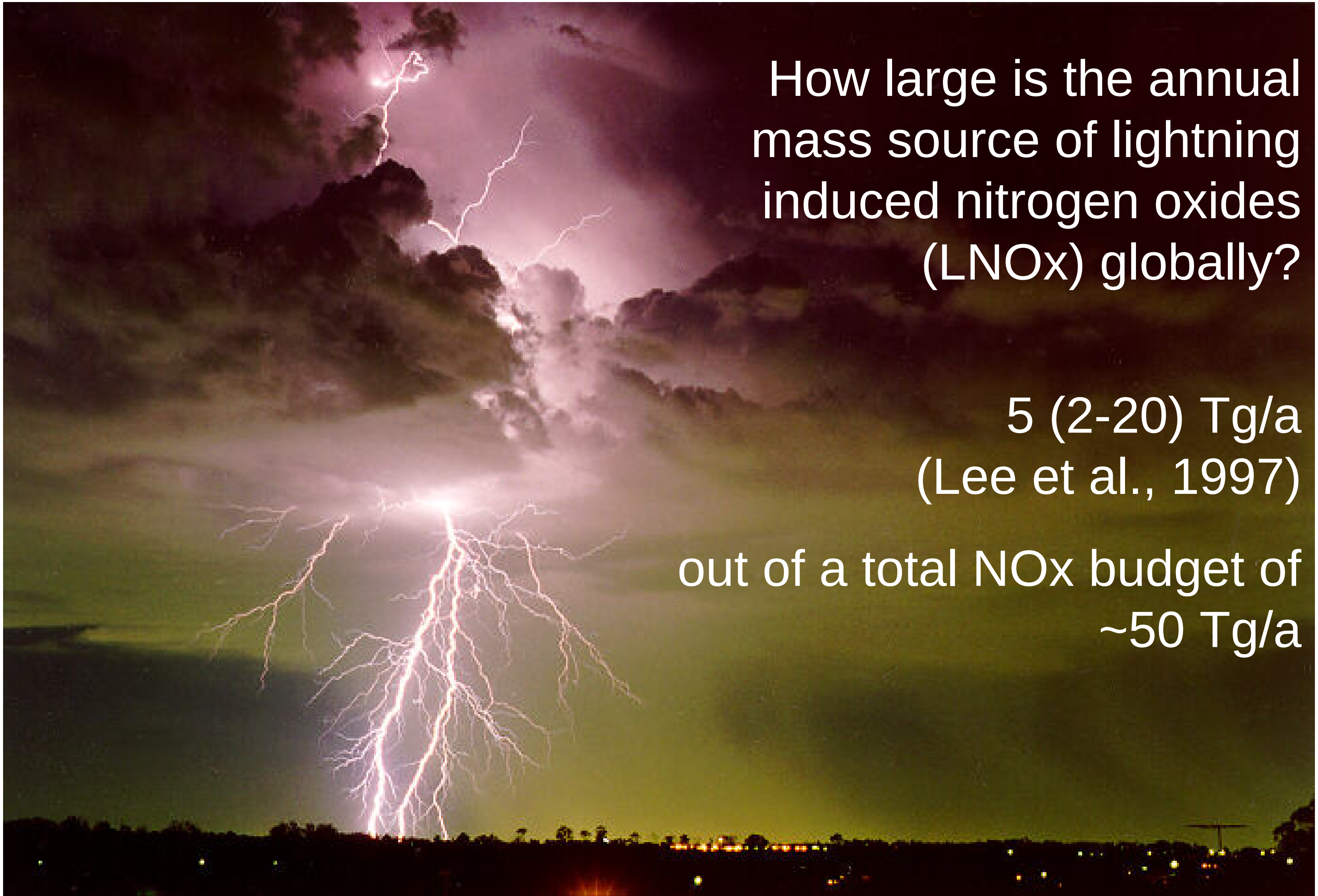
Towards a robust estimate of the global lightning nitrogen oxides source using in situ and remote sensing data and model results

Ulrich Schumann, Christian Kurz,
Hans Schlager, Heidi Huntrieser, Arthur Schady,
Louisa Emmons, Lorenzo Labrador, Ernst Meijer,
Alexej Ulanovsky, Silvia Viciani
and the TROCCINOX team

and Andreas Richter



How large is the annual mass source of lightning induced nitrogen oxides (LNOx) globally?

