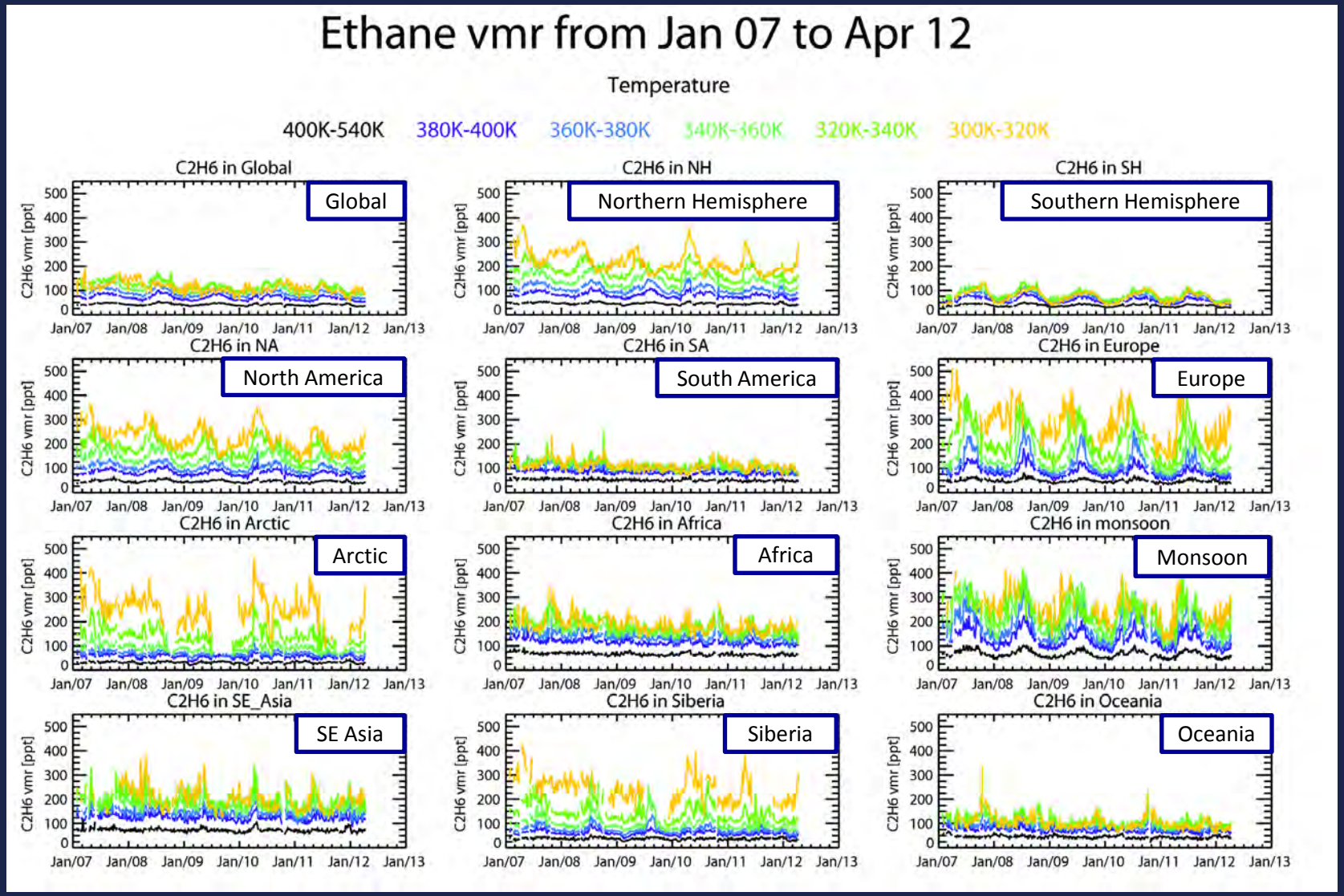
Ethane measurements from MIPAS (1)



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Deriving changes in ethane (2007-2012)

Ethane changes (ppt/year)

Temp (K)	Change in VMR (ppt/year)		
	Global	NH	SH
400-540	-1.0±1.5	-1.7±1.4	-1.2±1.3
380-400	-2.8±2.2	-3.6±1.8	-2.7±1.8
360-380	-4.1±2.7	-5.1±1.9	-4.0±2.2
340-360	-5.9±3.2	-7.8±2.0	-5.1±2.4
320-340	-7.3±3.7	-7.2±2.1	-4.2±2.4
300-320	-6.5±4.1	-5.7±1.9	-2.5±2.1

Northern Hemisphere

Temp (K)	Change in VMR (ppt/year)					
	NA	Europe	Arctic	Monsoon	SE Asia	Siberia
400-540	-1.0±1.3	-1.3±1.1	-0.2±0.9	-2.3±1.8	-1.4±1.9	-0.5±1.0
380-400	-2.9±2.0	-4.0±1.7	-1.4±1.3	-5.6±2.6	-3.2±2.4	-1.5±1.5
360-380	-4.4±2.4	-4.8±2.1	-2.1±1.5	-9.9±3.2	-5.1±2.8	-2.6±1.8
340-360	-7.3±3.0	-8.5±2.7	-4.0±1.9	-15.7±3.5	-8.3±3.3	-3.9±2.2
320-340	-10.9±3.8	-15.1±3.9	-10.9±2.9	-15.2±3.8	-8.5±3.3	-9.5±3.3
300-320	-14.9±4.2	-23.0±5.6	-17.4±4.9	-21.0±4.7	-7.3±3.0	-17.1±5.6

Southern Hemisphere

Temp (K)	Change in VMR (ppt/year)		
	SA	Africa	Oceania
400-540	-1.7±1.5	-2.1±1.7	-1.7±1.4
380-400	-3.9±2.0	-4.7±2.3	-3.6±1.8
360-380	-5.2±2.5	-6.8±2.9	-5.1±1.9
340-360	-6.0±2.6	-9.3±3.3	-7.8±2.0
320-340	-5.2±2.5	-9.1±3.5	-7.2±2.1
300-320	-4.0±2.7	-7.5±3.4	-5.7±1.9

Key

NH	Northern Hemisphere
SH	Southern Hemisphere
NA	North America
SA	South America
SE Asia	South-east Asia

change between -5 and -10 ppt/yr

change of over -10 ppt/yr

- Simple linear approximation, taking into account variability to define errors on the fit
- Globally, **largest changes occur** in upper troposphere (**300-340 K**)
  - Northern Hemisphere changes larger than Southern Hemisphere
  - Link to changes in sources in northern hemisphere (venting and flaring of natural gas)
  - Changes in Southern hemisphere also significant, minimal contribution from venting, so most likely linked to changes in biomass burning
- Global decrease compares very closely to Simpson et al. tropospheric decrease of -6.8±0.6 ppt/yr

Ema

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Methane measurements from MIPAS

Methane changes (ppt/year)

Temp (K)	Change in VMR (ppt/year)		
	Global	NH	SH
400-540	-10.1±13.2	-15.2±12.8	-4.4±13.0
380-400	-10.2±14.8	-10.5±14.2	-9.3±14.8
360-380	-8.9±13.7	-8.7±13.9	-9.2±13.1
340-360	-4.8±12.4	-5.0±13.2	-4.7±11.5
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300-320	-7.5±14.9	-6.7±15.8	-6.1±13.6

Northern Hemisphere

Temp (K)	Change in VMR (ppt/year)					
	NA	Europe	Arctic	Monsoon	SE Asia	Siberia
400-540	-10.9±12.0	-13.4±12.0	-23.2±11.9	-12.9±13.2	-12.9±12.3	-12.5±12.2
380-400	-8.6±11.7	-12.9±10.1	-14.9±9.8	-11.0±15.2	-12.2±13.7	-11.2±10.1
360-380	-8.1±12.0	-10.2±10.1	-11.7±9.9	-9.3±15.4	-6.7±14.1	-8.4±10.1
340-360	-6.9±11.8	-5.8±9.9	-8.4±10.6	-5.0±13.4	-4.1±13.3	-1.0±10.4
320-340	-7.9±12.0	-6.2±11.0	-4.2±13.4	-9.4±12.8	-6.3±13.5	-1.6±12.1
300-320	-2.6±13.0	-11.3±13.3	5.5±18.5	-13.5±15.5	-10.4±15.4	5.4±17.1

Southern Hemisphere

Temp (K)	Change in VMR (ppt/year)		
	SA	Africa	Oceania
400-540	-7.3±13.0	-10.4±12.9	-5.6±10.9
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300-320	-8.0±15.0	-12.2±14.3	-5.4±11.4

Key

NH	Northern Hemisphere
SH	Southern Hemisphere
NA	North America
SA	South America
SE Asia	South-east Asia

change between -5 and -10 ppt/yr
change of over -10 ppt/yr
Increase

- Methane also shows large scale decreases over 2007-2012
  - largest decreases in the stratosphere (but note large uncertainty on these data)
- Interestingly, whereas decreases occur in ethane over the Arctic and Siberia, methane shows a moderate increase (300-320)
  - Are changes in local sources (i.e. permafrost) affecting the Arctic and Siberia?

