ACE-FTS satellite measurements of HCN in the upper troposphere to N_2O in the lower thermosphere

Patrick E. Sheese[1], Kaley A. Walker[1,2], Chris D. Boone[2], Peter F. Bernath[3]

[1]University of Toronto, Toronto, ON, Canada

[2]University of Waterloo, Waterloo, ON, Canada

[3]Old Dominon University, Norfolk, VA, USA

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ACE-FTS

Atmospheric Chemistry Experiment – Fourier Transform Spectrometer

- Canadian satellite SciSat was launched into a circular, high-inclination orbit in August 2003
  - ACE-FTS and MAESTRO instruments on board
- ACE-FTS is a solar occultation instrument
  - High spectral resolution FTS in the 2.2 to 13.3 μm spectral range
  - 30+ trace species are retrieved, as well as 20+ subsidiary isotopologues
  - Vertical resolution of 3-4 km
- *NEW* ACE-FTS level 2 version 3.5/3.6 data were used in this study
  - Complete dataset spans Feb 2004 – yesterday
  - Download it today!

N_2O

Stratospheric N_2O

- Surface sources:
  - Ocean and soil emissions
  - Agriculture
  - Biomass burning and fossil fuel combustion

- Injected into the stratosphere via Brewer-Dobson circulation
  - Used as dynamical tracer in stratosphere

- Sinks:
  - $\mathrm{N_2O} + h\nu(\lambda < 200\ \mathrm{nm}) \rightarrow \mathrm{N_2} + \mathrm{O}(^1\mathrm{D})$
  - $\mathrm{N_2O} + \mathrm{O}(^1\mathrm{D}) \rightarrow 2\mathrm{NO}, or\ \mathrm{N_2} + \mathrm{O_2}$

Possible upper atmospheric sources of N_2O

$$N_2(A^3\Sigma_u^+) + O_2 \rightarrow N_2O + O$$

(~95 km, descent)

- **Zipf and Prasad [1982], Nature**
  - Predicted ~10^9 cm^{-3} (~1-10 ppm) in lower thermosphere during strong magnetic storms

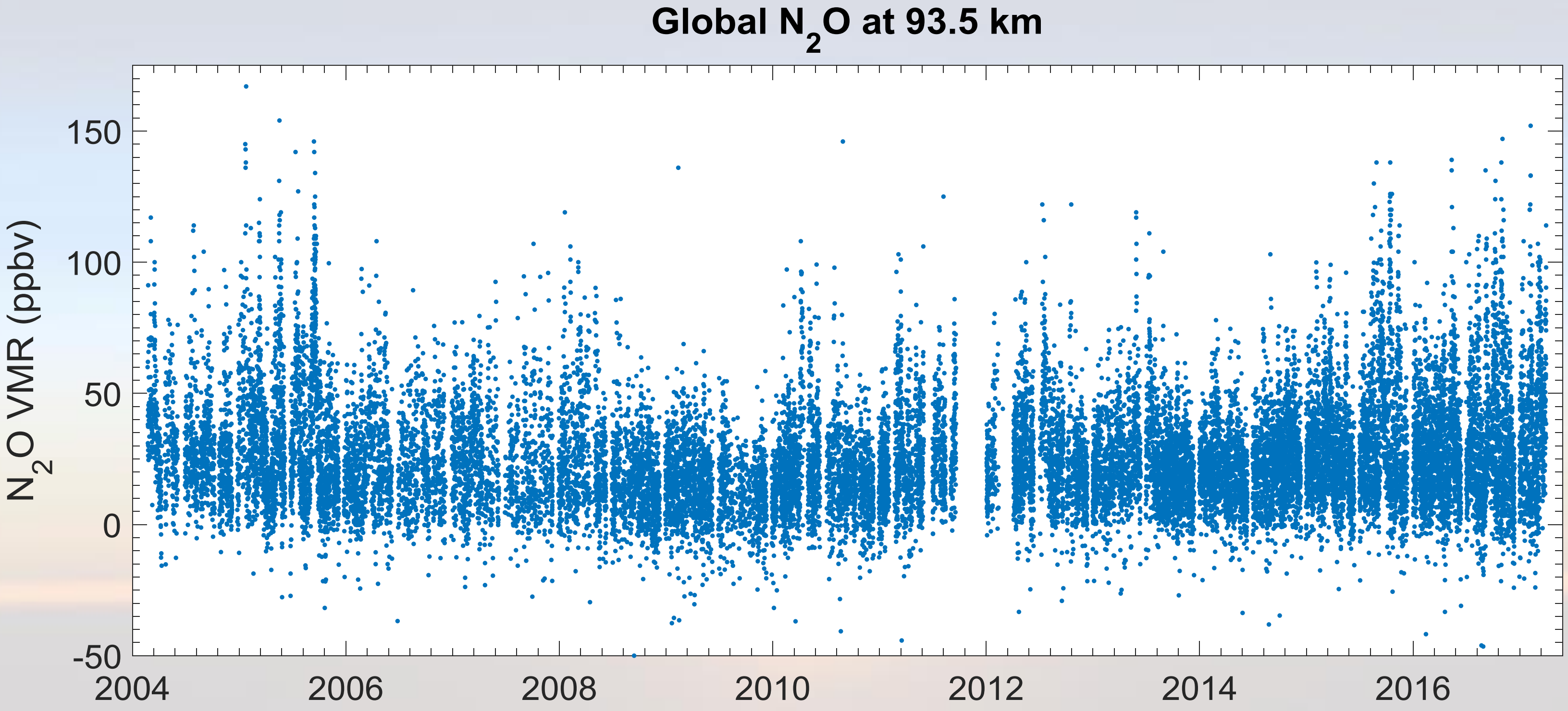
$$NO_2 + N(^4S) \rightarrow N_2O + O$$

(~75 km, descent)

- **Funke et al. [2008a], ACP**
  - MIPAS N_2O enhancement after 2003 solar proton event
- **Semeniuk et al. [2008], JGR**
  - CMAM reproduces 2004 ACE-FTS v2.2 N_2O enhancement in upper stratosphere

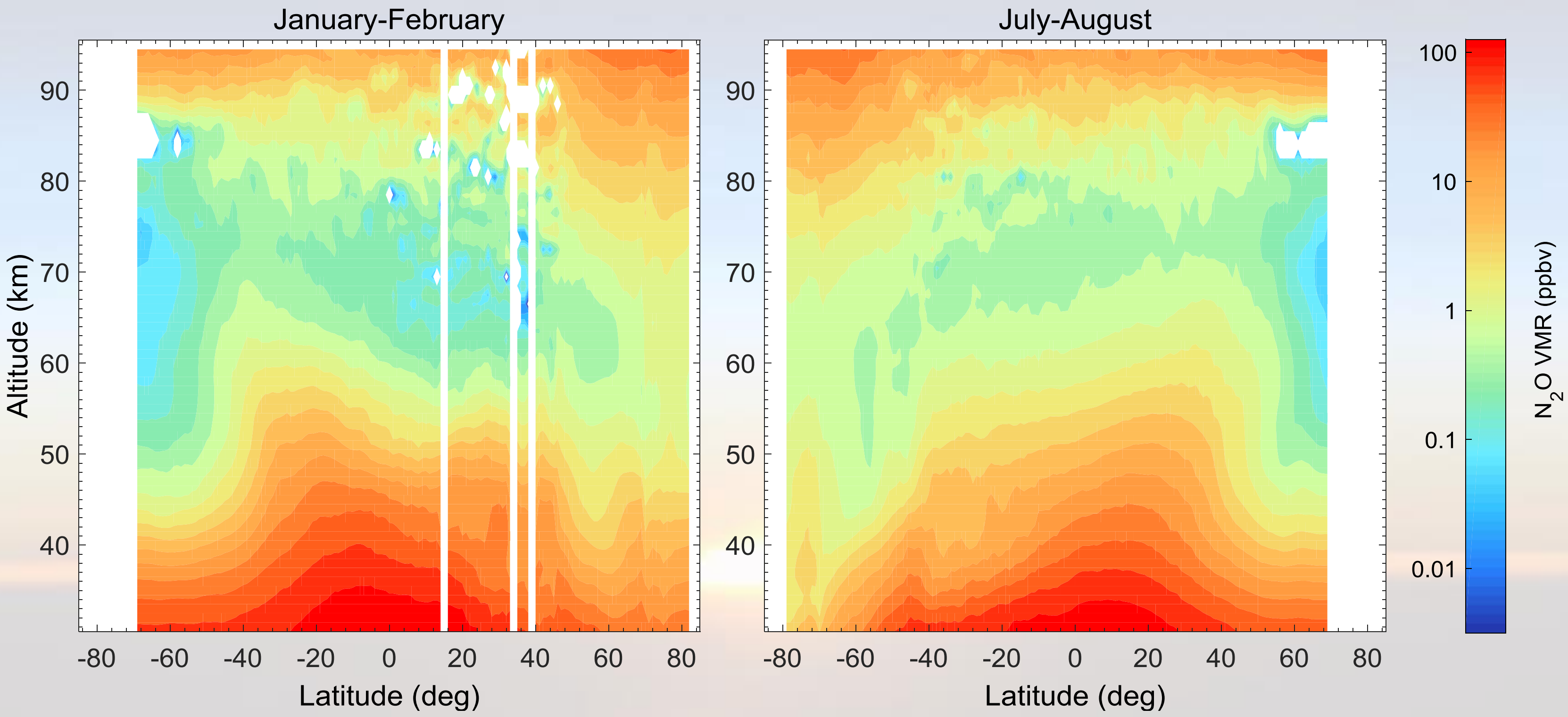
- Funke et al. [2008b], ACP
  - MIPAS mesospheric N_2O enhancements during polar winter
    - Predicted N_2O VMRs of ~100 ppb in lower thermosphere

ACE-FTS N_2O data in thermosphere



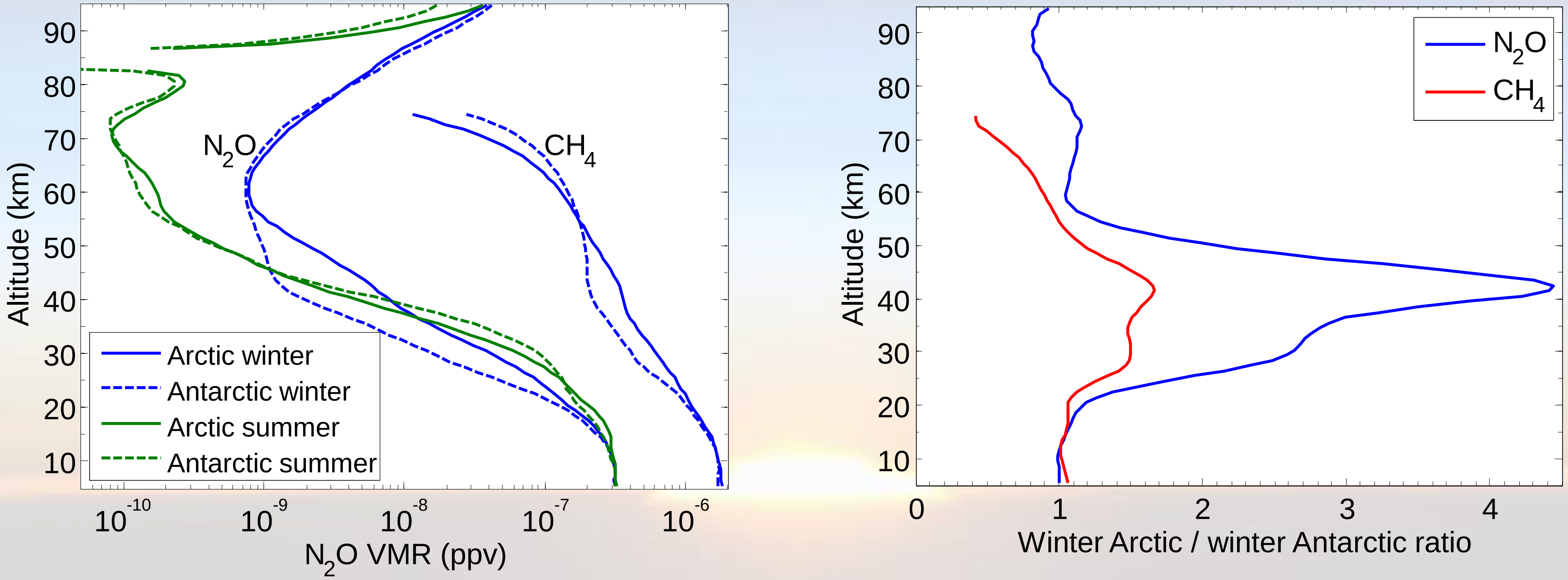
- Typically in the 10's of ppb
- Reaches over 100 ppb during times of strong solar activity