5 (2-20) Tg/a
(Lee et al., 1997)

out of a total NOx budget of ~50 Tg/a

LNOx influences
NOx concentration in tropical troposphere: up to 70%
Tropospheric O_3: up to 30 % in the tropics due to LNOx
Radiative forcing due to tropospheric O_3
OH concentration and lifetime of CO, CH_4, etc.
Radiative forcing due to CH_4
NOx concentration in upper mid-latitudinal troposphere
Relative importance of aviation NOx contribution
Climate change feedback (LNOx-O_3-RF-Temp.)
Acid rain (possibly 50% over the tropics)

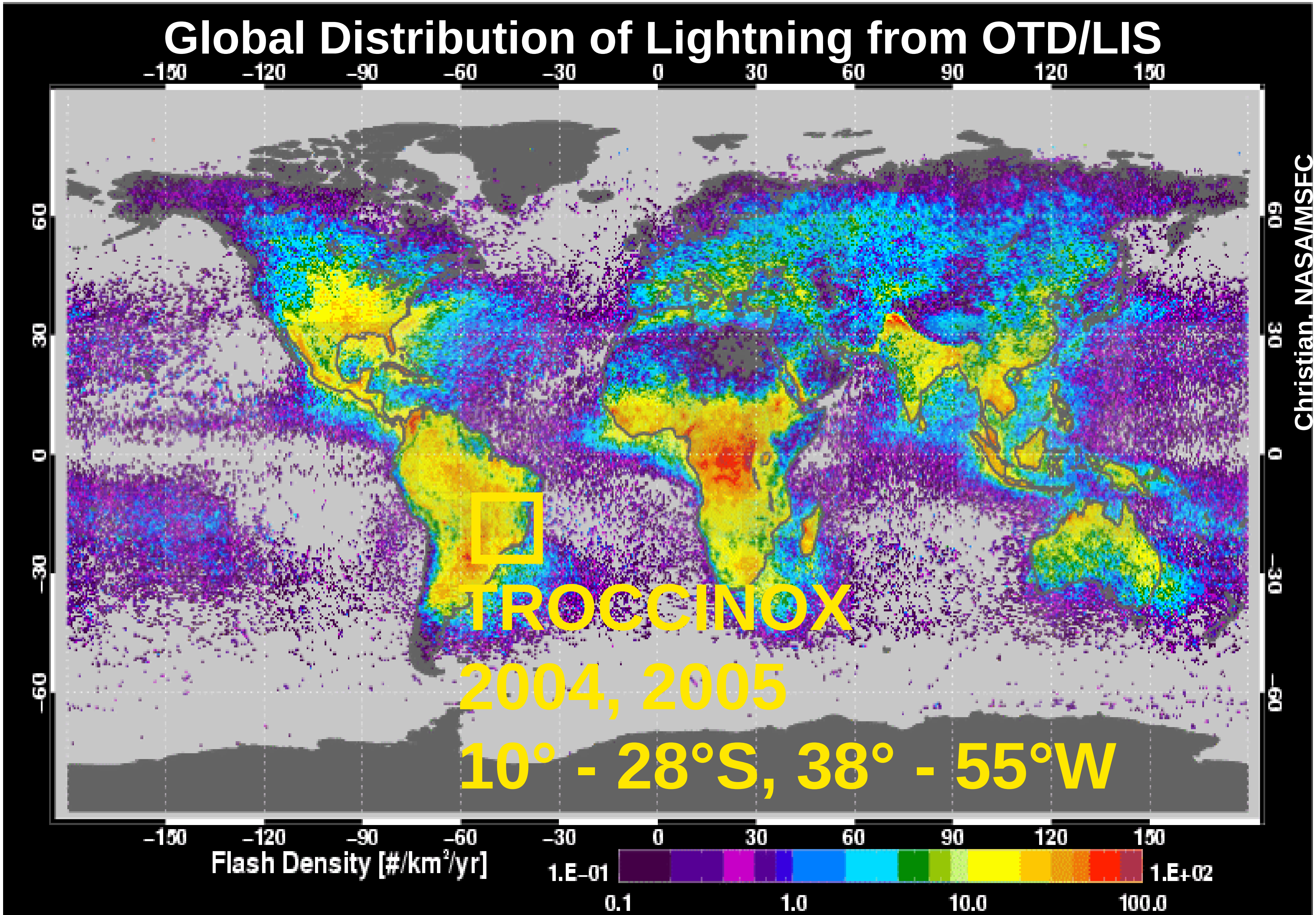
Required accuracy better than 50 % or < 3 Tg/a.

