Modelling disturbed stratospheric Chemistry during solar induced NOx Intrusions observed by MIPAS-ENVISAT

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Motivation

- Possible solar influence on climate via amplification mechanism within the stratosphere
- Interference through radiative active gases, especially ozone
- **Direct effects through heating**
- **Indirect effects through change in chemistry**
  - **Direct by radiation and particle interaction**
  - **Indirect by mesospheric NOx intrusions**
- Change in dynamics to be translated to the troposphere

Evaluation through combination and comparison of

- **MIPAS-ENVISAT observations**
- **CTM-modelling**

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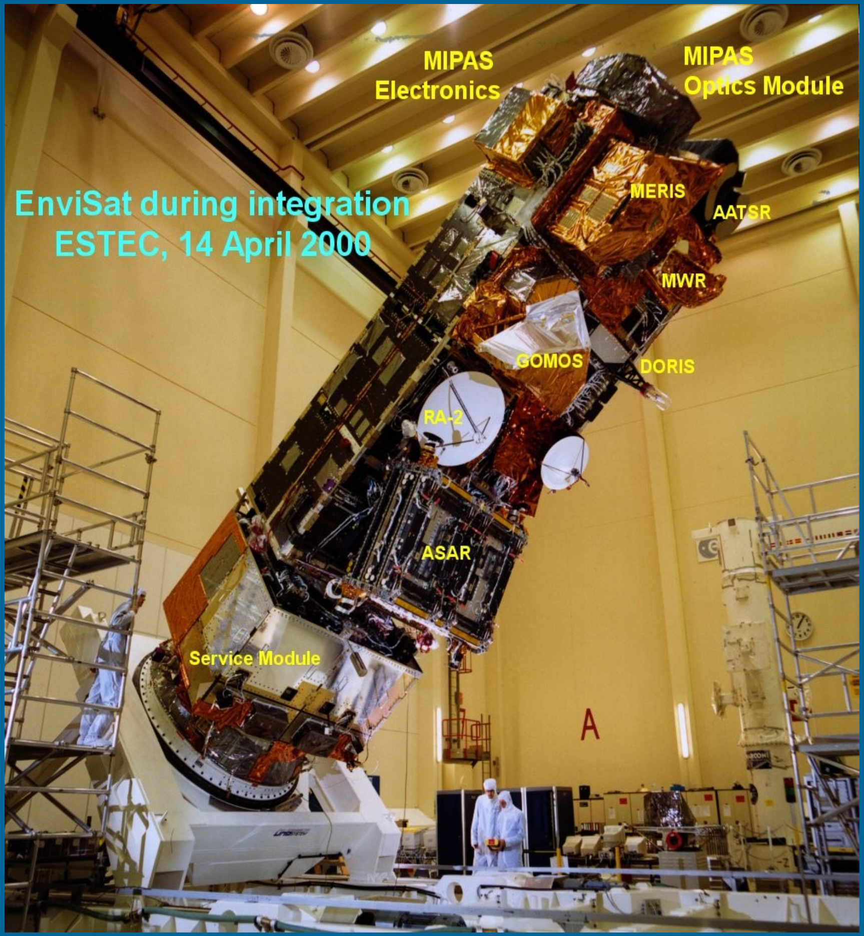
MIPAS instrument



- Michelson interferometer launched on 1 March 2002 on ENVISAT (sun-synchronous polar orbit, 98.5º).
- Spectra in 4.15-14.6μm (685- 2410 cm^{-1}) at 0.035 cm^{-1} in 4.5 s.
- Sensitivity: 4-30 nW/(cm^2 sr cm^{-1}).
- Radiometric accuracy: 1-3 % (4-15μm).
- Limb, altitude coverage: 6-68 km at 3 km steps in 85 s. (6-170 km)
- Global latitude (Day/Night) coverage
- 72 profiles/orbit; ~1000 profiles/day
- Continuous measurements Sep 2002-Mar 2004.
- Campaigns mode in 2005 (RR) (30-40%)
- Nominal obs. 2006 on … (40-80%)

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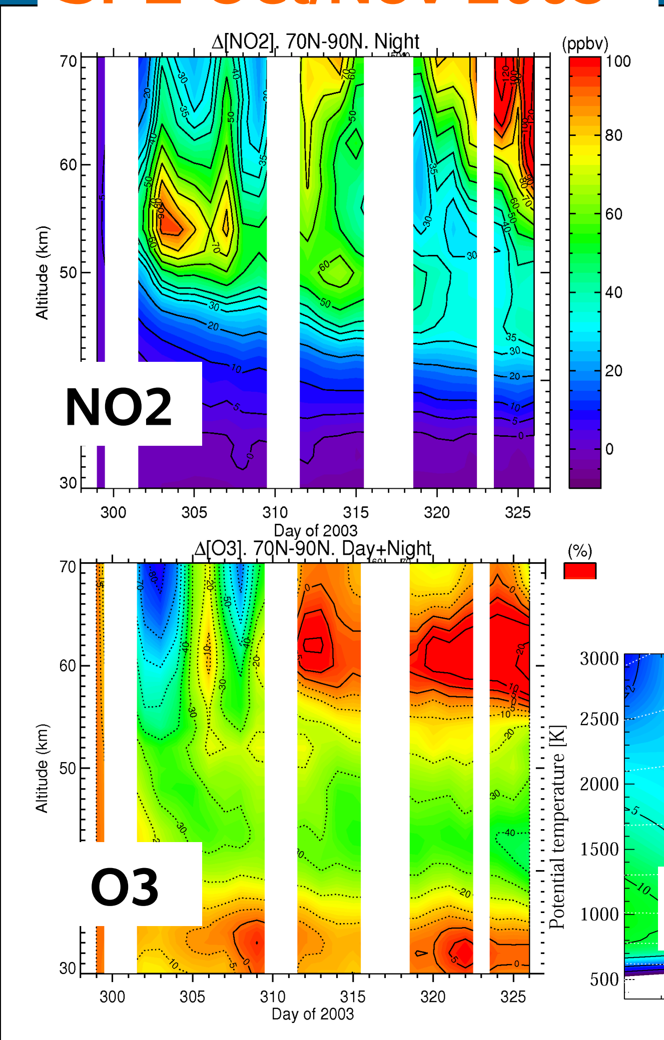
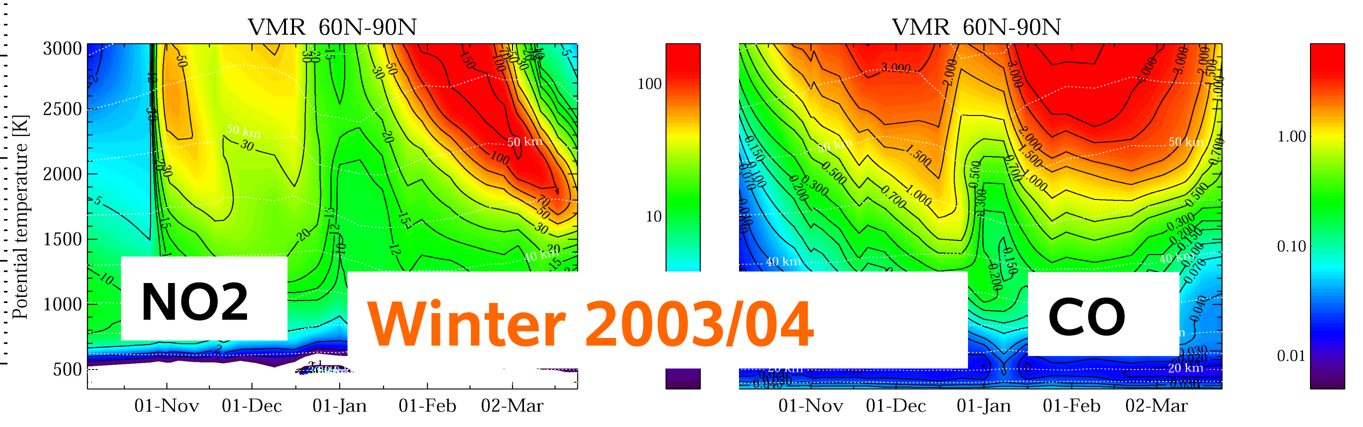
MIPAS sees particle effects in the strato-/mesosphere

SPE Oct/Nov 2003



- MIPAS observes effects related to solar proton events (SPEs) (in-situ particle effects) and NOx intrusions transported downward during polar winter
- Good spatial and time coverage with **standard product of ESA**
- More species and higher accuracy with **IMK/IAA retrieval**



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Description of the KASIMA model

- Euler and spectral, diagnostic and prognostic
- vertical range 7 - 120 km, vertical resolution variable with height
- horizontal resolution T21 (5.6° x5.6°)
- ECMWF analyses in combination with trend of prognostic part (T, Div, Vort nudged to analyses; below 17.5 km CTM); operational analyses after Sep 2003, ERA-40 before
- prognostic part uses physical parameterisation of net heating rates and breaking gravity waves
- Chemistry (108 reactions, 38 substances, heterogeneous reactions) up to 90 km
- On-line fast-j2 (vectorized) + Lyman-a
- Mixing ratios of source gases according WMO

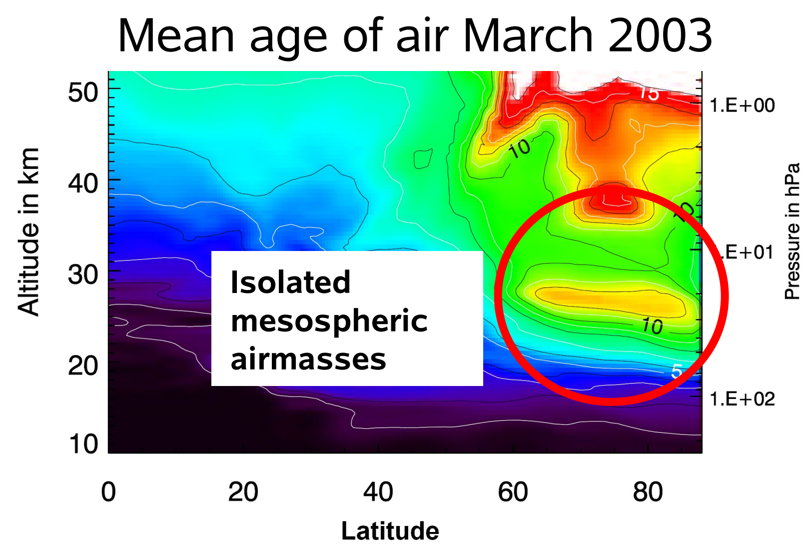

Figure 4: Meridional plot for l = 20., day 64

Applications

- Seasonal studies of transport and comparison with observations
- Polar chemistry,
- NOy budget in the lower stratosphere
- Multi-annual integrations with long-lived tracers
- Studies of SF6 with mesospheric chemistry

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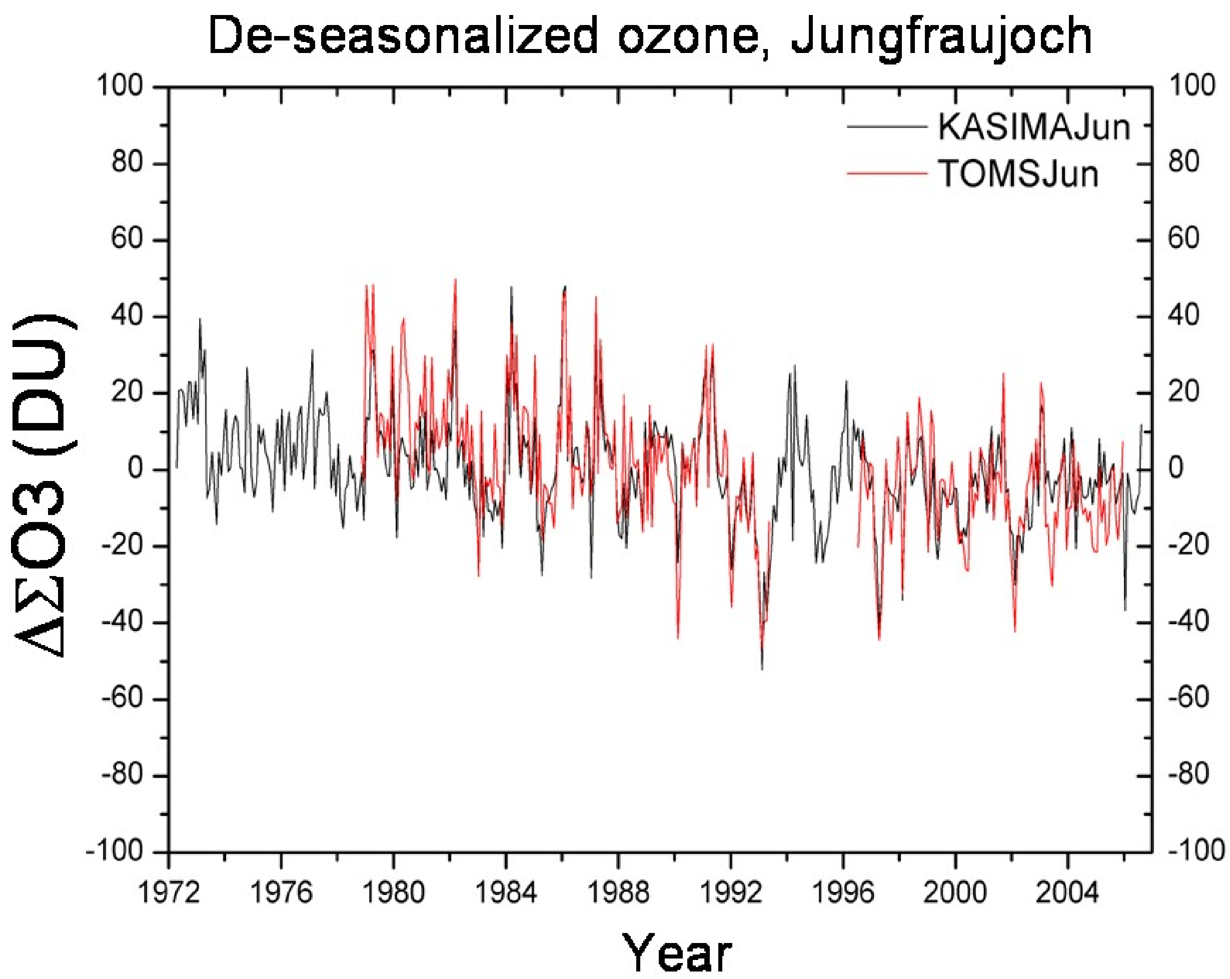
Performance of the model: long term simulations of ozone



FIMK

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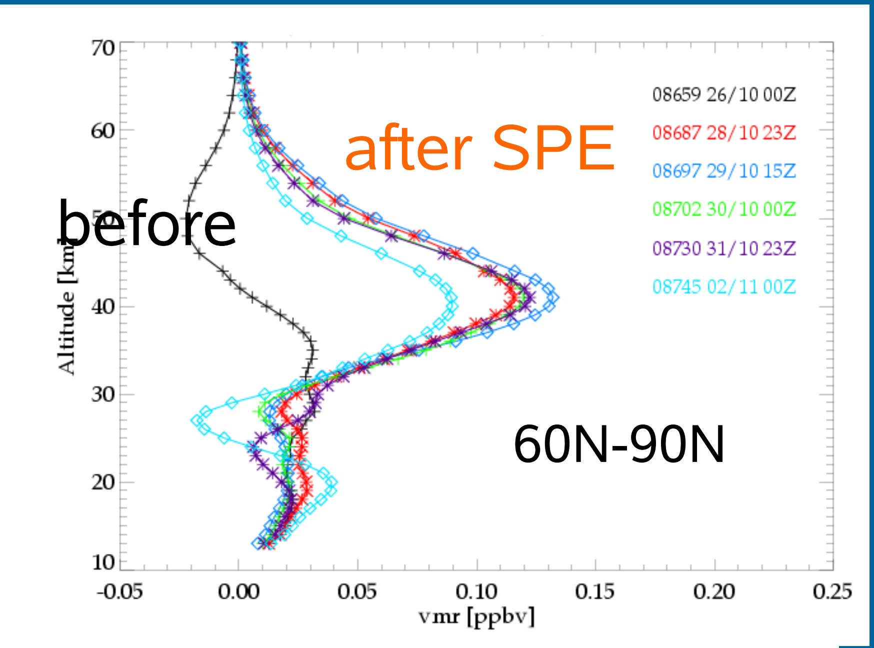
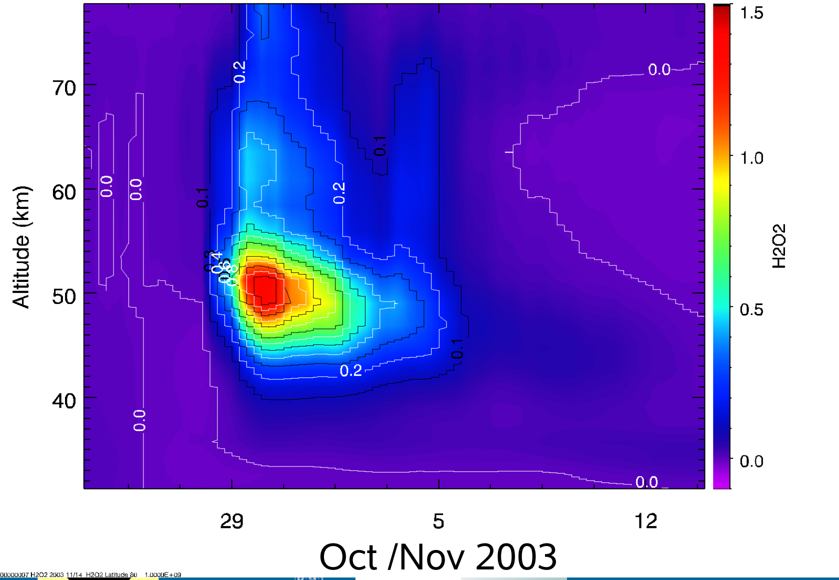
Solar Proton Event(s)

HOx related chemistry: observation of H_2O_2





- Retrieval of H_2O_2 for better estimation of HOx related chemistry effects (preliminary results)

  HOx cycle. Photochemical equilibrium with OH:

  $OH + OH + M \longrightarrow \mathbf{H_2O_2} + M$

  $H_2O_2 + hv$ (longwave) $\longrightarrow 2\ OH$

- Comparison with model: production of HOx via ionisation rates calculated on basis of GOES extrapolated flux, Bethe-Bloch and standard efficiencies for HOx/ionis.

