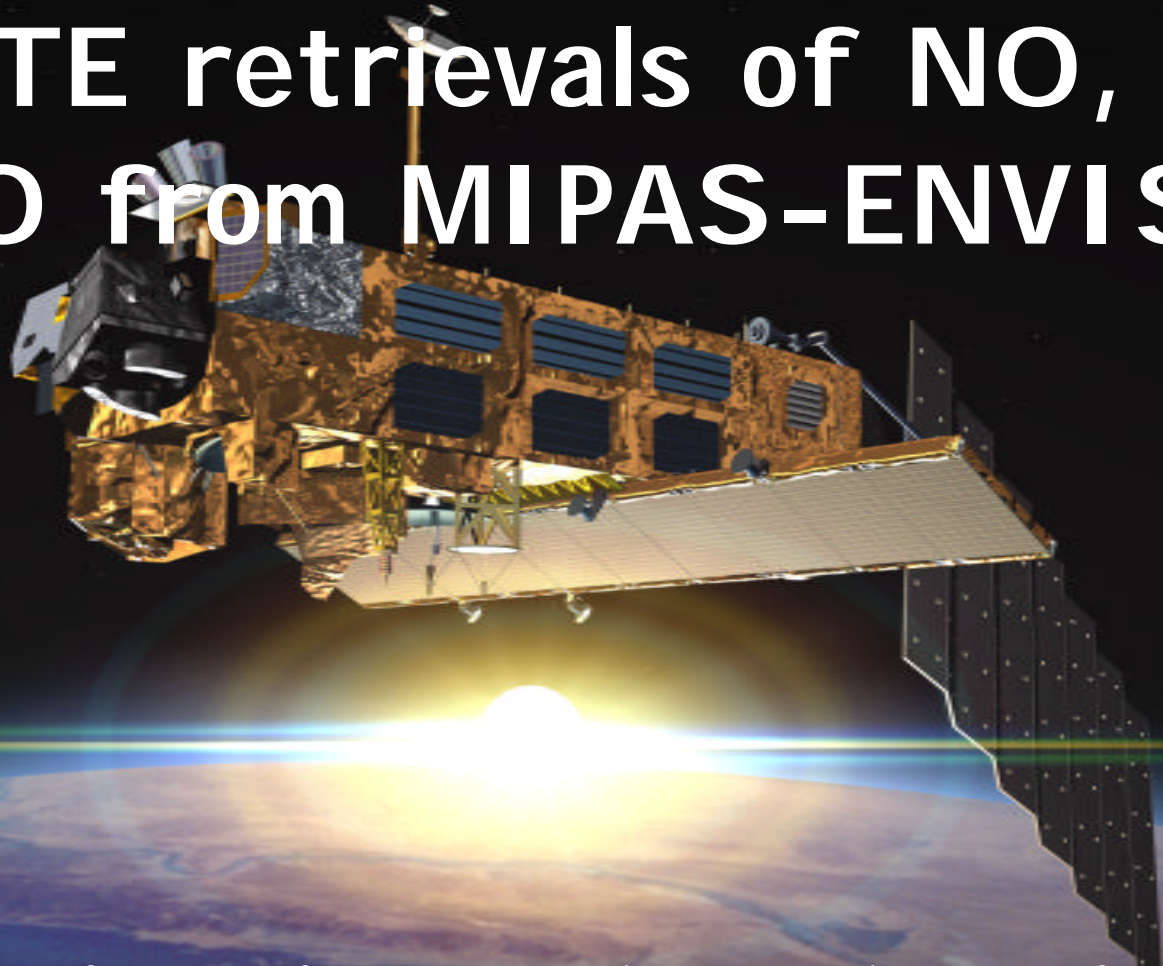


# Non-LTE retrievals of NO, NO<sub>2</sub>, and CO from MIPAS-ENVISAT



B. Funke<sup>1</sup>, T. von Clarmann<sup>2</sup>, H. Fischer<sup>2</sup>, M. García-Comas<sup>1</sup>, S. Gil-López<sup>1</sup>, N. Glatthor<sup>2</sup>, U. Grabowski<sup>2</sup>,  
M. Höpfner<sup>2</sup>, S. Kellmann<sup>2</sup>, M. Kiefer<sup>2</sup>, A. Linden<sup>2</sup>, M. López-Puertas<sup>1</sup>, M.Á. López-Valverde<sup>1</sup>,  
G. Mengistu Tsidu<sup>2</sup>, M. Milz<sup>2</sup>, T. Steck<sup>2</sup>, G.P. Stiller<sup>2</sup>, D.Y. Wang<sup>2</sup>

<sup>1</sup> Instituto de Astrofísica de Andalucía (CSIC), Apdo. 3004, 18080 Granada, Spain.

<sup>2</sup> Institut für Meteorologie und Klimaforschung, Forschungszentrum Karlsruhe GmbH, Karlsruhe, Germany



## Outline

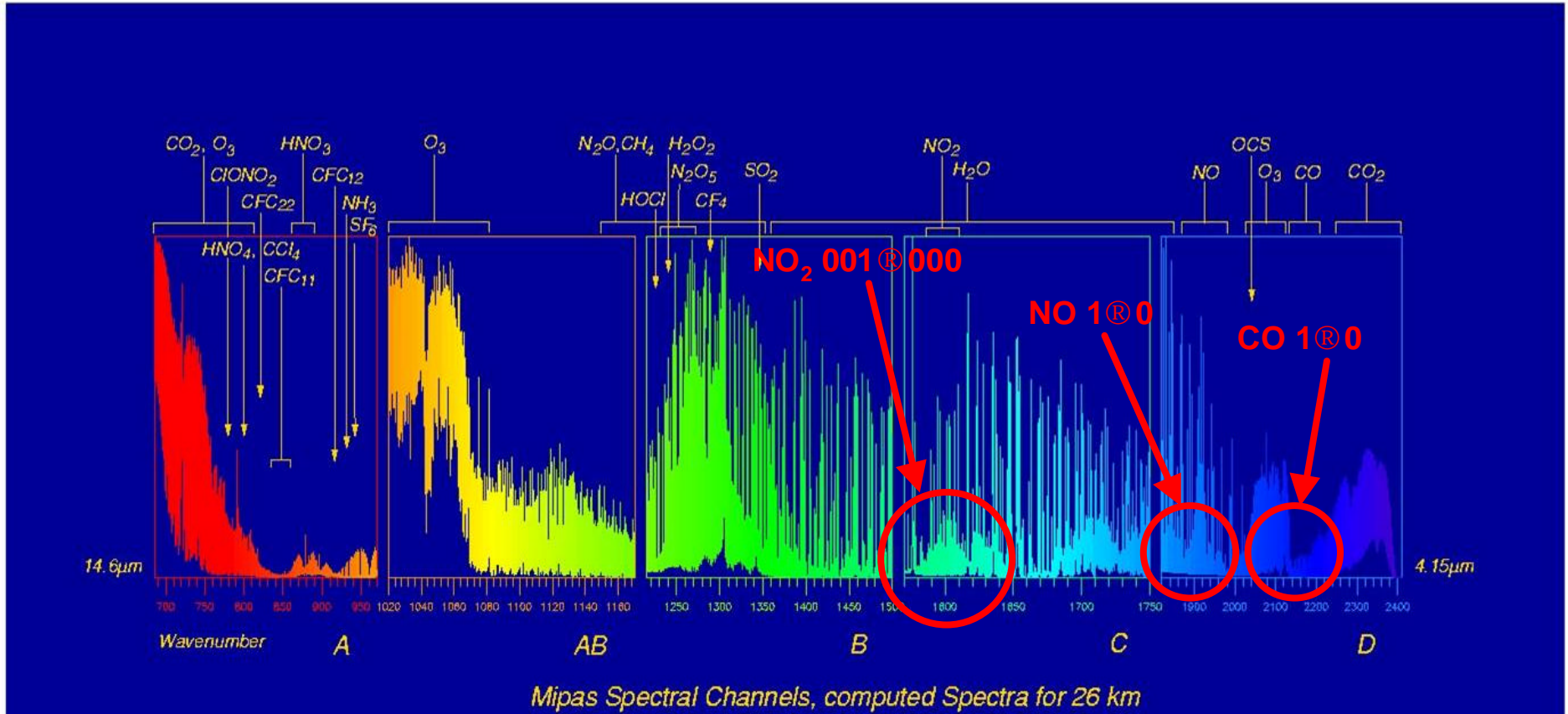
- The non-LTE retrieval scheme
- Retrieval setup & accuracy of data products
- **Validation**
- **Measurements of NO, NO<sub>2</sub>, and CO in July, September, and October 2002 (including split-up of S polar vortex at 24-26 September): preliminary results**



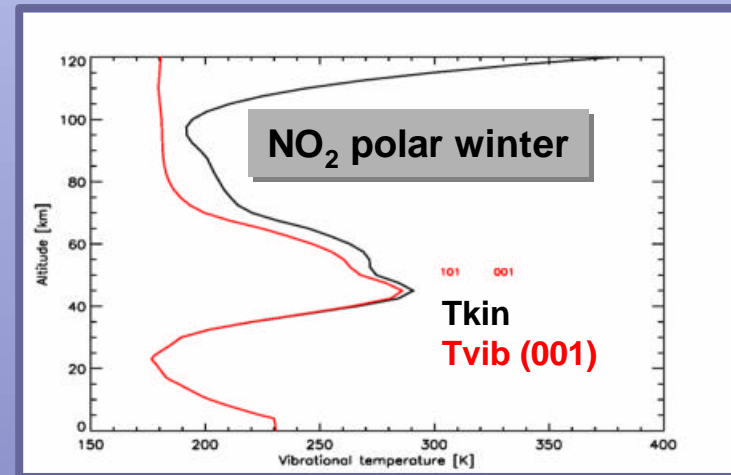
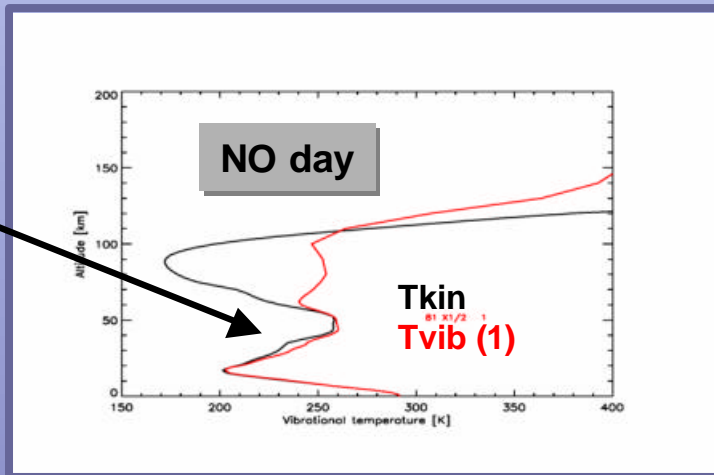
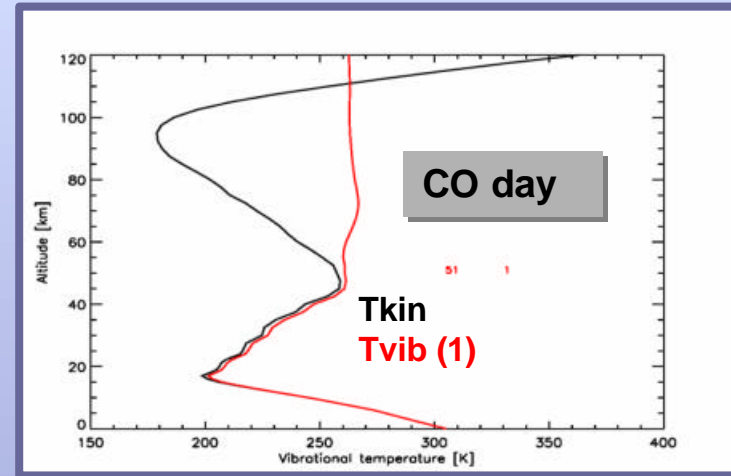
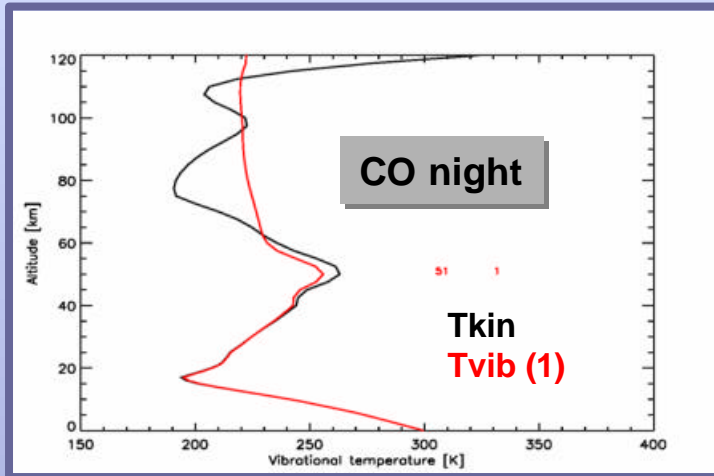
## Scientific objectives