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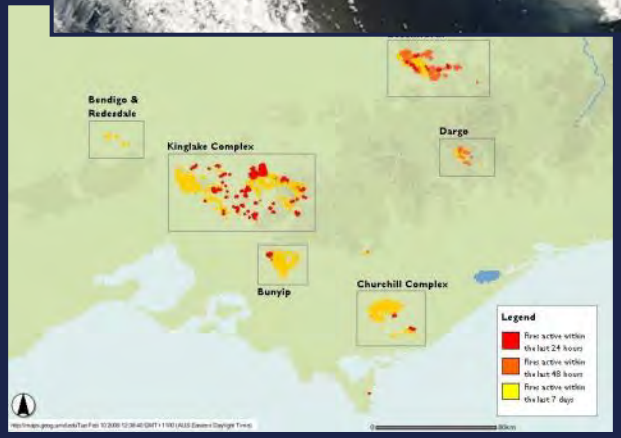
Short-term stratospheric perturbations caused by wildfires (1)

- A series of bushfires which started in South-Eastern Australia on February 7th 2009
- Over 450000 hectares burnt
- Fires burned over the period 7/2 – 14/3
- Other work determines stratospheric aerosol/gas injection



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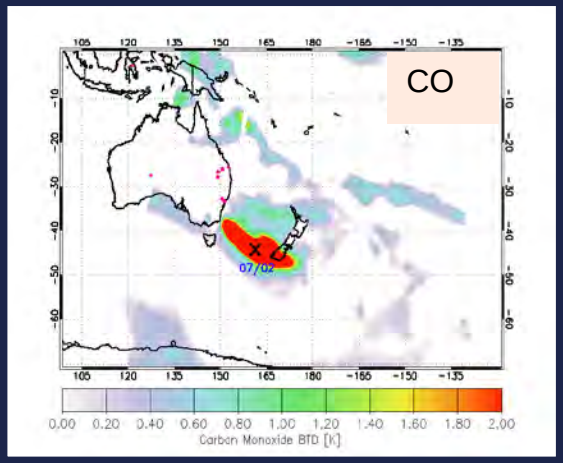
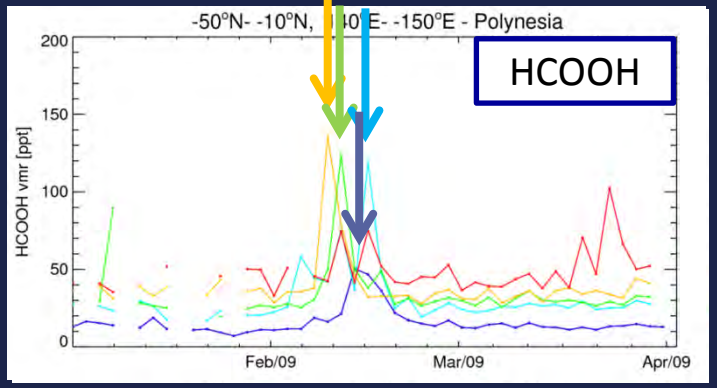
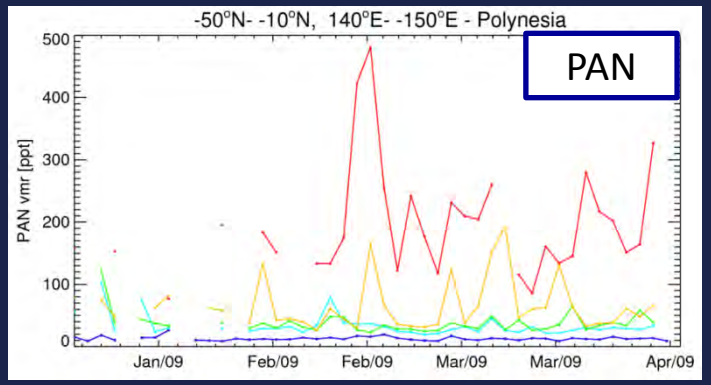
Short-term stratospheric perturbations caused by wildfires (2)



- IASI measurements (left) show large total column perturbations in pollutants CO and HCOOH in the total column
  - Rapid horizontal transport away from the fires
- 5-year HCOOH time series shows stratospheric feature
  - Up to x4 increase in UTLS HCOOH
- Perturbation in PAN only visible in 280-300 K layer
- Peak in HCOOH averaged concentrations propagates through the UT between 9th-15th
  - unclear as to why the stratospheric peak (400-540 K) is before the lowermost stratosphere peak (380-400K)





400 K to 540 K
380 K to 400 K
360 K to 380 K
340 K to 360 K
320 K to 340 K

Temp (K)	340-360	360-380	380-400	400-540
Peak day (HCOOH)	09/02	11/02	15/02	13/02

MIPAS observes perturbations on very short time-scales

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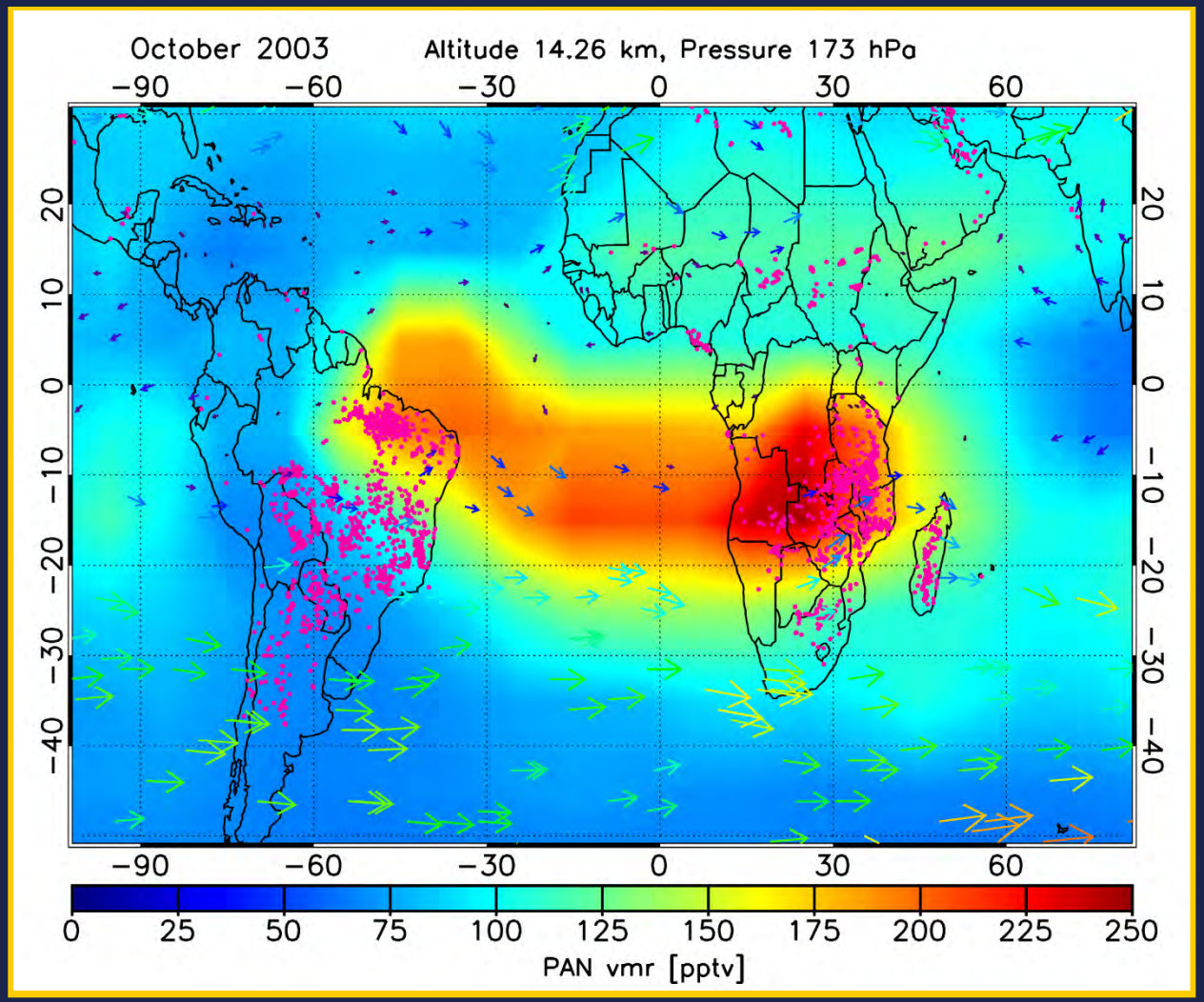
Investigating seasonality with MIPAS



- PAN measurements during October 2003
- Transport of pollution across Southern Atlantic Ocean
- Taken from: Moore and Remedios, Seasonality of Peroxyacetyl nitrate (PAN) in the upper troposphere and lower stratosphere using the MIPAS-E instrument, Atmos. Chem. Phys., 10, 6117-6128, 2010

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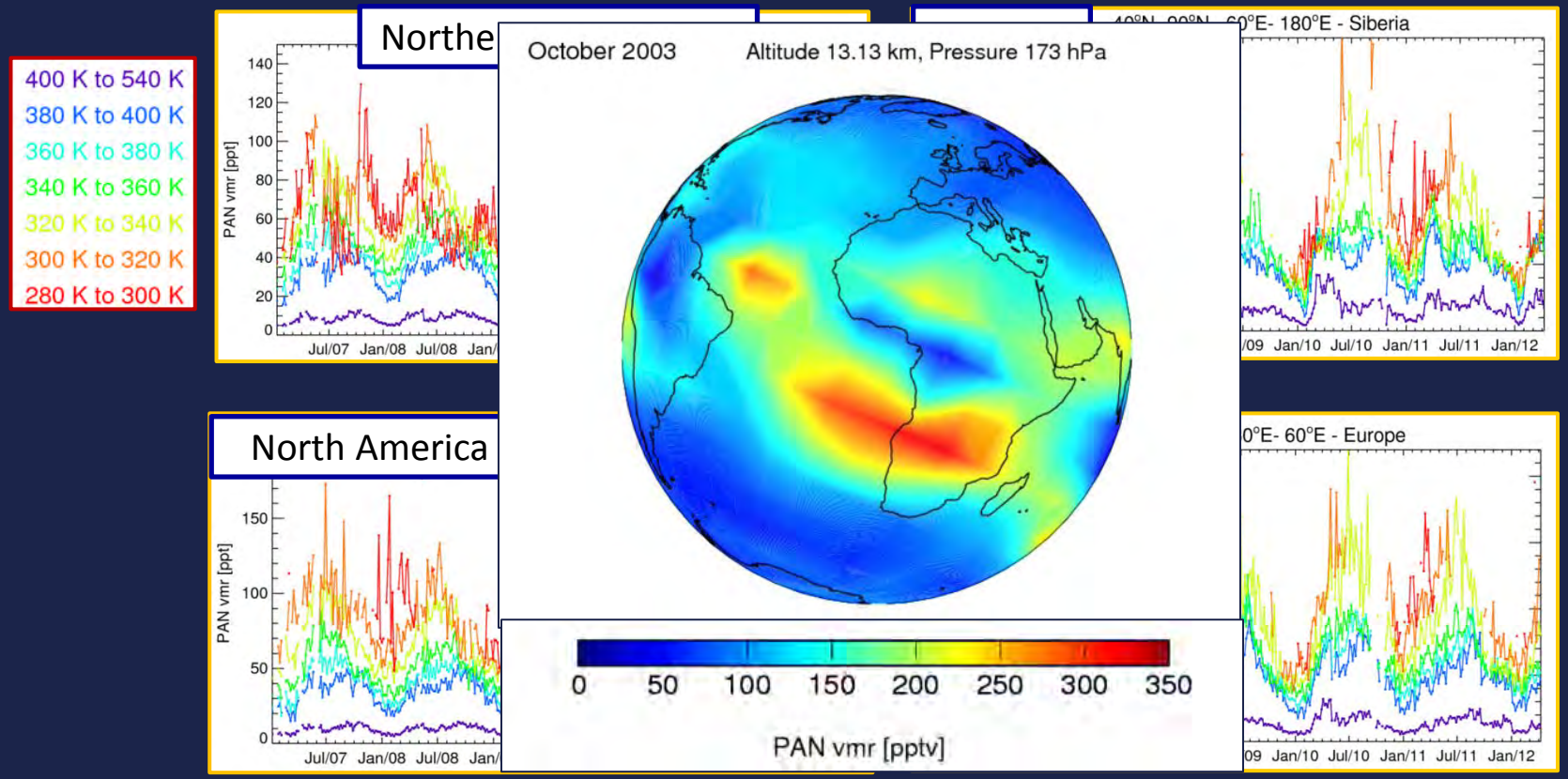
Seasonality from MIPAS - PAN