Methods to estimate the global LNOX production rate S in mass of fixed nitrogen induced per year

Method	Typical reference	Range S, Tg/a
Flash extrapolation: S = Production per flash $\times$ global flash rate	Lawrence et al., (1995)	2 (1-8)
Storm extrapolation: S = Production per thunderstorm $\times$ global thunderstorm rate	Huntrieser et al. (2002)	3 (1-20)
Model fit: S in global chemical transport model fitted to observations	Levy et al. (1996, 1999)	4 (2-6) (actually: 1.4-21; model sensitivity - OH, resolution?)

Methods to estimate the global LNOX production rate S in mass of fixed nitrogen induced per year

Method	Reference using GOME data	Range S, Tg/a
Flash extrapolation: S = Production per flash × global flash rate	Beirle et al. (2004, 2006) (Australia, Gulf of Mexico)	2.8 (0.8-14) 1.7 (0.6-4.7) (GOME + local flash rates)
Storm extrapolation: S = Production per thunderstorm × global thunderstorm rate	Huntrieser et al. (2002) In-situ	3 (1-20)
Model fit: S in global chemical transport model fitted to observations	Boersma et al. (2005), tropics	3.5 (1.1-6.4) (TM4, 2 versions, GOME-NO2)


Global Distribution of Lightning from OTD/LIS
Christian, NASA/MSFC
TROCCINOX
2004, 2005
10° - 28°S, 38° - 55°W
Flash Density [#/km²/yr]
1.E–01
1.E+02
0.1
1.0
10.0
100.0

Falcon, Geophysica and Bandeirante during TROCCINOX in Araçatuba, S.P., Brasilien, February 2005

Thank you to EU, UNESP/IPMET and DLR-FB

Data, TROCCINOX 2004 and 2005

Measurement Data

Falcon: CO, O_3, NO, NOy (Hans Schlager et al., DLR-IPA)

Geophysica:

NO, NOy (H. Schlager et al., SIOUX)

O_3 (A. Ulanovsky, CAO &
Fabrizio Ravegnani, CNR, et al. FOZAN)