- Dynamical coupling of stratosphere and mesosphere  
P CO
- Descent of mesospheric/thermospheric  $\text{NO}_x$  down to stratosphere & implications to stratospheric chemistry  
P  $\text{NO} + \text{NO}_2$ , CO

# MIPAS spectral channels

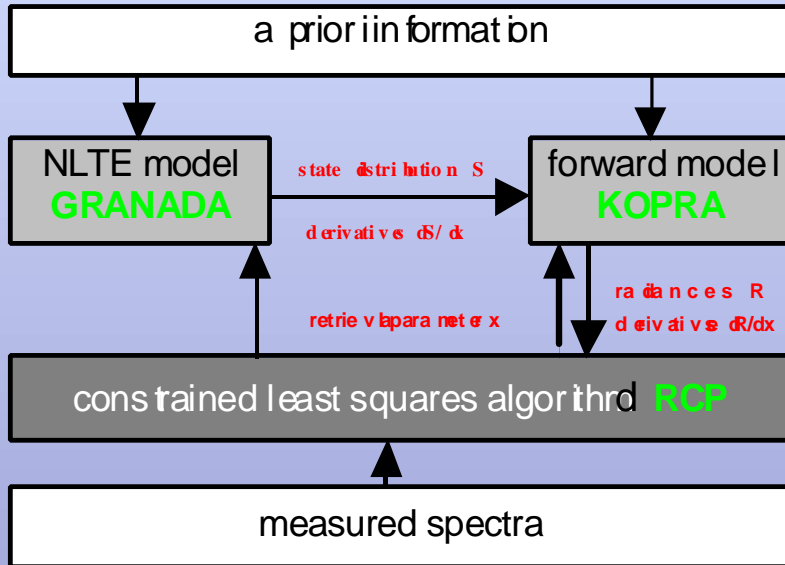


# Vibrational temperatures of CO, NO, and NO<sub>2</sub>



vmr - dependent !!!! .....recursive non-LTE retrieval scheme required

# The non-LTE retrieval processor



## Forward model: Karlsruhe Optimised and Precise Radiative transfer Algorithm (KOPRA)

- Line-by-line radiative transfer model
- Interface for generic NLTE-model GRANADA
- supports vibrational and rotational non-LTE
- Computes spectra and Jacobians for LTE and non-LTE

## Retrieval Control Program (RCP)

- global fit least squares algorithm + userdefined regularisation

## Non-LTE model: Generic Radiative transfer And non-LTE population Algorithm (GRANADA)

- Calculation of vibrational and rotational populations and their derivatives wrt the NLTE retrieval parameters
- Generalized scheme: same algorithm used for populations of CO<sub>2</sub>, O<sub>3</sub>, CO, NO, NO<sub>2</sub>, H<sub>2</sub>O, OH, etc.
- Userdefined (states and transitions, altitude range, iteration strategies, process definition, etc.)
- Rotational (and spin-orbit) non-LTE
- Line-by-line and line independent radiative transfer (KOPRA)
- Inversion of multilevel steady state equation with the Lambda iteration or Curtis matrix formalisms



## Retrieval setup & performance: **nominal** mode

	<b>CO</b>	<b>NO</b>	<b>NO<sub>2</sub></b>
<b>tangent heights</b>	12 -70 km	20 -70 km	15-70 km
<b>microwindows</b>	2024 - 2217 cm <sup>-1</sup>	1840 -1920 cm <sup>-1</sup>	1580 - 1630 cm <sup>-1</sup>
<b>non-LTE</b>	vibrational	vibrational +rotational	vibrational
<b>retrieval grid</b>	0-120km	0-200 km	0-120 km
<b>regularisation</b>	Tikhonov 1st order	Tikhonov 1st order	Tikhonov 1st order
<b>vertical resolution</b>	10km @ <40 km 7 km @ 40 - 70 km	10km @ 20 km 5 km @ 40 km >50 km @ > 100 km	7 km @ 20 km 4 km @ 40 km
<b>noise error</b>	20 ppb @ 30 km 100 ppb @ 50 km	1-3 ppb in statosphere	0.2-0.8 ppb
<b>systematic error sources</b>	<ul style="list-style-type: none"><li>•temperature</li><li>•ILS</li><li>•terminator (non-LTE)</li></ul>	<ul style="list-style-type: none"><li>•temperature</li><li>•thermospheric horizontal structure</li></ul>	<ul style="list-style-type: none"><li>•temperature</li><li>•ILS</li></ul>



## Validation

### **SPIRALE $\text{P}$ CO**

in situ balloon measurements (1)

Kiruna ( $68^{\circ}\text{N}$  /  $25^{\circ}\text{E}$ ), 21/01/2003

### **MIPAS-B $\text{P}$ $\text{NO}_2$**

Limb emission balloon measurements (2)

Air s/ Ladour ( $41^{\circ}\text{N}/0^{\circ}\text{E}$ ,  $45^{\circ}\text{N}/1^{\circ}\text{E}$ ), 24/09/2002

### **HALOE $\text{P}$ $\text{NO}+\text{NO}_2$**

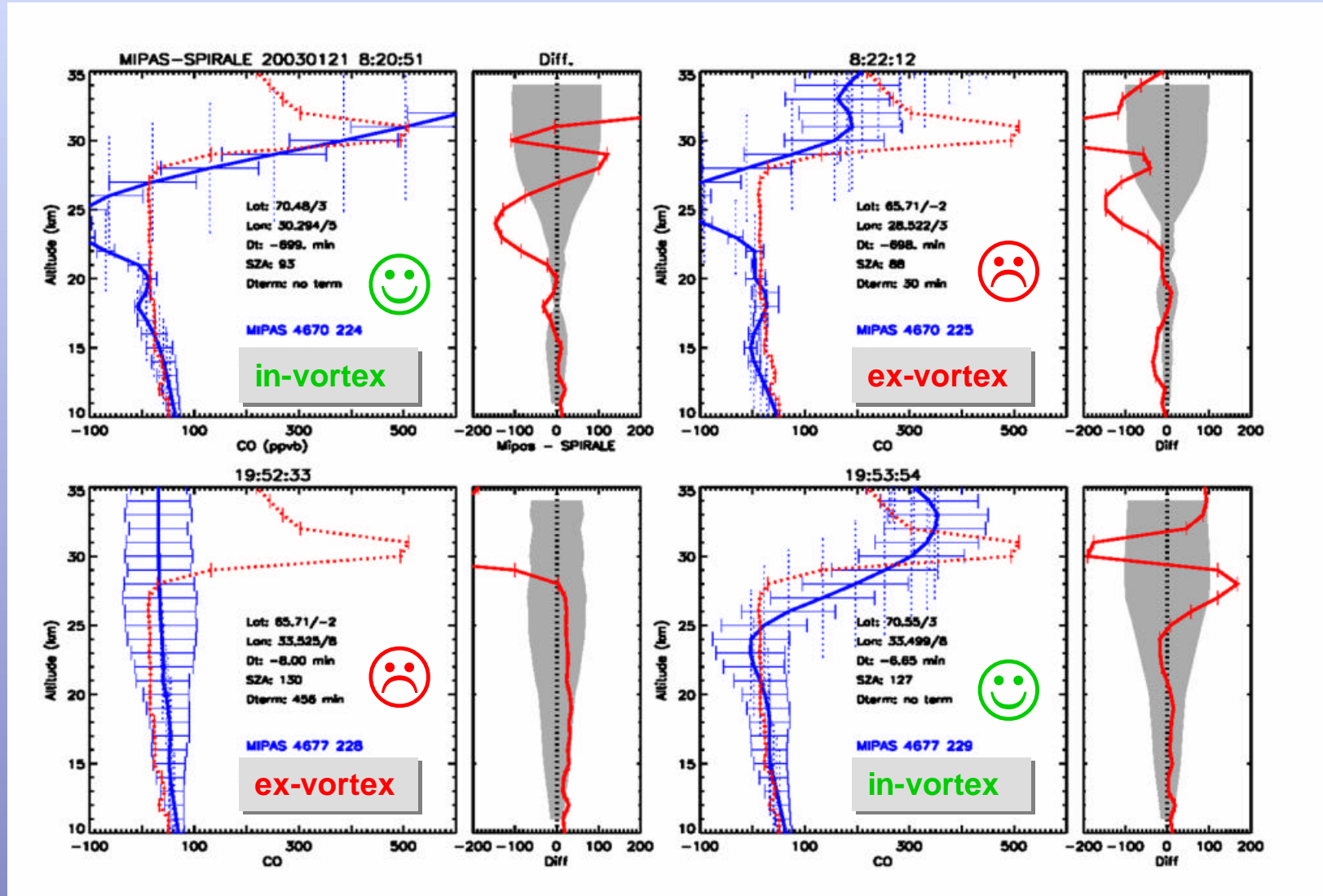
Solar occultation satellite measurements (~ 120 coinc.)

mainly N hemisphere, July - October 2002





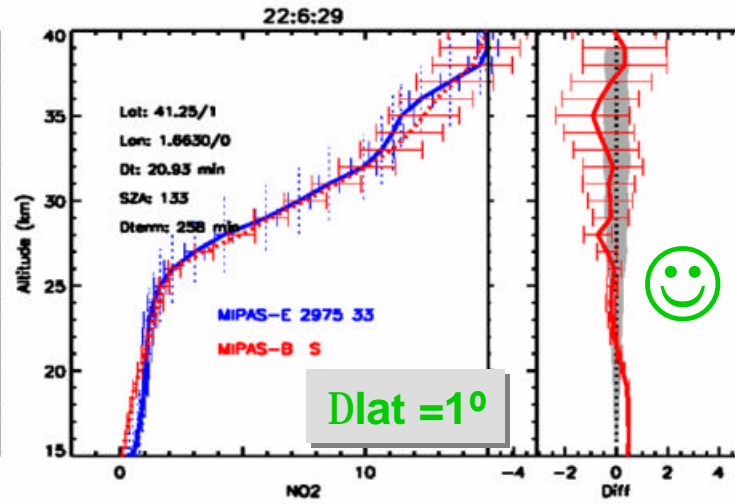
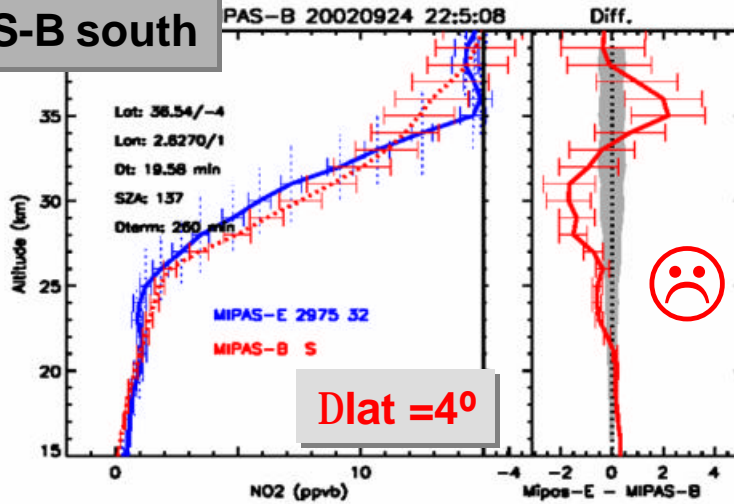
# CO validation with SPIRALE (in situ, 1 profile at 67°N)



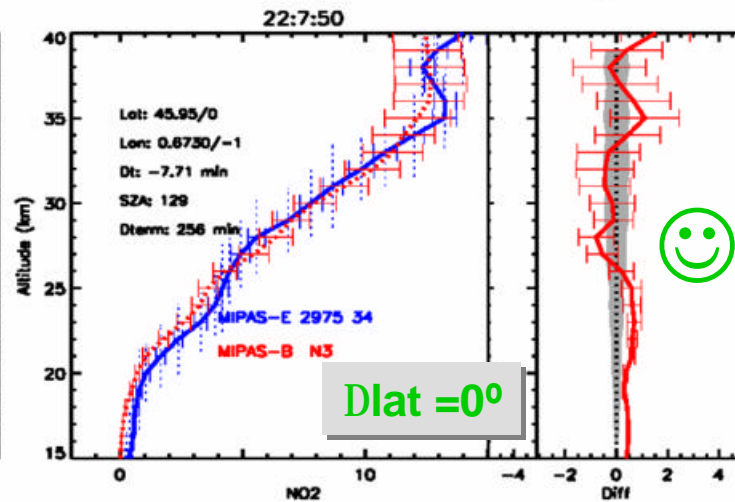
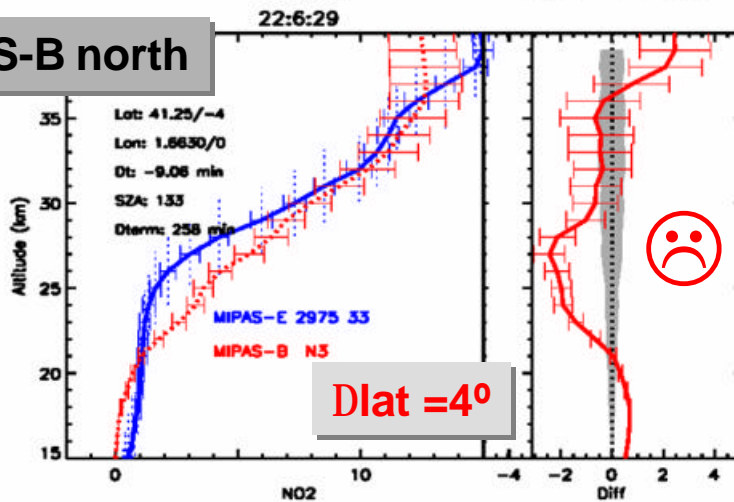