ACE-FTS N_2O climatology



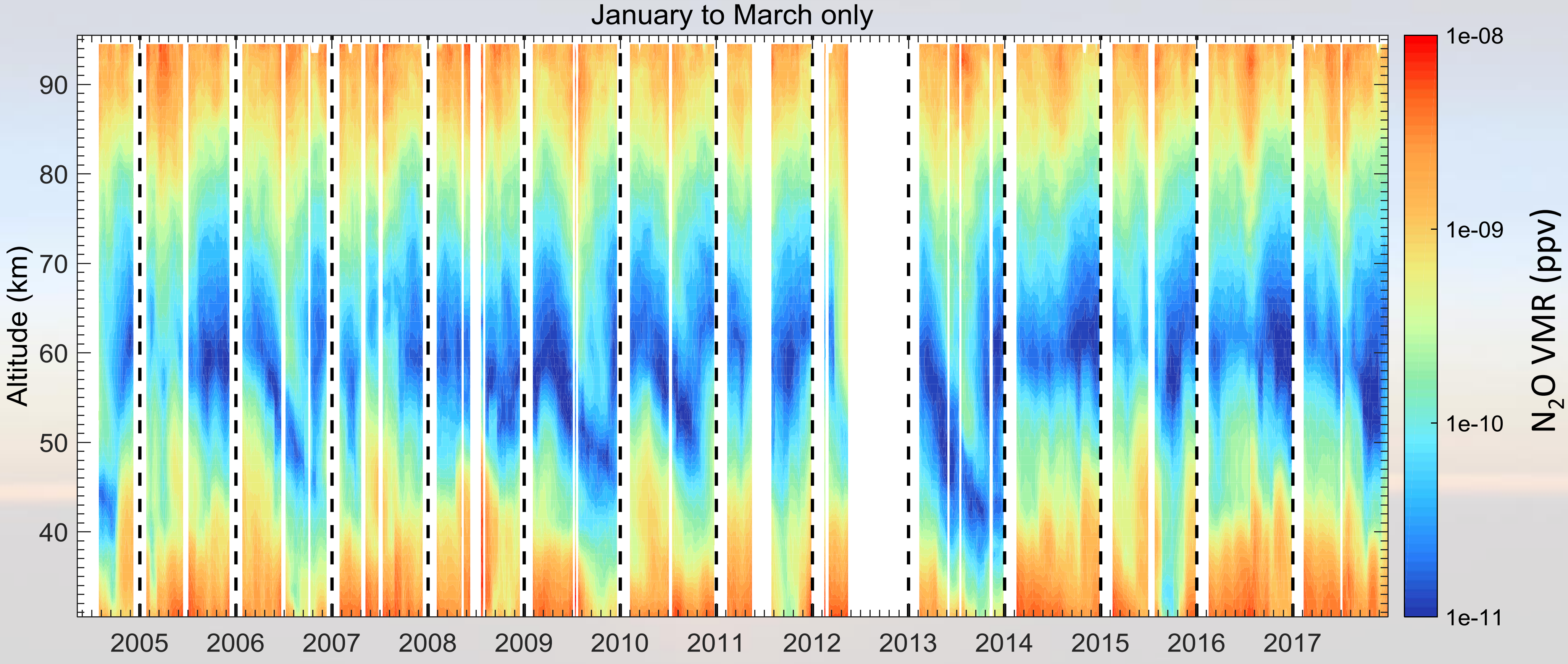
- Stratospheric region exhibits Brewer-Dobson circulation
- Clear thermospheric N_2O source

Mean ACE-FTS profiles



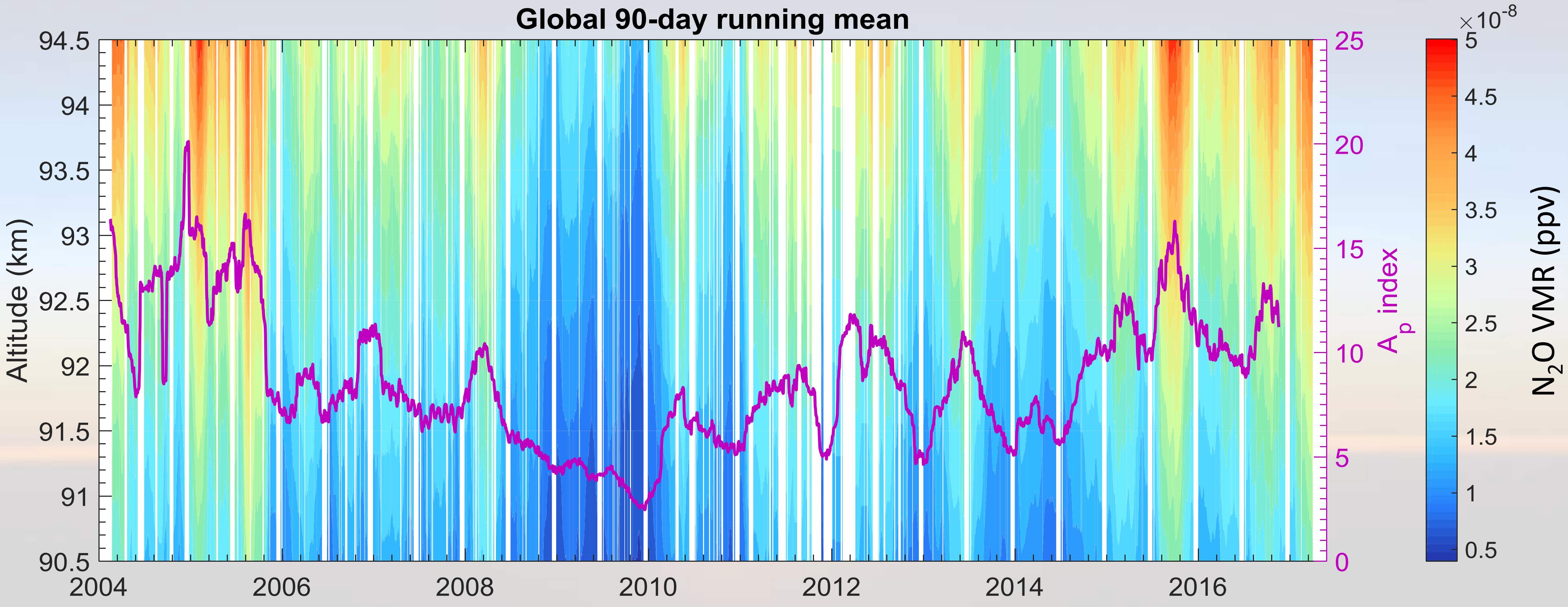
- **Arctic winter and Antarctic summer – Dec-Feb**
- **Arctic summer and Antarctic winter – June-Aug**

ACE-FTS Arctic time series



- **Regular Arctic winter N_2O intrusions in stratopause region**
  - **Especially during sudden stratospheric warmings**