CO (S. Viciani, P. Mazzinghi et al., INOA, CO-TDL

Model Data

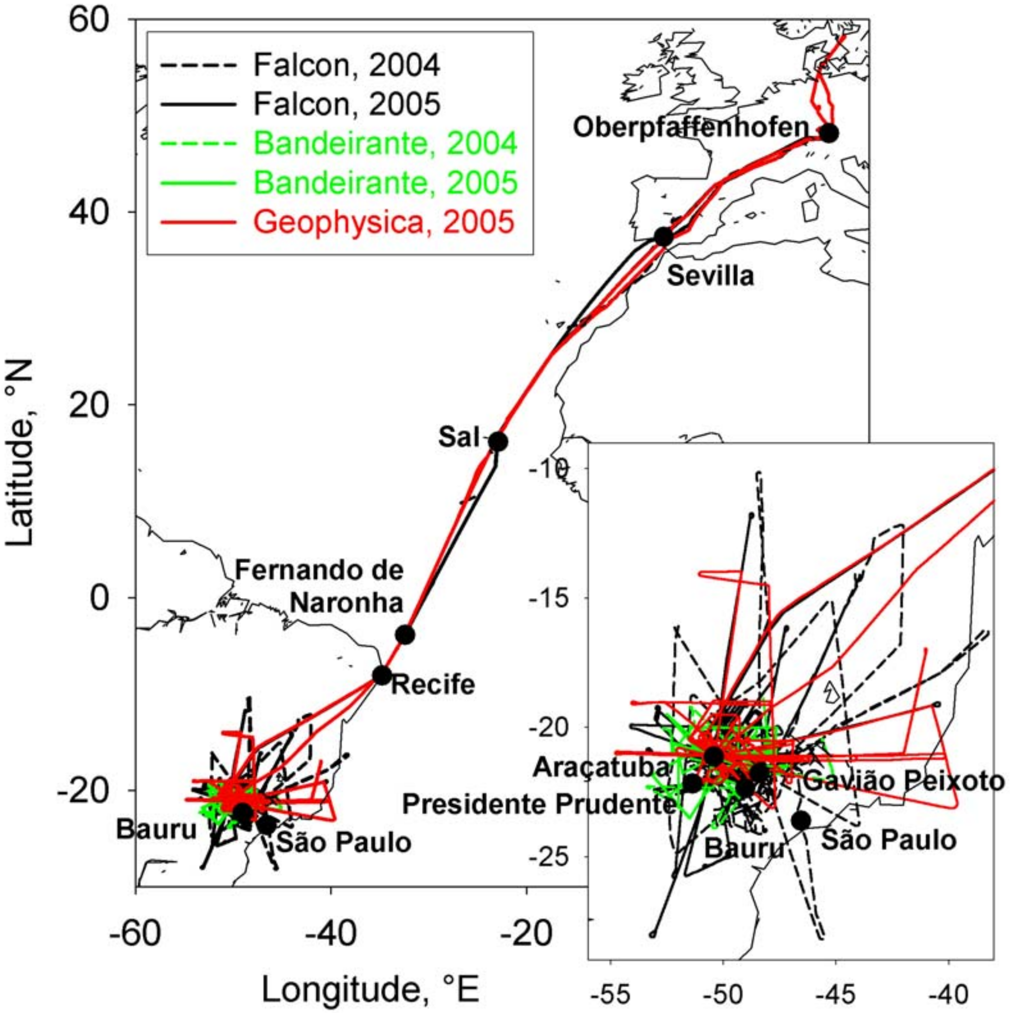
ECHAM5/MESSy (MPI-C + C. Kurz et al, DLR-IPA)

TM4 (Ernst Meijer, KNMI)

MOZART-4 (MZ4, Louisa Emmons, NCAR)

MPIC MATCH (L. Labrador, M. Lawrence, MPI-C)

Flights



Flight data used for this analysis (SH):