# NO<sub>2</sub> validation with MIPAS-B (2 profiles, night)

MIPAS-B south

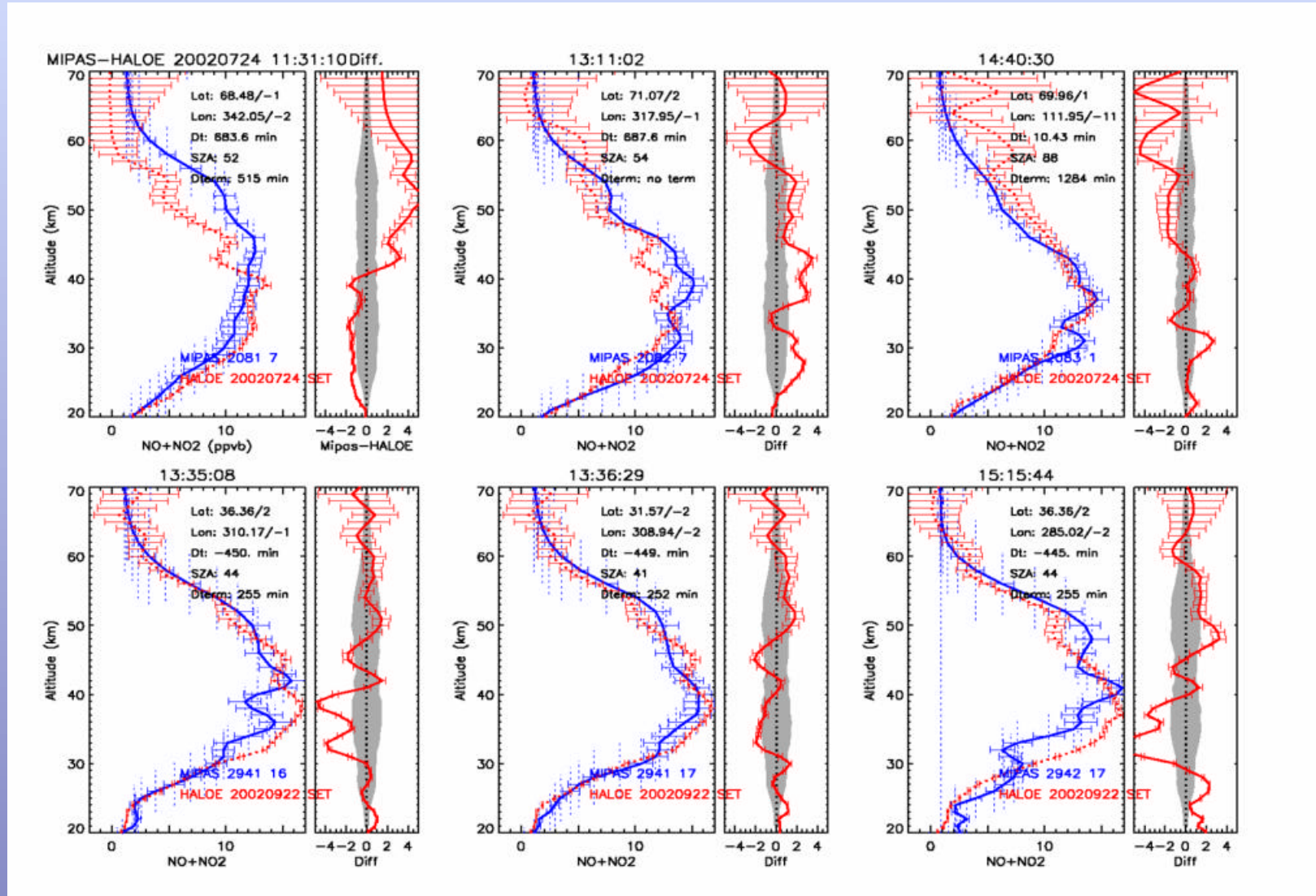


MIPAS-B north

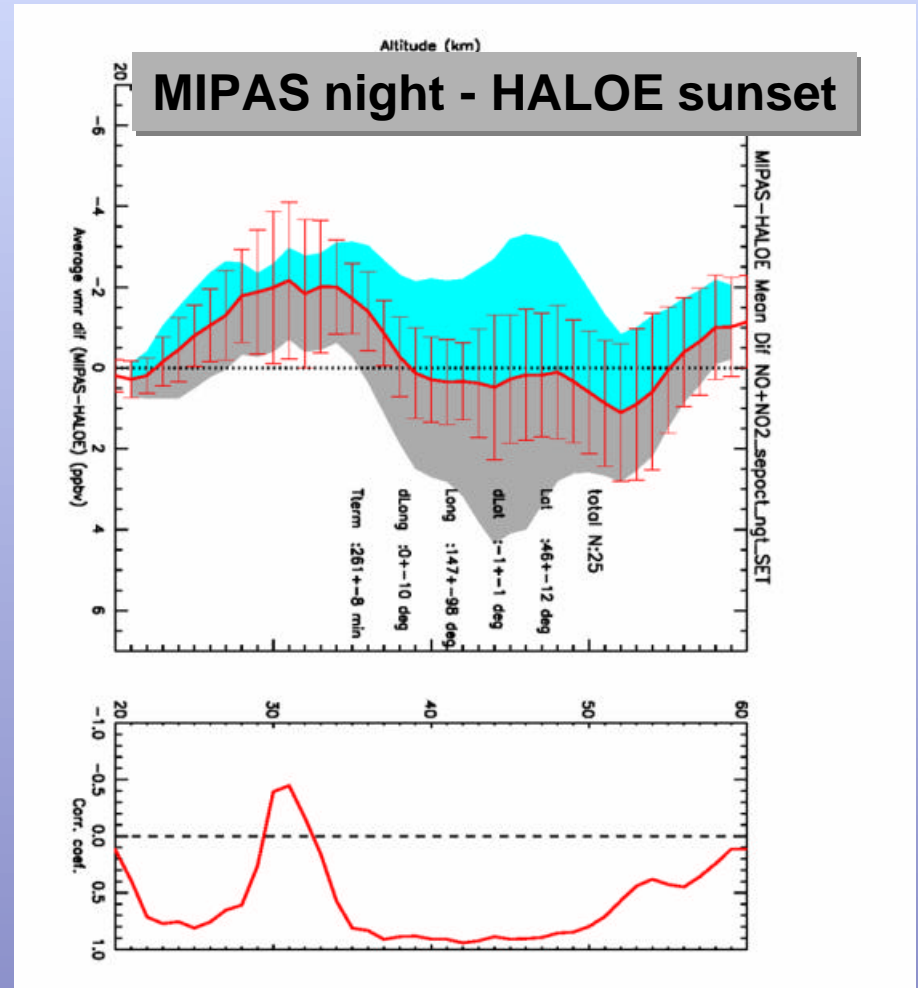
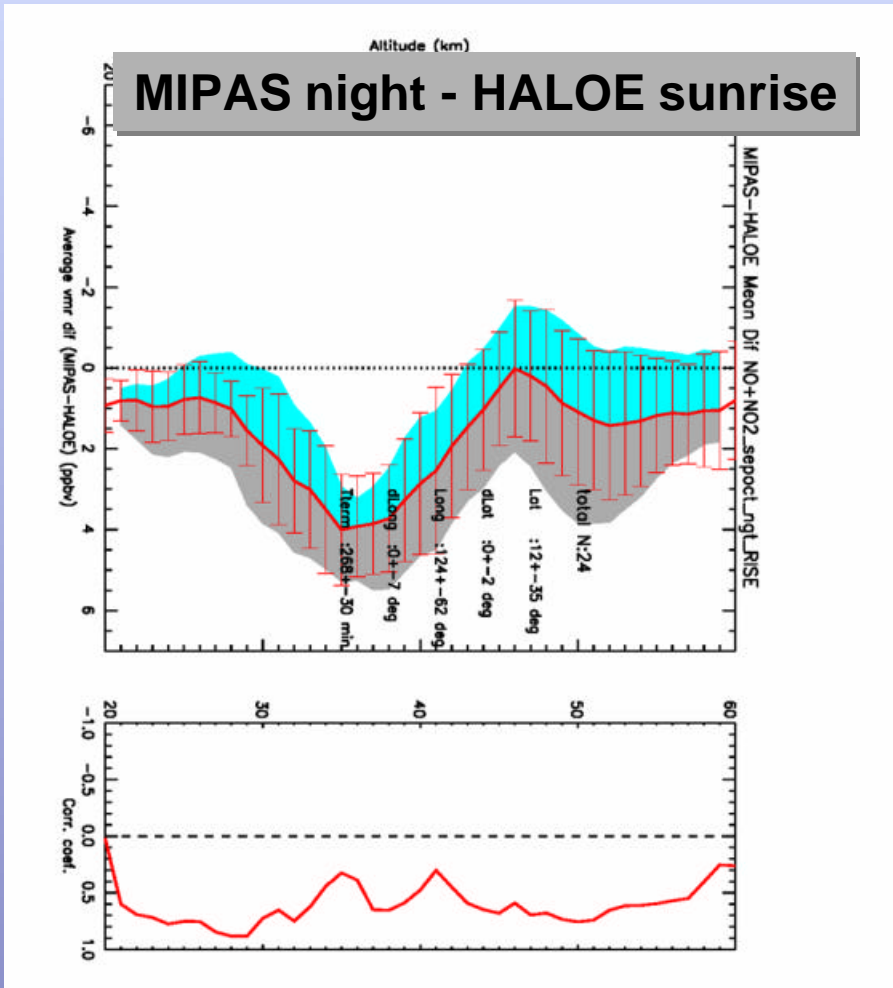