- Northern Hemisphere seasonal cycle in the upper troposphere/lower stratosphere PAN, peaks in June/July, lowest in December/January
- Siberian peaks often linked with local fire activity over the boreal forests and is a good indicator of fire strength
- Biogenic sources over Europe drive the changes in the PAN vmrs

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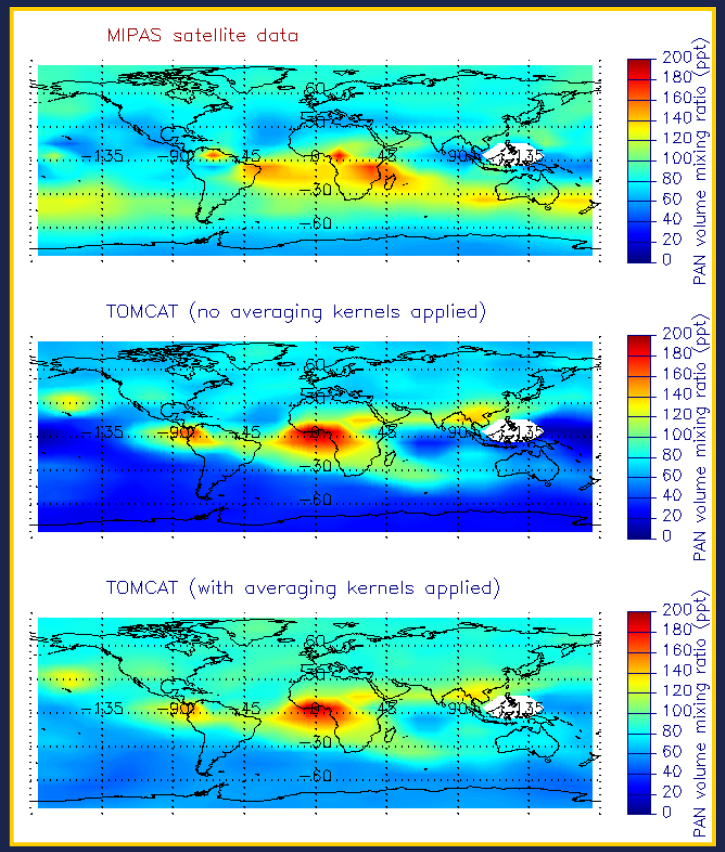
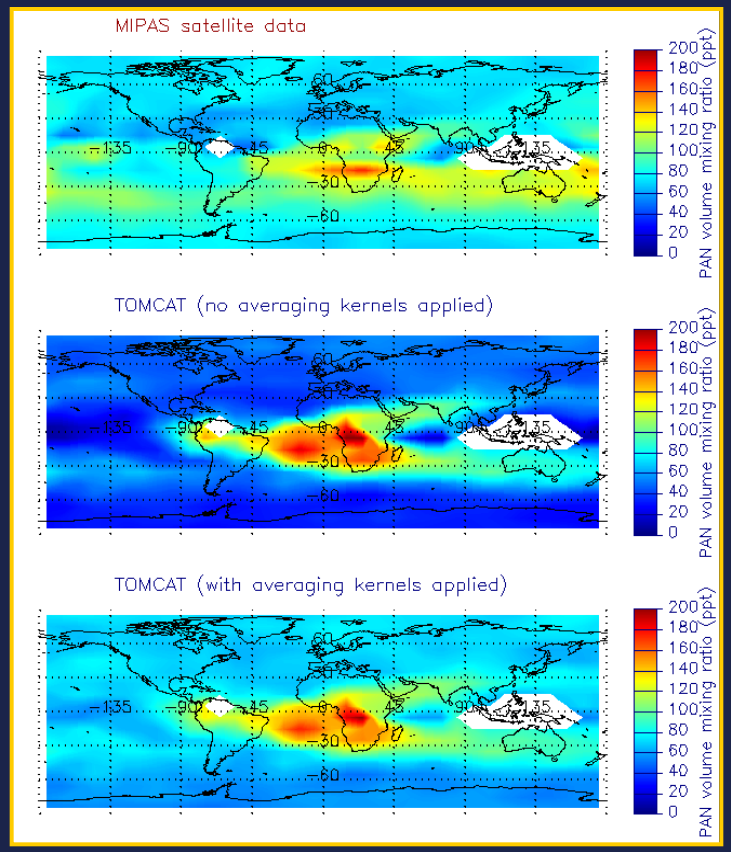
MIPAS and TOMCAT PAN comparisons

October 2003



November 2003



- Look at Southern Hemisphere "burning" season
- Plume (likely mix of Southern America and Southern Africa emissions) seen in MIPAS data for both October and November but not as clear in TOMCAT – lifetime issue?

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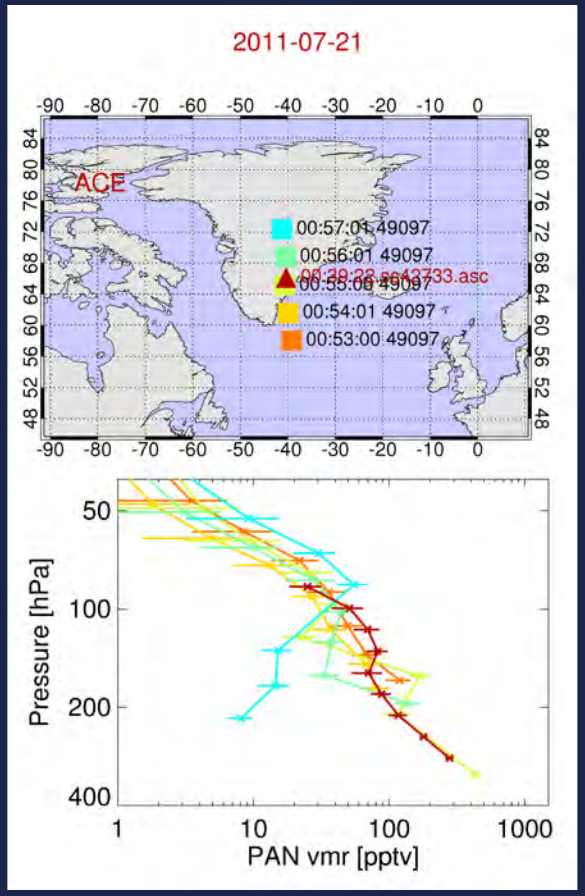
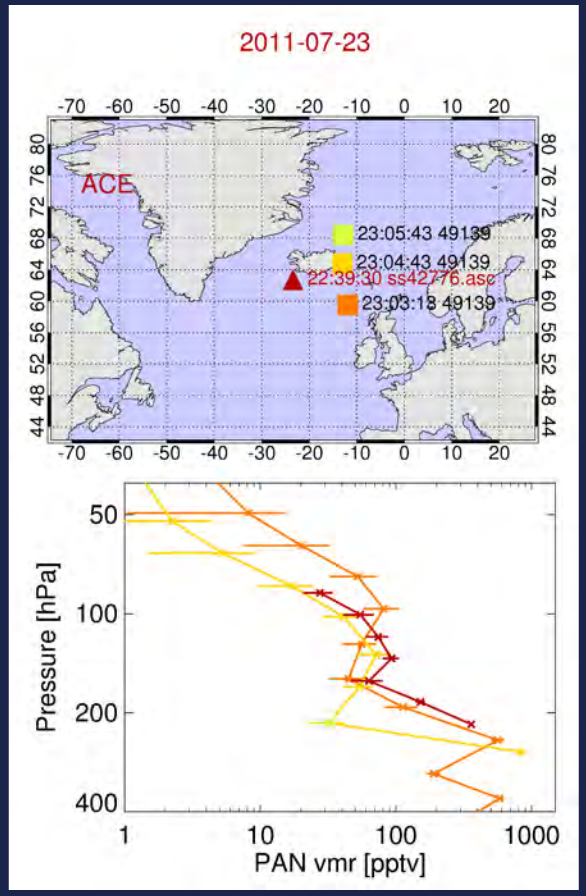
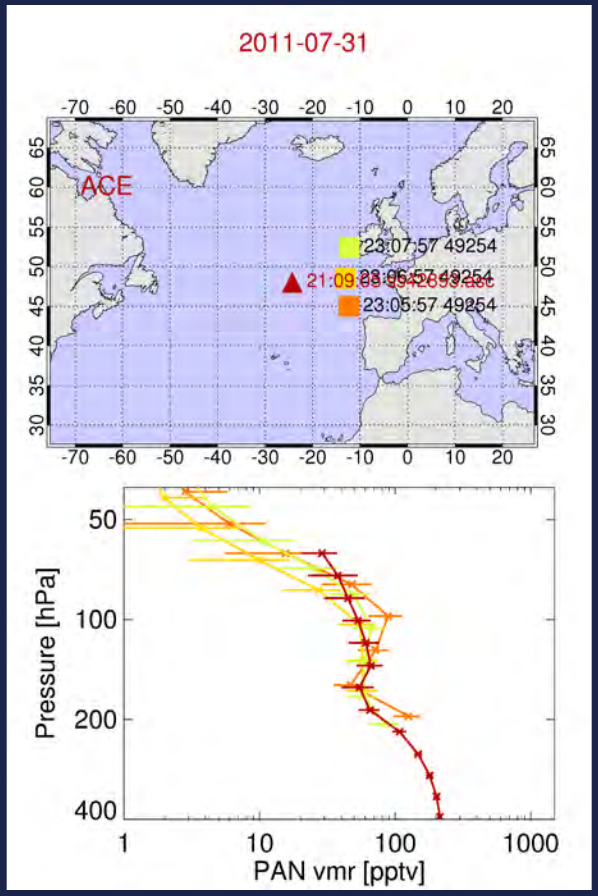
Peroxyacetyl Nitrate: ACE-MIPAS comparisons