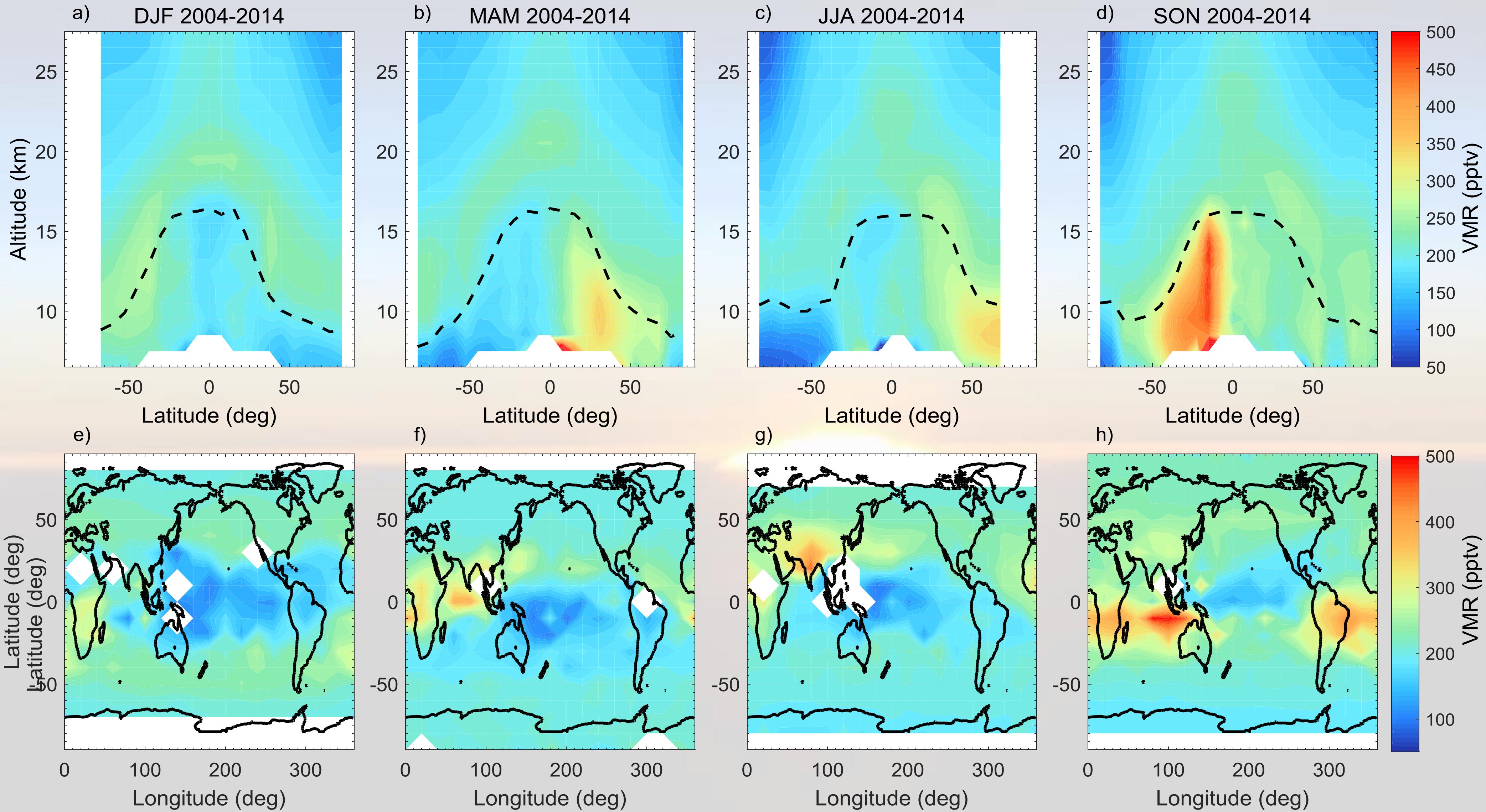
11-year solar cycle



- A_p index as a proxy for energetic particle precipitation

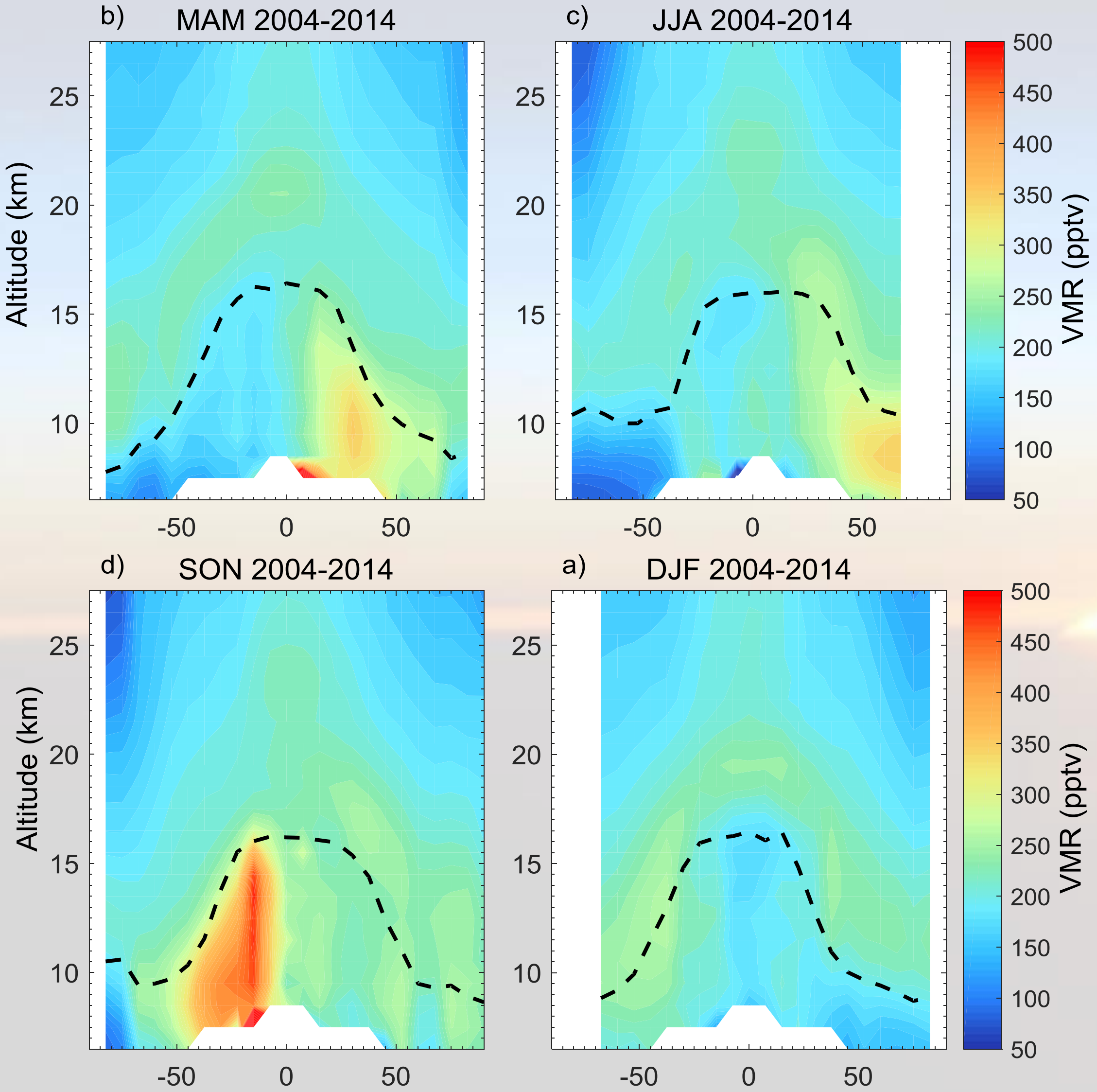
HCN in the UTLS

HCN seasonal climatology



14.5 km

ACE-FTS and MIPAS HCN climatology



N. Glatthor et al.: Variations in MIPAS HCN amounts

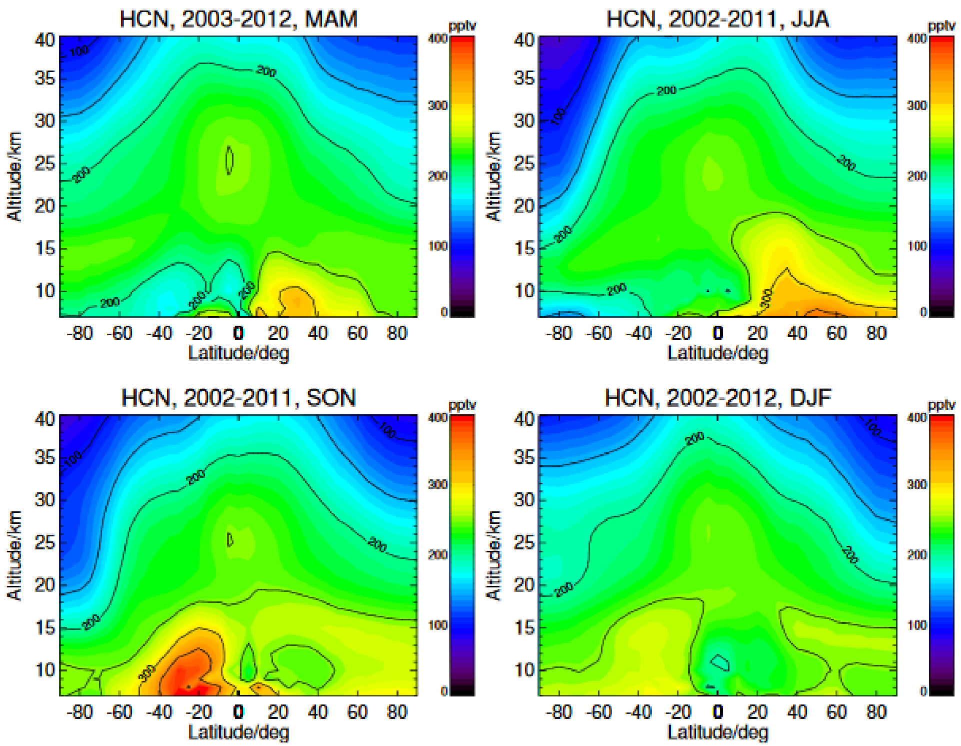


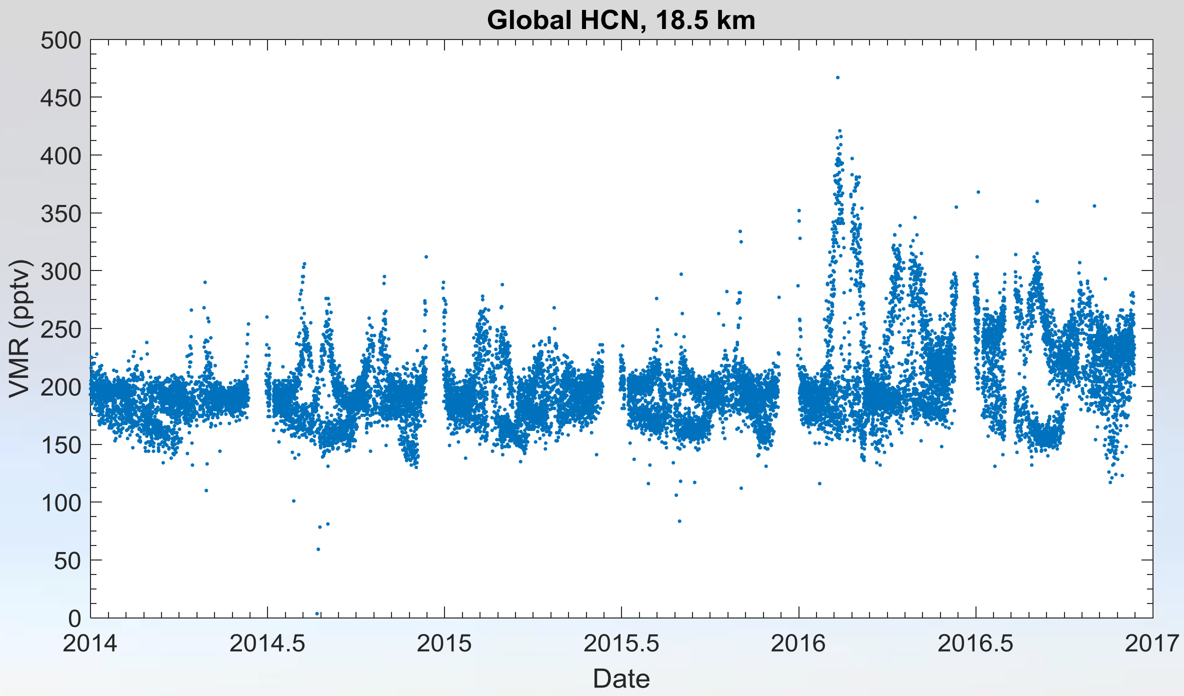
Figure 2. Climatological latitude–height cross sections of HCN volume mixing ratios measured by MIPAS during March to May (top left), June to August (top right), September to November (bottom left) and December to February (bottom right). The distributions are averaged over the time period 2002 to 2012.

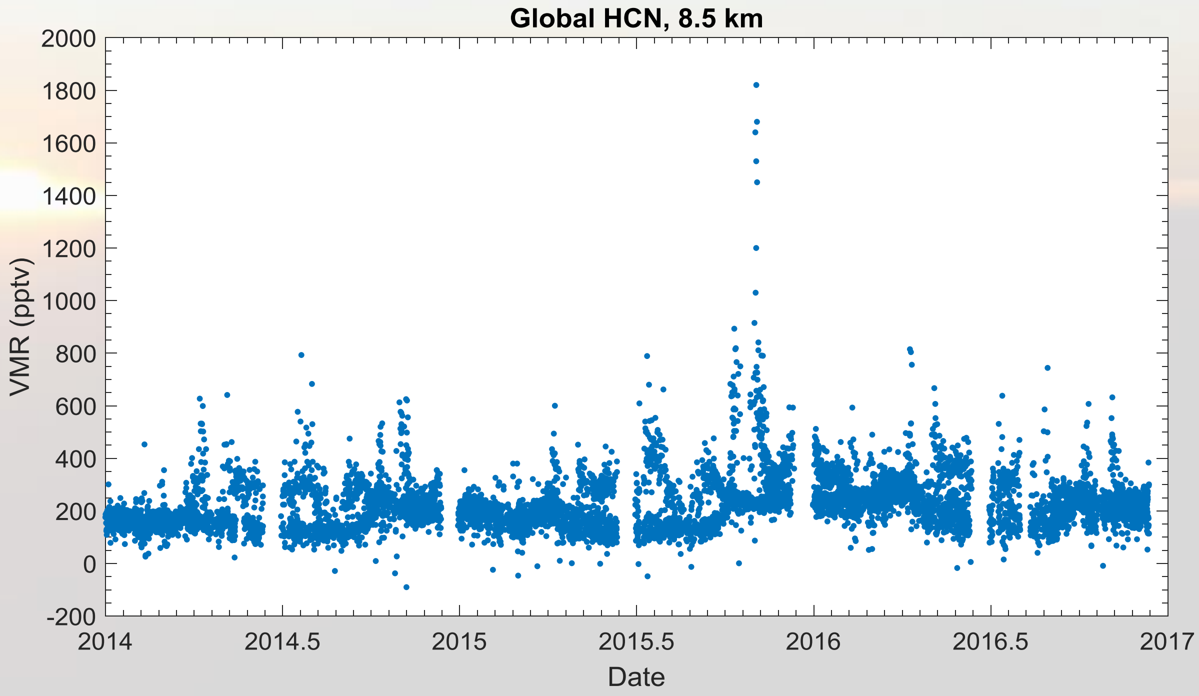
- **Good qualitative agreement**
- **ACE-FTS exhibits a positive bias**
- **(or MIPAS exhibits negative bias)**
- **MLS instrument on NASA's Aura satellite also measures HCN, but not below 16 km**