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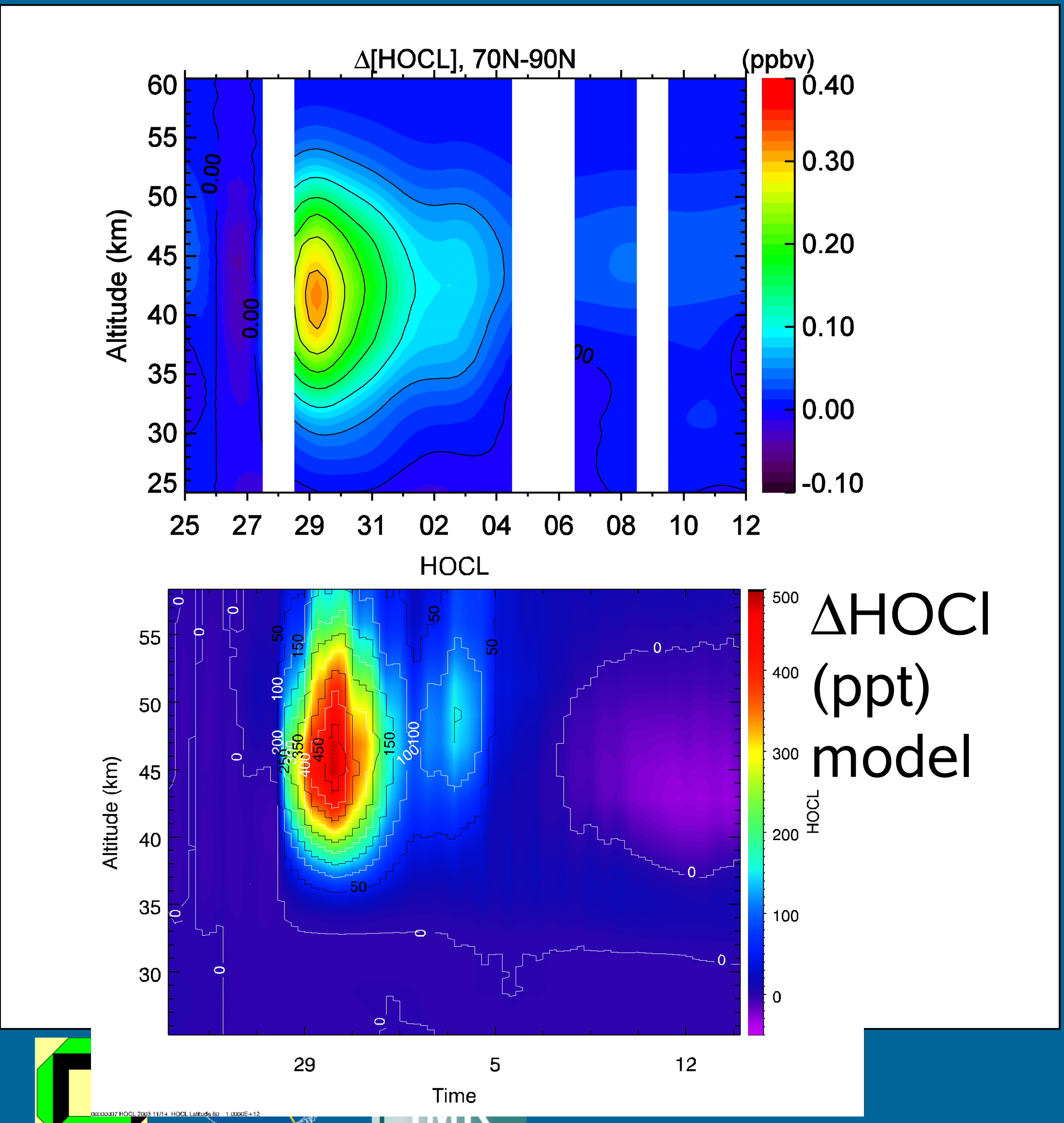
HOCl: comparison with MIPAS



$OH + HCl \rightarrow H_2O + Cl$

$Cl + O_3 \rightarrow ClO + O_2$

$ClO + HO_2 \rightarrow HOCl + O_2$

- possible estimation of HOx
- good agreement with HOCl
- problems with HNO3

NOx intrusions and long term effects

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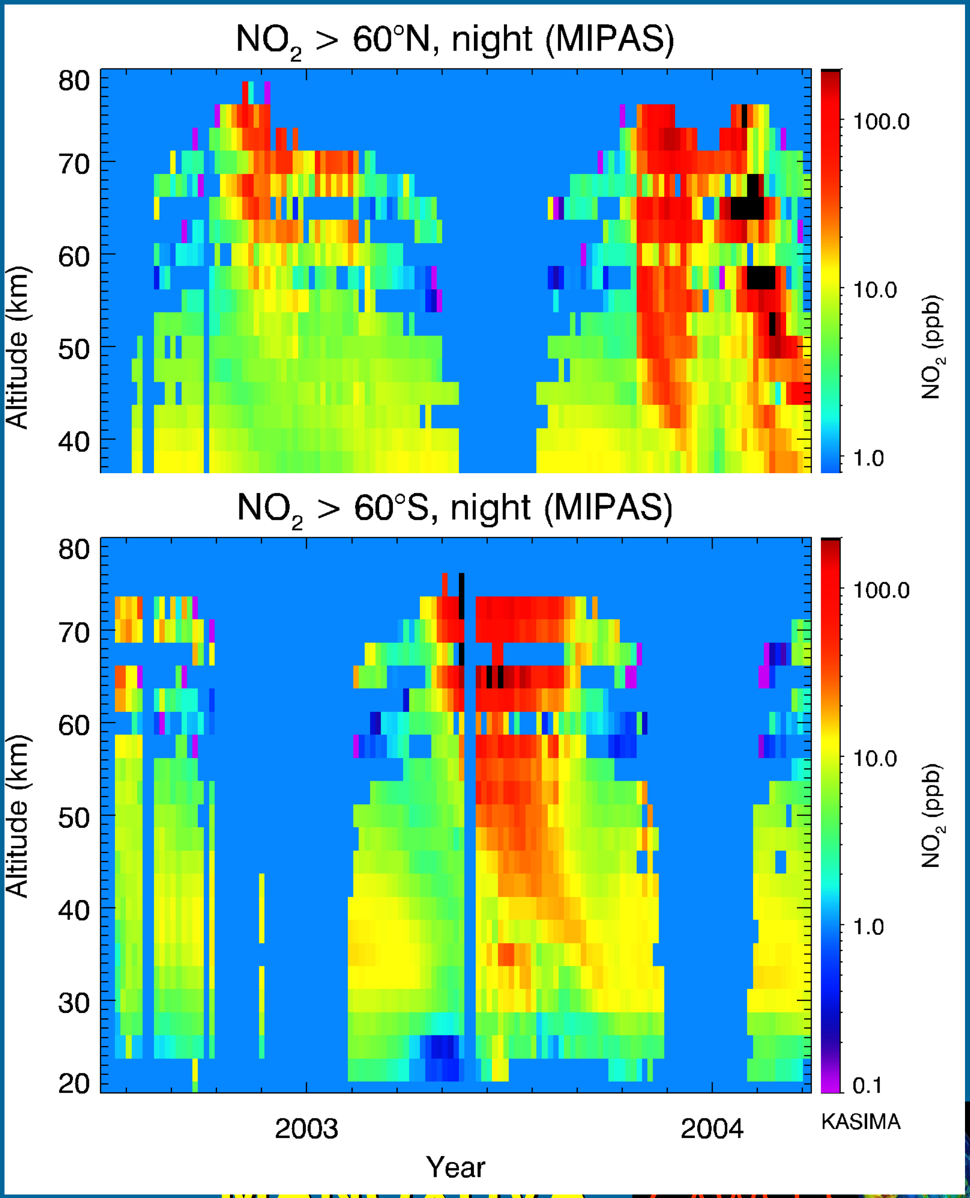
NOx intrusions and long term effects

- Two types of NO_2 enhancements are seen in data:
  - thermospheric origin with enhancements in the mesosphere only in the arctic night
  - global enhancements in the mesosphere and stratosphere as a aftermath of solar proton events (SPE)
- ESA data in the mesosphere only available during phases of enhanced NOx
- Use MIPAS NO2 night to prescribe NOx
- Only above ~55 km as a disturbance
- Compare with reference run
- Fixed heating rates (chemistry effects)



Using data in the model

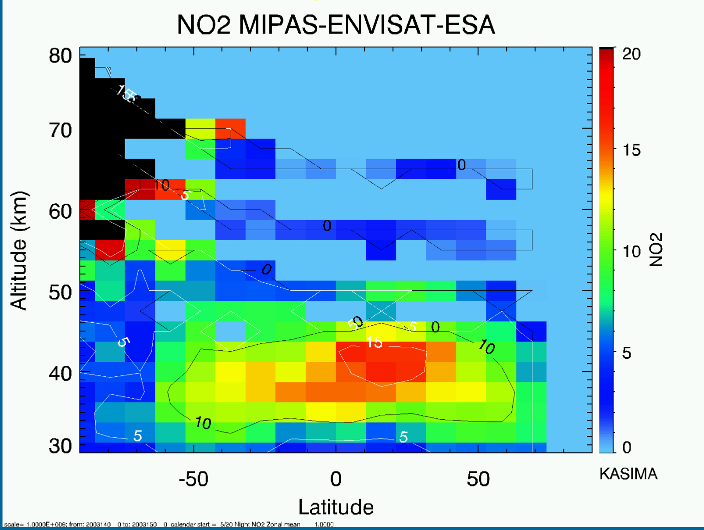
- We use NO_2 observation during night as a proxy for NO by sampling several days
- Because of the low frequency of MIPAS observations, as a compromise 5 days averages are used; not zonal means
- Data are interpolated to a grid
- Interpolation is done in a form of modified inverse distance weighting:

  $g_j = \sum w_{ij}\, y_i$ g: gridded data at gridpoint x_j ; y_i observation at x_i

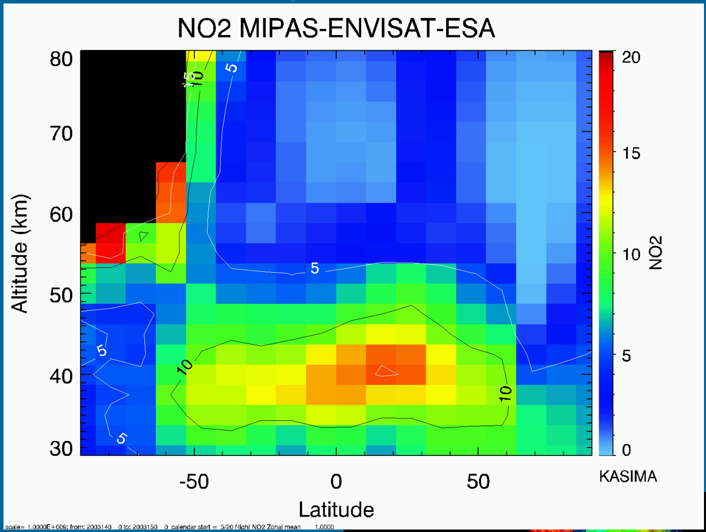
  $w_{ij} \sim 1 / 1 + (d_{ij} / l)^p$

  $d_{ij} = d(x_j,x_i)$: distance on sphere of gridpoint and observation, aspect ratio vertical to horizontal about 100, p some exponent (here p=8), l some characteristic length (about 200 km) parameters used result from tests and somewhat subjective