Young plume (< 24 hours) older plume (24-48 hr) aged plume (48-72 hr)







Dark red – ACE
"Other" colours - MIPAS

Em

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Summary

- A unique dataset with which to study chemical and dynamical processes in the UTLS
- As part of NCEO we've been able to produce mature datasets of volatile organic compounds from MIPAS
  - "Official" release V1.0 to be made available on CEDA (2007-2012 nominal mode)
  - Please contact me if you're interested in receiving the data
- Long-term changes in trace gases are observable in MIPAS measurements
  - Significant decline in ethane in the UTLS
- Observing atmospheric thermal emission and therefore day/night monitoring, allows short-term events to be picked up in the longer time series (i.e. significant wildfires)
- The long dataset shows important seasonal features in the MIPAS data, driven by a combination of chemical and dynamic processes.
- Comparisons have been made to models, to critically test their quality
- Comparisons have also be made to other satellite instruments such as ACE

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Outlook

- MIPAS data being used in conjunction with IASI to improve tropospheric IASI retrievals
  - Used to constrain HCOOH and C_2H_2
  - Large impact on tropospheric columns where VOCs show spike in the UTLS
  - Potential future application to a “convoy”-type mission, with nadir and limb viewing instruments working in conjunction to measure the atmosphere
- University of Leicester working with the University of Oxford to deliver novel MIPAS cloud products to CEDA (including H. Sembhi and R.Graves)
  - Including measurements of volcanic and wildfire-smoke plumes
- In the short- to mid-term future, there will be no other instrument like it in space. Although some of the data are now up to 12 years old, they still offer a unique measurement of the upper troposphere and stratosphere, which is why ESA will soon be continuing to support reprocessing and validation activities until 2017.

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Acknowledgements

- National Centre for Earth Observation for funding
- European Space Agency (for provision of MIPAS data)
- Eumetsat (for provision of IASI data)
- Anu Dudhia – MIPAS retrieval scheme (MORSE), and forward model (RFM)
- Sam Wharton at the University of Leicester

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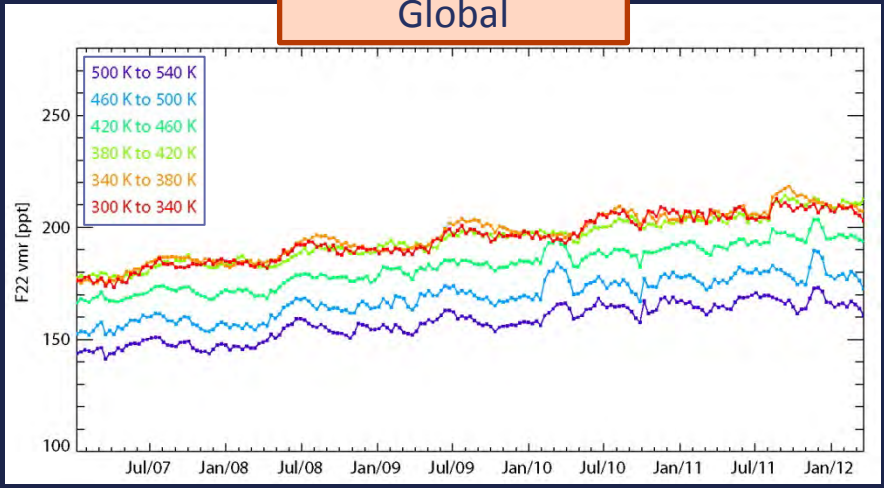
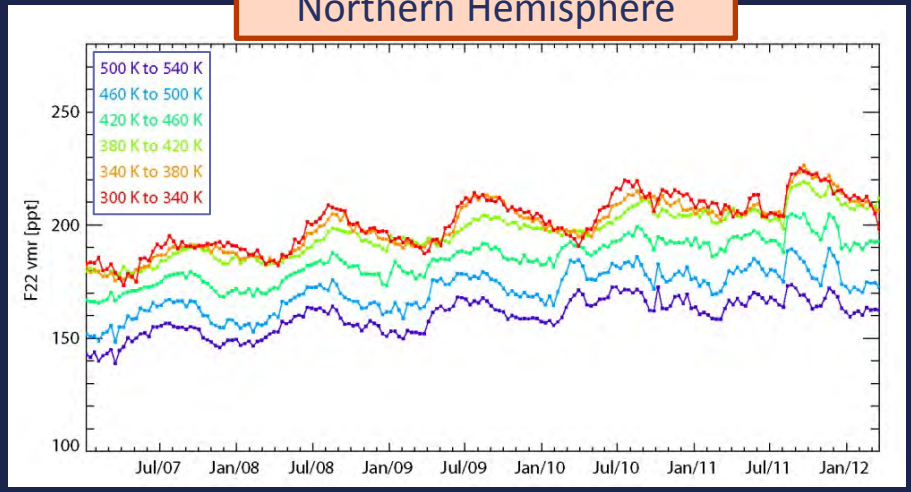
Measuring HCFC-22 in the UTLS

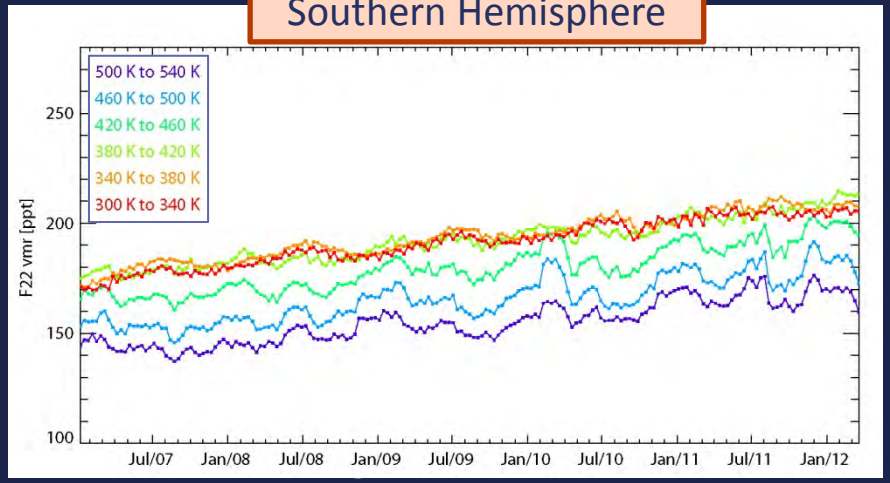
Global



Northern Hemisphere



Southern Hemisphere



- Hydrogenated chlorofluorocarbons (HCFCs) were seen as a viable alternative to CFCs, which destroy stratospheric ozone
- However, HCFC–22 is an efficient greenhouse gas
  - 1700 times stronger as a greenhouse gas than an equivalent volume mixing ratio (vmr) of CO_2
- Seasonal cycles in Northern Hemisphere linked to transport processes
- Fortems-Cheiney et al., JGR 2013 report a continuously rising global emission from 182±11 Gg in 1995 to 410±9 Gg in 2009, mainly driven by rises over Eastern Asia.

Courtesy of David Moore – Leicester NCEO

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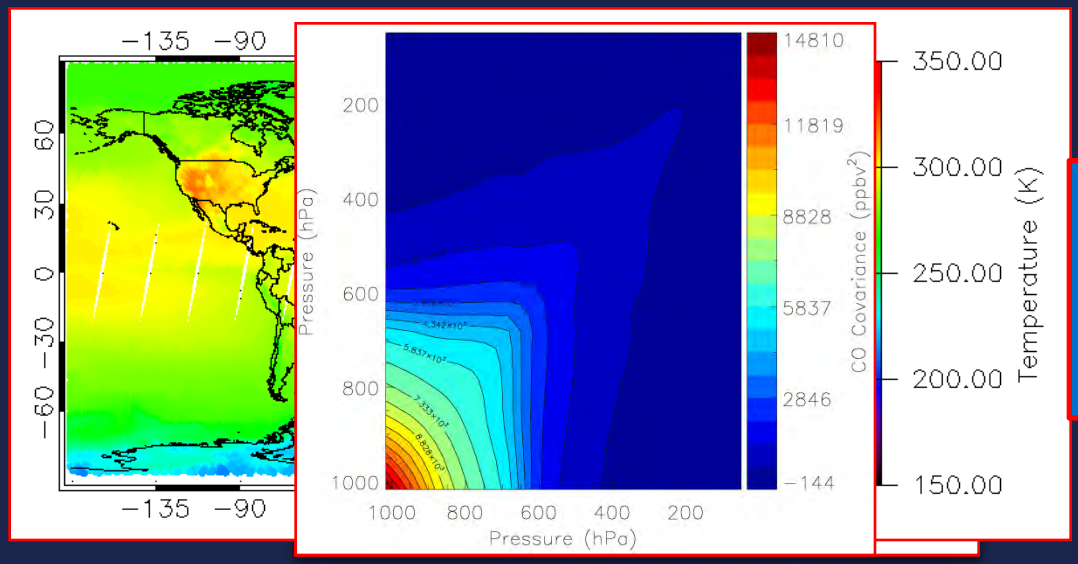
ULIRS – University of Leicester IASI Retrieval Scheme

Pre-process and Cloud Clear IASI Data

Get ECMWF T, Altitude and H_2O Profiles

Get Surface Emissivity from CIMSS



Calculate a priori statistics from TOMCAT (for CO) or mix INTEX-B/ACE/MIPAS for organics.