Looking at ACE-FTS HCN

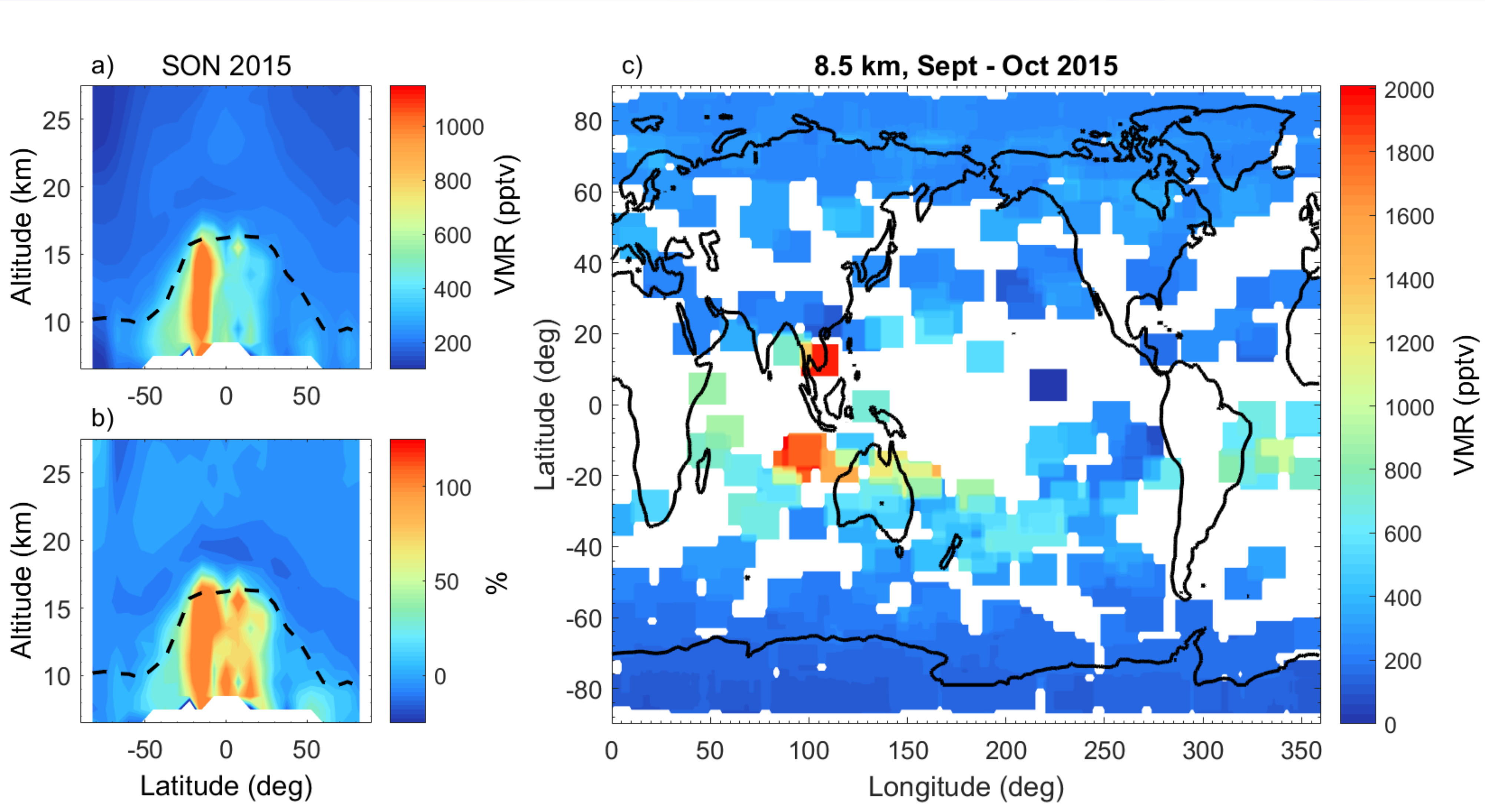
- Clear structure in 2016 stratospheric data
  - Likely not just increase noise (not instrument or processing error)