Sampled data



Interpolated data



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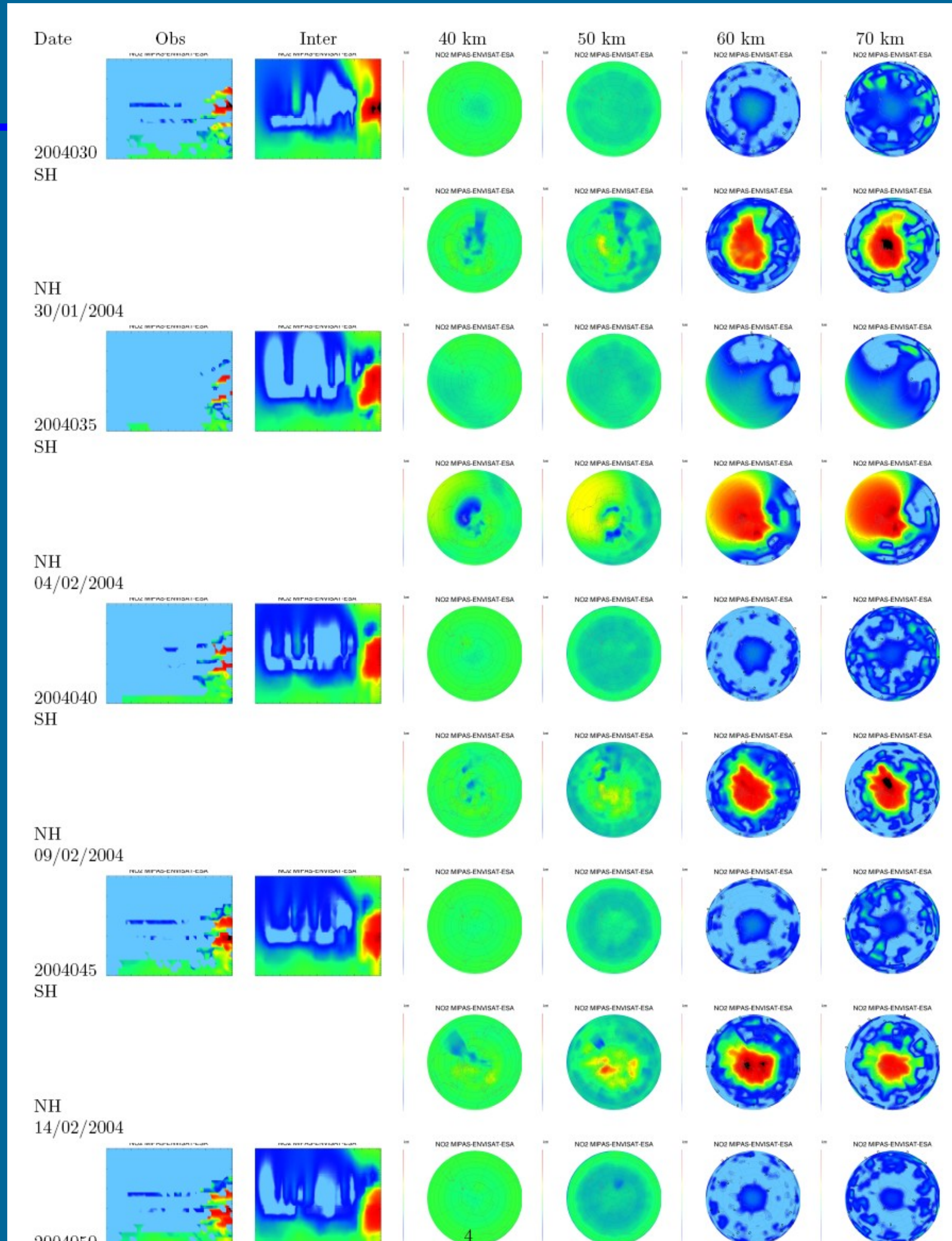
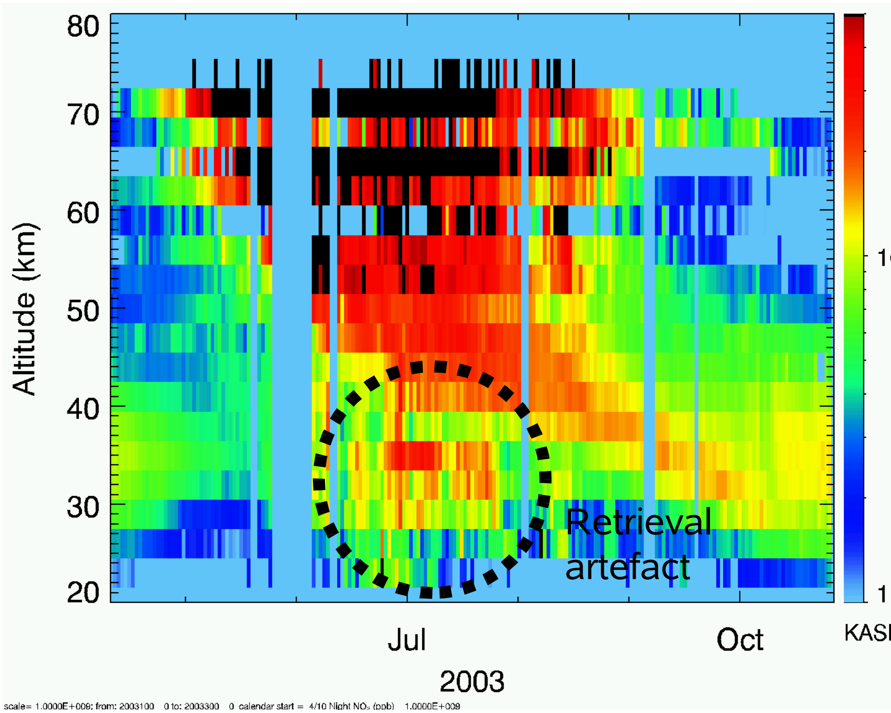
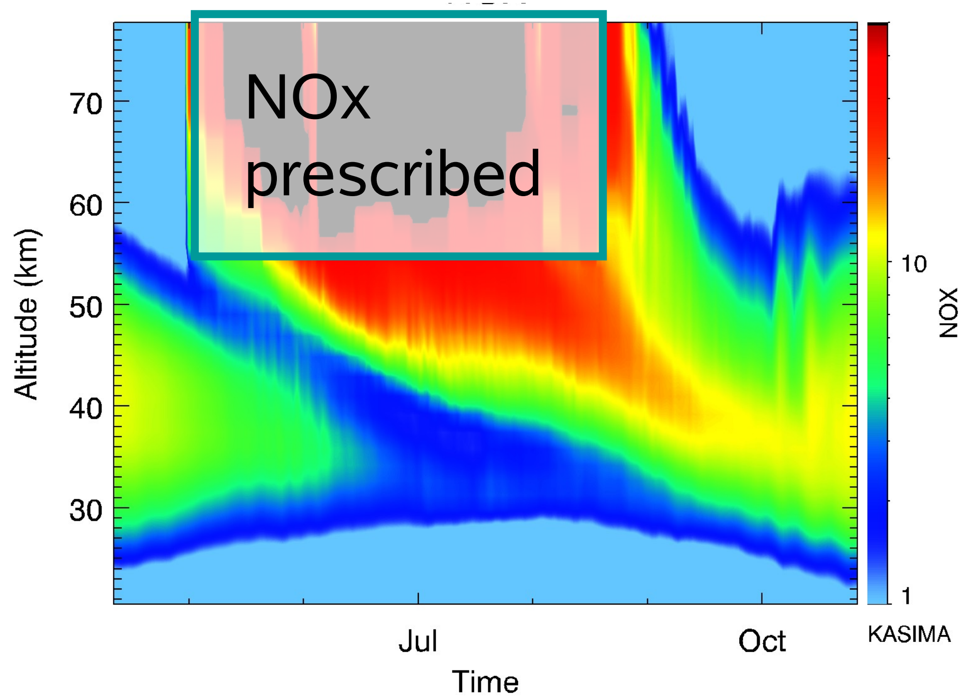

Date
Obs
Inter
40 km
50 km
60 km
70 km
NO2 MIPAS-ENVISAT-ESA
2004030
SH
NH
30/01/2004
2004035
SH
NH
04/02/2004
2004040
SH
NH
09/02/2004
2004045
SH
NH
14/02/2004
2004050
4

NOx in the model and in observations

Observations
night NO_2 MIPAS-ENVISAT > 60°S



Model NOx > 60°S



Antarctic winter 2003

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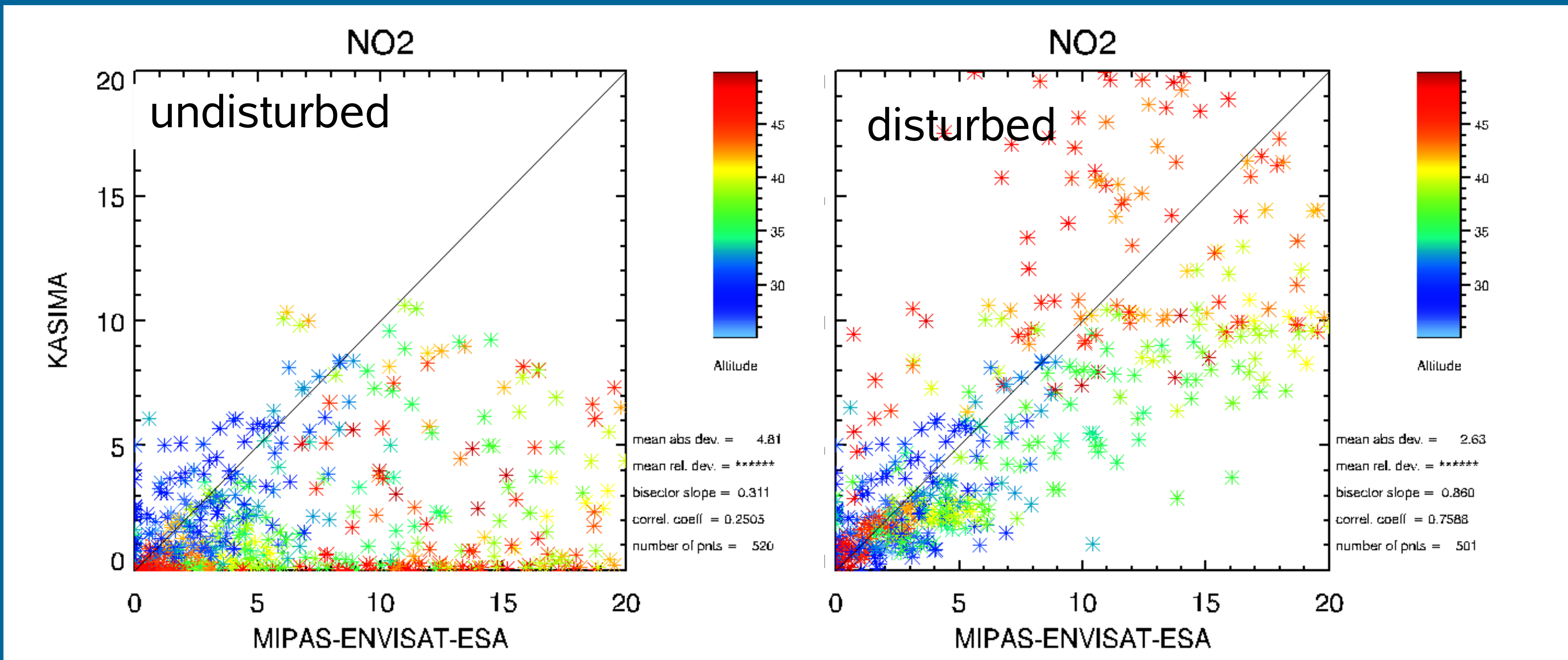
Correlations Observations vs. Model

Effect of NOx intrusion on correlation for stratosphere 50 days after initialization 2003 DOY 180. Only coincident data are used (|Δt| < 30 min)



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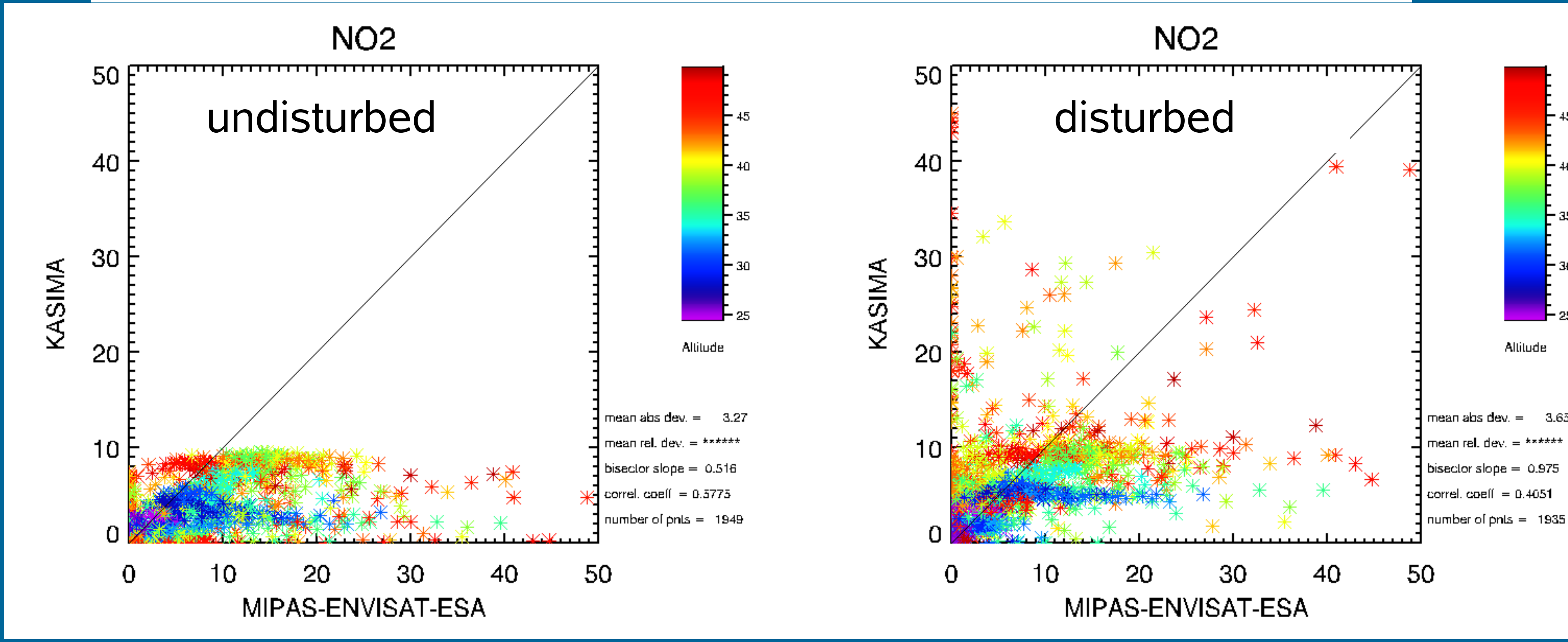
Correlations Observations vs. Model

Effect of NOx intrusion on correlation for stratosphere winter 2003/2004 DOY 30 -80. Only coincident data are used (|Δt| < 30 min)



Problems with dynamically very active winter 2003/2004

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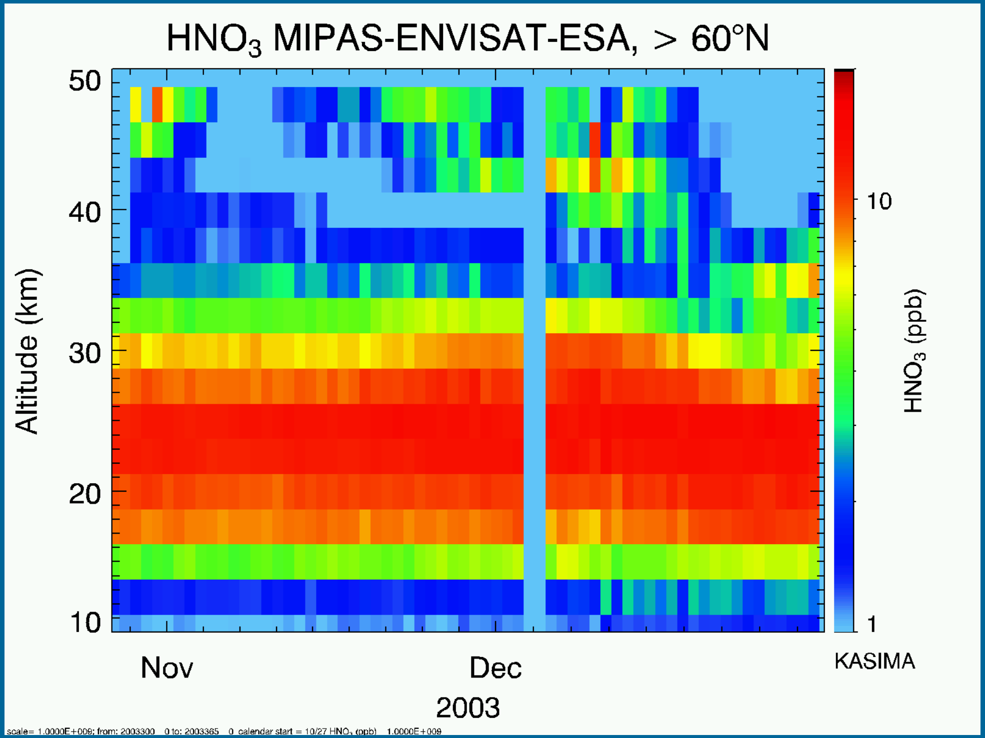
HNO_3 buildup through ion cluster chemistry

- MIPAS data show strong HNO_3 enhancement in Antarctic and Arctic winters (see analysis of Orsolini et al. 2004 for winter 2003/2004)
- Has been also observed by CLAES
- Even in zonal mean present till January
- Missing in model
- Ion mechanism (Böhringer etal)?