# NO<sub>x</sub> validation with HALOE (examples)



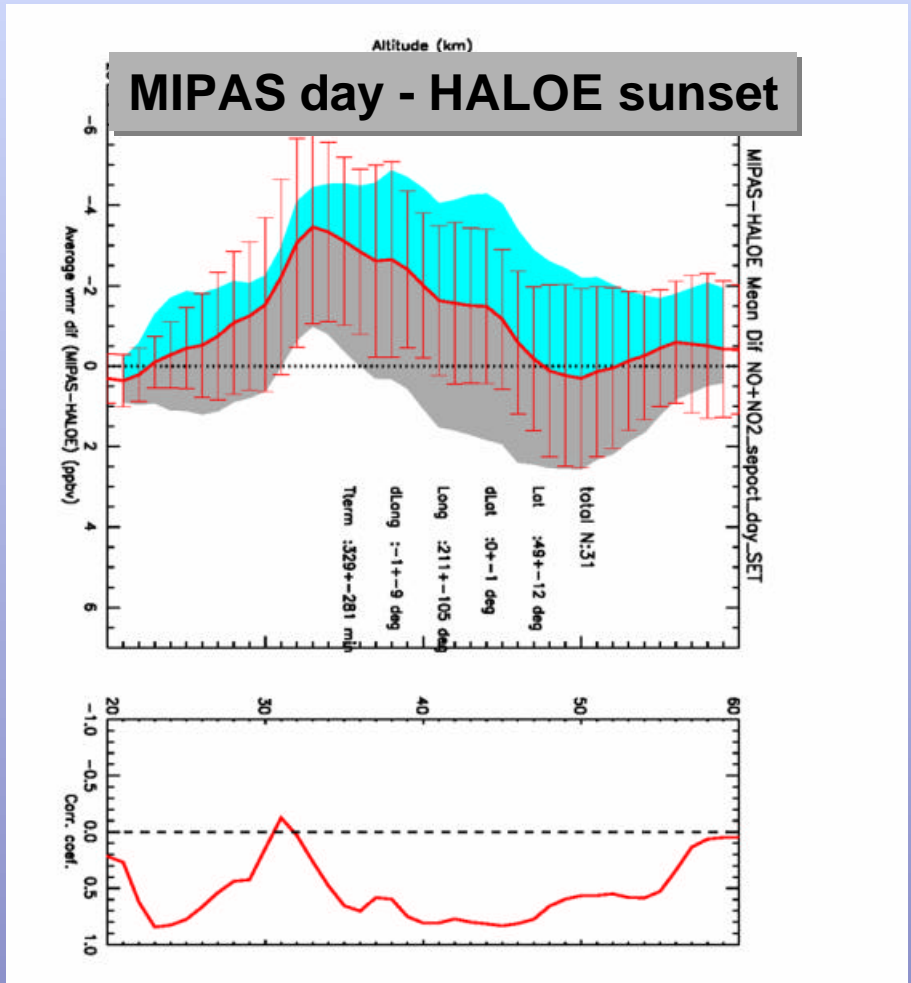
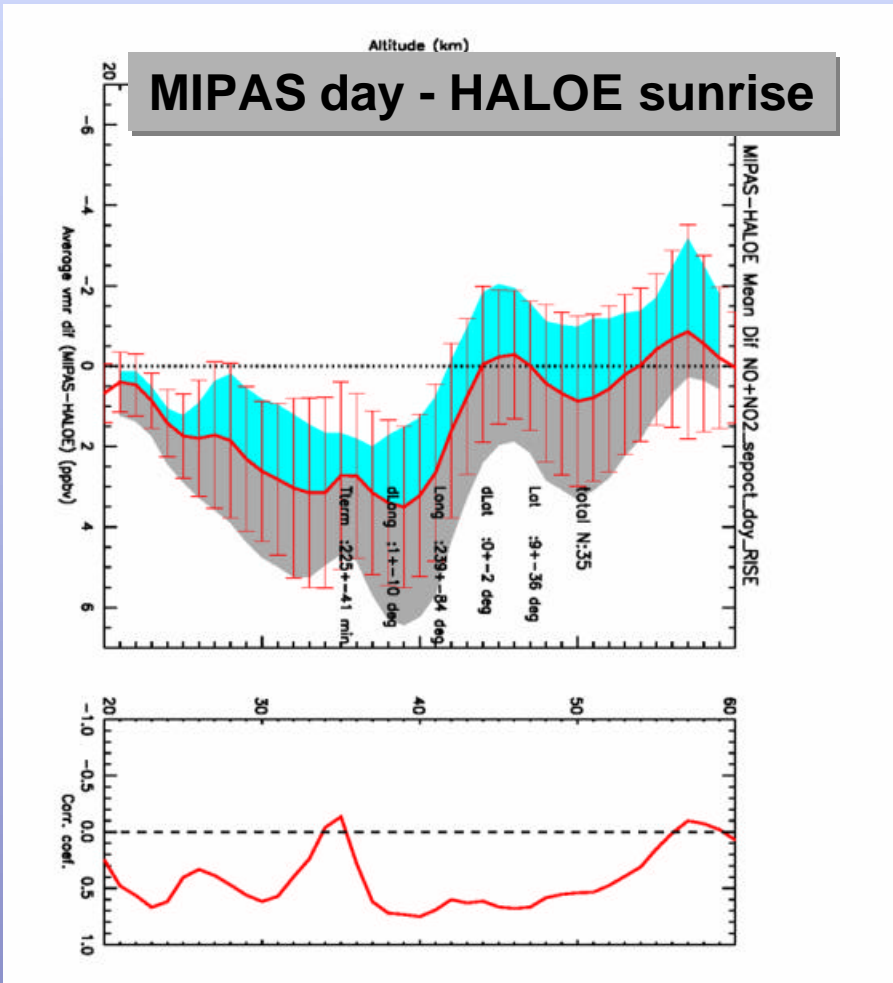
# Nighttime $\text{NO}_x$ compared to HALOE



± 3 ppbv difference due to  $\text{NO}_2 \leftrightarrow \text{N}_2\text{O}_5$  conversion between sunset and sunrise



# Daytime NO<sub>x</sub> compared to HALOE



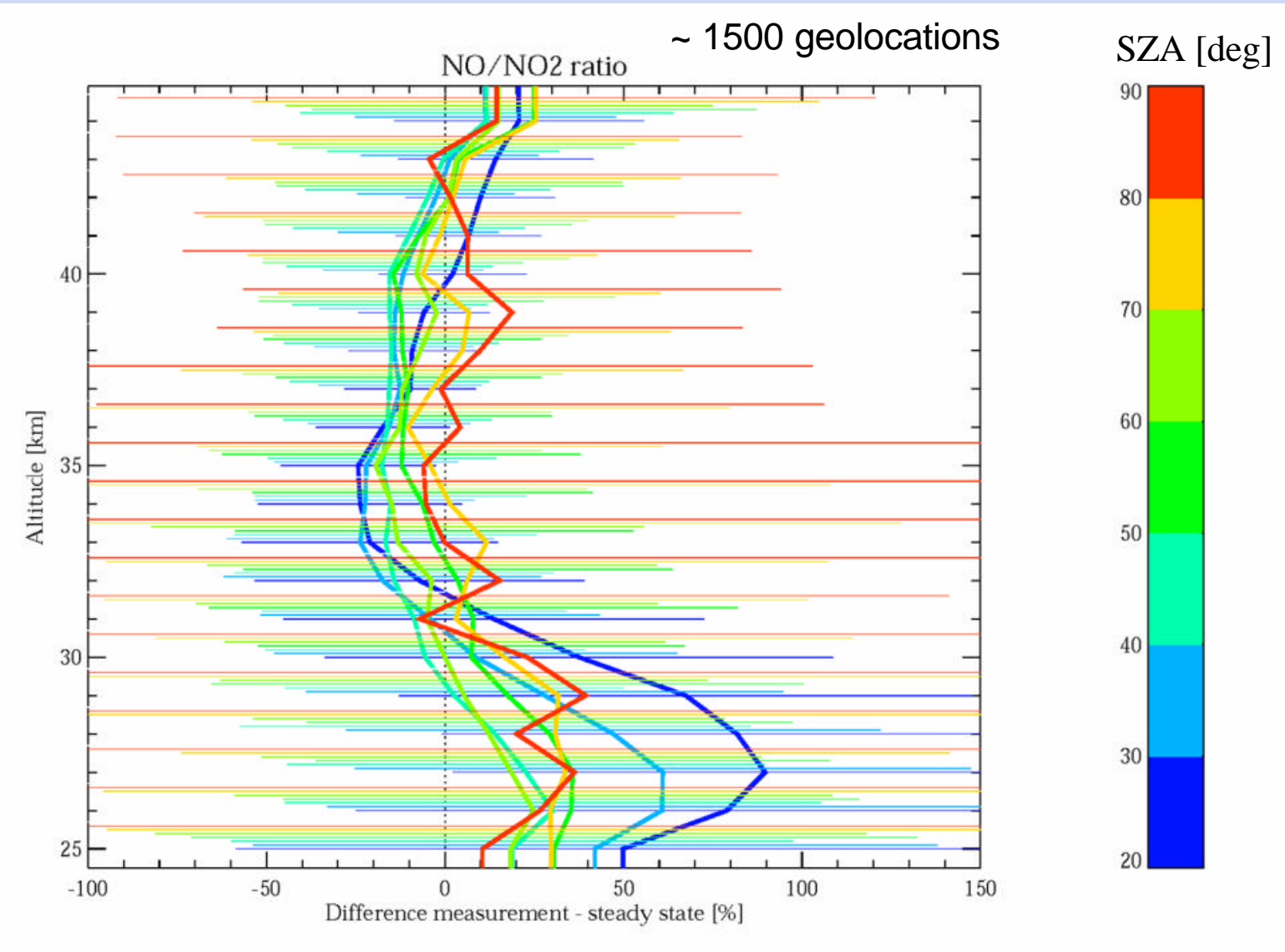
± 3 ppbv difference due to NO<sub>2</sub>↔N<sub>2</sub>O<sub>5</sub> conversion between sunset and sunrise

# Is measured NO and NO<sub>2</sub> consistent with steady state?

Steady state:

$$\frac{[NO]}{[NO_2]} = \frac{J_{NO_2} + k_{NO_2+O}[O]}{k_{NO+O_3}[O_3] + k_{NO+ClO}[ClO]} \quad \text{with} \quad [O] = \frac{J_{O_3}[O_3]}{k_{O+O_2+M}[O_2][M]}$$

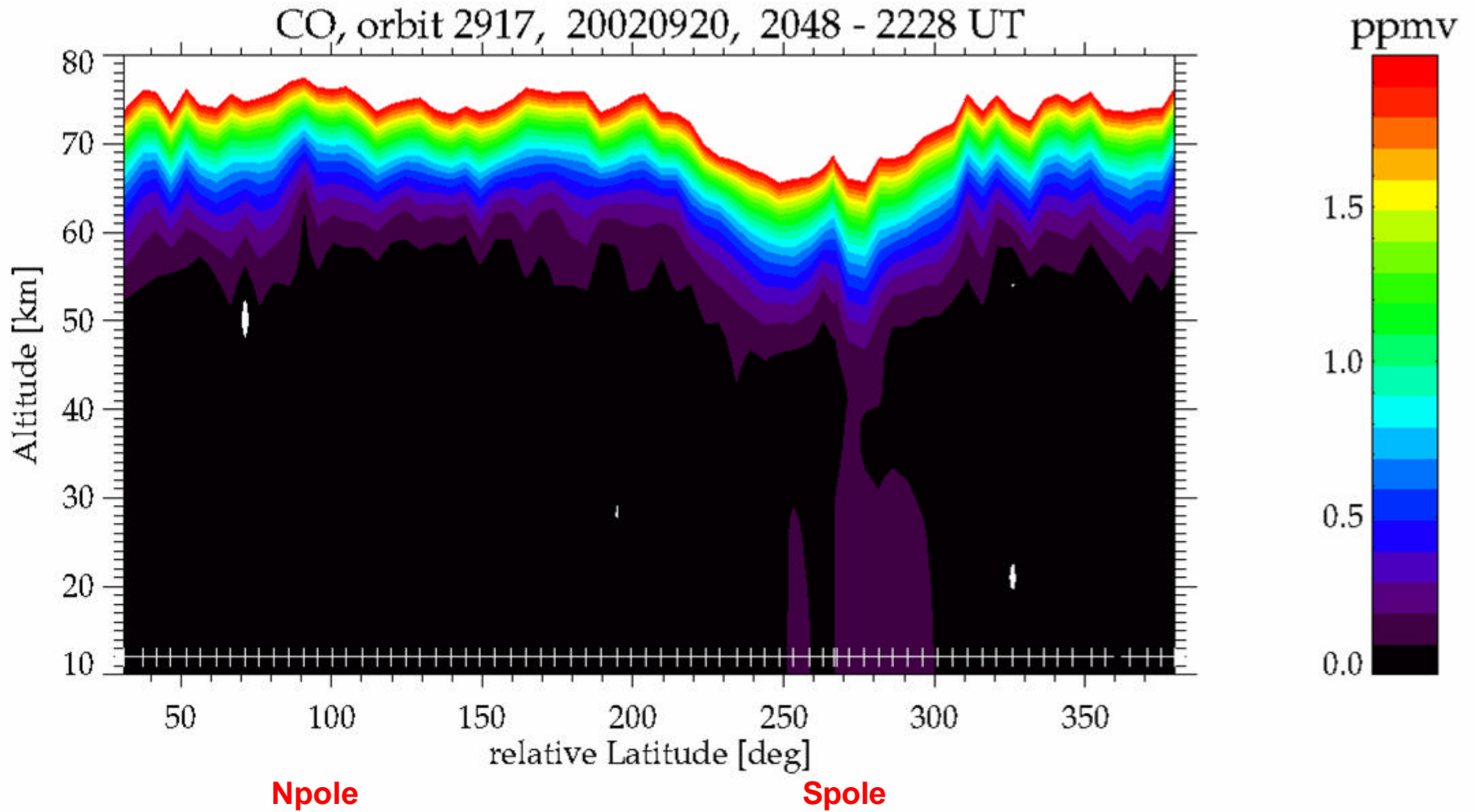
- MIPAS O<sub>3</sub>, ClO, p, and T
- Photolysis rates from TUV model