- Strong enhancement in upper tropospheric data just before 2016

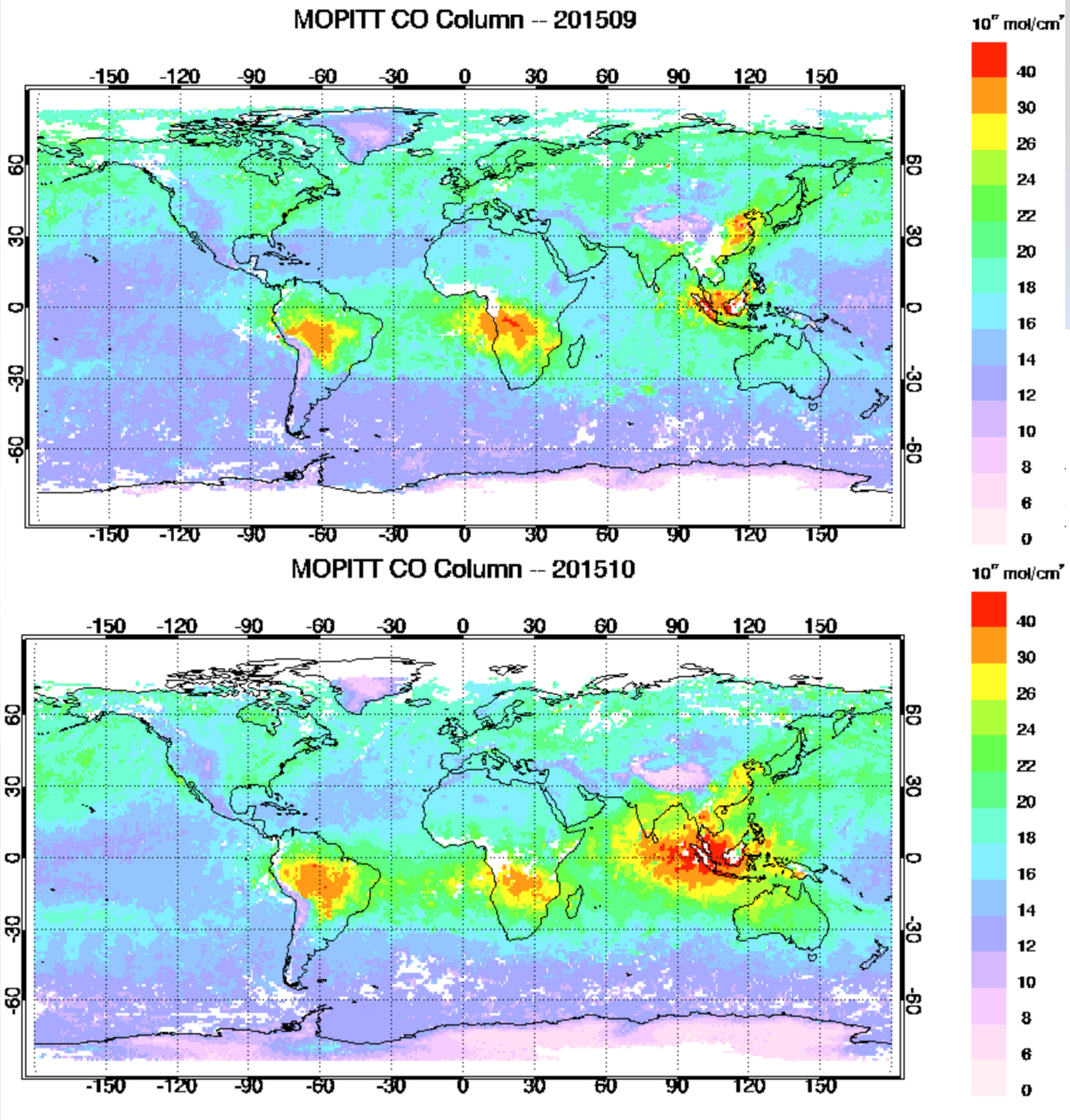


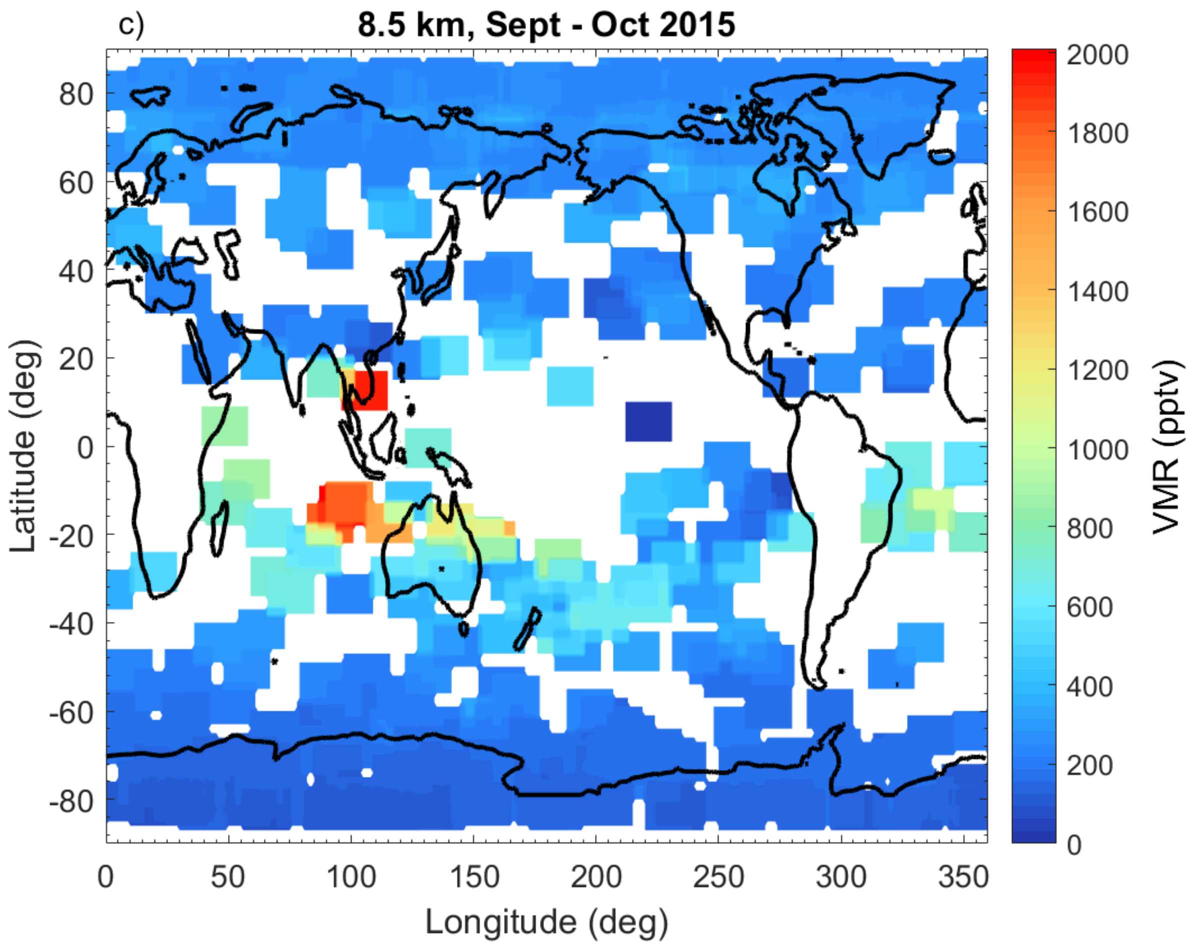


2015 enhancement



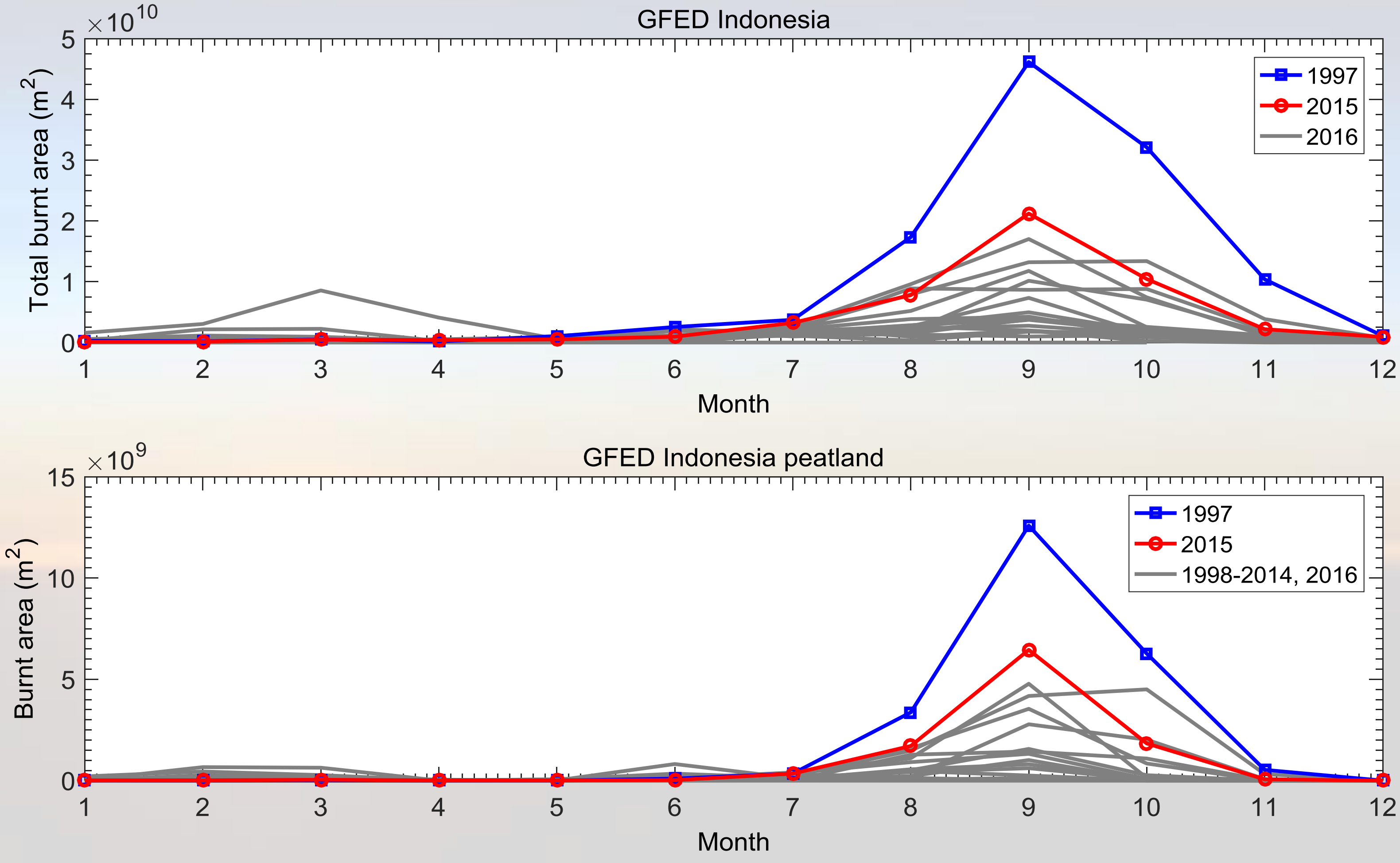
2015 enhancement seen in MOPITT CO data



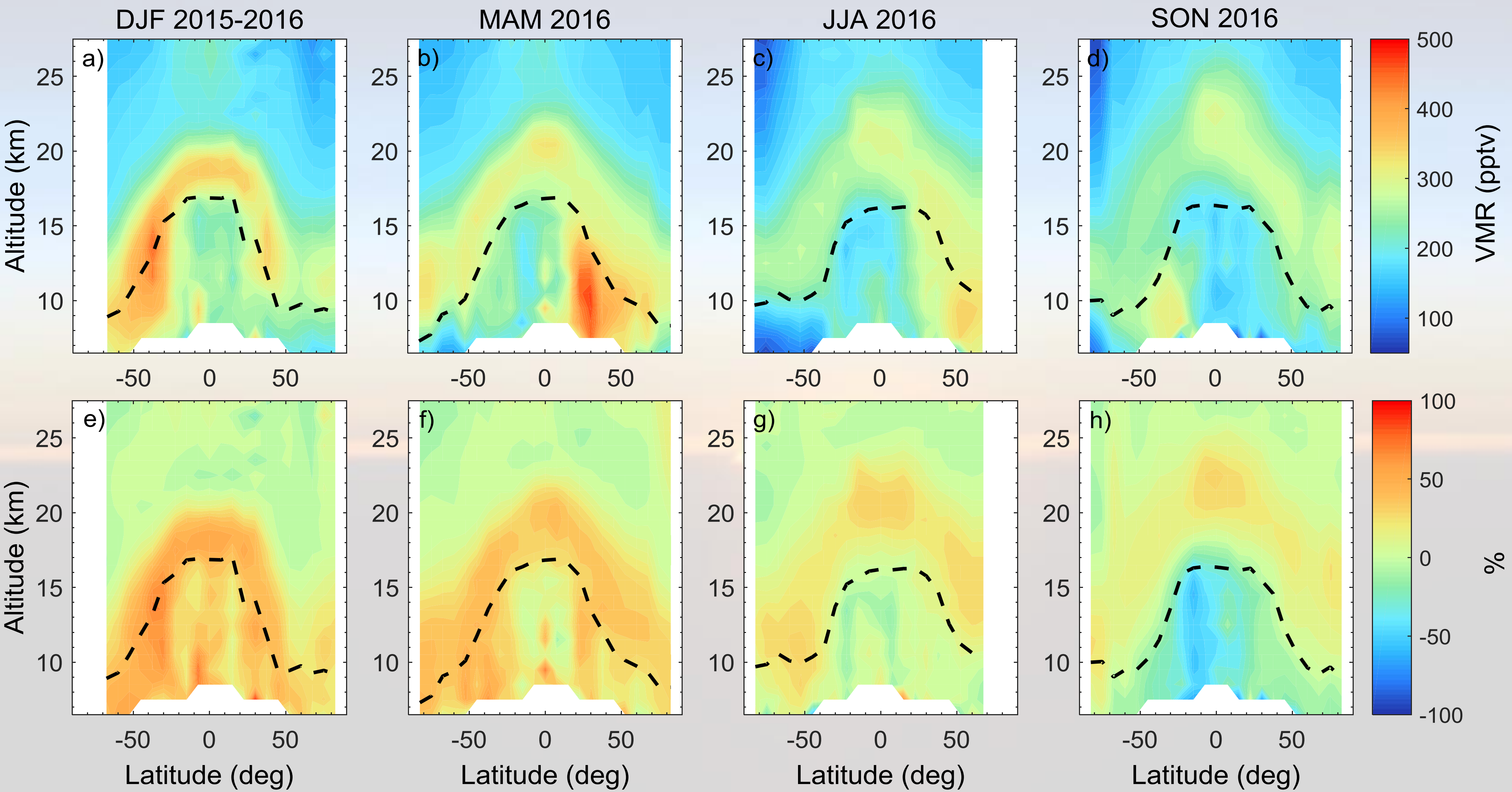


GFED

- Global Fire Emissions Database
  - Burnt area, including small fires
  - Includes different land types, e.g. peatland
  - Burning peat emits ~5-10x more HCN than other typical biomass



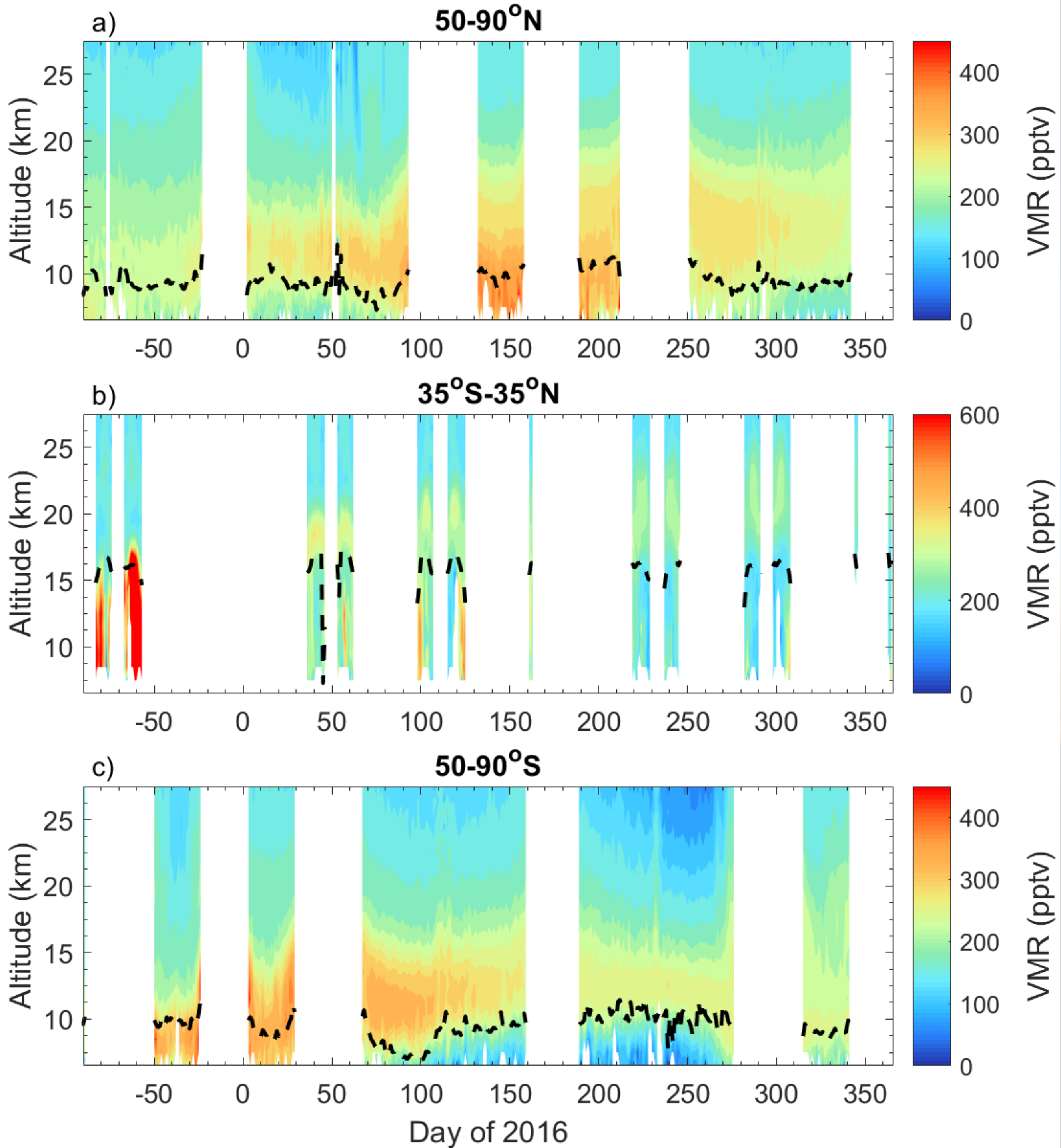
HCN seasonal time series



HCN profile time series

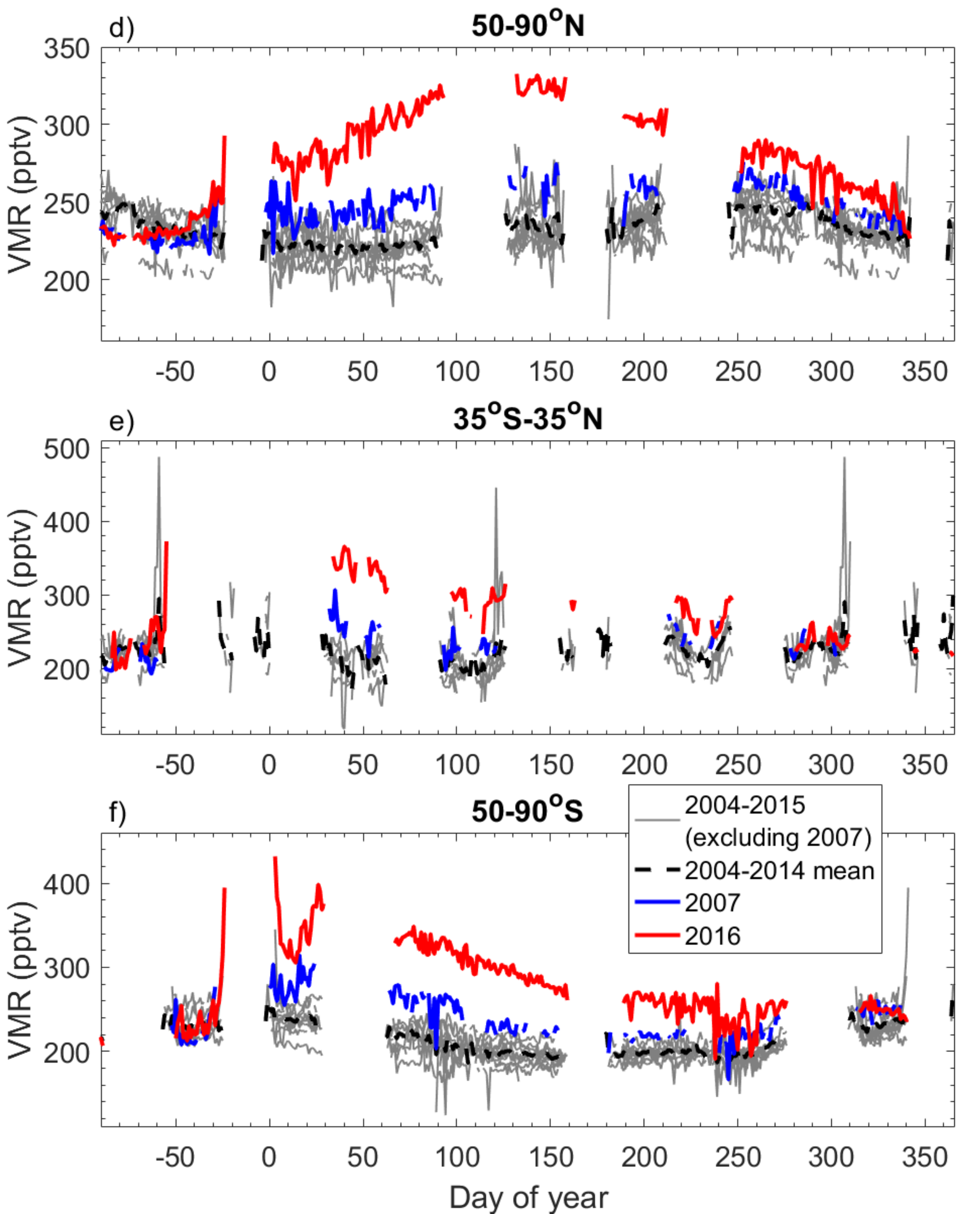
- **50-90°N**: lower stratospheric HCN values increase in early 2016 with no similar increase in upper troposphere
  - Enhancement in May in UTLS
- **30°S-30°N**: extreme increase in September-October 2015 in upper troposphere
  - Enhancements in April in upper troposphere
- **50-90°S**: from Novermber to April HCN enhancement transported from upper troposphere to lower stratosphere