- Simplified approach of de Zafra et al.
- Ion reaction $N_2O_5 + H^+.(H_2O)_n \rightarrow H^+(H_2O)_{n-1} + 2HNO_3$ (only for z > 40km)

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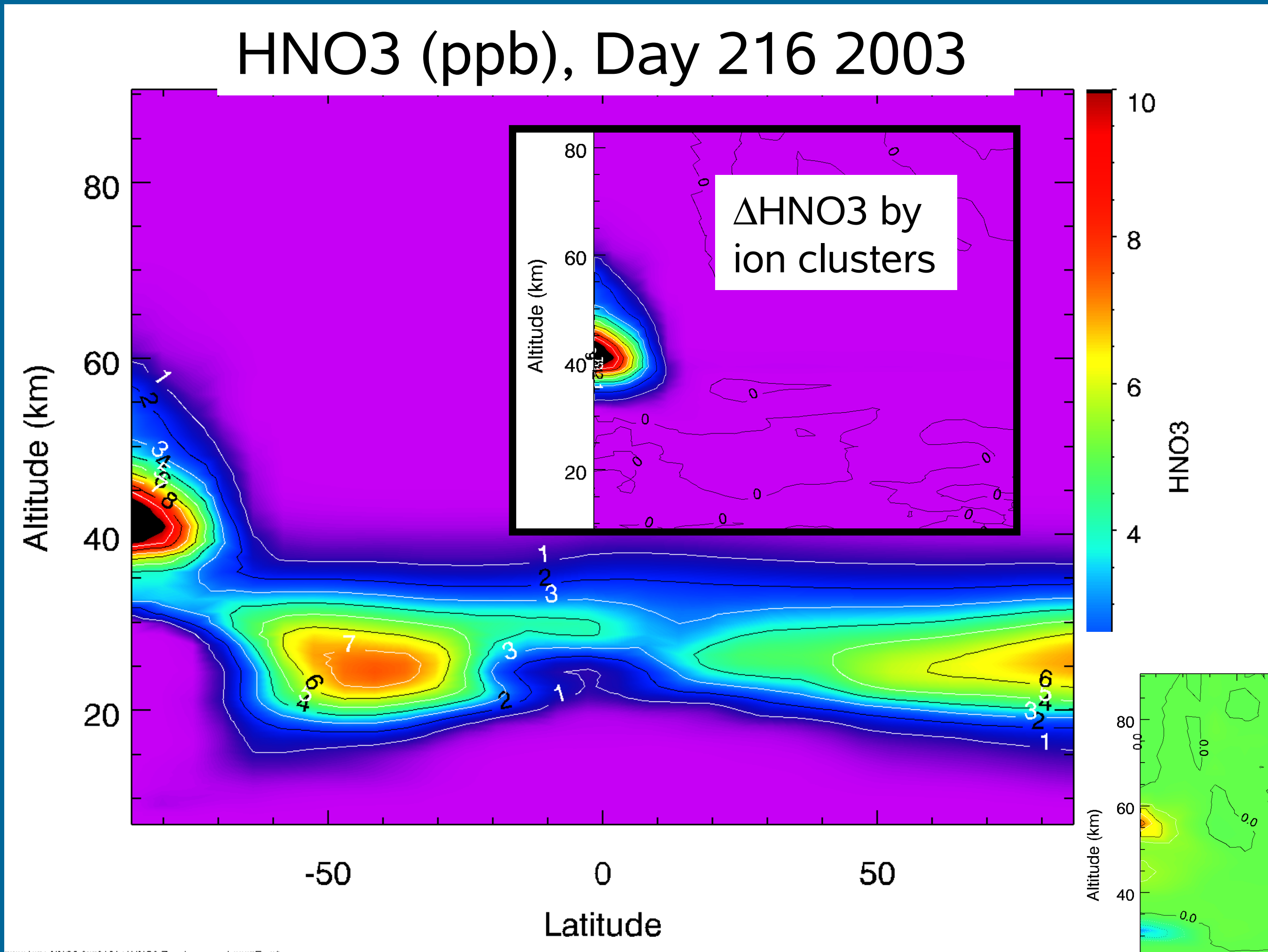
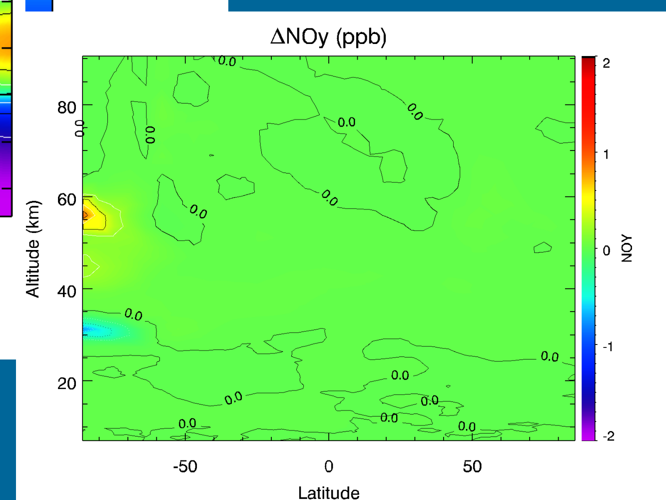
Effects of water ion clusters on NOy



- Ion clusters can provide HNO3 secondary maximum
- Small effects on total NOy and ozone



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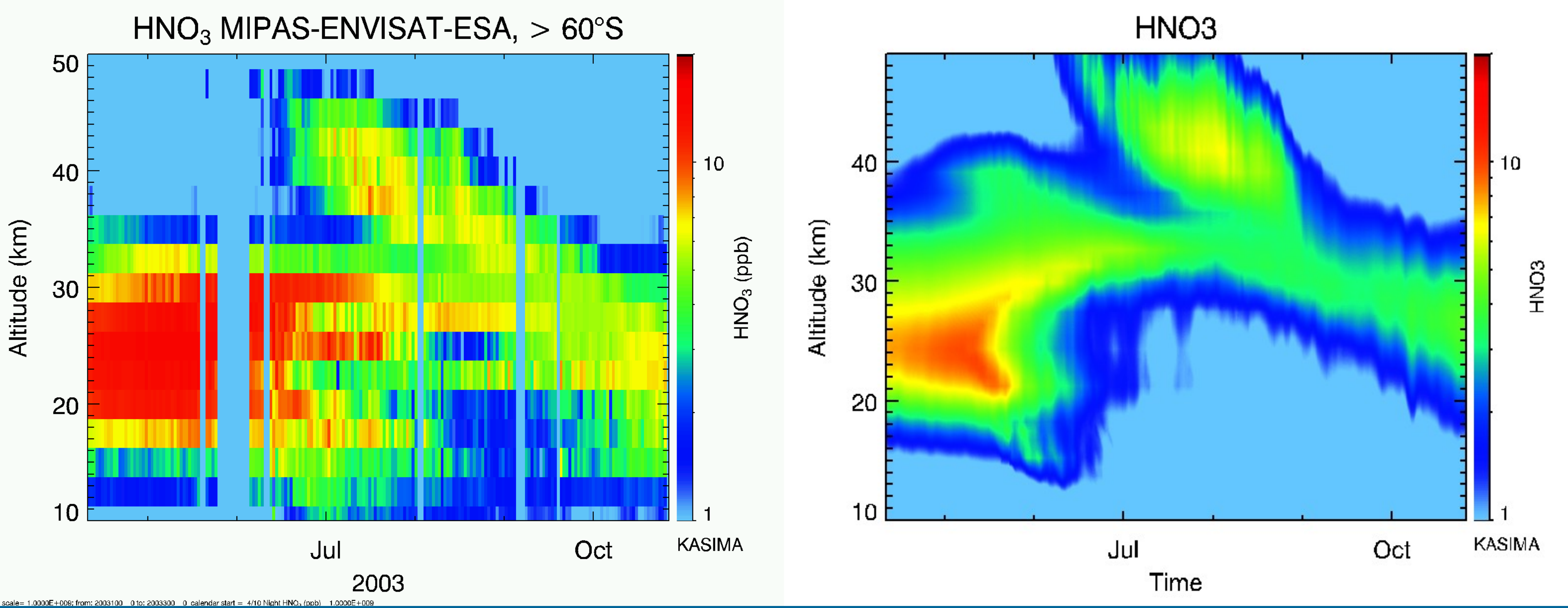
HNO3 in the model and in observations



Antarctic winter 2003 (see also analysis of Funke et al. and Stiller et al.)

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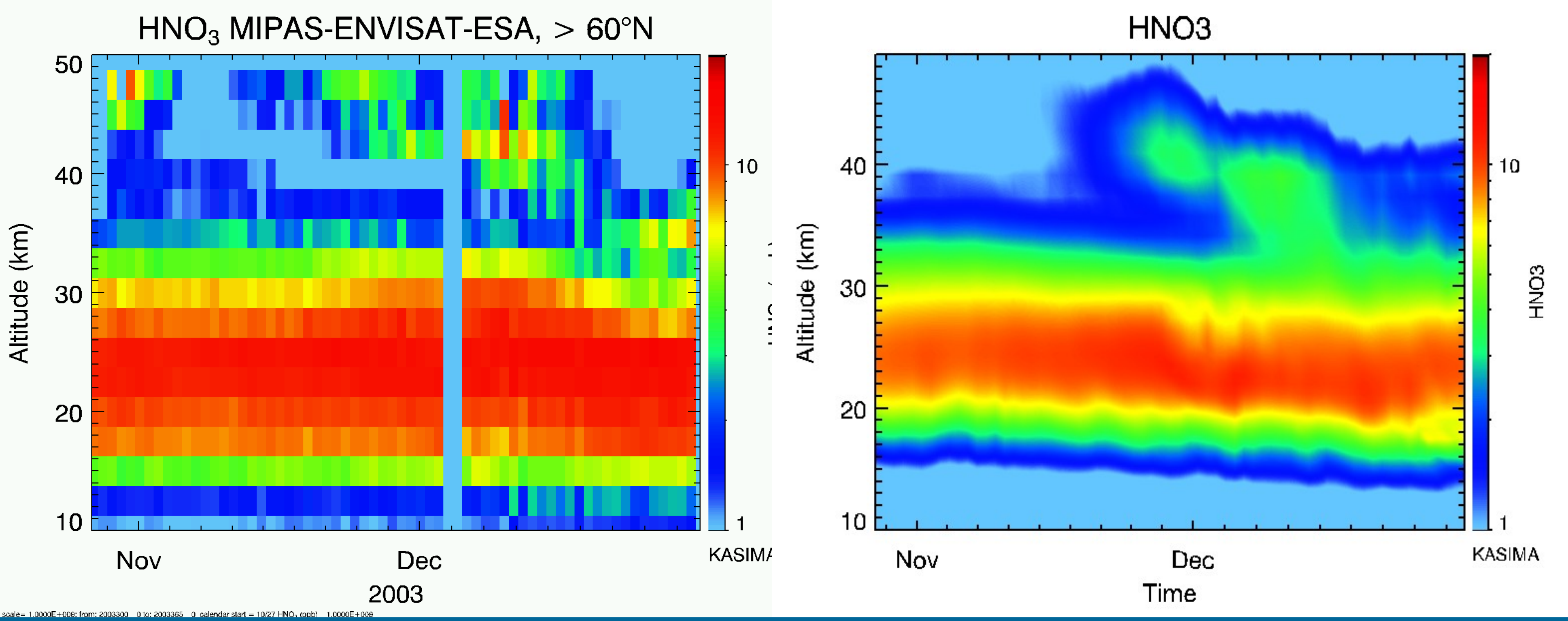
HNO3 in the model and in observations



Early Arctic winter 2003/2004

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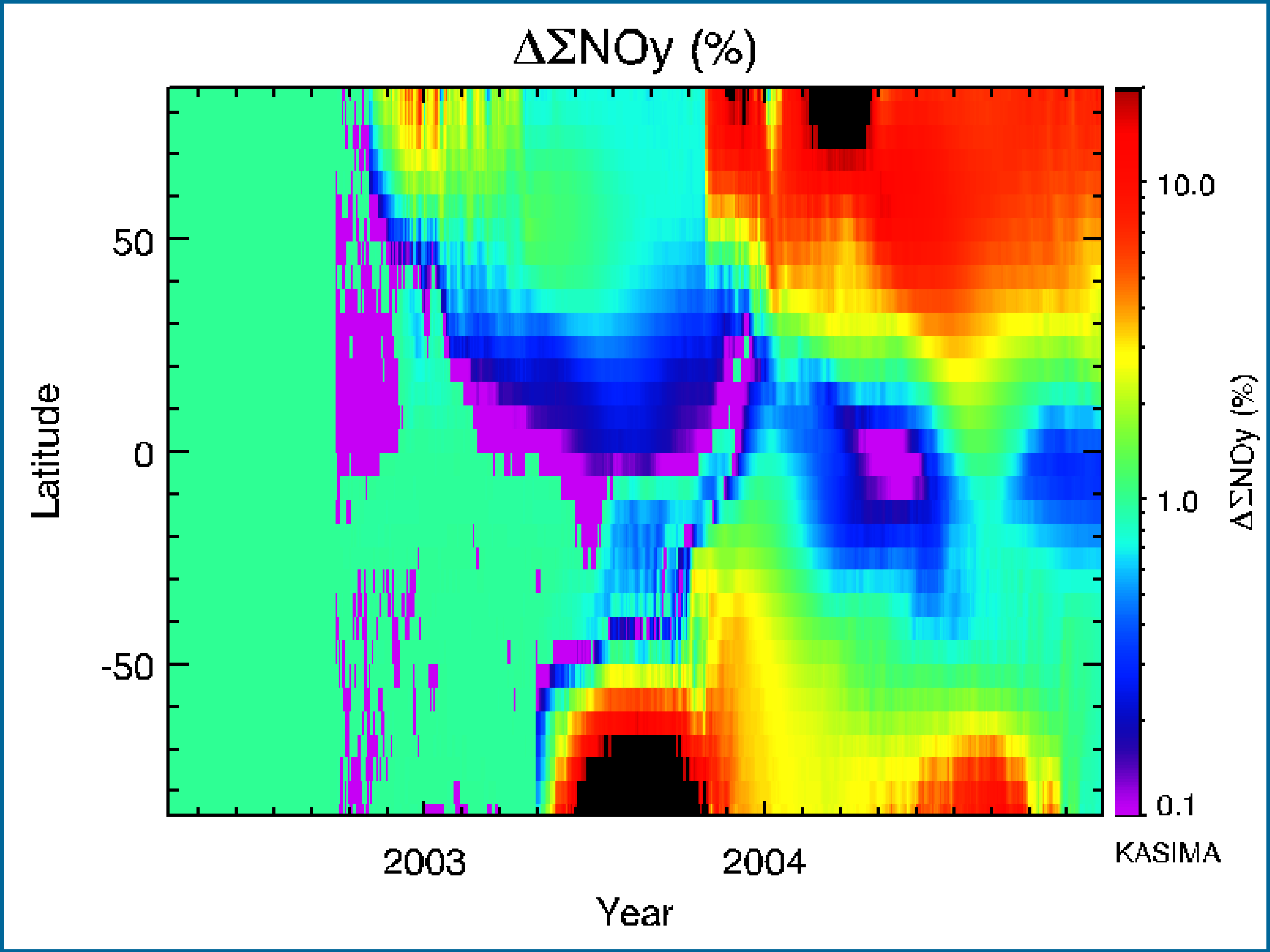
Effect on total NOy



- NOy enhancements lasting more than one winter
- Mostly confined to polars latitudes
- Strong in Arctic winter 2003/04

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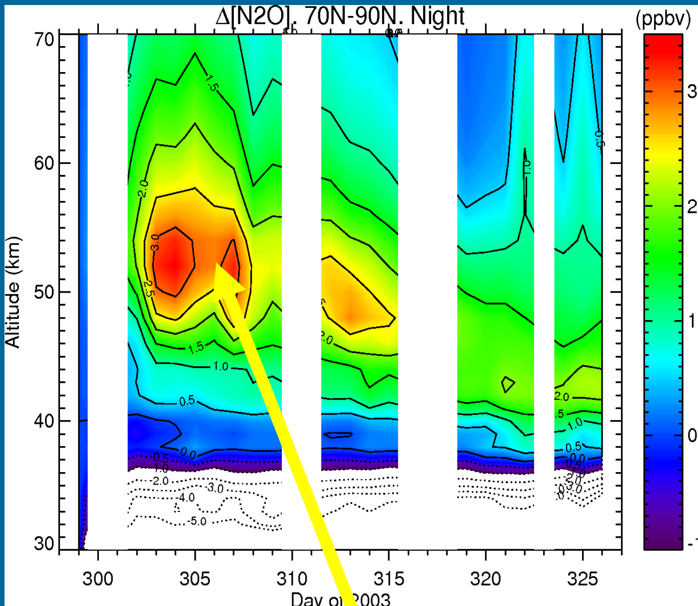
NOx as a source for N2O