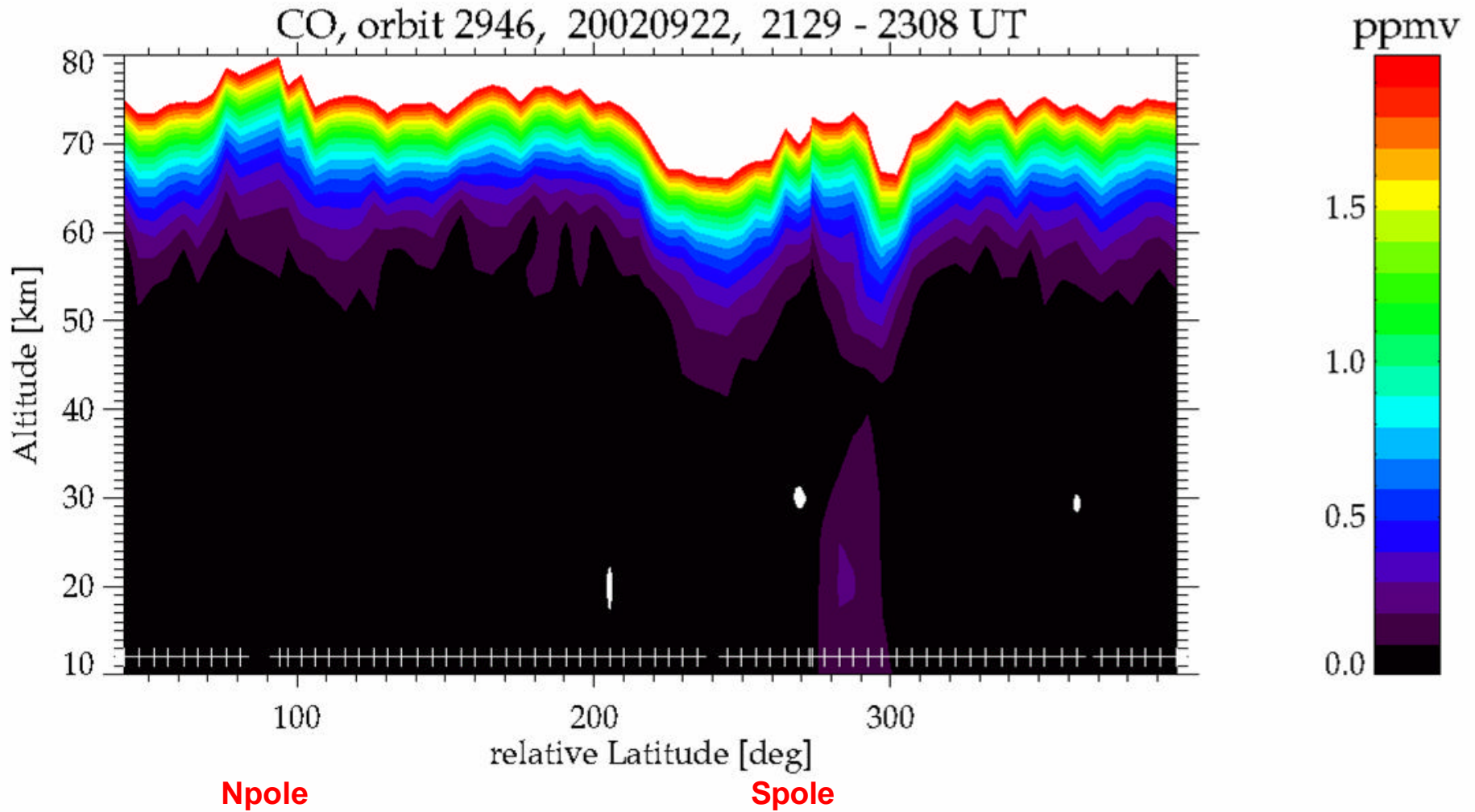


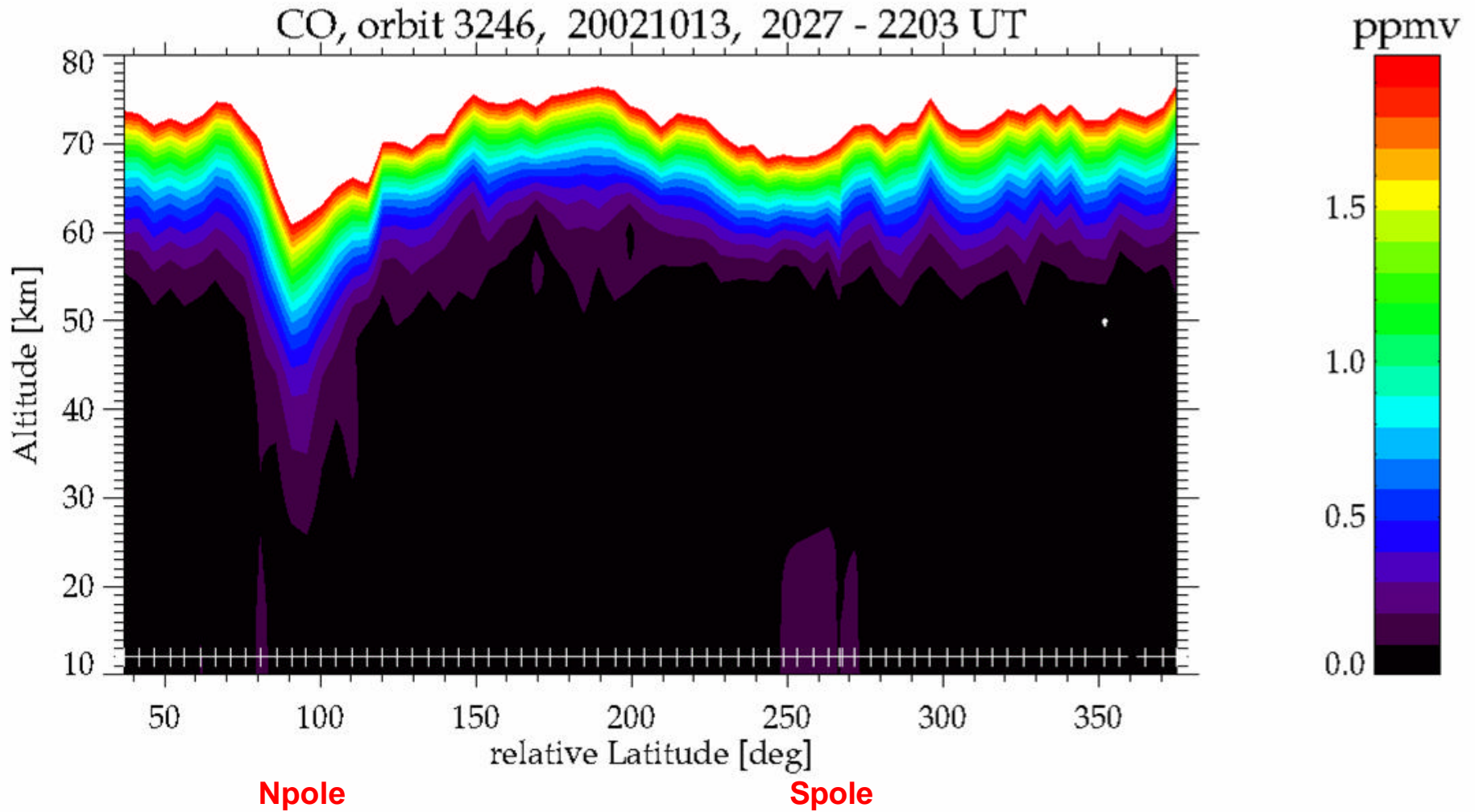


# CO measurements







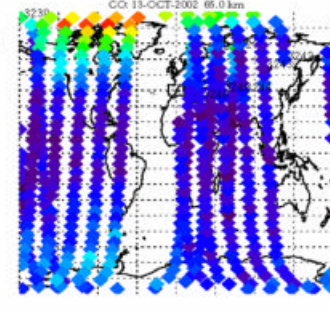
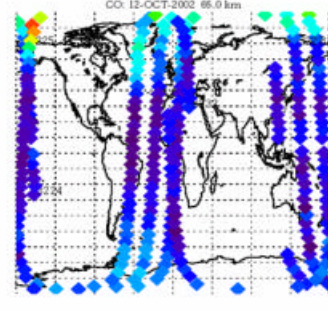
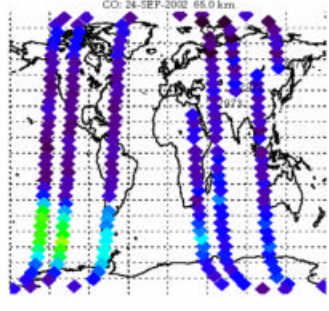
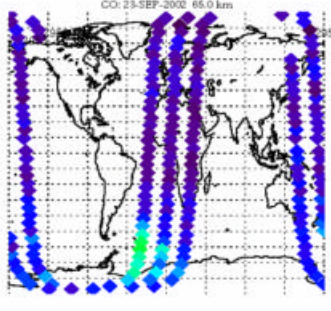
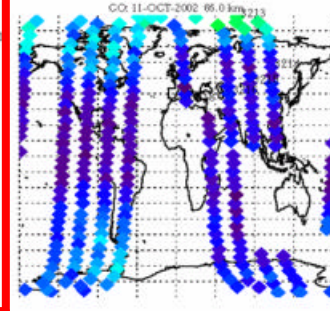
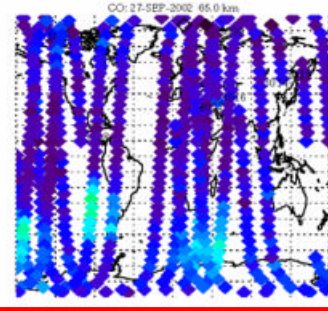
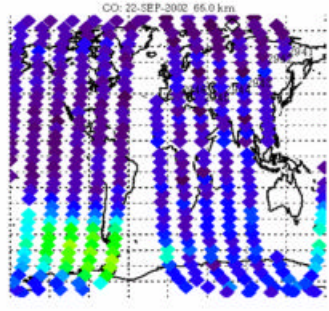
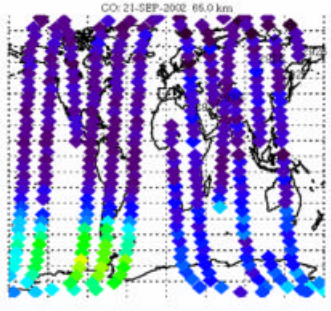
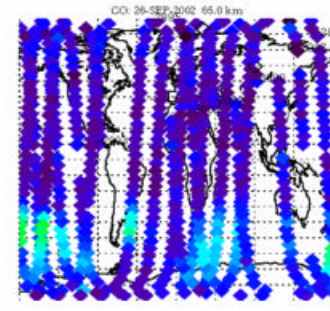
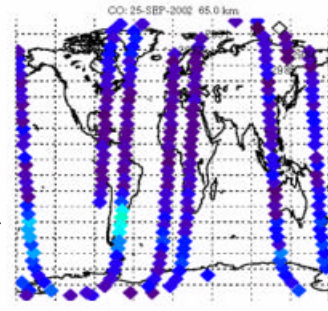
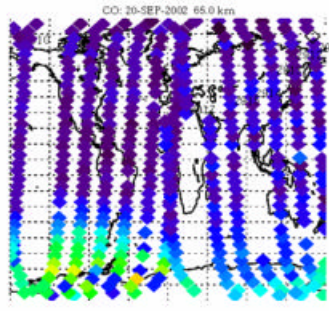
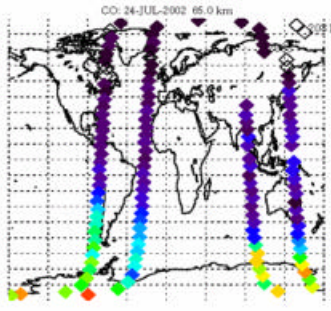




- ⊕ Mesospheric air in stratosphere over South pole.
- ⊕ Turnover of meridional circulation in September/October.