daily mean values



HCN 2 km above tropopause

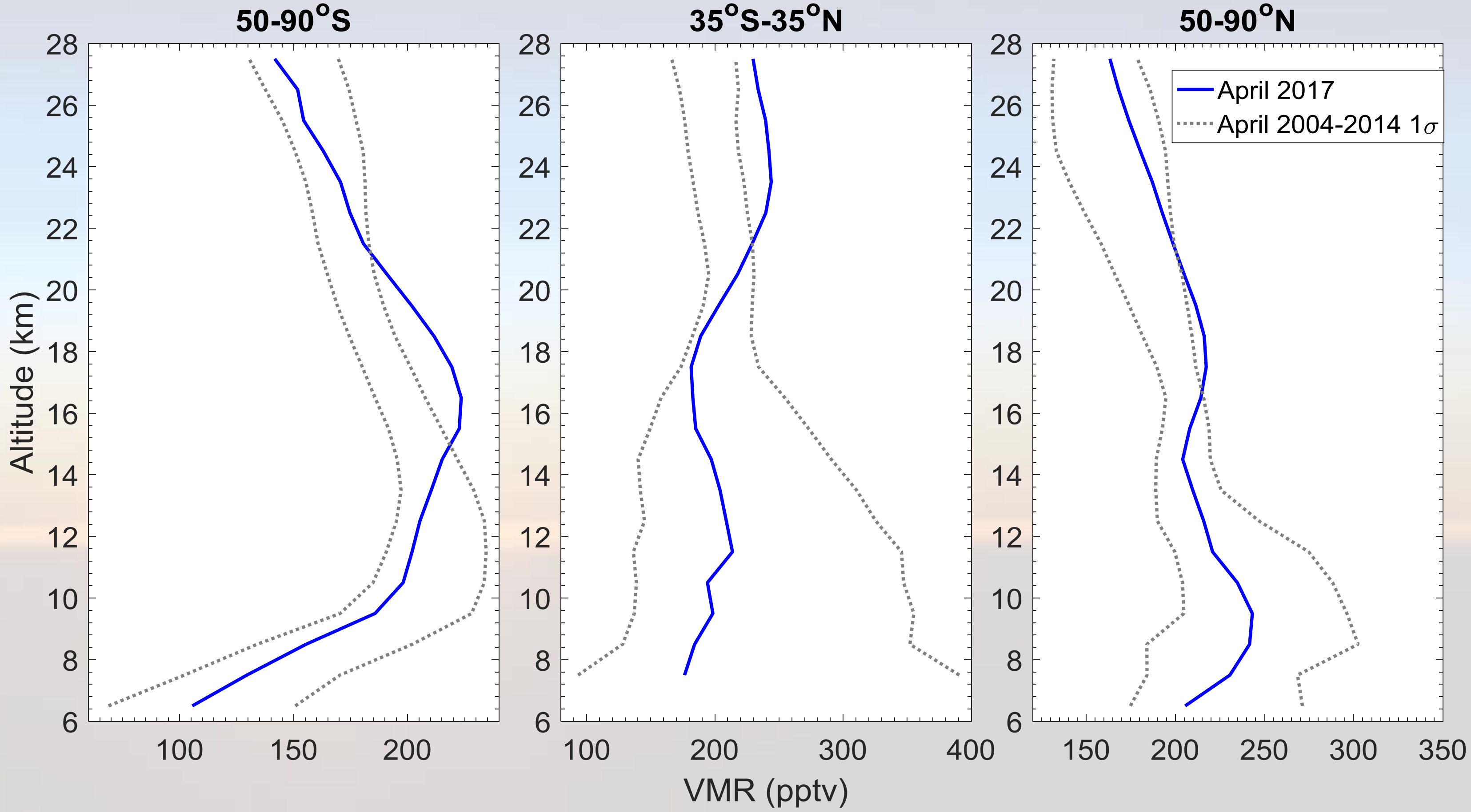
- Values are not just above average, they are greater than all previously measured values
- Possible that lower stratospheric HCN concentrations were greater in 1997



daily mean values

April mean HCN profiles

- **Most recent data still shows enhancement (greater than 1σ from climatological mean) in lower stratosphere**



Summary

- N_2O
  - ACE-FTS has the only measurements of N_2O in the upper mesosphere – lower thermosphere
  - Clear EPP source in lower thermosphere
    - Continual source throughout all seasons
      - ~10 ppb near equator
      - ~30 ppb near poles
  - N_2O Transported down into upper stratosphere in winter
  - In Arctic winter 40-50 km region, N_2O can be predominantly thermospheric
    - In summer, purely tropospheric
- HCN
  - Strong El Niño conditions in 2015 led to increased temperatures and drought conditions in Southeast Asia, including Indonesia, leading to most intense biomass burning season since 1996 (which also was a strong El Niño year)
    - Peatland fires were intense and emitted highest levels of HCN in the UTLS in the ACE-FTS record
  - HCN was transported from troposphere into lower stratosphere in late 2015/early 2016, and transported to higher latitudes
    - 2016 had enhanced levels HCN throughout the year at all latitudes

Thanks!

The extra bits

Literature!

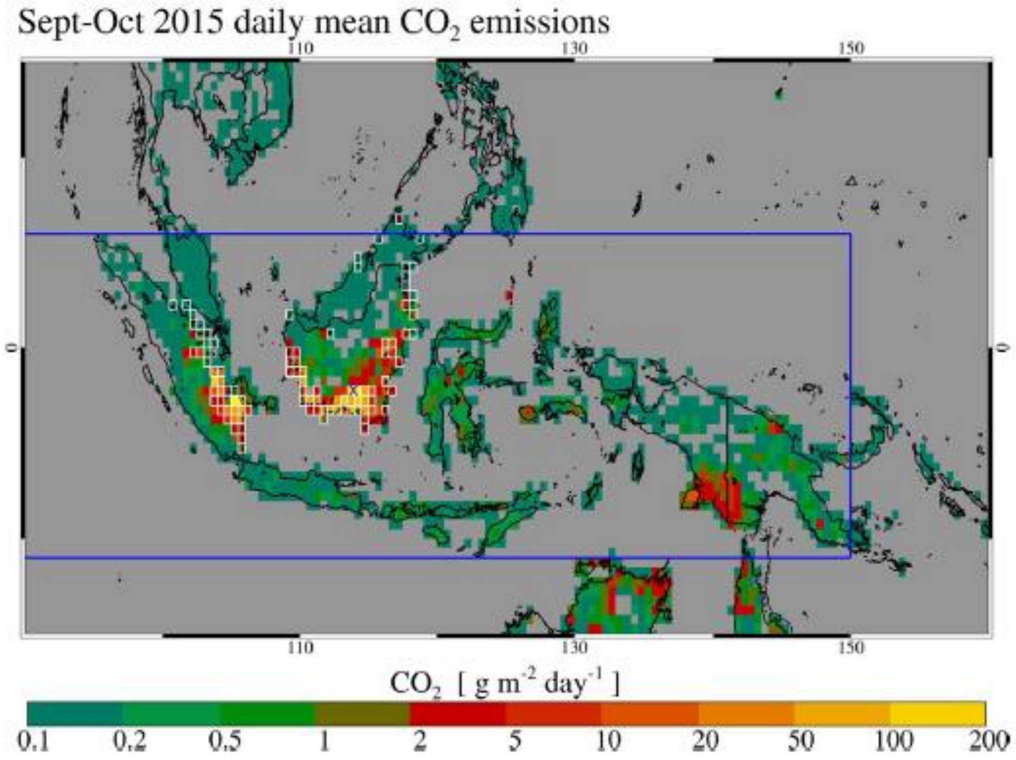


Figure 1. Daily mean CO_2 emissions from peat and vegetation fires burning across maritime southeast Asia in Sept-Oct 2015, presented in 0.5° × 0.5° grid cells. Cells containing peat soils according to landcover data used in GFAS[23] are outlined in white (see Supplementary Information B). Locations of our *in situ* trace gas measurements lie close to the Central Kalimantan Capital of Palankaraya, Kalimantan (113.92°E, 2.21°S), indicated with the blue cross (See also Fig. 3). The thick blue line indicates the border of the study domain (east part only shown, full range 70°E–150°E; 11°S–6°N). Map was generated using IDL v8.4 software, http://www.exelisvis.com.

Fire carbon emissions over maritime southeast Asia in 2015 largest since 1997

V. Huijnen[1], M. J. Wooster[2,3], J. W. Kaiser[4], D. L. A. Gaveau[5], J. Flemming[6], M. Parrington[6], A. Inness[6], D. Murdiyarso[5,7], B. Main[2] & M. van Weele[1]

Literature!

Atmospheric CH_4 and CO_2 enhancements and biomass burning emission ratios derived from satellite observations of the 2015 Indonesian fire plumes

Robert J. Parker[1,3], Hartmut Boesch[1,3], Martin J. Wooster[2,3], David P. Moore[1,3], Alex J. Webb[1], David Gaveau[4], and