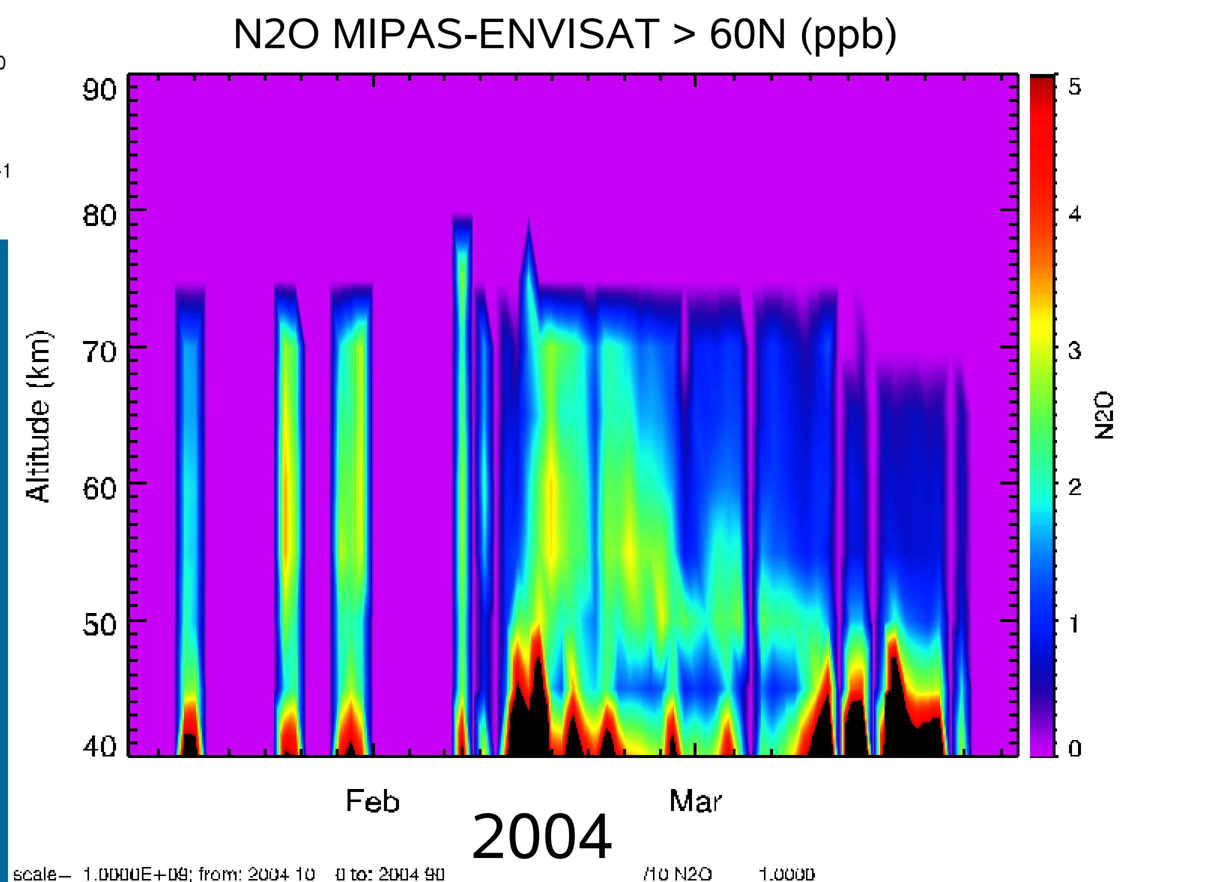


- Quasi In-situ produced N during aurora events?
- Effect for tracer-tracer correlation studies?

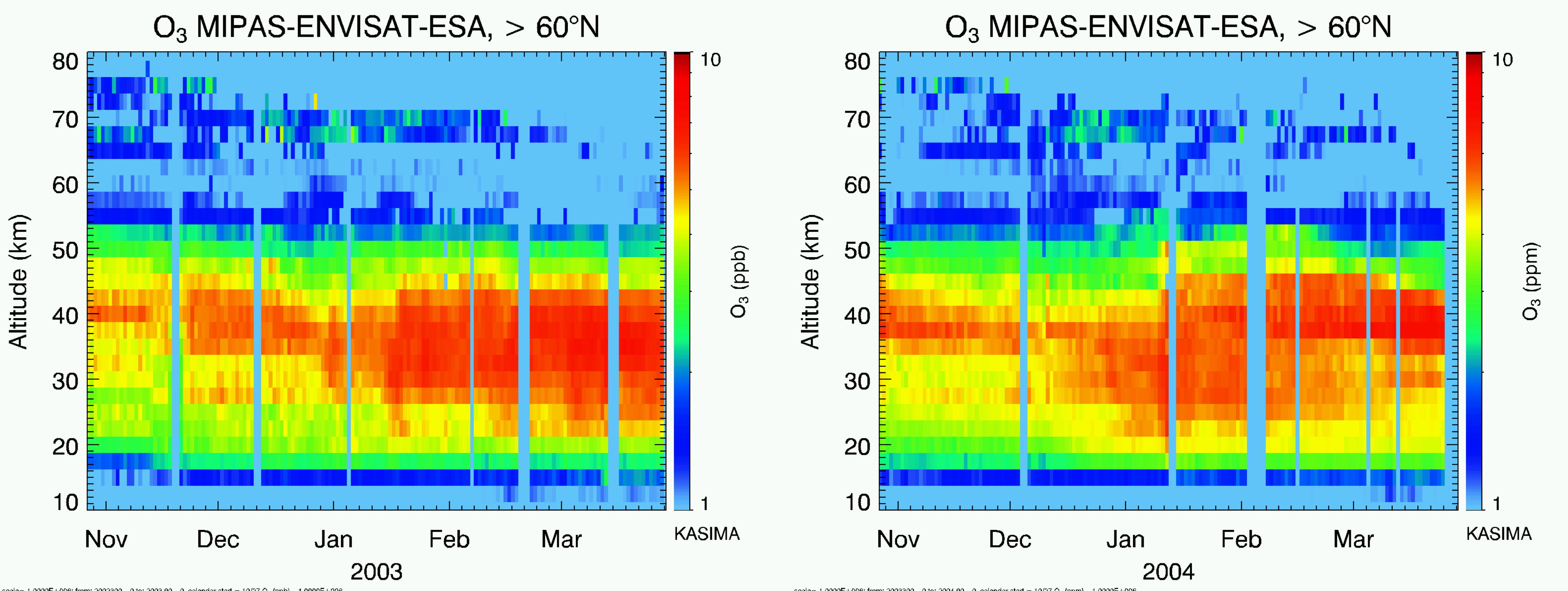
N2O from

N + NO2 -> N2O + O

In-situ production of N during SPE



Ozone



Effect on ozone is not very obvious on the long term

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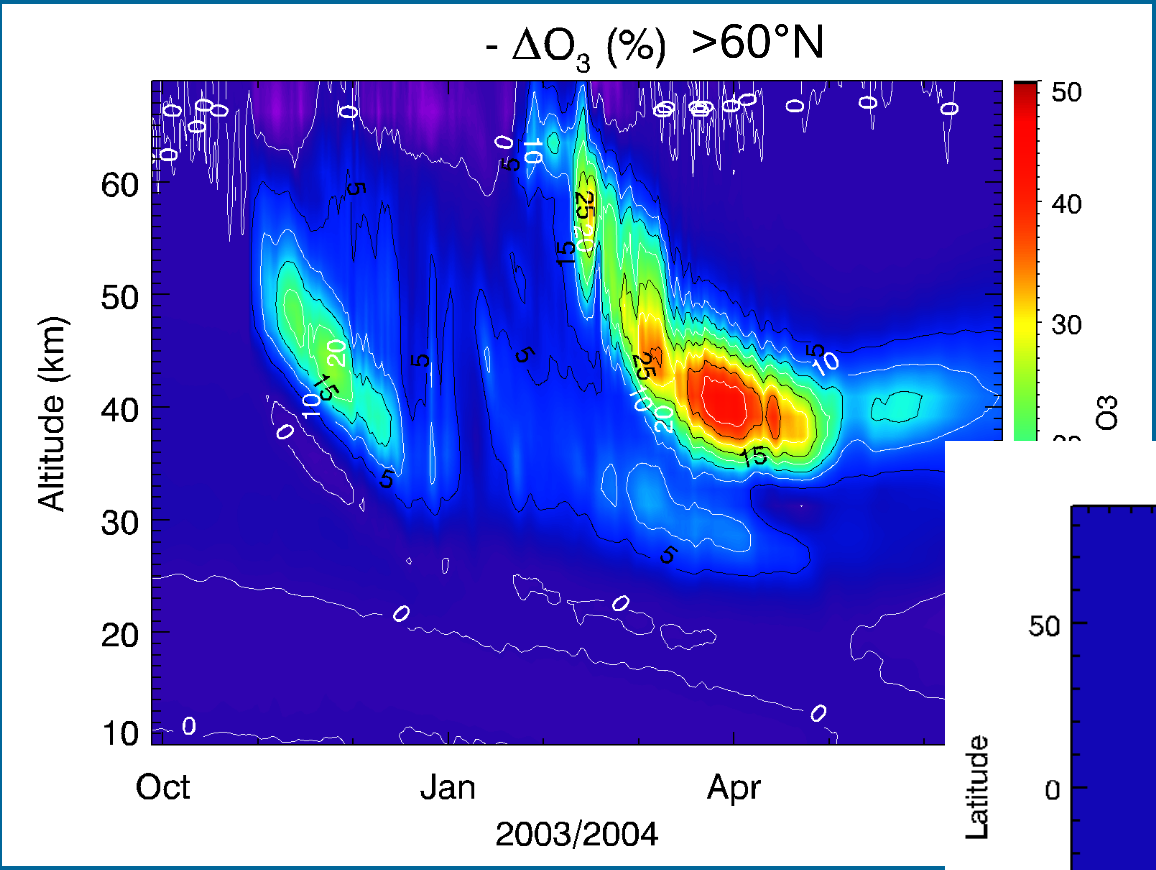
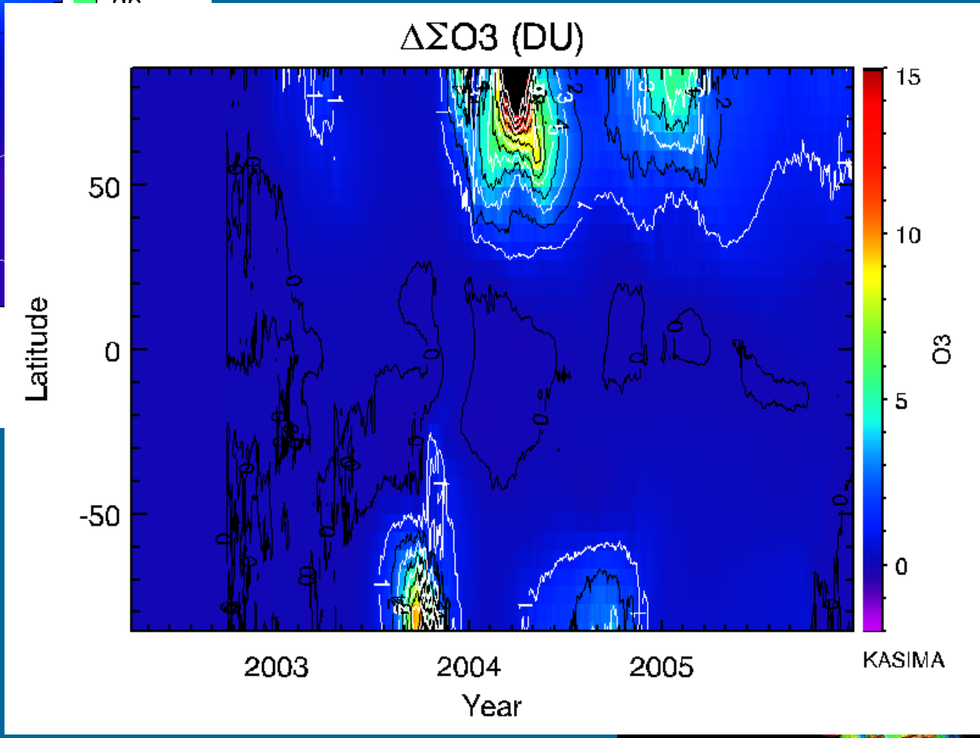
Additional ozone loss



- Use reference run to estimate additional ozone loss as difference between the two runs



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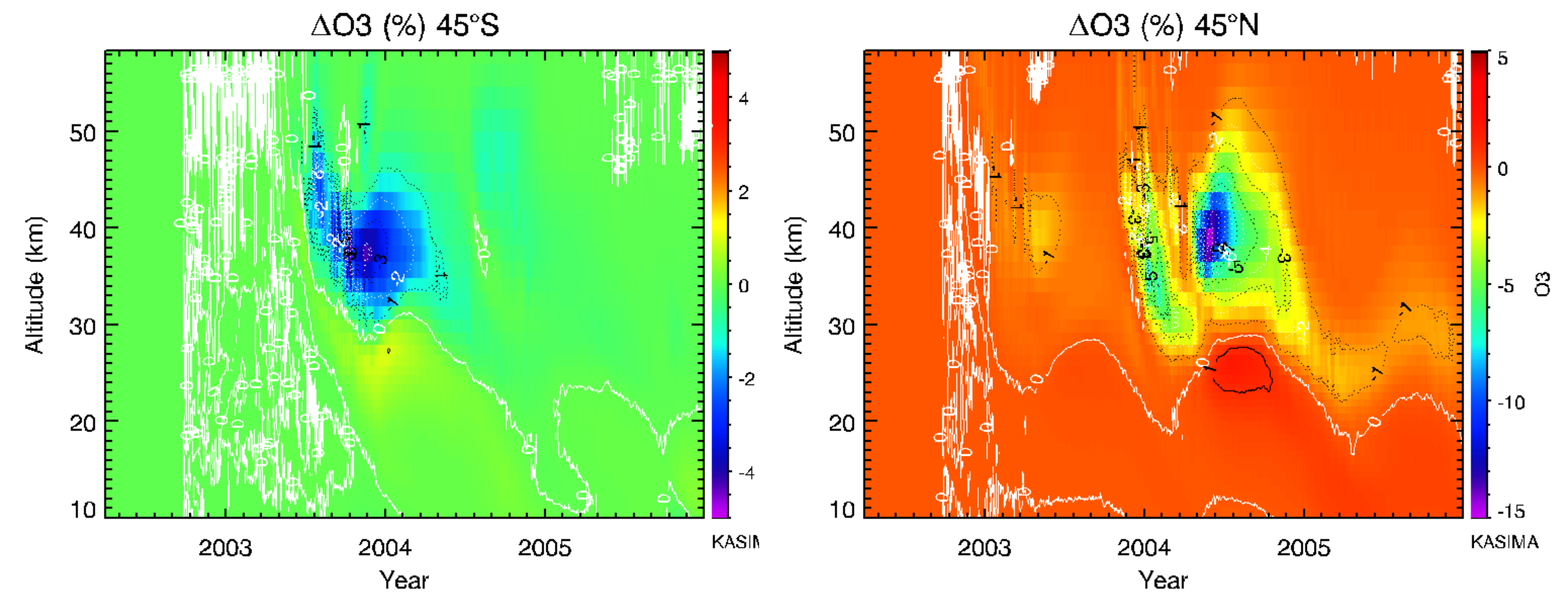
Midlat effects in ozone