# Mesospheric CO at 65km

24 July



Vortex split-up

20 -27 Sept

11-13 Oct



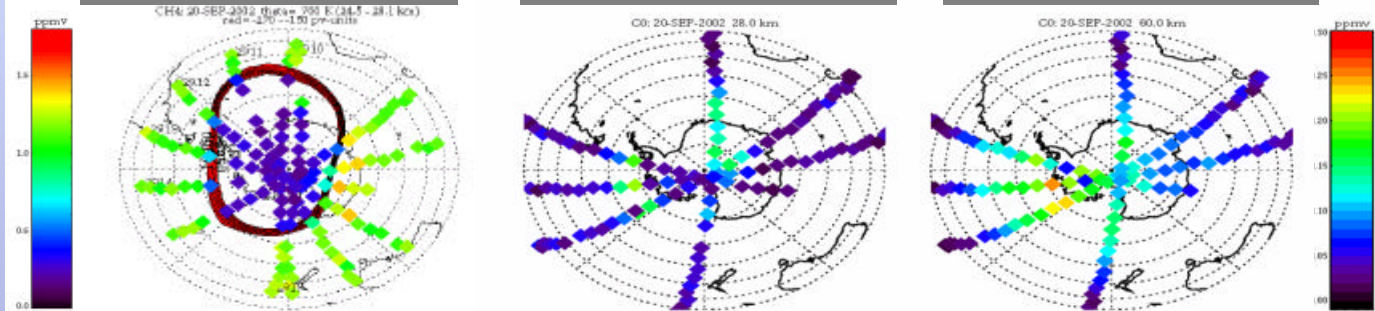
# Comparison of CO, CH<sub>4</sub>, and pV

CH<sub>4</sub> @ q=700 K

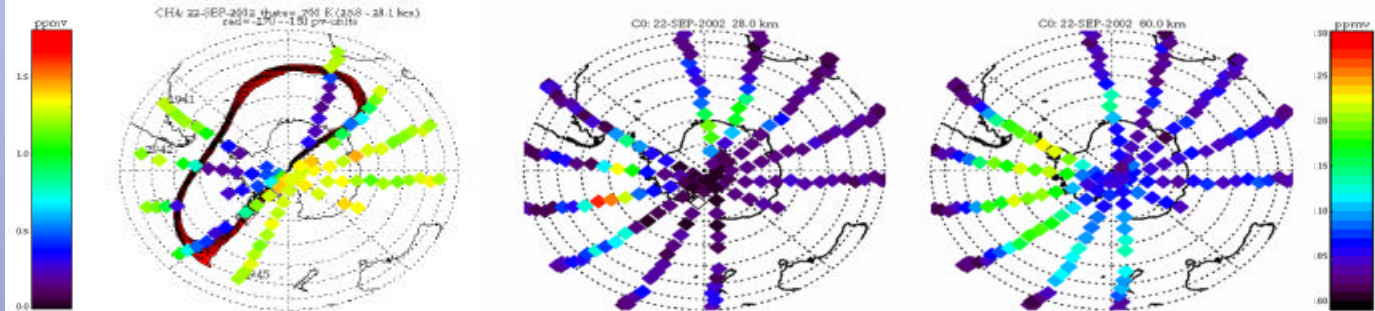
CO @ 28 km

CO @ 60 km

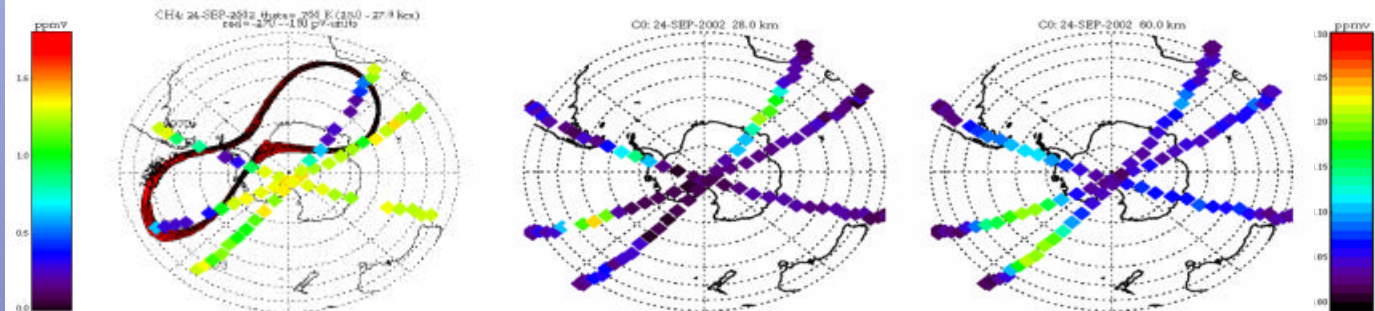
20 Sept.



22 Sept.



24 Sept.





# Dynamical connection between stratosphere and mesosphere:

Is turnover of circulation induced by vortex split ??????

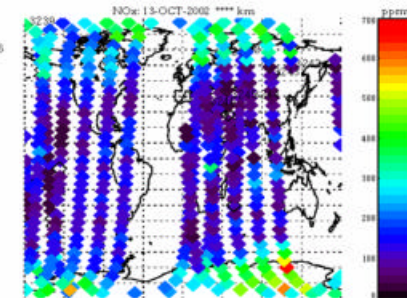
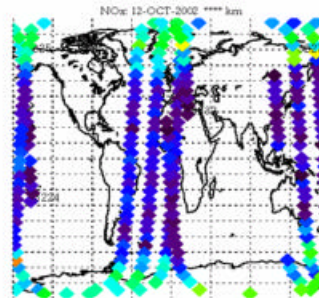
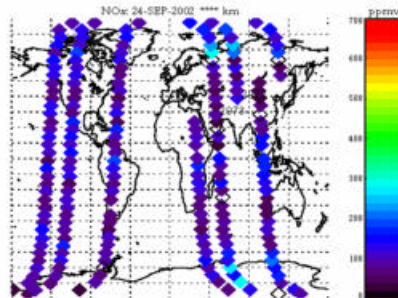
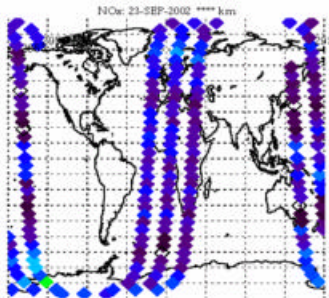
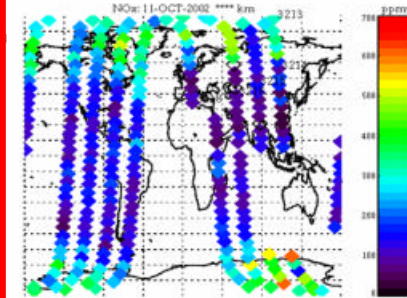
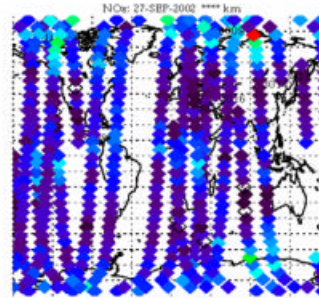
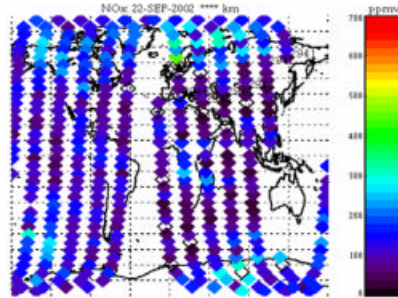
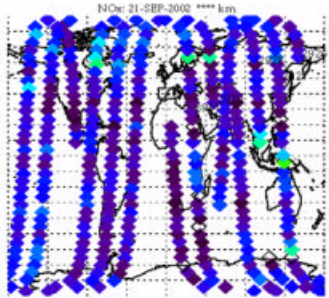
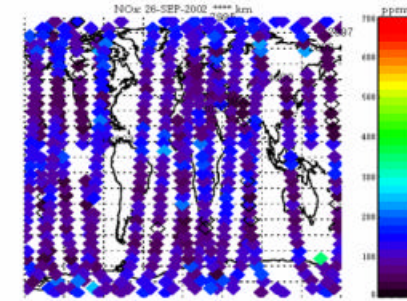
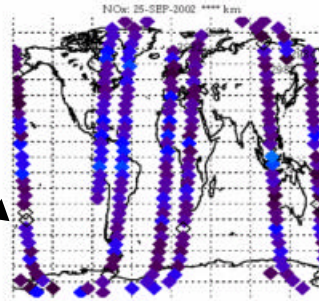
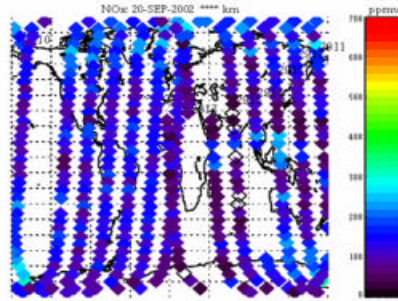
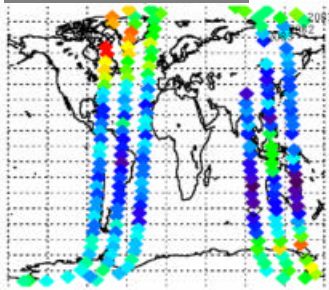