Select Climatology

Perform OEM Retrieval Using Oxford RFM

Convert to Total Column Using DEM Data

Full details of ULIRS in Illingworth et al., AMT, 2011

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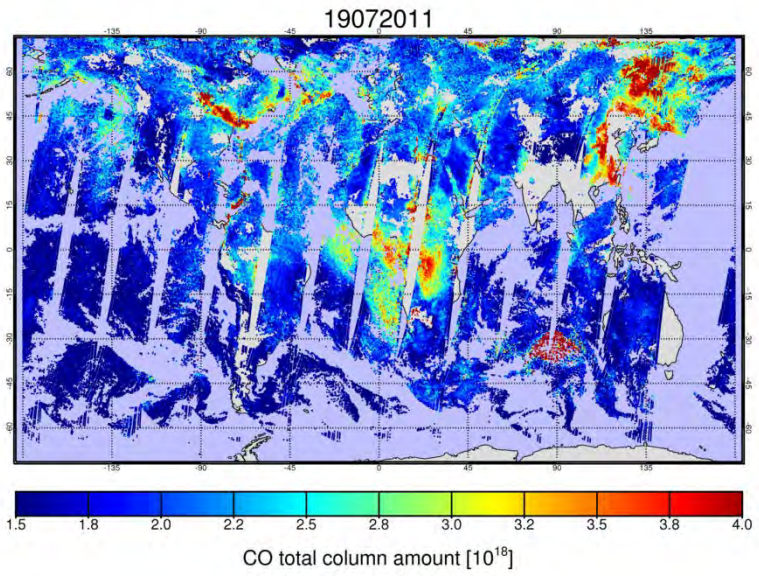
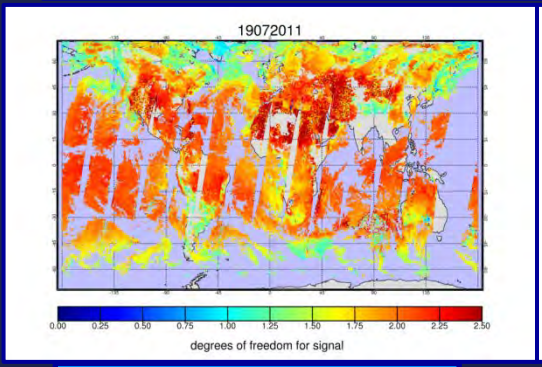
ULIRS – University of Leicester IASI Retrieval Scheme - updates

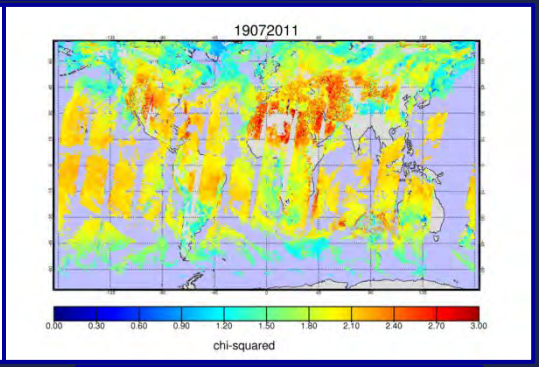
Global total column CO from ULIRS – 19/07/2011



- adapt IDL retrieval scheme for UoL high-performance computing cluster
  - Rewrite "maths" part in fortran
  - Split job into 10x10 degree latitude bins
  - **1 day (global) takes ~ 3 days to run**, reduced from several weeks
- RTTOV incorporated in a "testing" phase
  - May improve run speed further



Degrees of freedom



Chi-squared

- Considering to include an aerosol retrieval for VOCs
  - More susceptible to cloud contamination (800-1100 cm^{-1})
  - Worked successfully for adapted scheme for ARIES aircraft instrument (Illingworth et al. 2013, AMTD)

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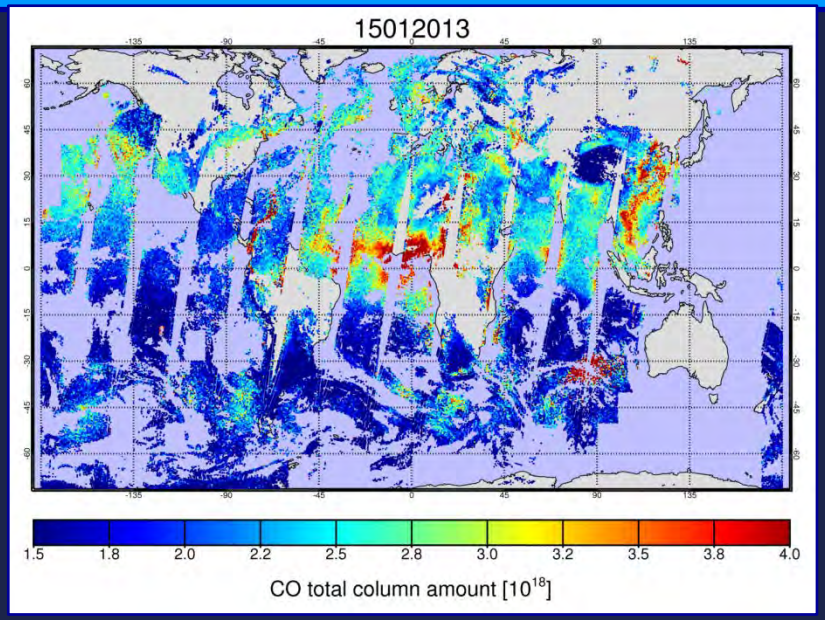
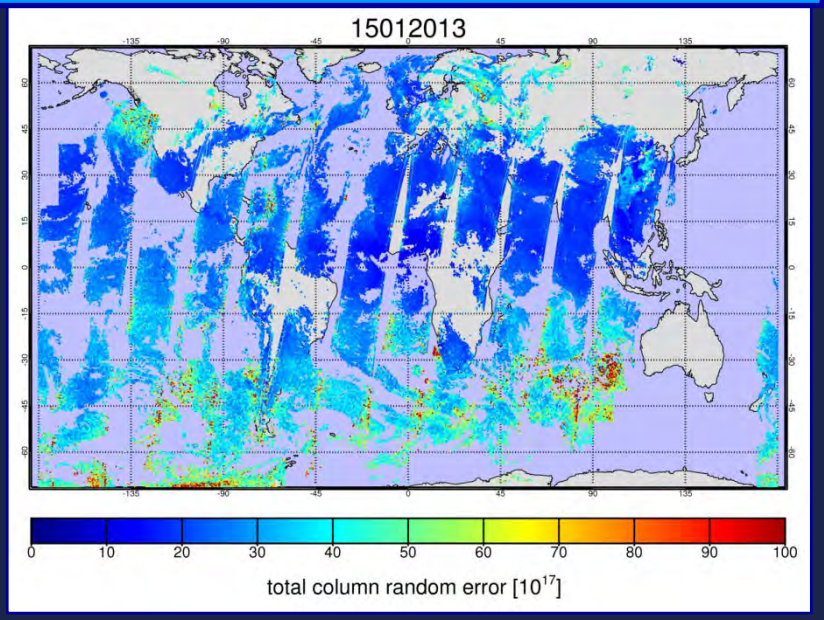
ULIRS CO- updates

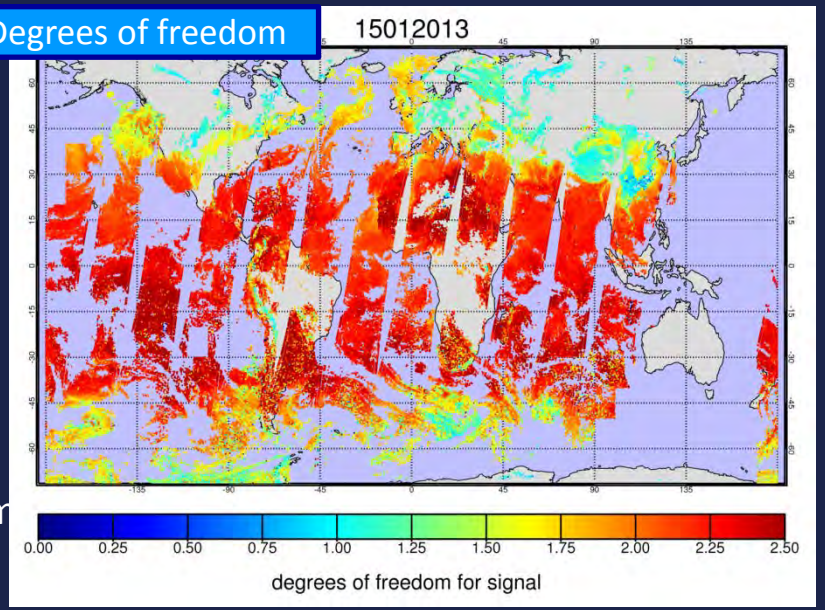
Global total column CO from ULIRS – 15/01/2013