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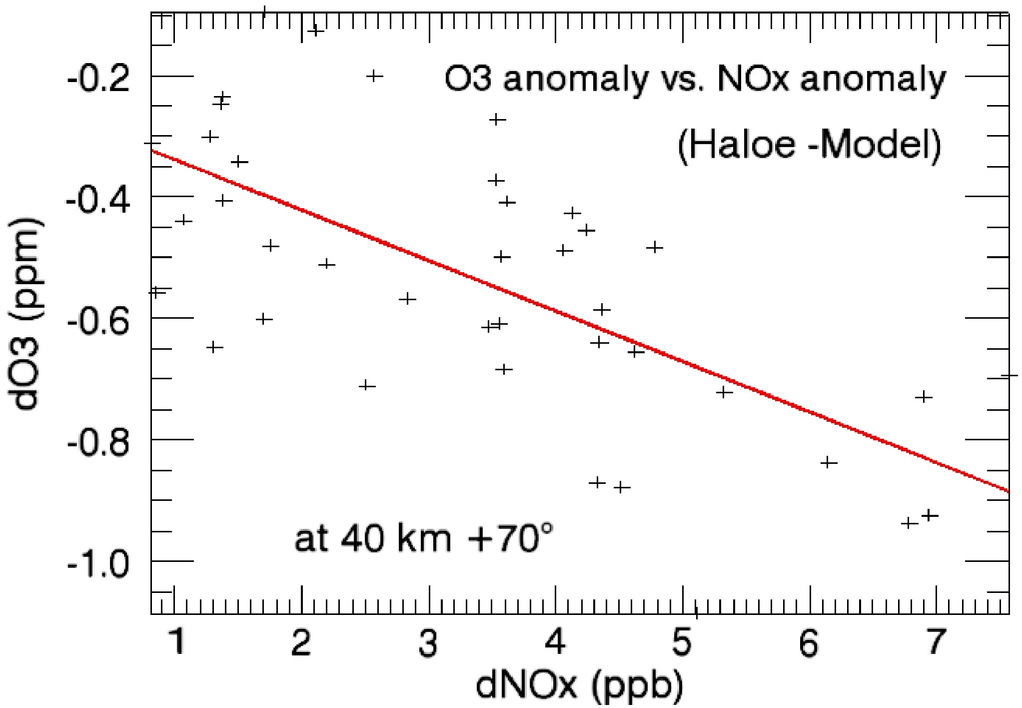
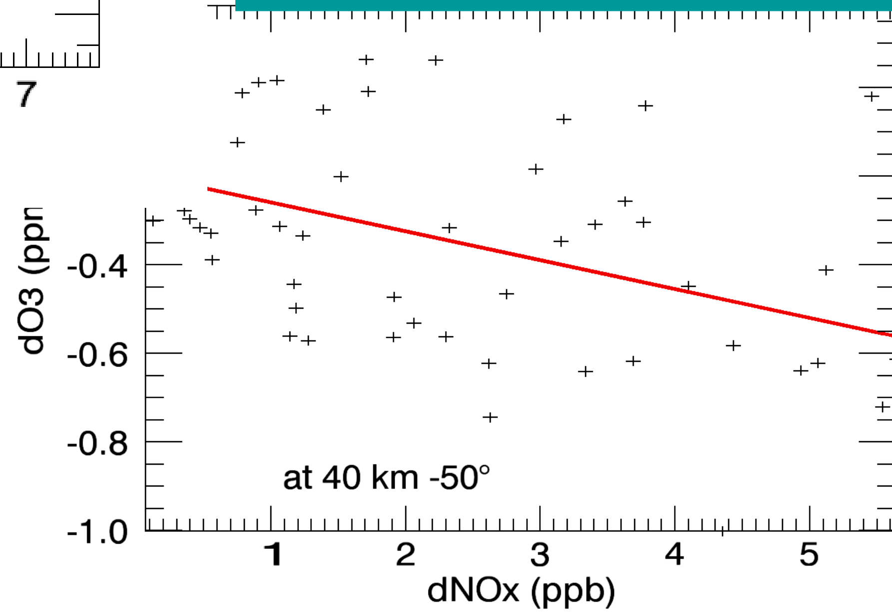
Multidecadal simulations and search for solar signal

Long-term variation of NOx and effects on O3

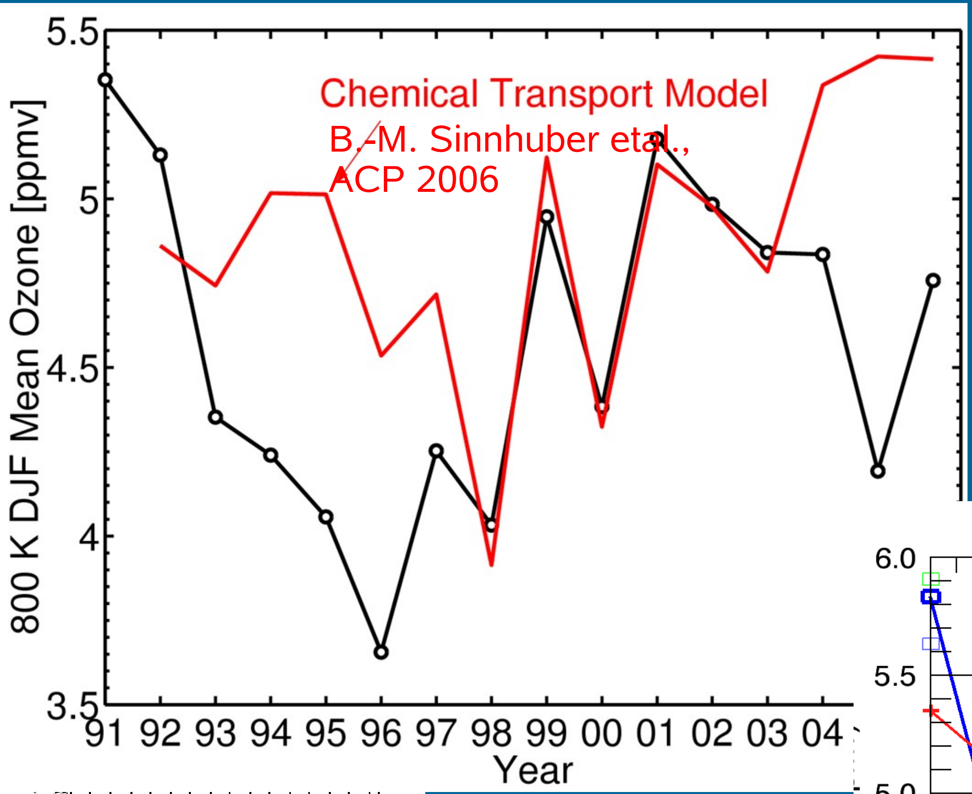


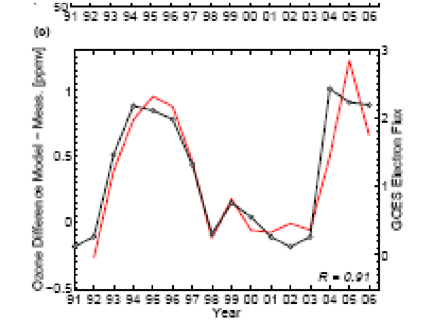
- Use multidecadal run to search for solar related effects
- As no solar signal is included in the run, look for anomalies when comparing with data
- Use HALOE O3 and NOx observations in altitude and latitude bands

NOx related O3 signal!

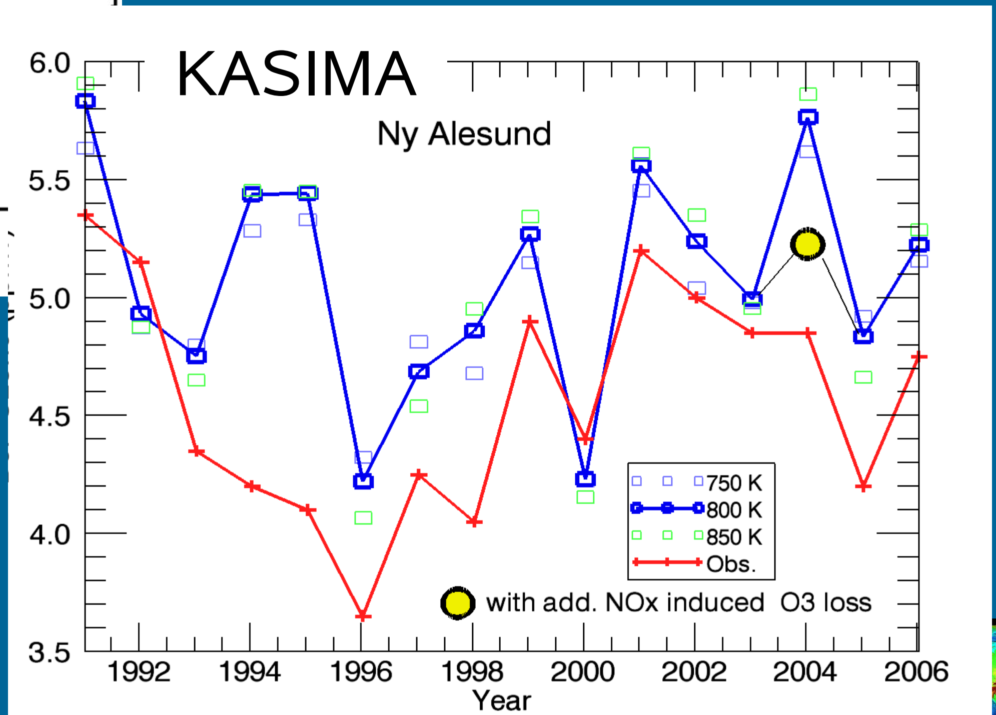


Another mystery



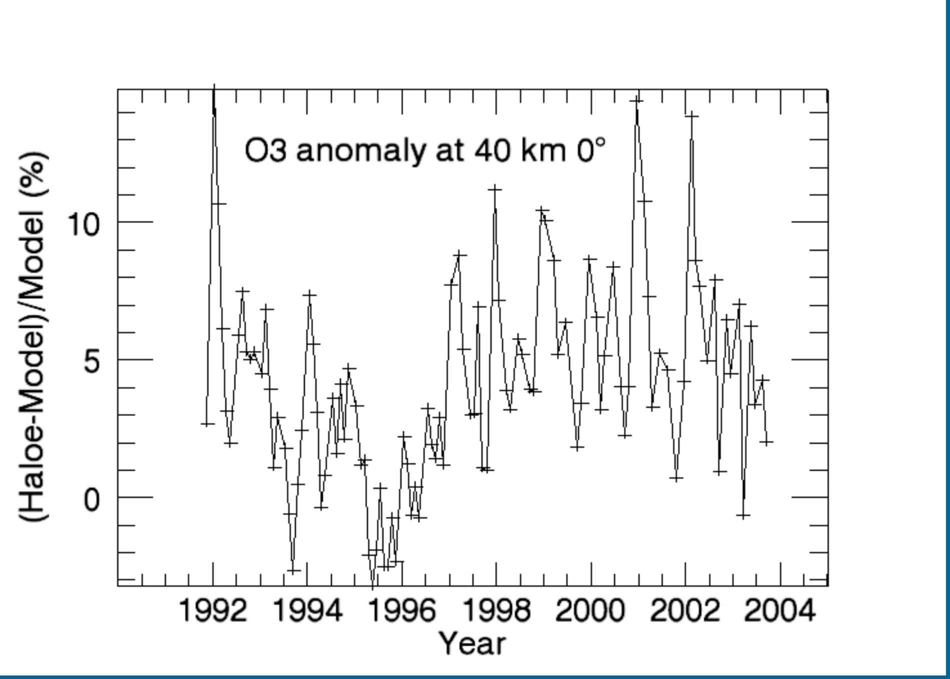


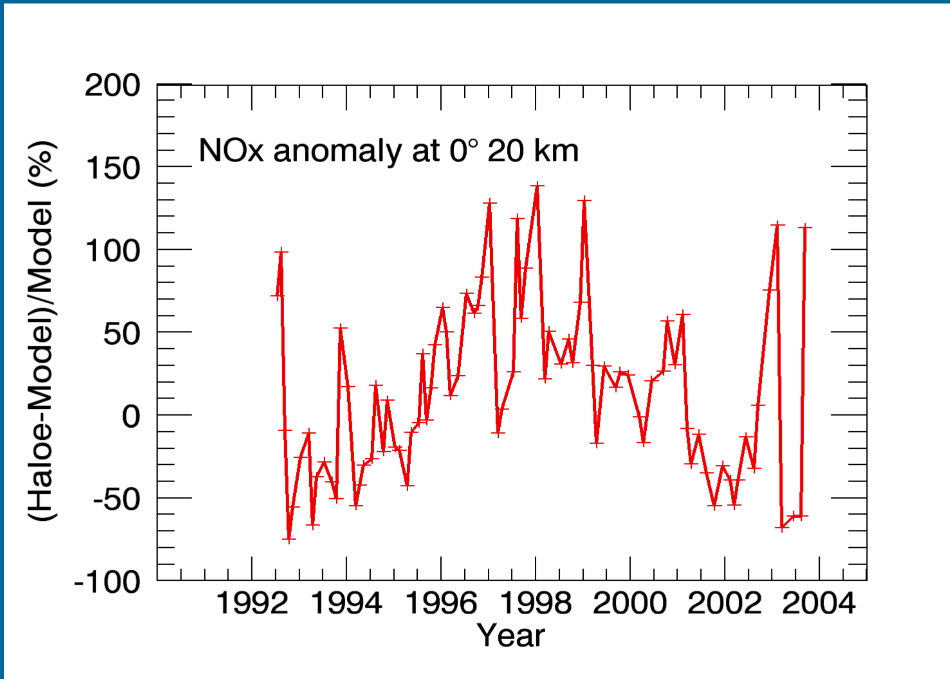
- Sinnhuber etal found anomaly in Spitzbergen O3 data
- Anomaly may be partly explained by NOx intrusion
- Correlation with GOES lost



Signatures of UV variability?

HALOE - MODEL





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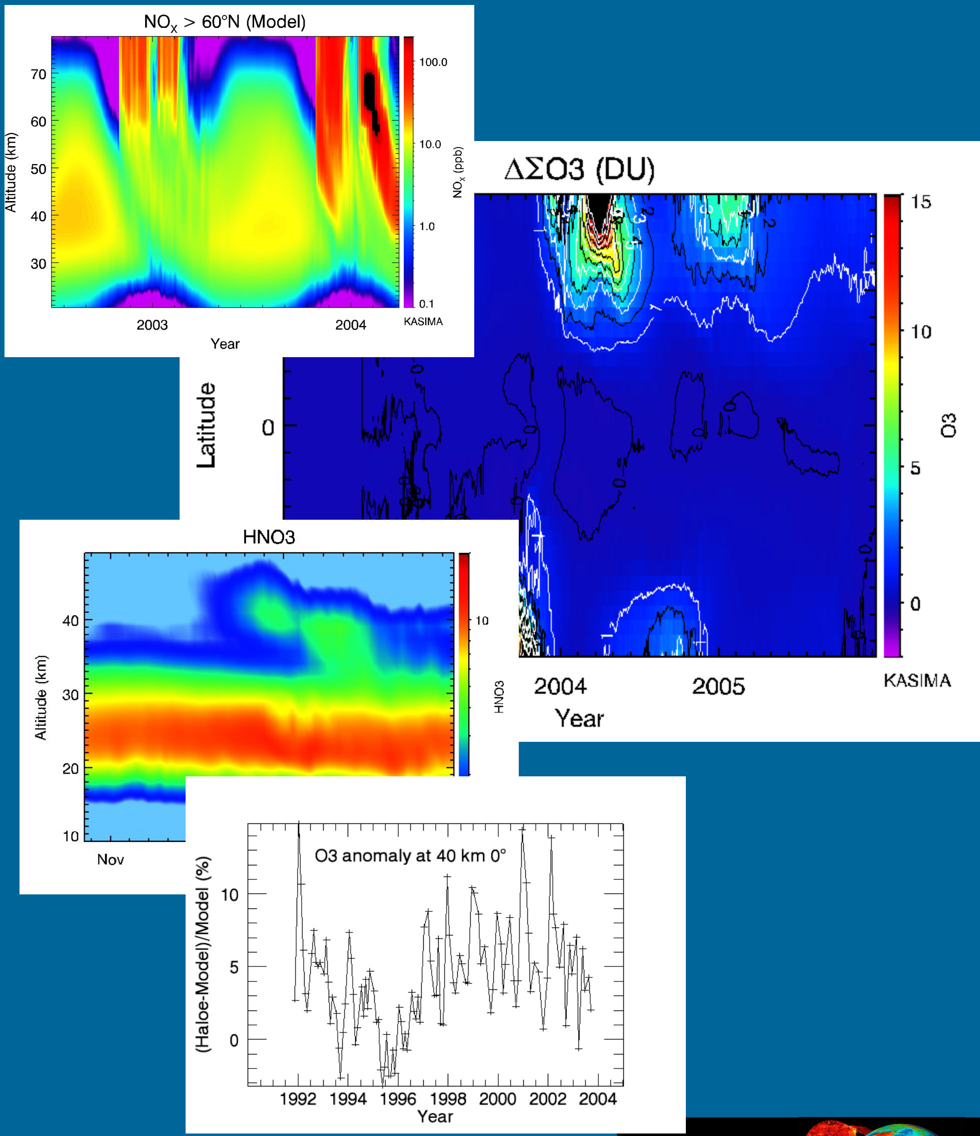
Summary:

- MIPAS provides an extensive dataset for chemistry studies after SPEs
- MIPAS data can be used to prescribe NOx in KASIMA's polar meso/stratosphere and to estimate influence of NOx intrusions on chemistry
- Important effects on ozone restricted to polar latitudes. Trend analysis for high latitudes may be affected!
- Observed HNO_3 enhancements can be qualitatively reproduced when introducing HNO_3 buildup via H_2O cluster ions
- Open questions:
  - Interplay of in-situ production and dynamics in MLT for aurora effects
  - Downward transport during winter
  - Understand buildup of HNO3 quantitatively
  - The buildup of N2O
  - Relative contribution of EPP and UV for ΔO3 on longer timescales



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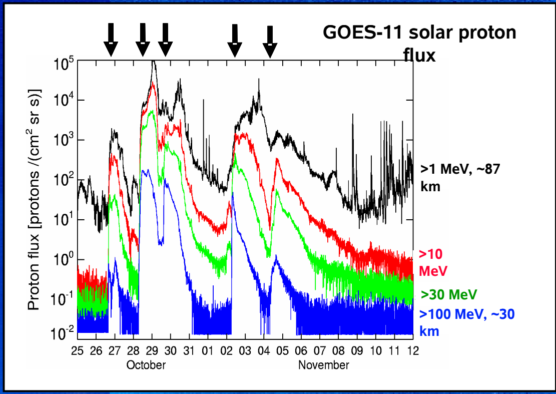
Thank you !