# NO<sub>x</sub> measurements



# Thermospheric NO at 120 km

24 July  
F10.7 » 210



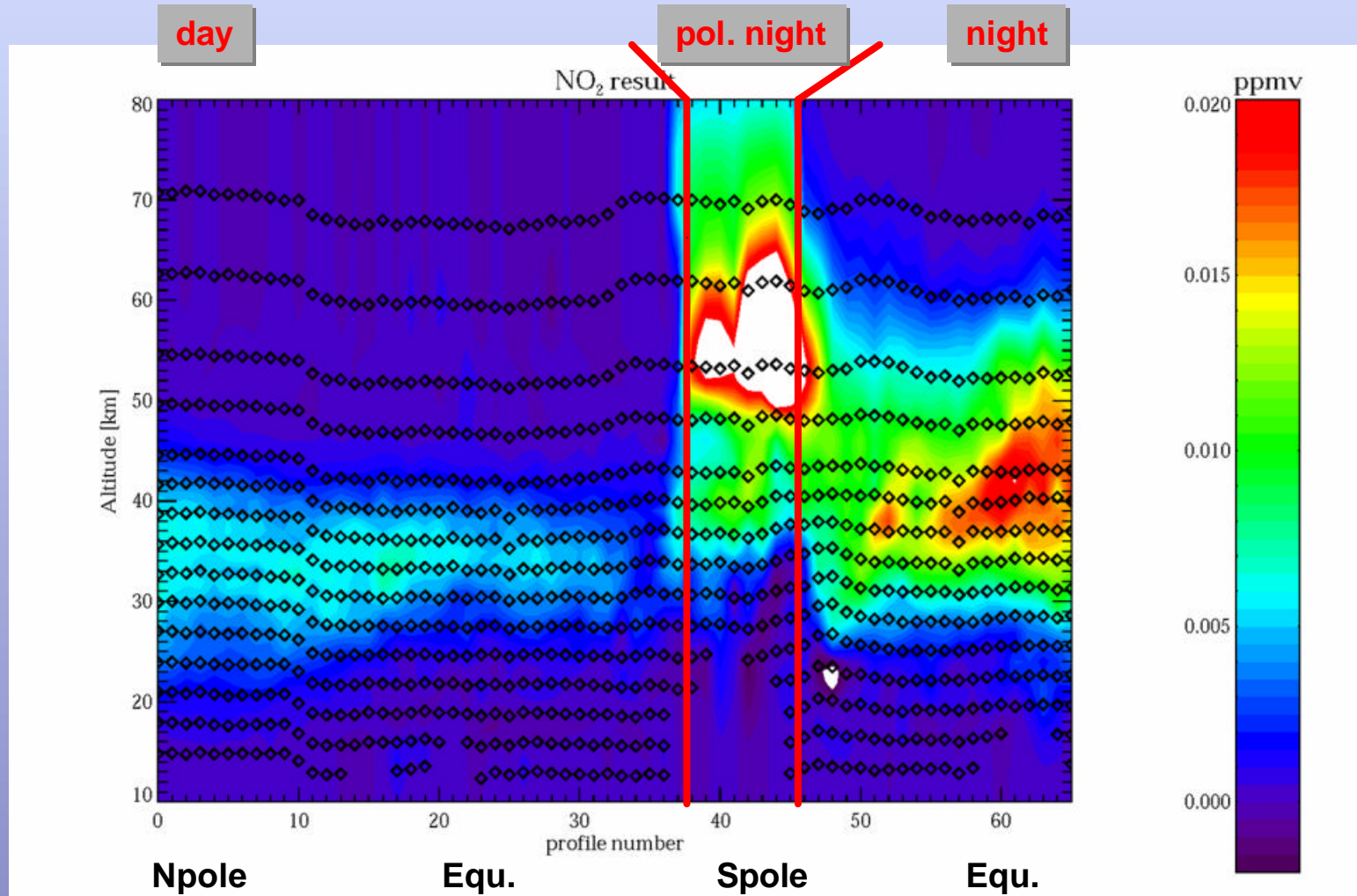
20 -27 Sept  
F10.7 » 155

11-13 Oct  
F10.7 » 180



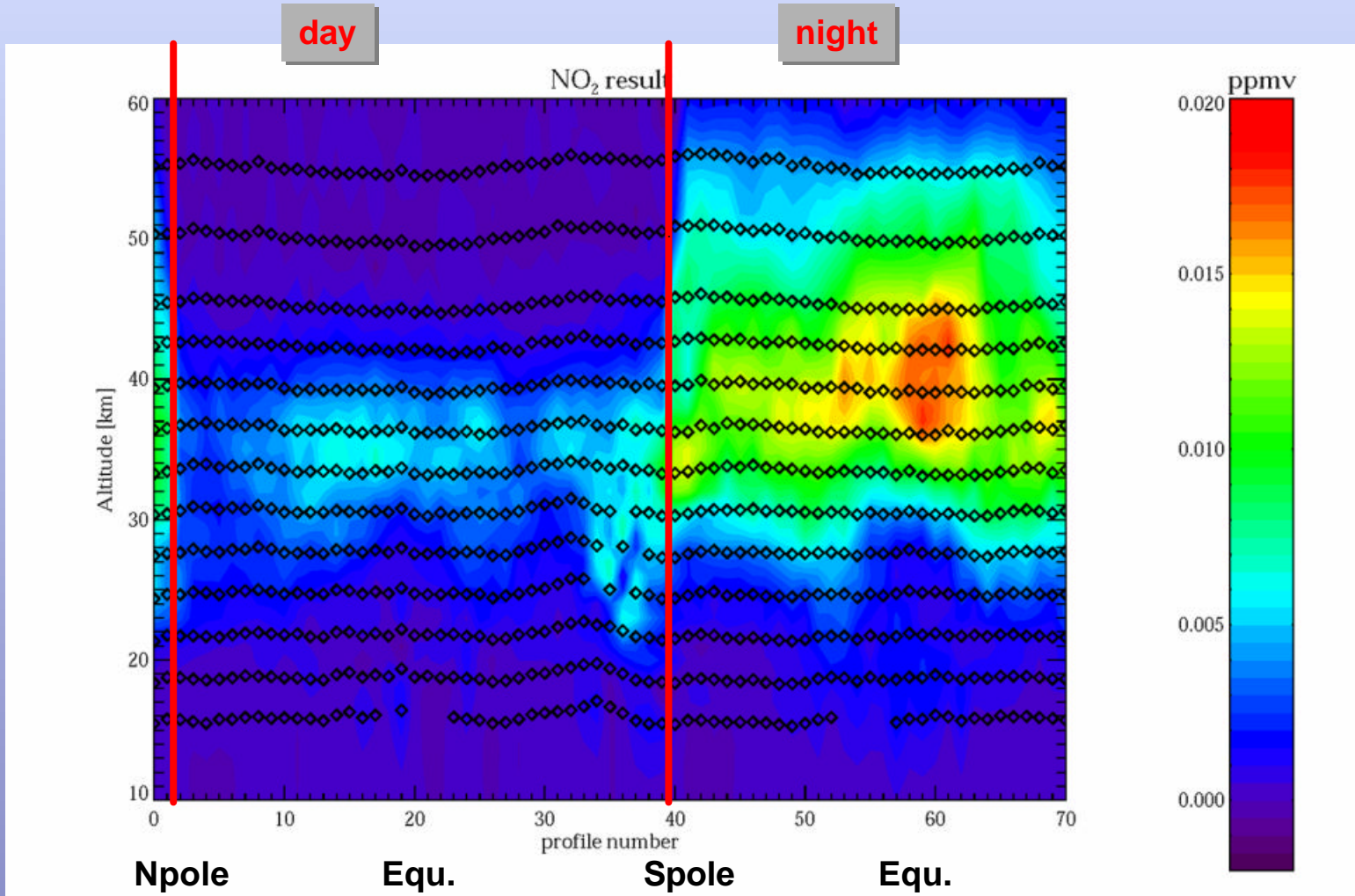
**...is thermospheric NO<sub>x</sub>  
descending to the  
stratosphere?**

# NO<sub>2</sub> measured at 24 July 2003



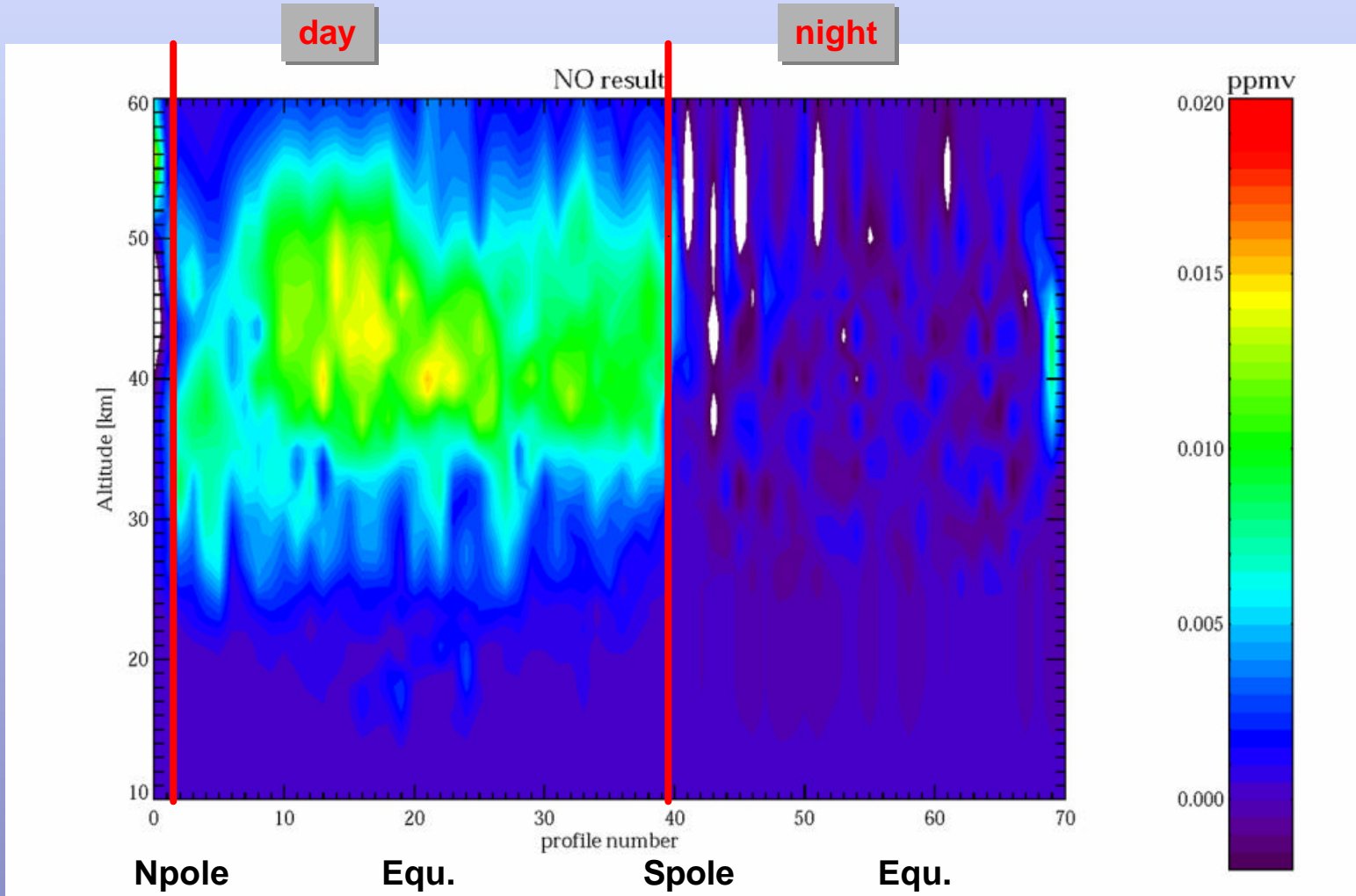
...over 50 ppb NO<sub>2</sub> over the S pole at 60 km !!!!

# NO<sub>2</sub> measured at 24 September 2002



... but not in September !!!!

# NO measured at 24 September 2002

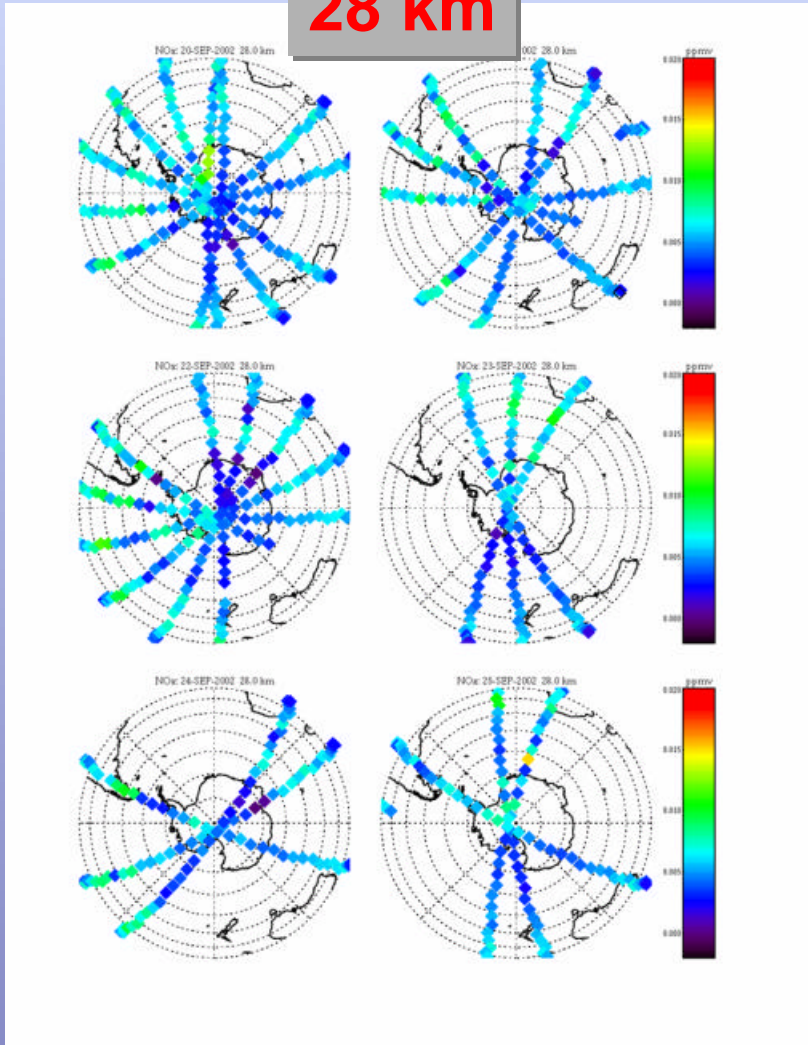




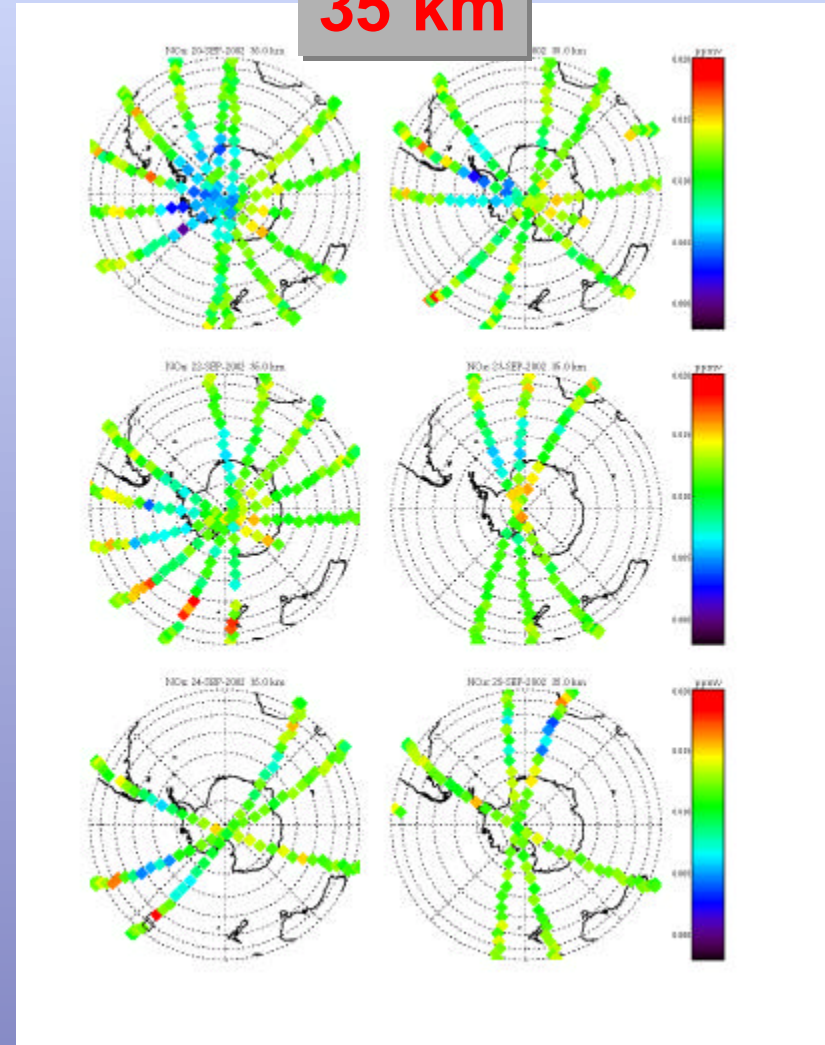
**Enhanced** stratospheric **NO<sub>x</sub>**  
transported down from the  
upper atmosphere can only be  
detected at **polar night**, when  
mesospheric NO photolysis  
barreer is not active.