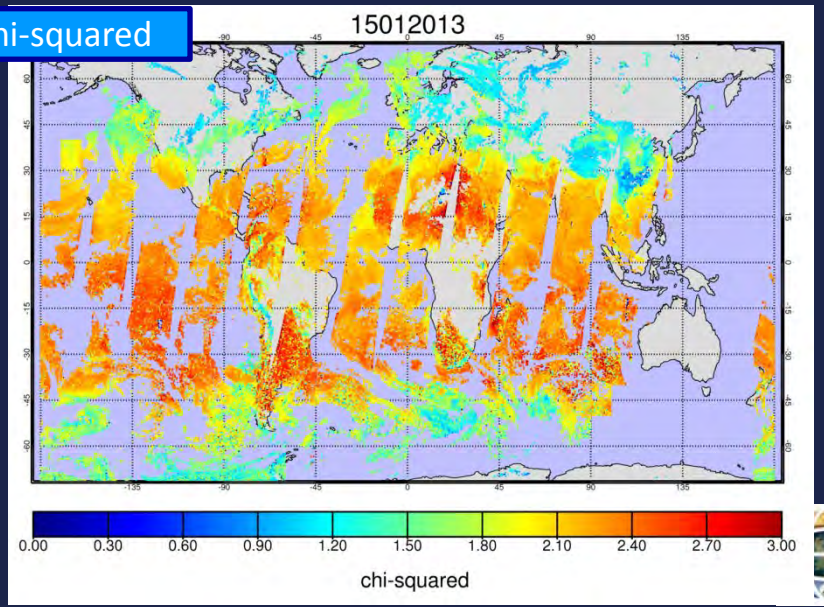
Random retrieval error on total column CO



Degrees of freedom



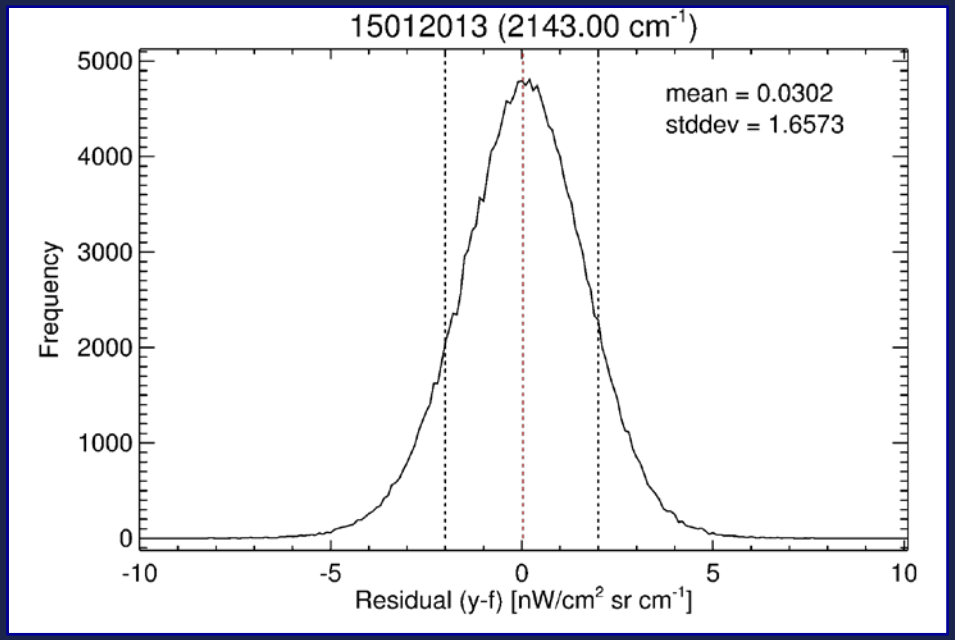
Chi-squared



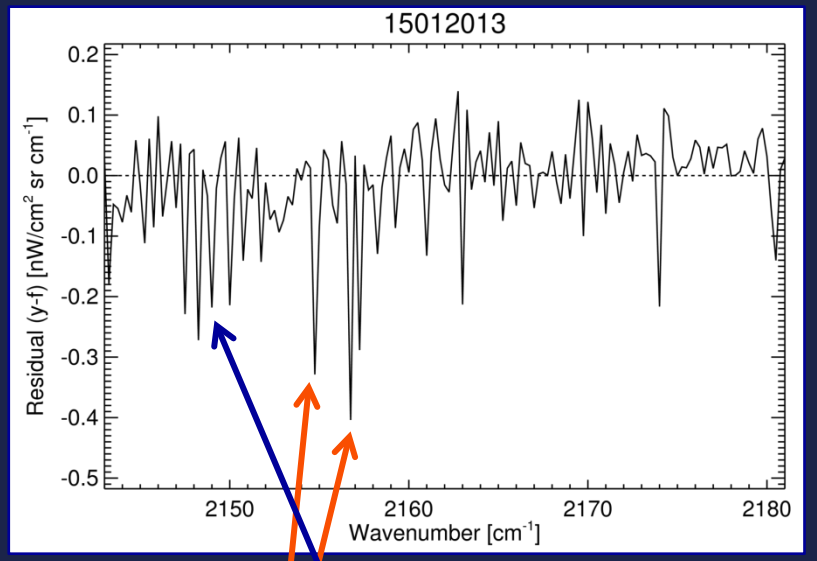
Em

ULIRS CO- residual fits

Histograms of ULIRS CO residuals at each spectral point used in the retrieval (2143-2181 cm^{-1} range)



Average CO residuals from ULIRS – 15/01/2013



- Histograms of ULIRS CO residuals
- Mean very close to zero in majority of cases with a standard deviation of < 2 nW/cm^2 sr cm^{-1}
  - IASI noise ~ 2 nW/cm^2 sr cm^{-1} in this spectral range)

- Small structure in average residuals
  - A few water lines still cause residual fit bias
  - Similarly some small structure with CO_2 fit
- Overall, excellent fit to CO signal, within IASI noise constraints
  - Confidence in global CO retrievals

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Relationship between ULB-LATMOS CO and ULIRS CO

Comparison of FORLI and ULIRS CO retrievals		
1)	Forward modelling using look up table approach (pre-calculated table of absorbances)	line-by-line forward model approach (RFM Oxford)
2)	2143 -2181.25 cm^{-1} CO microwindow	2143 -2181.25 cm^{-1} CO microwindow
3)	retrievals on 19 altitude levels (1 km steps up to 18 km, then 18 km to top of atmosphere)	retrievals on 30 equidistant pressure levels (surface to 50 hPa)
4)	Temperature and humidity from the EUMETSAT level 2 product	Temperature and humidity from the ECMWF 1.125 degree 6-hourly data
5)	cloud fraction < 25 % used	two cloud threshold tests: a) Threshold test comparing measured BT to the Earth's skin temperature (Hadji-Lazaro et al. [2001]) b) 8-11 micron difference (Strabala et al. [1994])

- First evaluation of the ULB-LATMOS FORLI scheme CO and the ULIRS CO products
- The Fast Optimal retrievals on Layers for IASI (FORLI) was developed at the Iniversite Libre de Bruxelles (ULB)
  - Outlined in Hurtmans et al. 2012
- Daily (ascii) files containing CO products available from http://ether.ipsl.jussieu.fr/ether/pubipsl/iasi_CO_uk.jsp
- Files also include information on partial column averaging kernels and a priori CO

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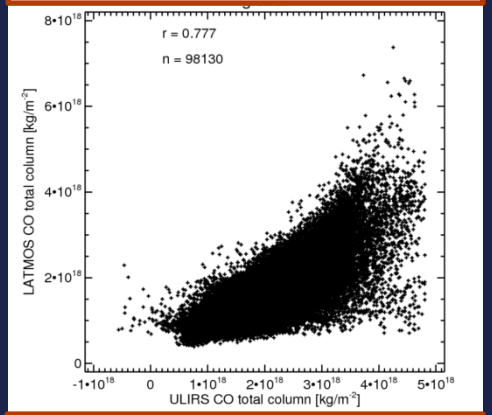
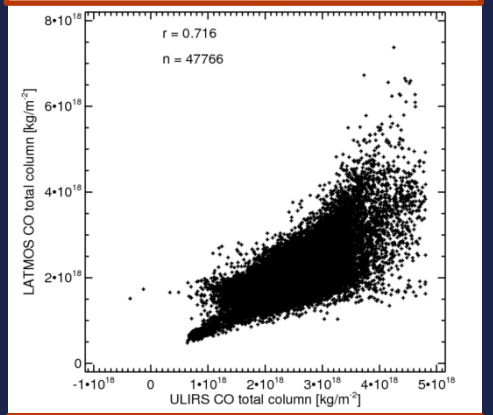
Relationship between ULB-LATMOS CO and ULIRS CO (part 1)