Particle precipitation, ionization and radical production

- Energy loss of energetic particles in the atmosphere by ionization, N2+, and subsequent production of NO+
- Recombination of N2+ or NO+ or charge exchange reaction yield N(2D, 4S)
- N(2D) + O2 -> NO + O

  N(4S) + NO -> N2 + O fast
  N(4S) + O2 -> NO + O slow
- Net effect is the production of NOx in the lower thermosphere/mesosphere
- Production of HOx in the meso-/stratosphere via water cluster ions



See Brasseur Solomon 2005

Result: NOx and HOx enhancements in the middle atmosphere disturbing regular chemistry

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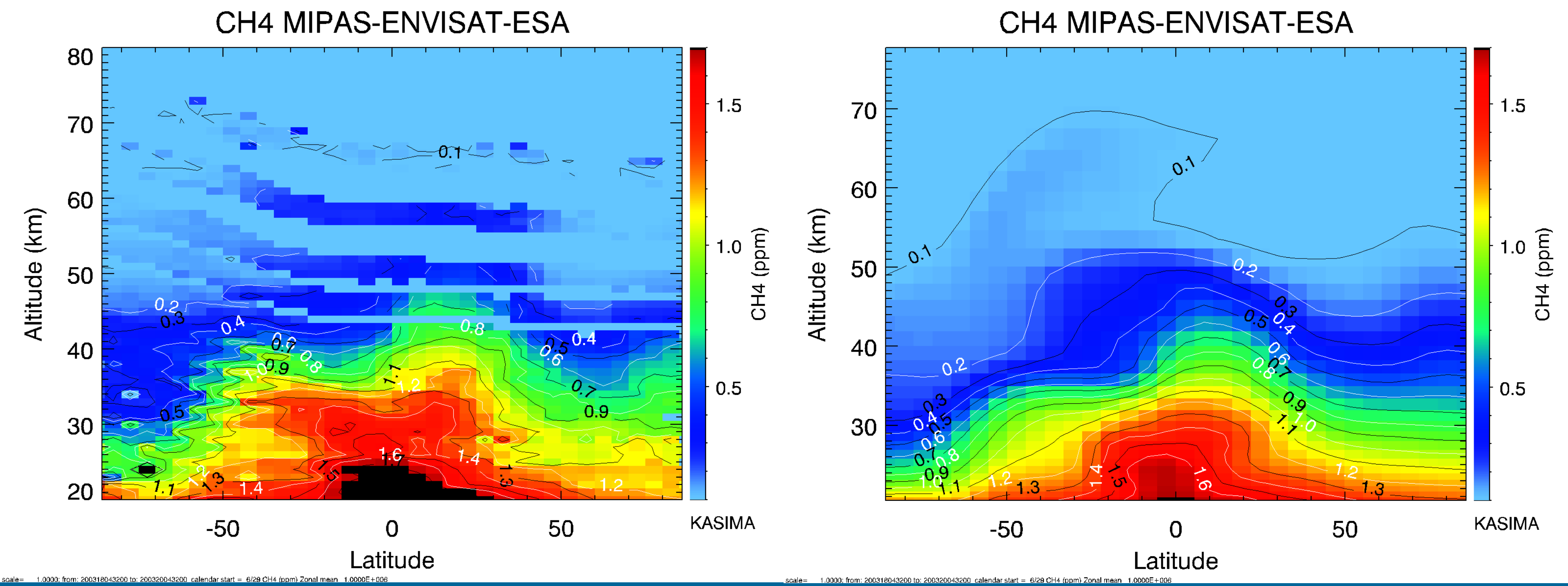
General transport properties of the model



Zonal mean CH4 (ppm) for period 2003 DOY 180-200

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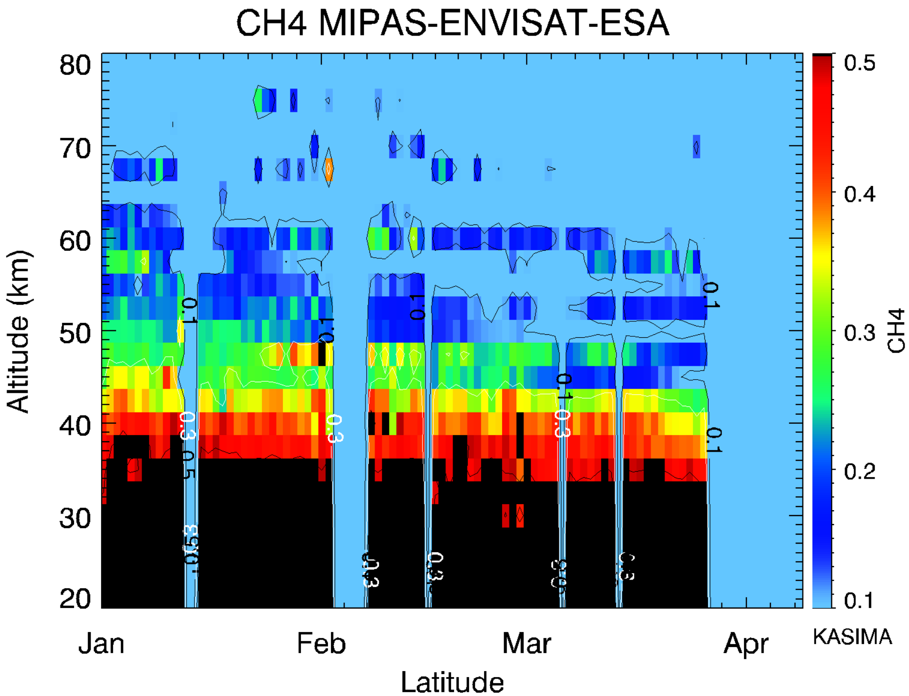
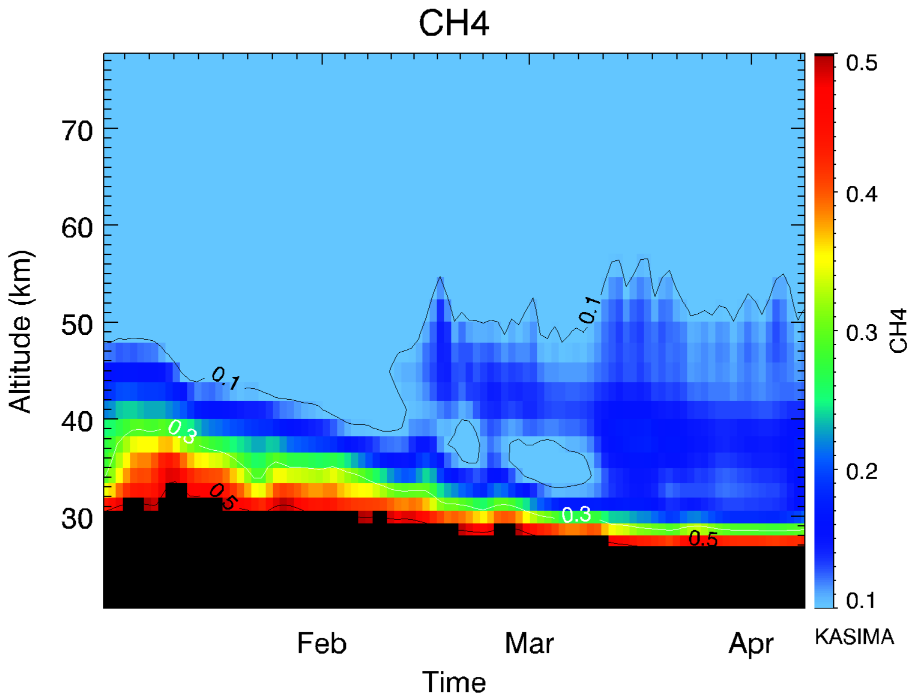
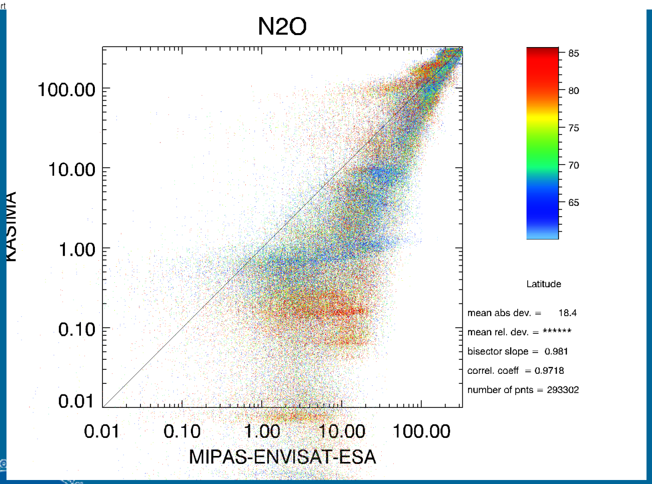
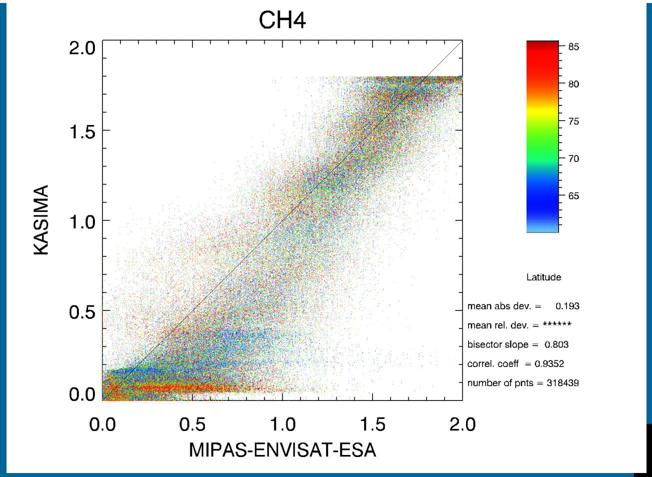
Descent in Arctic vortex winter 2003/2004(> 60N)





scale= 1.0000; from: 2004 1 0 to: 2004100 0 calendar start





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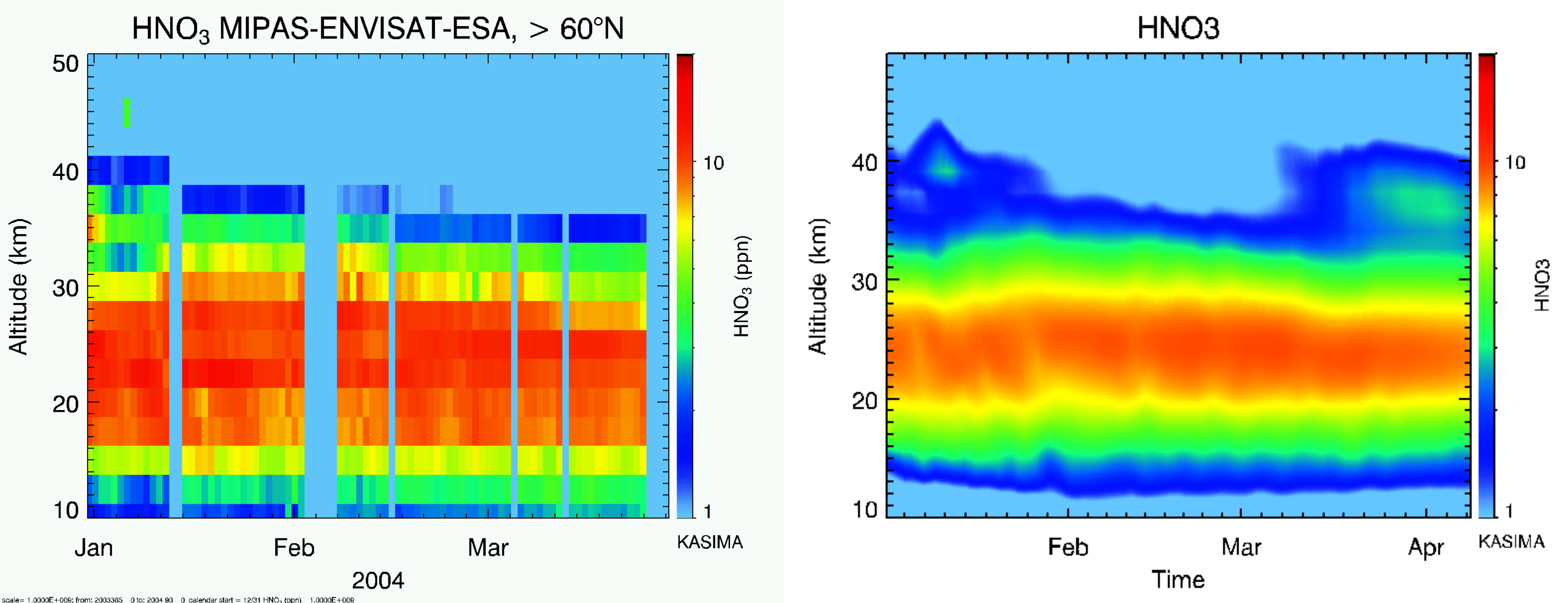
HNO3 in the model and in observations



Late Arctic winter 2003/2004

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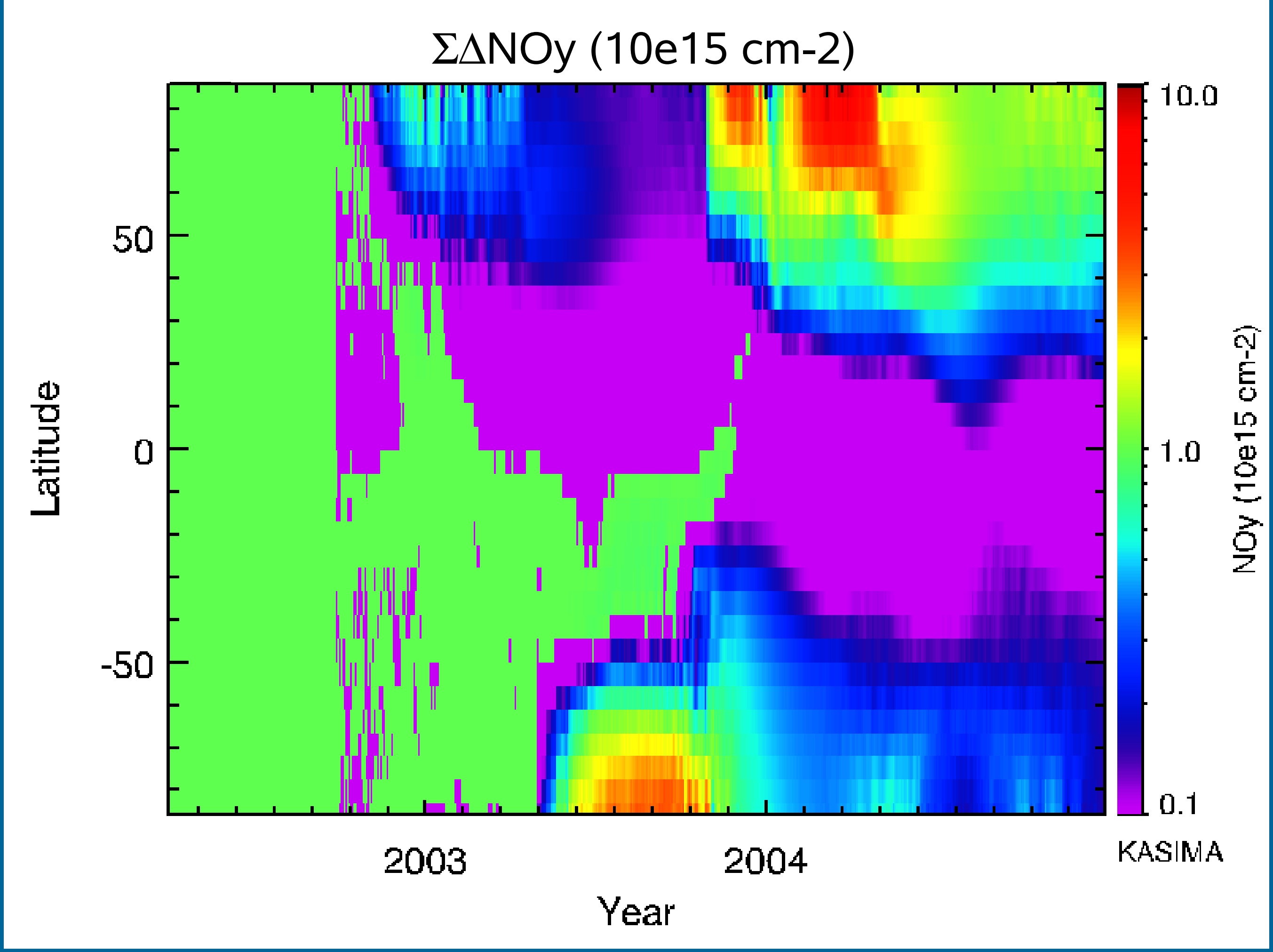
Effect of NOx intrusions on total NOy