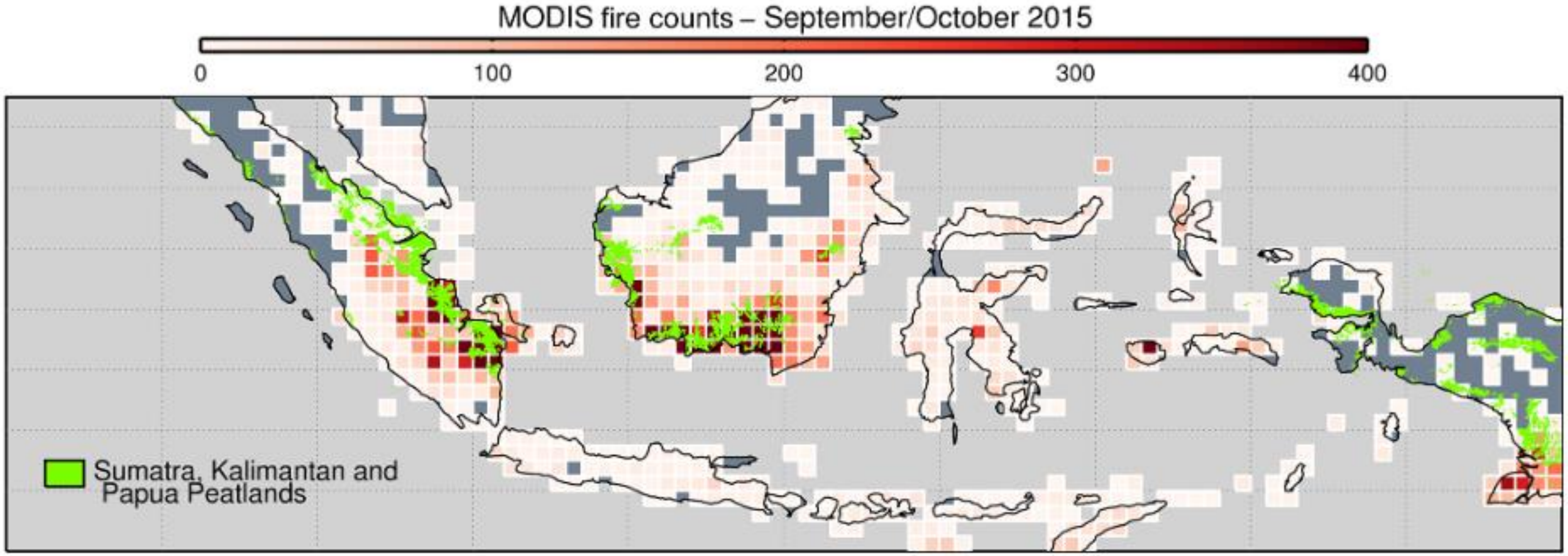
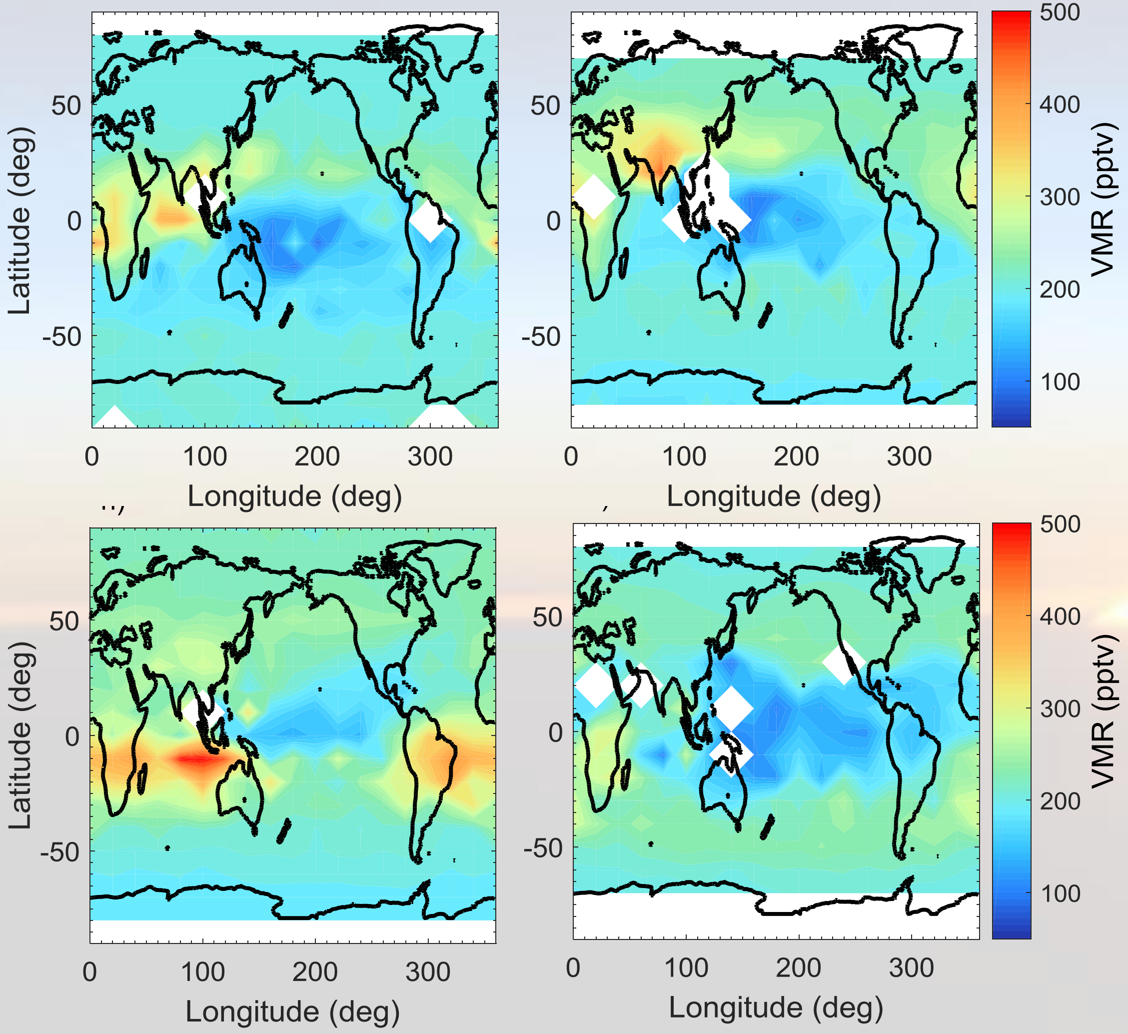


Figure 2. MODIS fire counts for September–October 2015 over the Indonesia, gridded into 0.5° × 0.5° boxes. Also overlaid are the locations of known peatlands in Sumatra (left), Kalimantan (centre) and Papua (right).

ACE-FTS and MIPAS HCN climatology



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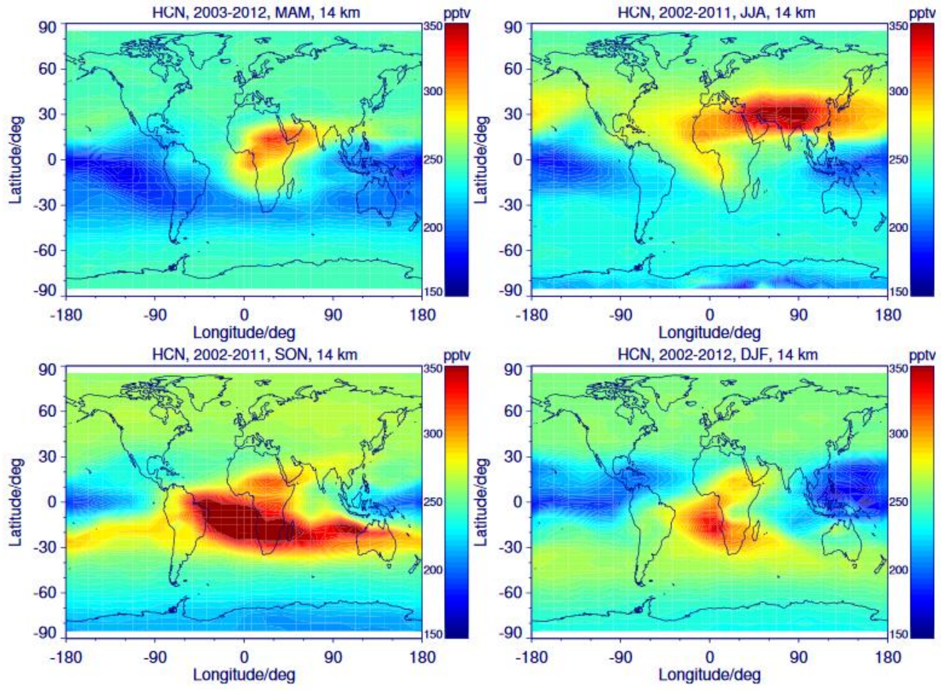
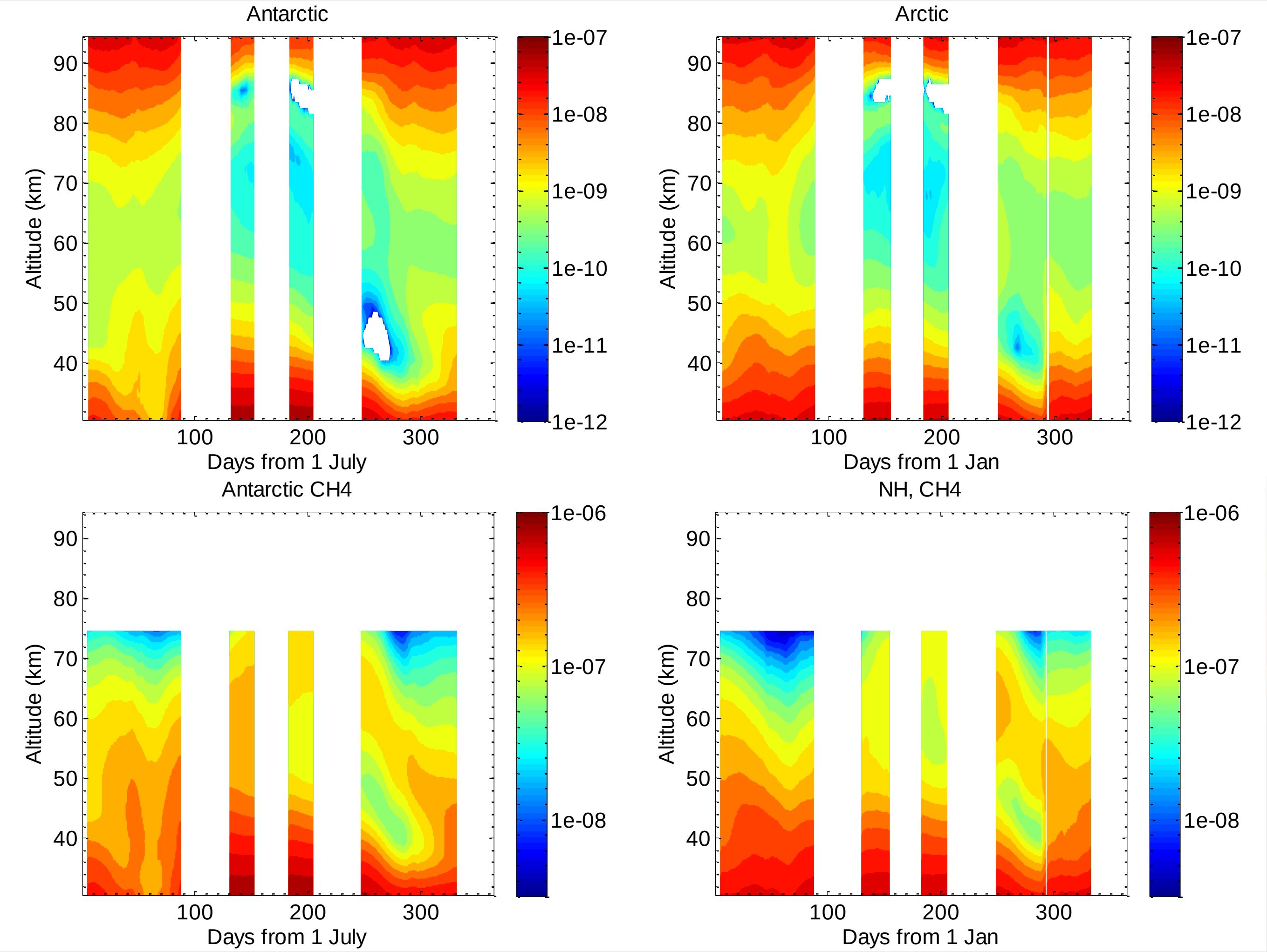


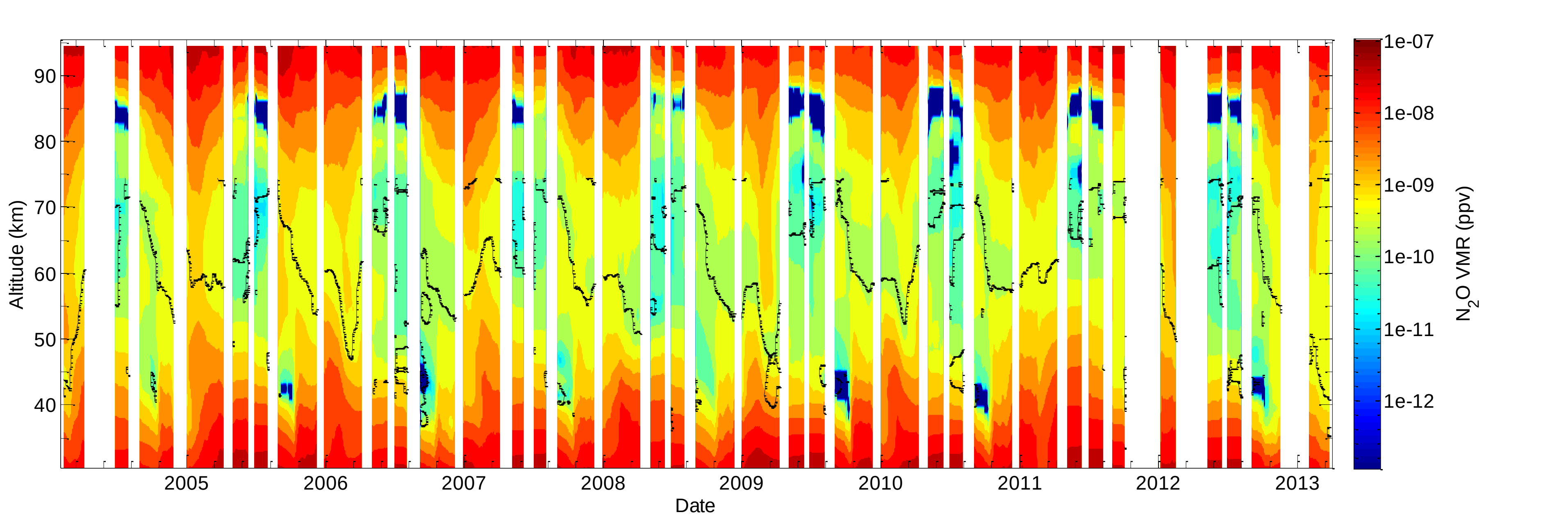
Figure 3. Climatological global HCN distributions measured by MIPAS during March to May (top left), June to August (top right), September to November (bottom left) and December to February (bottom right) at 14 km altitude. The distributions are averaged over the time period 2002 to 2012. Here and in subsequent contour plots values exceeding the displayed VMR range are also displayed in dark red.