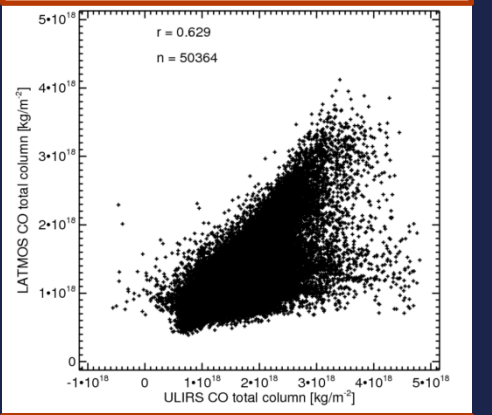
Global



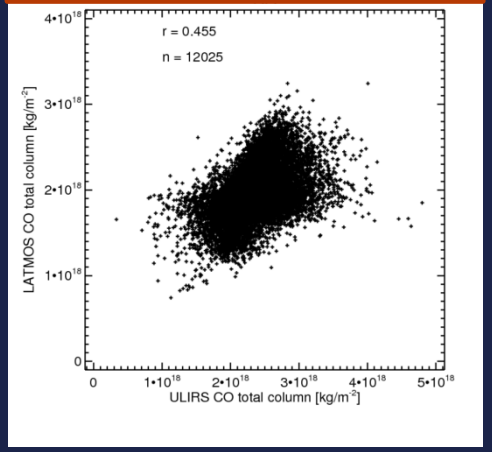
Northern Hemisphere



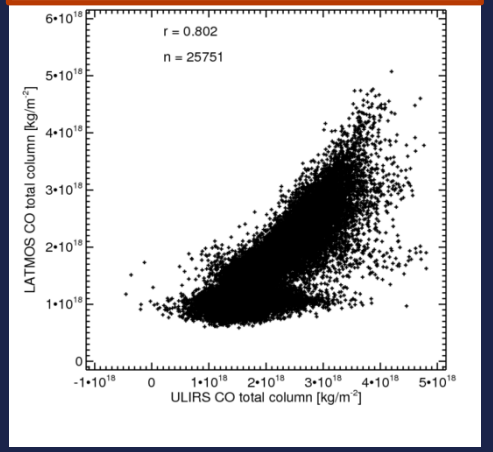
Southern Hemisphere



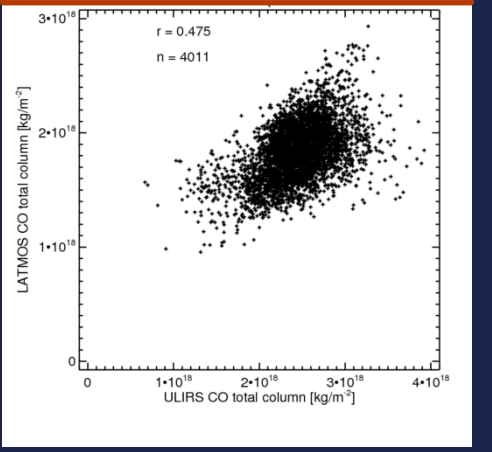
North America



South America



Europe



CO data provided by LATMOS/CNRS & ULB

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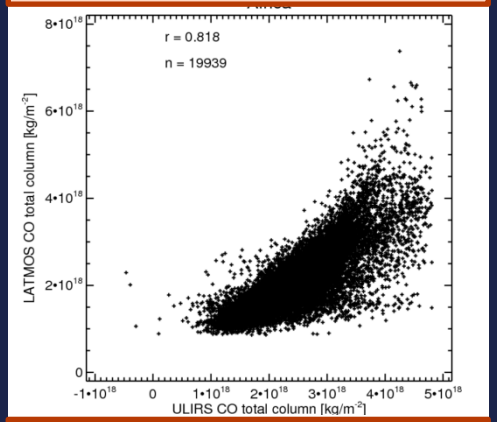
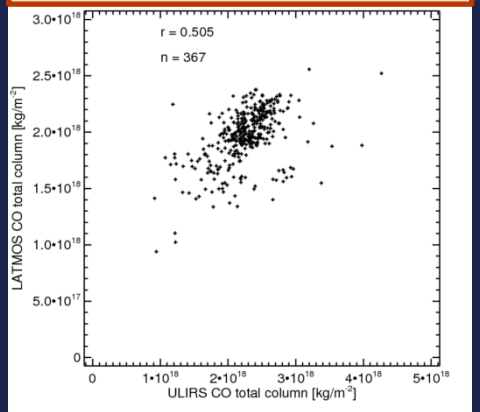
Relationship between ULB-LATMOS CO and ULIRS CO (part 2)

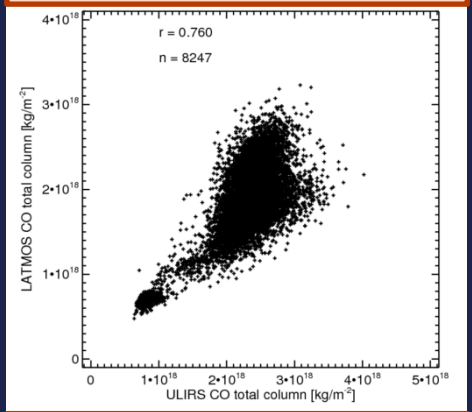
Africa



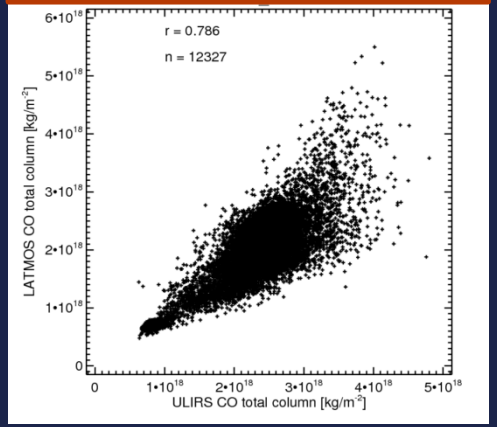
Arctic



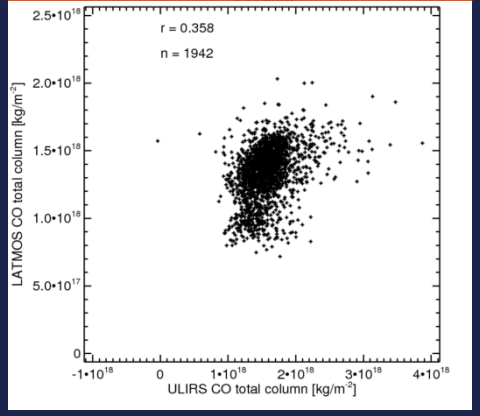
Indian Monsoon



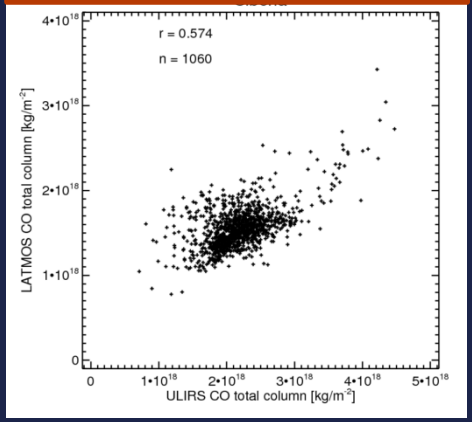
South-east Asia



Australasia



Siberia



CO data provided by LATMOS/CNRS & ULB

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EXTRA SLIDES

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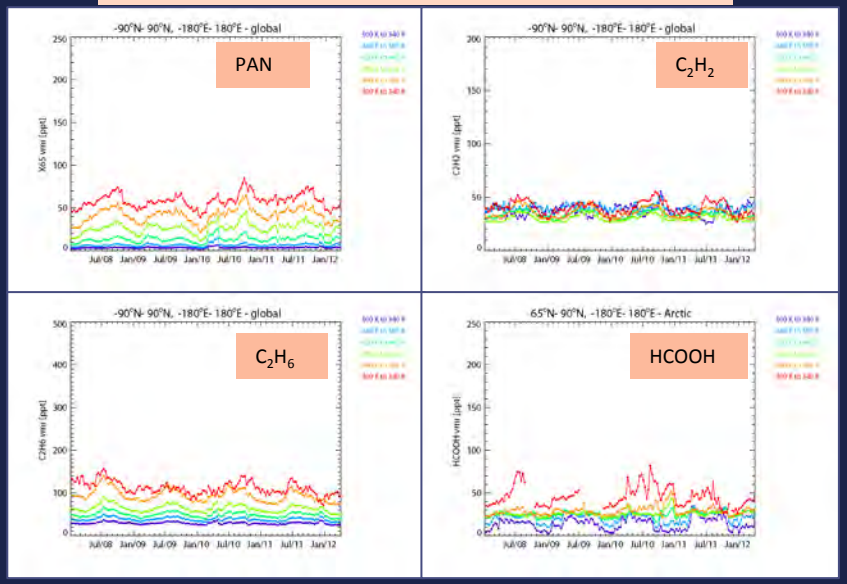
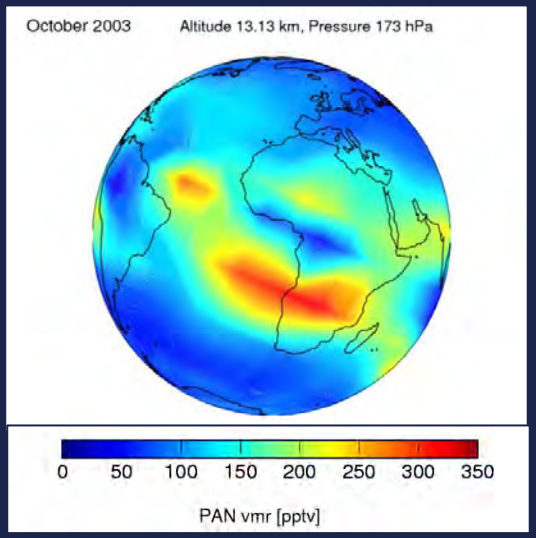
Multi-year volatile organic compound (VOC) products from MIPAS

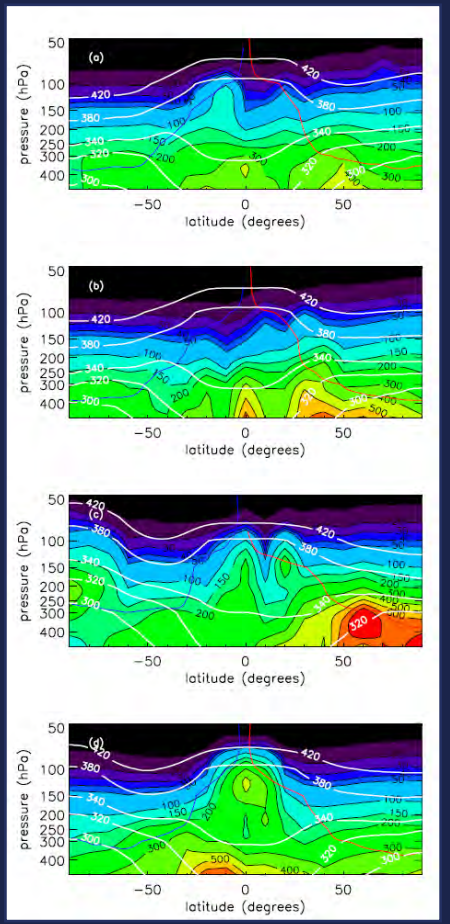
University of Leicester

Novel, multi-year, datasets of VOCs



First multi-seasonal PAN measurements from space





- MIPAS volatile organic compound products, developed through NCEO (Phase 1) at the University of Leicester, have aided understanding of chemical processes in the upper troposphere and lower stratosphere
  - Multi-year VOC datasets, available to modelling community
  - First global measurements of acetone from space
  - First multi-seasonal and multi-year PAN datasets in the upper troposphere and lower stratosphere

PAN cross-sections taken from : Moore, D. P. and Remedios, J. J.: Seasonality of Peroxyacetyl nitrate (PAN) in the upper troposphere and lower stratosphere using the MIPAS-E instrument, Atmos. Chem. Phys., 10, 6117-6128, doi:10.5194/acp-10-6117-2010, 2010.
Acetone cross-sections taken from: Moore, D. P., Remedios, J. J., and Waterfall, A. M.: Global distributions of acetone in the upper troposphere from MIPAS spectra, Atmos. Chem. Phys., 12, 757-768, doi:10.5194/acp-12-757-2012, 2012.