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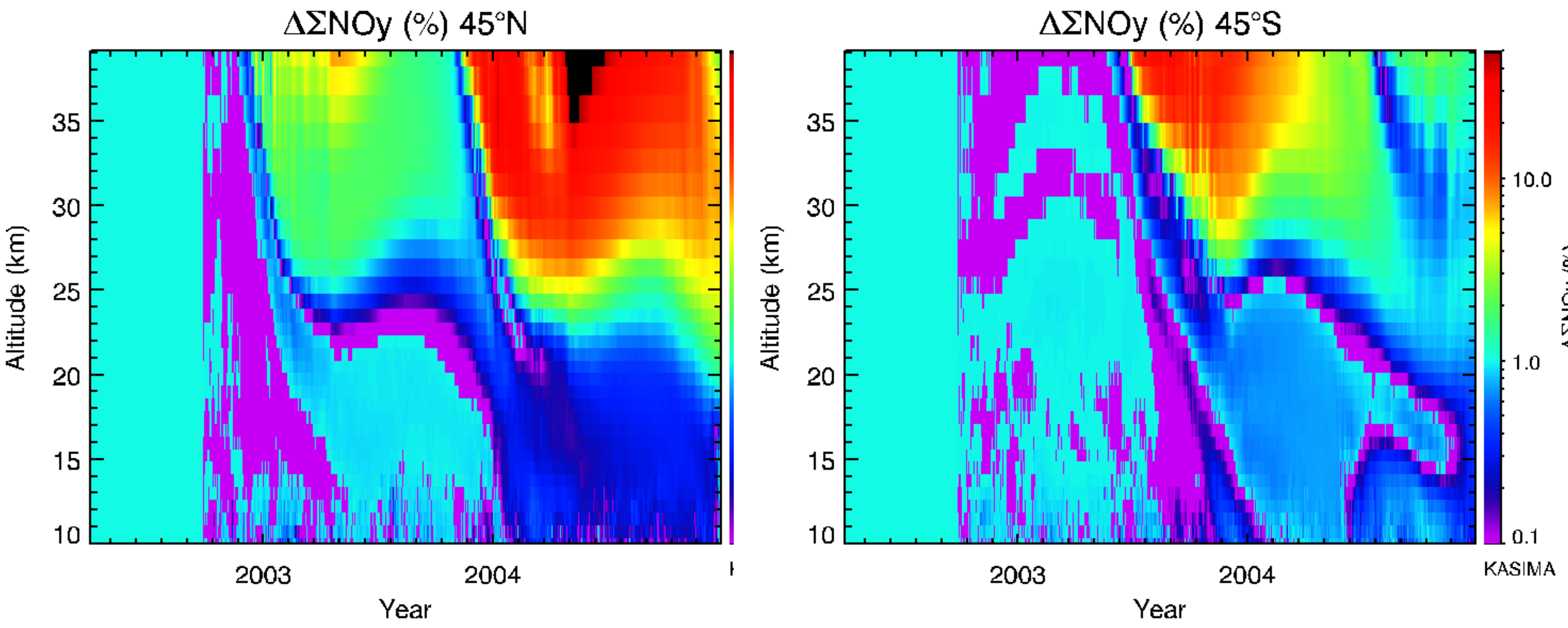
NOy



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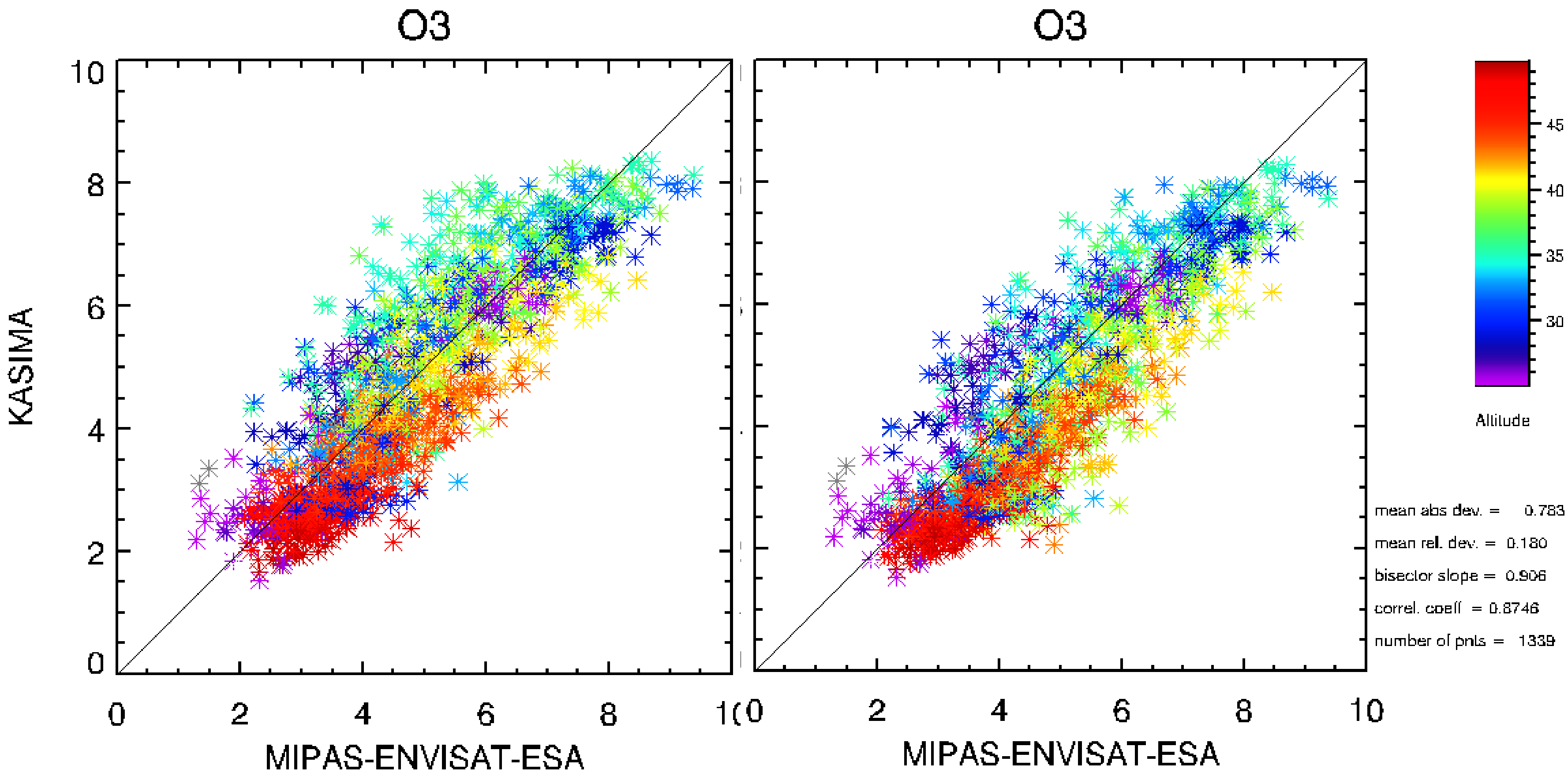
Ozone: Simulations vs. Observations
example antarctic winter, DOY 240 - 270



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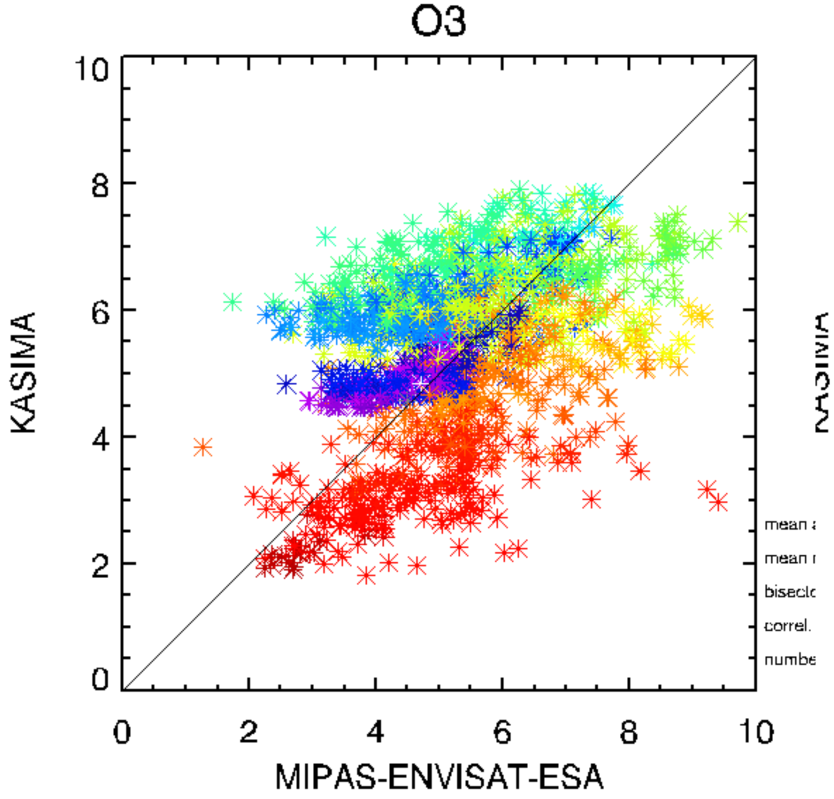
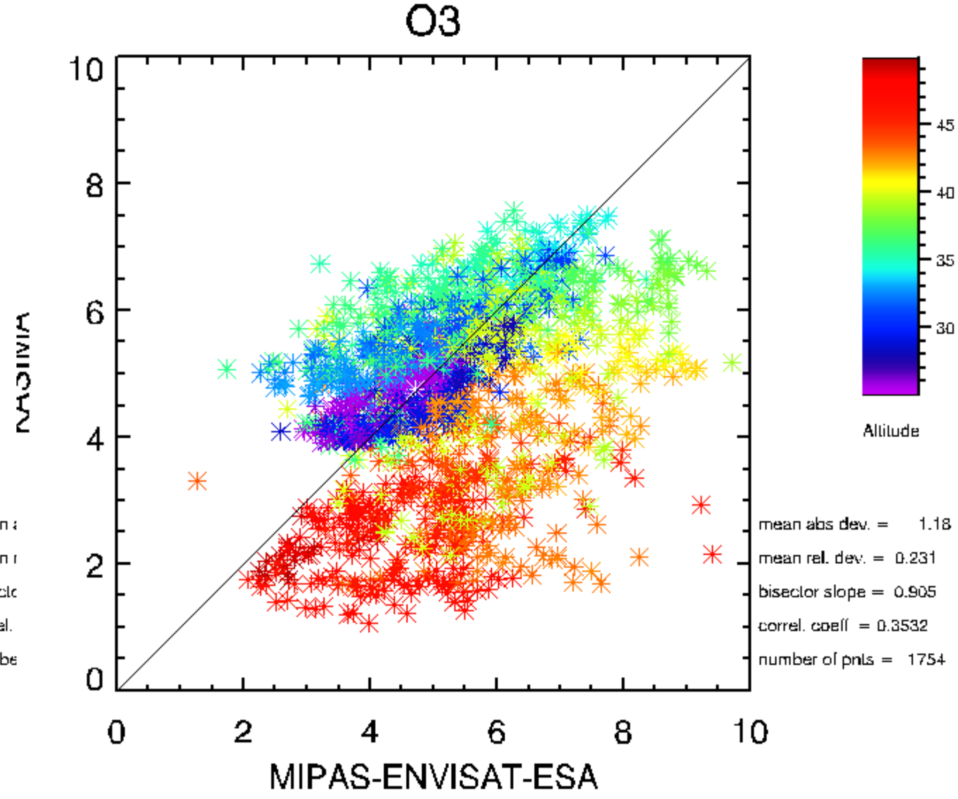
Correlation late winter 2003/2004 > 60°N
DOY 40 - 80

undisturbed



disturbed



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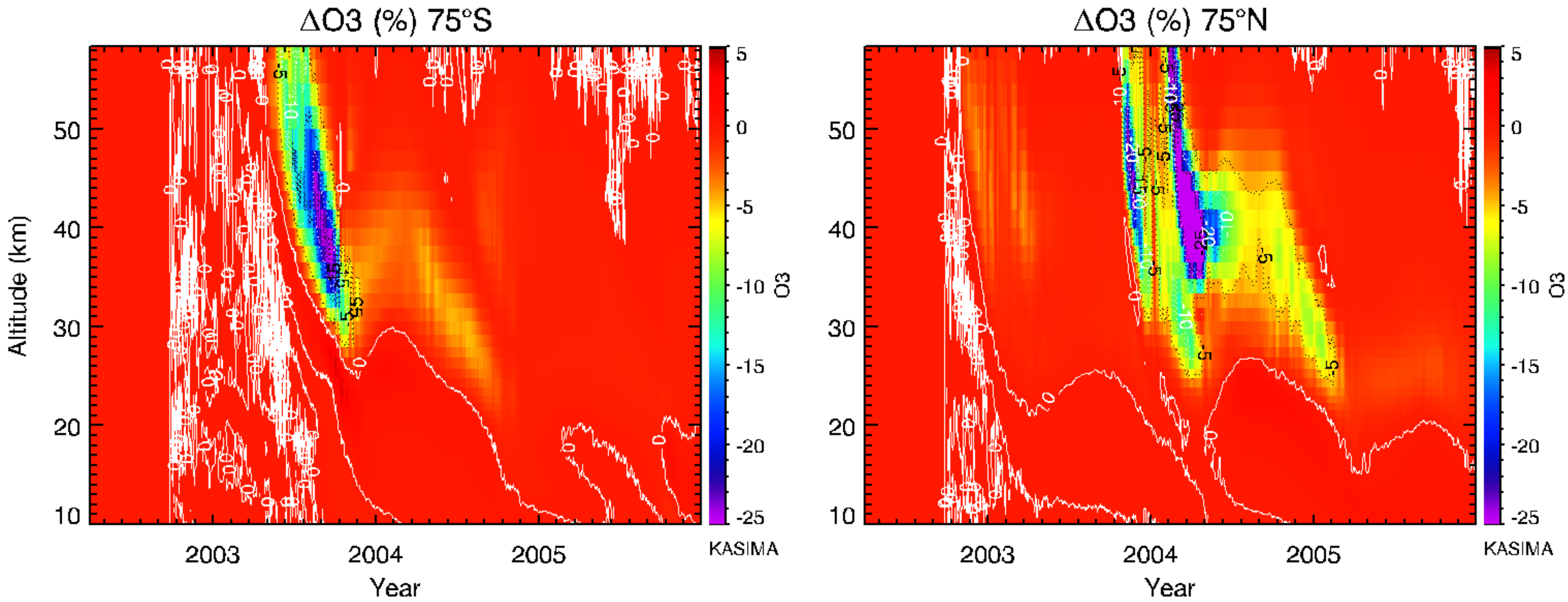
Polar ozone



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