# NO<sub>x</sub> in S Hemisphere in September (20-26/9/2002)

28 km



35 km





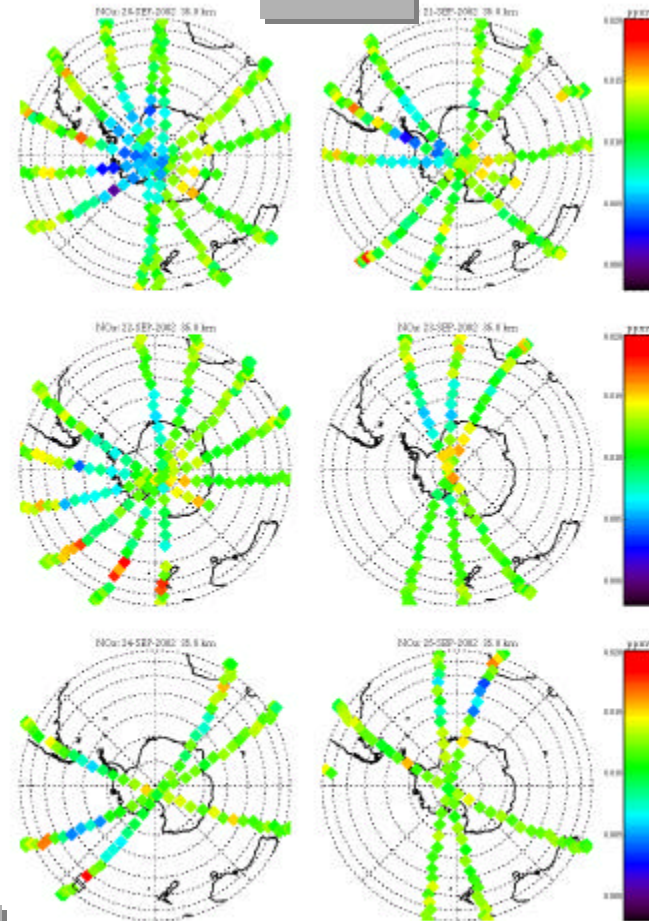
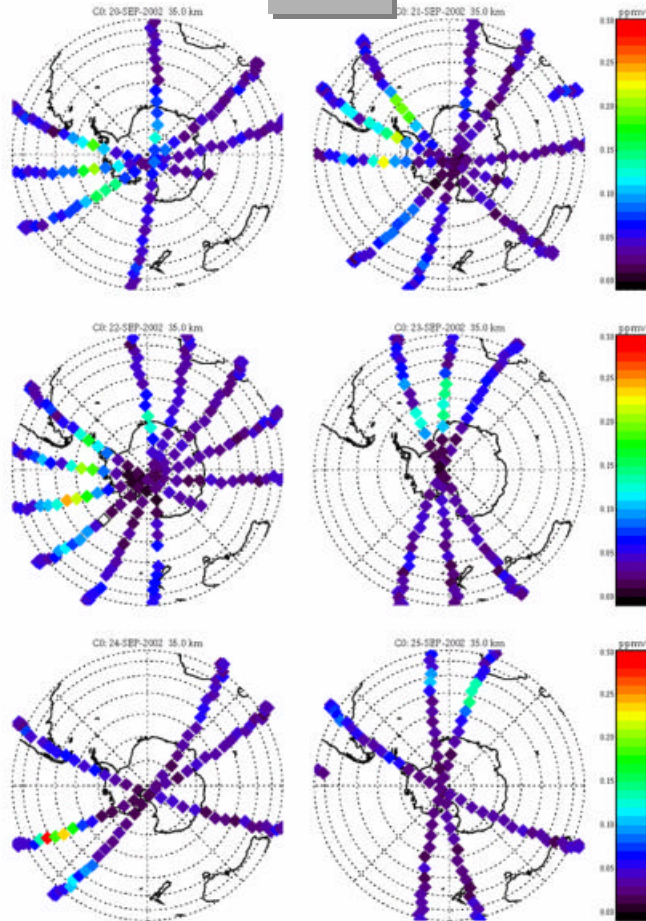
...distribution of  $\text{NO}_x$  above  
30 km is different to below  
(pronounced minima).  
Is this mesospheric  $\text{NO}_x$ ??



# NO<sub>x</sub> and CO in S Hemisphere at 20-26/9/2002

CO

NO<sub>x</sub>

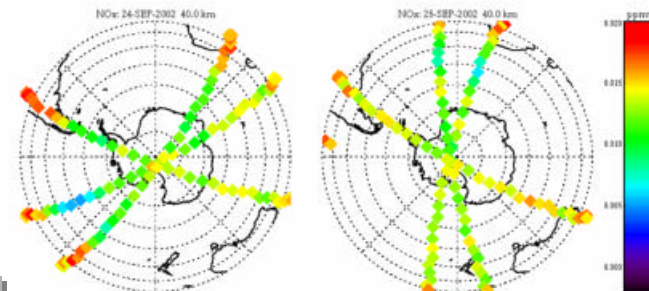
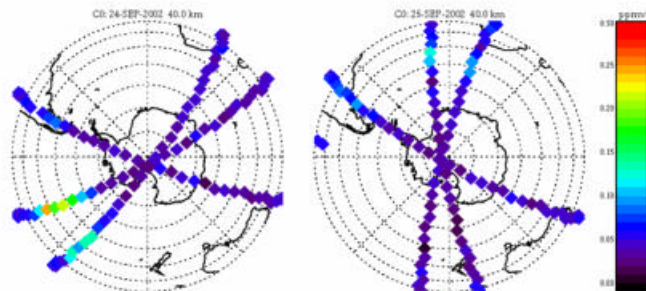
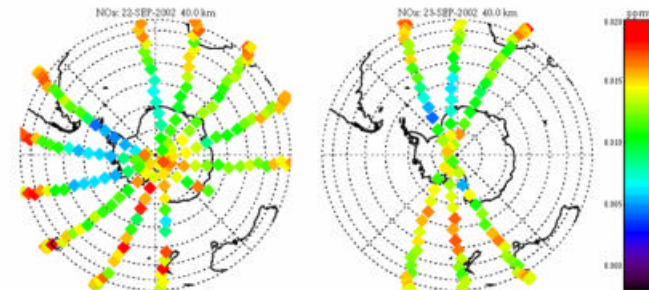
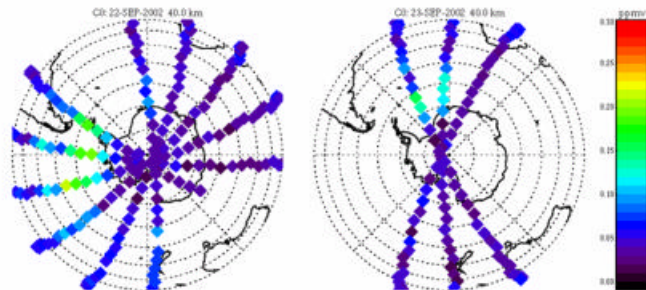
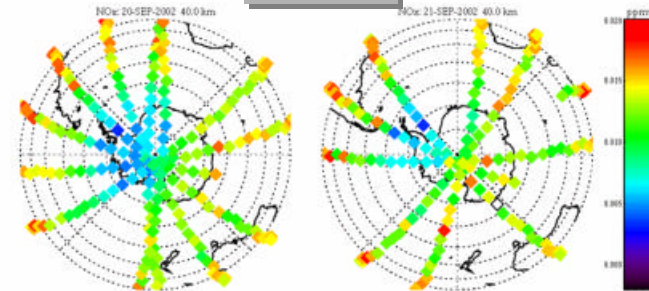
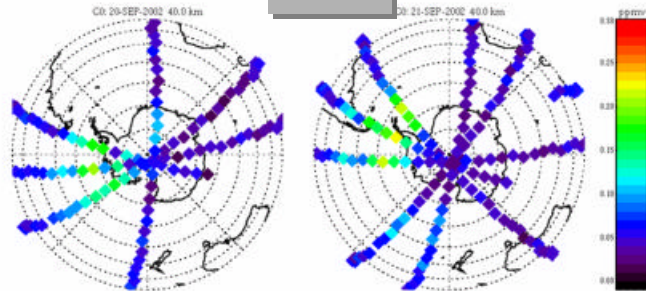


35 km

# No<sub>x</sub> and CO in S Hemisphere at 20-26/9/2002

CO

NO<sub>x</sub>

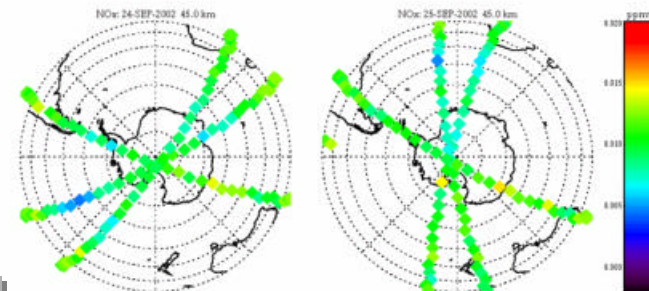
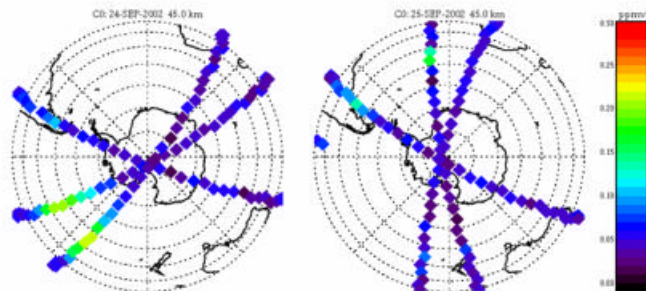
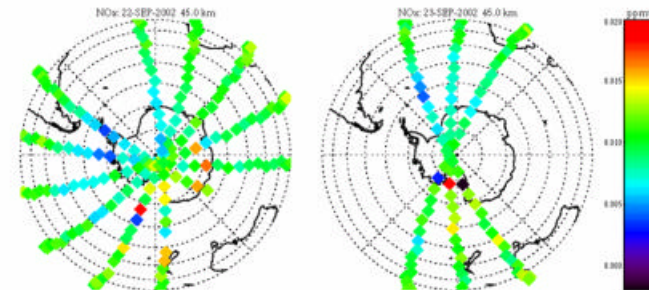
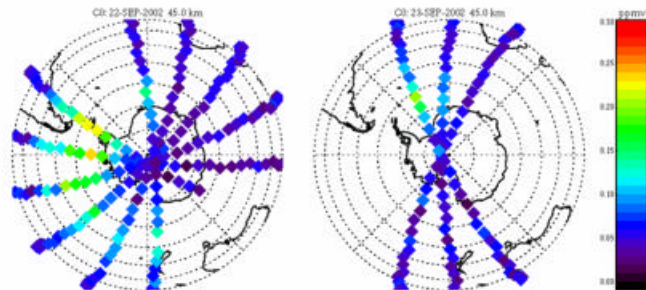
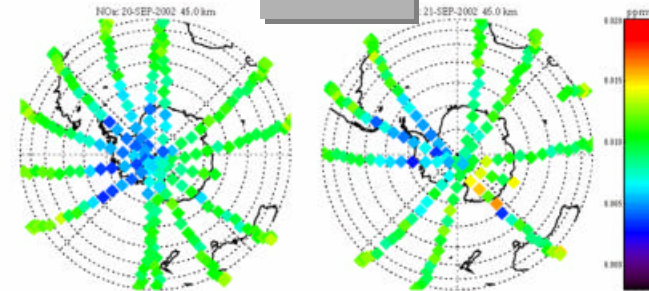
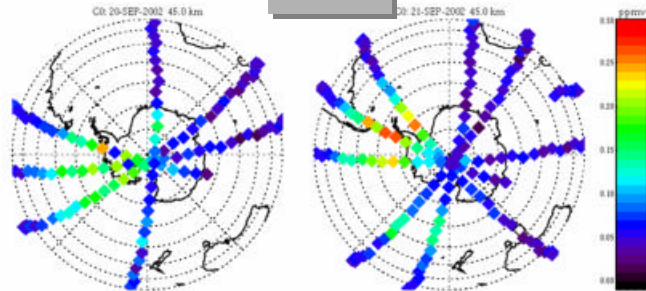


40 km

# No<sub>x</sub> and CO in S Hemisphere at 20-26/9/2002

CO

NO<sub>x</sub>



45 km



$\text{NO}_x$  vs. CO distributions over South pole give evidence for mesospheric  $\text{NO}_x$  in upper stratosphere with low vmr's in **September.**



## Conclusions

- non-LTE retrieval processor allows for NO, NO<sub>2</sub>, and CO retrieval with high accuracy and vertical resolution.
- Validation: good agreement of MIPAS/SPIRALE CO, MIPAS/MIPAS-B NO<sub>2</sub>, and MIPAS/HALOE NO<sub>x</sub>.
- Measurements of NO, NO<sub>2</sub>, and CO in July - October 2002:
  - Ⓡ Dynamical interaction of stratosphere and mesosphere.
  - Ⓡ Change of meridional circulation coincides with vortex split !?
  - Ⓡ Mesospheric air in stratosphere over South pole.
  - Ⓡ HIGH NO<sub>x</sub> in upper stratosphere during polar night BUT
  - Ⓡ LOW NO<sub>x</sub> in upper stratosphere over South pole in Sept.