- **Good qualitative agreement**
- **ACE-FTS exhibits a positive bias**
  - **(or MIPAS exhibits negative bias)**
  - **MLS instrument on NASA's Aura satellite also measures HCN, but not below ~16 km**

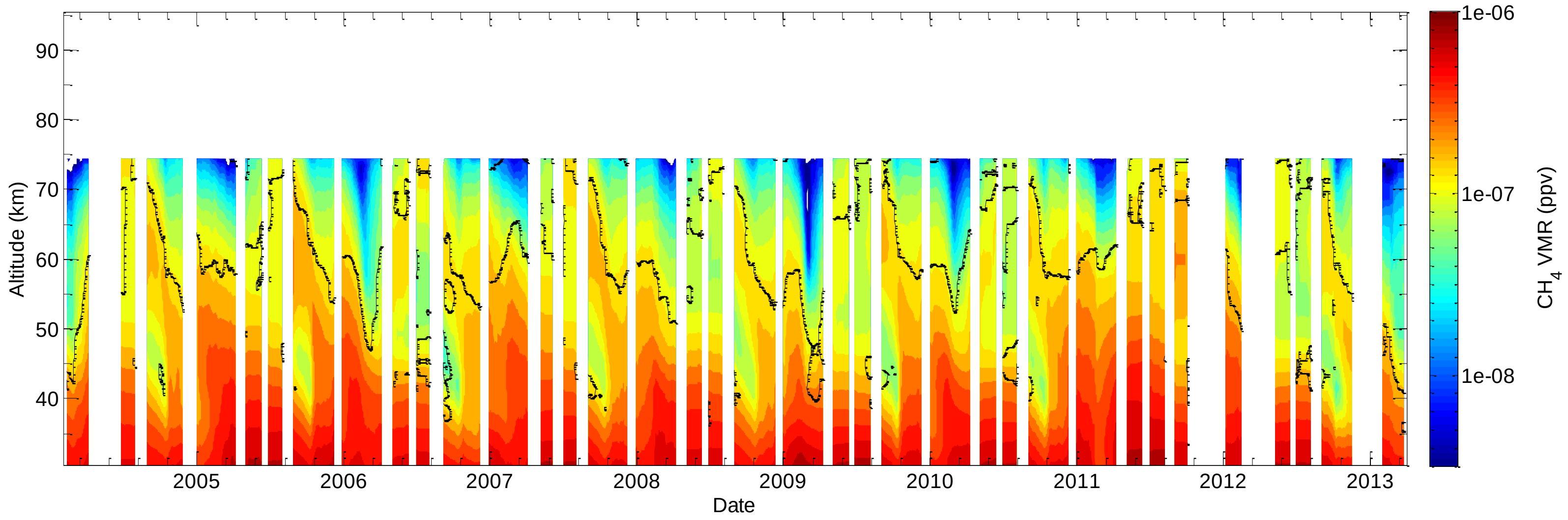


Antarctic
Arctic
Altitude (km)
Days from 1 July
Days from 1 Jan
1e-07
1e-08
1e-09
1e-10
1e-11
1e-12
Antarctic CH4
NH, CH4
1e-06
1e-07
1e-08

N2O time series

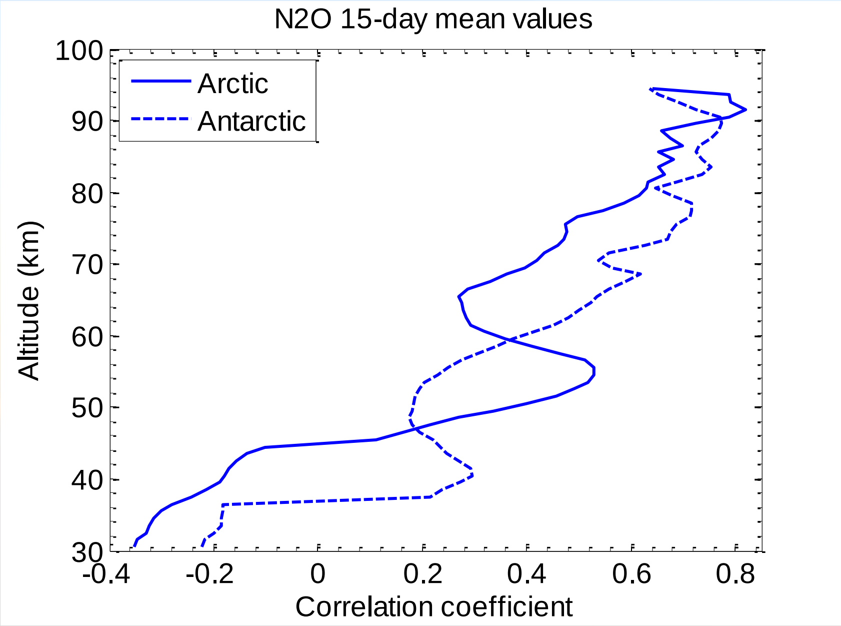


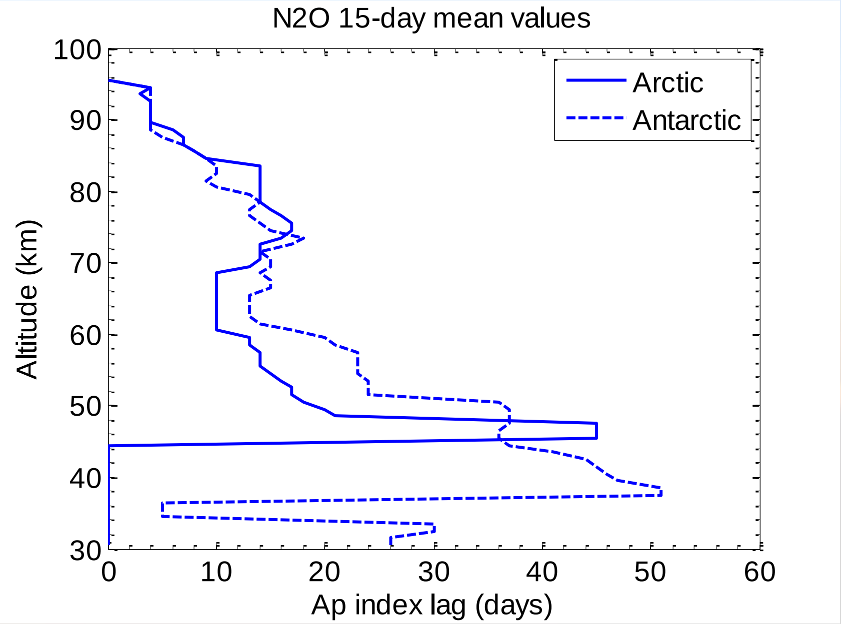
CH4 time series



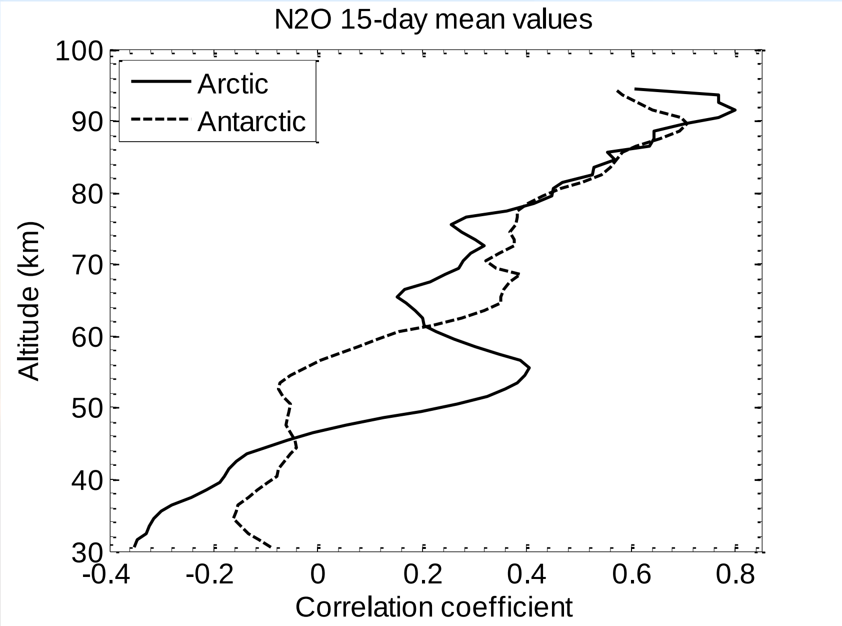
Winter N_2O correlation with A_p index

0-60 day lags added to Ap index time series



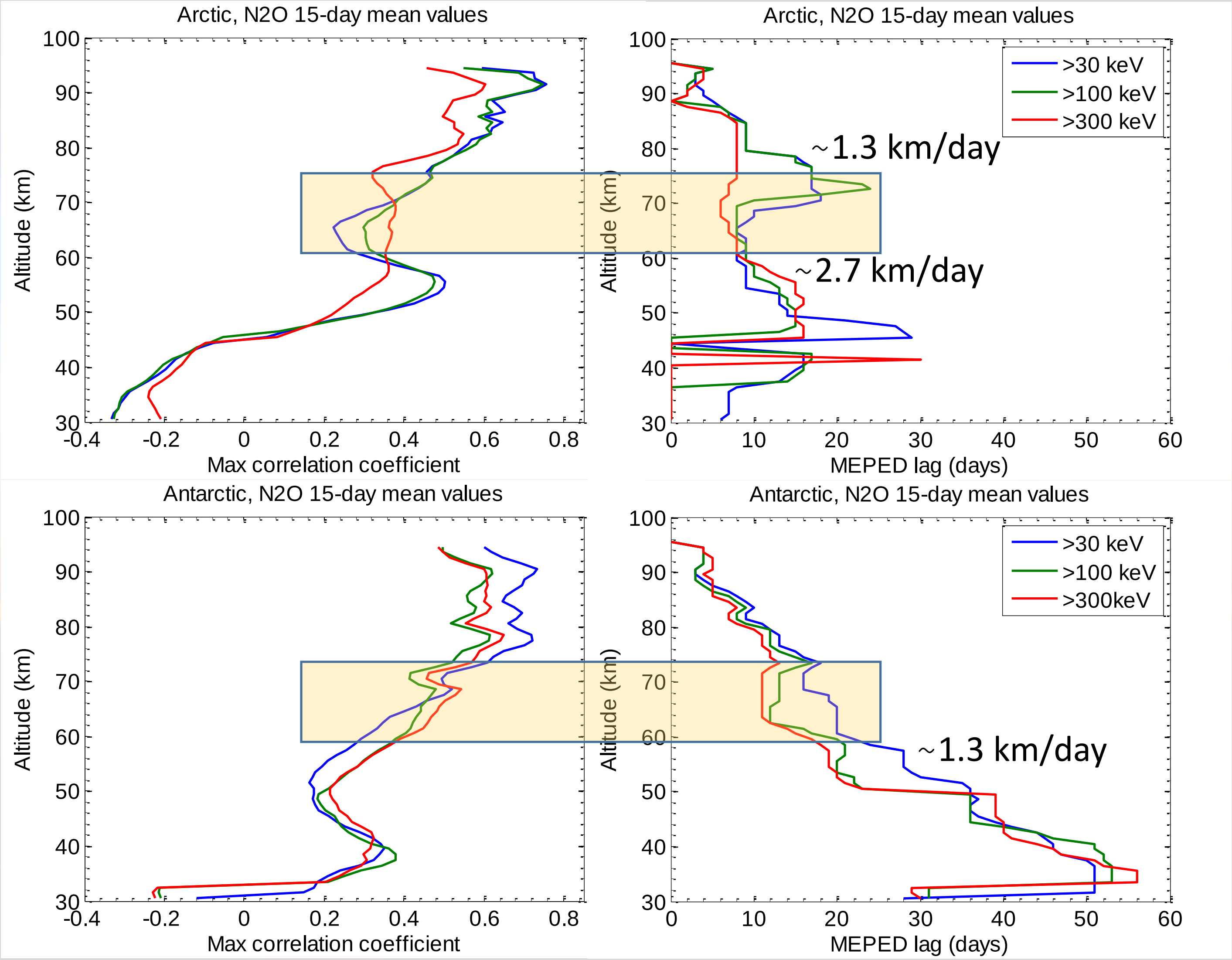


Winter N_2O correlation with A_p index



Winter correlation with MEPED

- MEPED (on NOAA-16 POES) measures electron fluxes at top of atmosphere
  - Ionization/dissociation due to precipitating electrons peak near
    - ~90 km for > 30 keV
    - ~75 km for > 100 keV
    - ~60 km for > 300 keV



- 0-60 day lag introduced in MEPED data