Seasonal latitudinal PAN distribution (units of pptv), averaged zonally for **(a)** January 2003, **(b)** March 2003, **(c)** August 2003 and **(d)** October 2003. The white lines indicate theta levels (K) and the red and blue lines represent the +/− 2 PVU levels.

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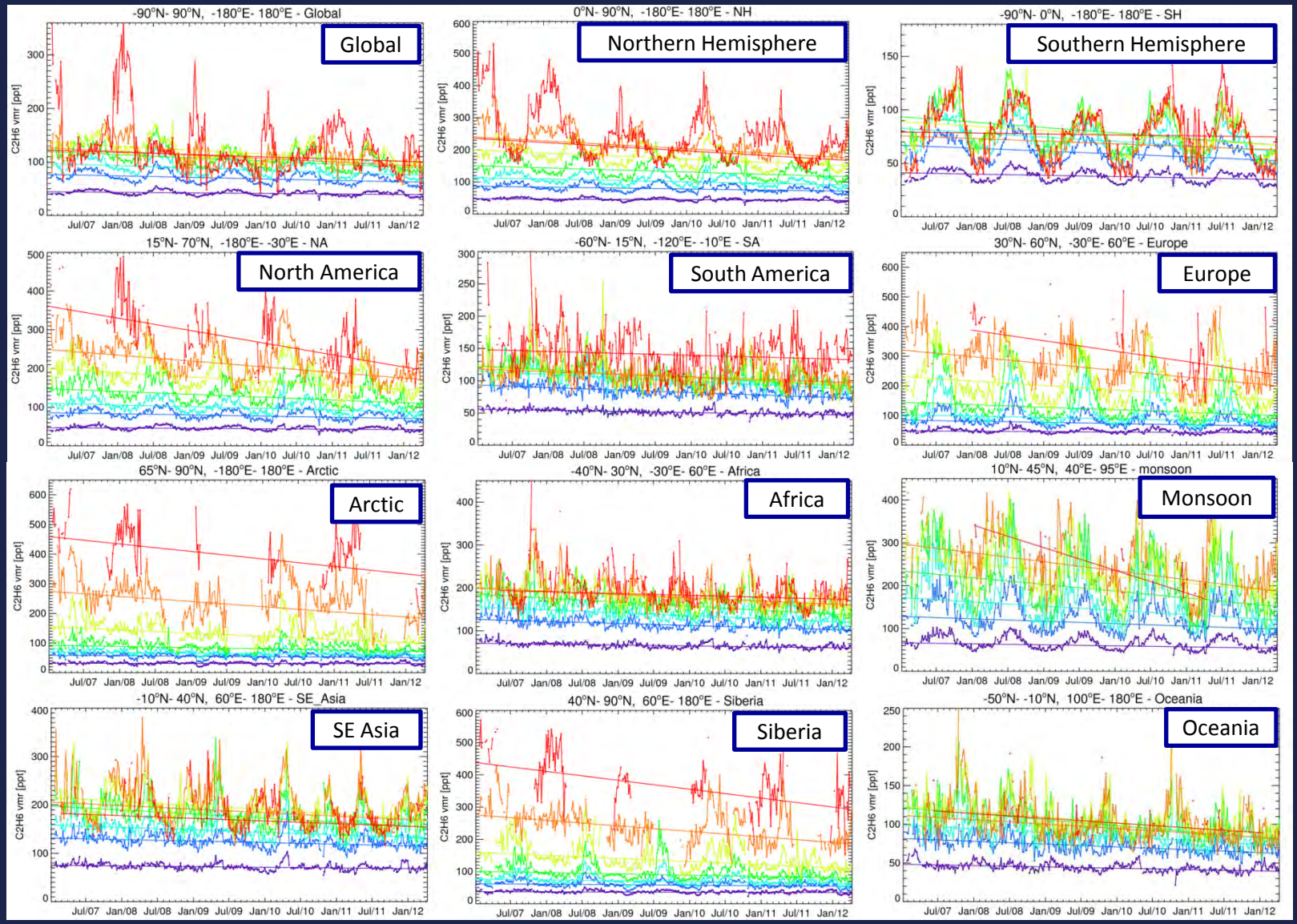
Ethane measurements from MIPAS (2)



400 K to 540 K
380 K to 400 K
360 K to 380 K
340 K to 360 K
320 K to 340 K
300 K to 320 K
280 K to 300 K

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Introduction

- **Suite of species we observe with MIPAS** provide information on biomass burning and plumes including:
  - Plume composition and chemistry
  - Intercontinental transport

Species	Spectral range [cm^{-1}]	Feature	Source	Importance	Reference
PAN	775- 800	Broad	Secondary pollutant	Ozone production in remote regions	Moore et al., ACP, 2010
Acetone	1215 -1220	Broad	Biomass burning/ biogenic	Upper tropospheric source of OH	Moore et al., ACP, 2012
C_2H_2	766 – 767	Strong line	Biomass burning	Role in glyoxal formation	Wiegele et al., 2012
C_2H_6	815-830	Strong line	Fossil fuels, biomass burning, biofuel	Precursor to tropospheric ozone, reacts with OH	Simpson et al., 2012
HCN	747 – 748	Strong line	Biomass burning	Tracer of biomass burning	Glattor et al., ACP, 2009
HCOOH	1105 – 1105.25	Strong line	Vegetation	Contribute to acidity of precipitation	Grutter et al., JGR, 2010

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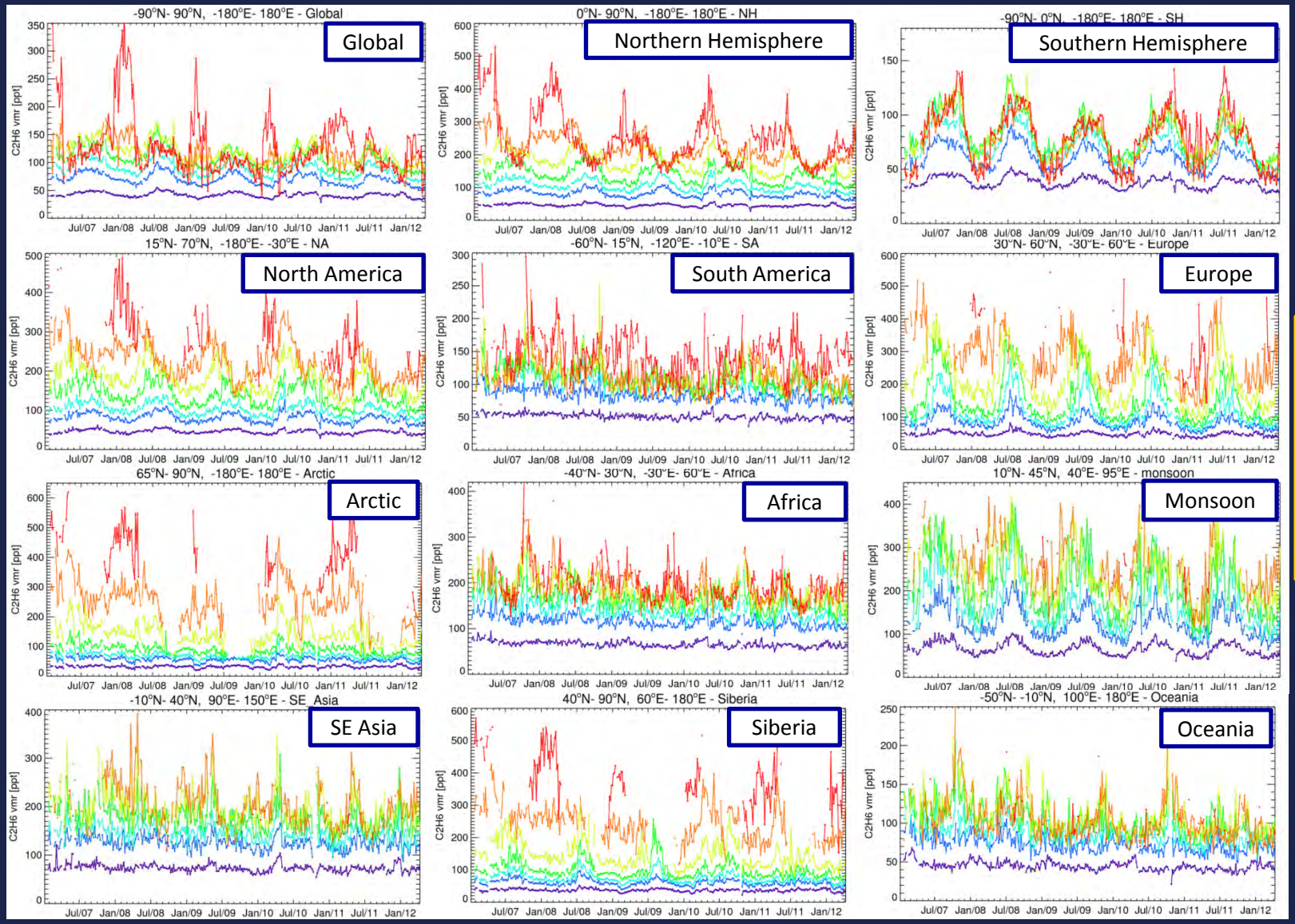
Ethane measurements from MIPAS (1)



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