12 Falcon flights in 2004 (up to 12.8 km altitude)

11 Falcon 2005

10 Geophysica 2005 (up to 20 km altitude)

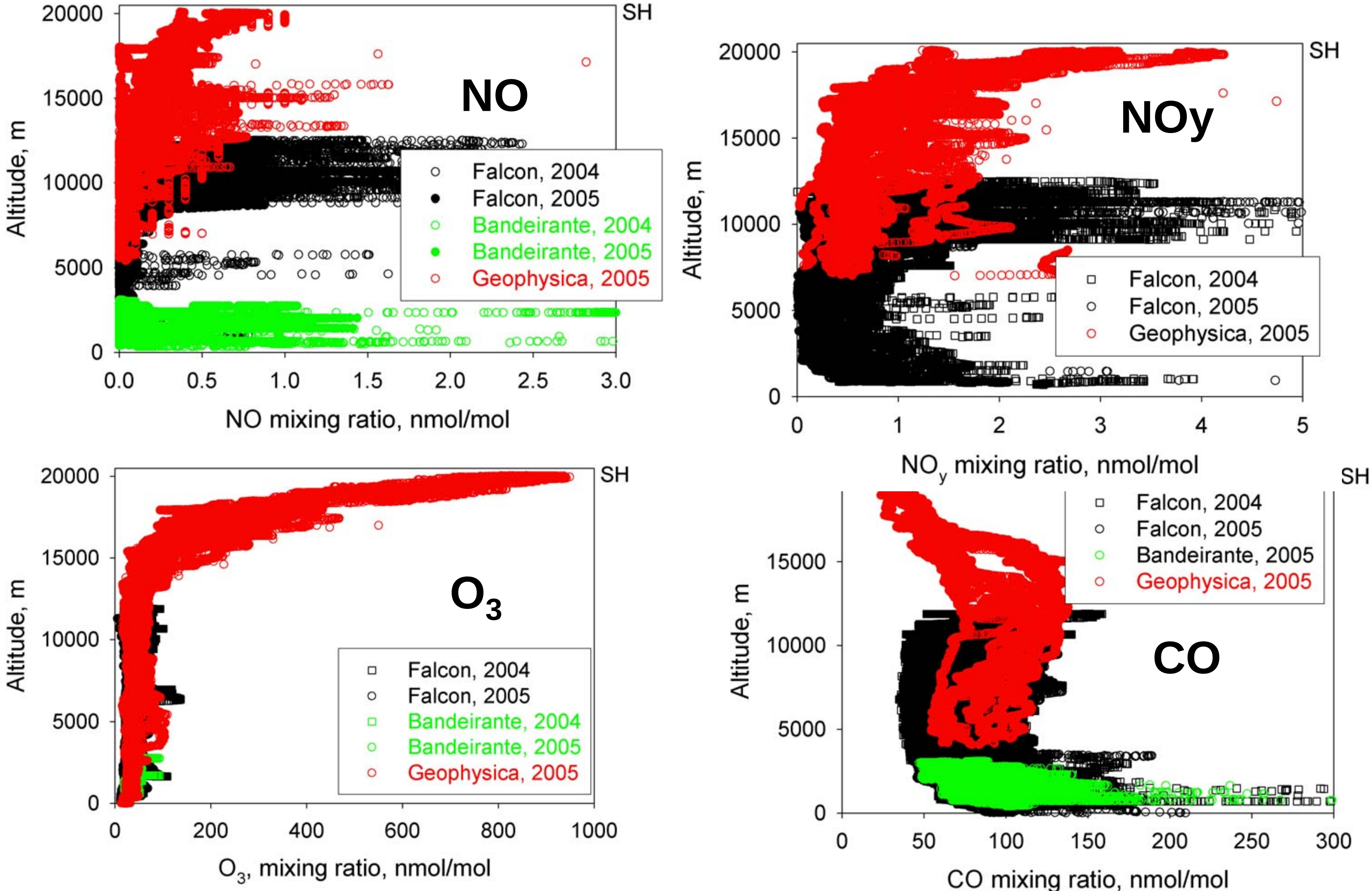
Flight types:

local near storms / nonlocal

with/without convection

in tropical/subtropical air masses

NO, NOy, O_3 and CO from TROCCINOX: LNOX signature



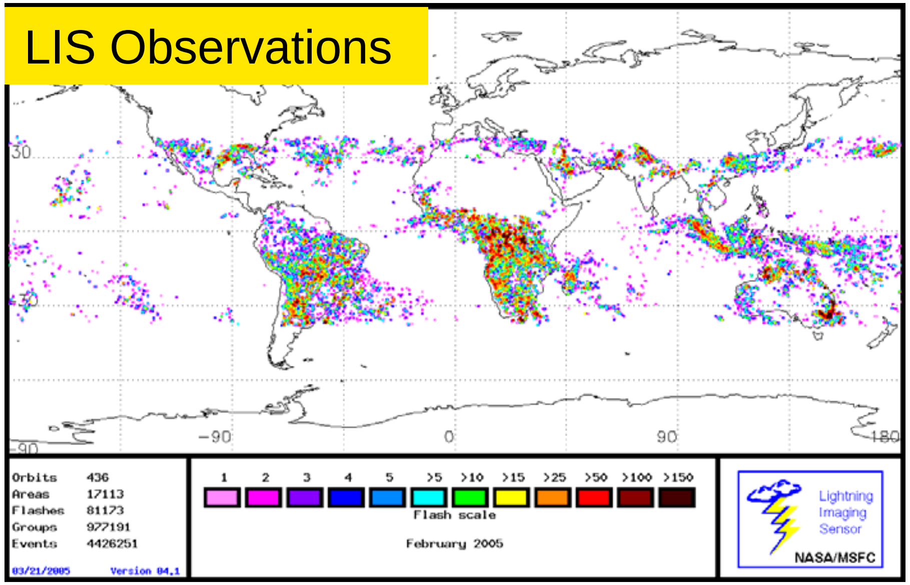
Models

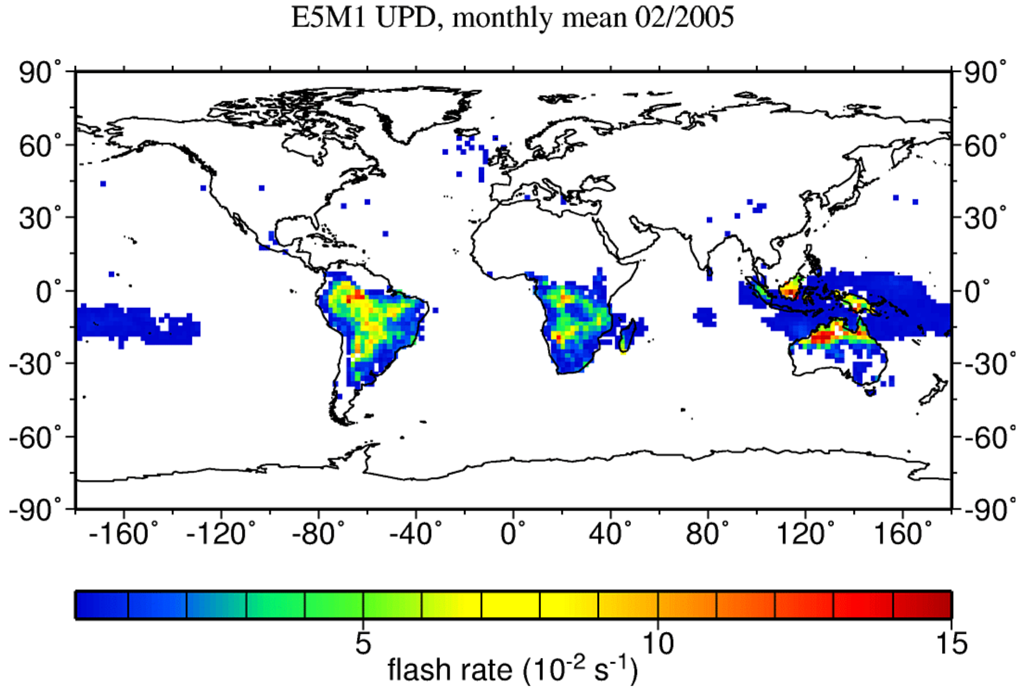
Model Name	Resolution	Meteorology, time intervals	Initialisation	LNOX model	Tg /a	Reference
<u>E5M1:</u> ECHAM5/ MESSy1	T63 (1.9° x 1.9°), 19 levels, Δt = 20 min	GCM nudged ECMWF, 6 h, 2005 only	1. Nov 2004	mass flux (UPD) (Grewe et al., 2001; Kurz et al., 2006)	0.75	Lelieveld et al. , Kurz et al. (2006)
<u>TM4</u>	3° x 2°, 31 levels, Δt = 30 min	CTM ECMWF, 6 h	1. Nov 2003	convective precipitation (Meijer et al., 2001)	0, 2, 5, 10	Meijer et al., (2000)
<u>MZ4</u>: MOZART-4	T42 (2.8°), 28 levels, Δt = 20 min	CTM NCEP reanalysis, 6 h	1. Nov 2003	Cloud height (Price et al., 1997; Pickering et al., 1998)	2, 5, 10	Horowitz et al., (2003), Emmons et al., in prep.

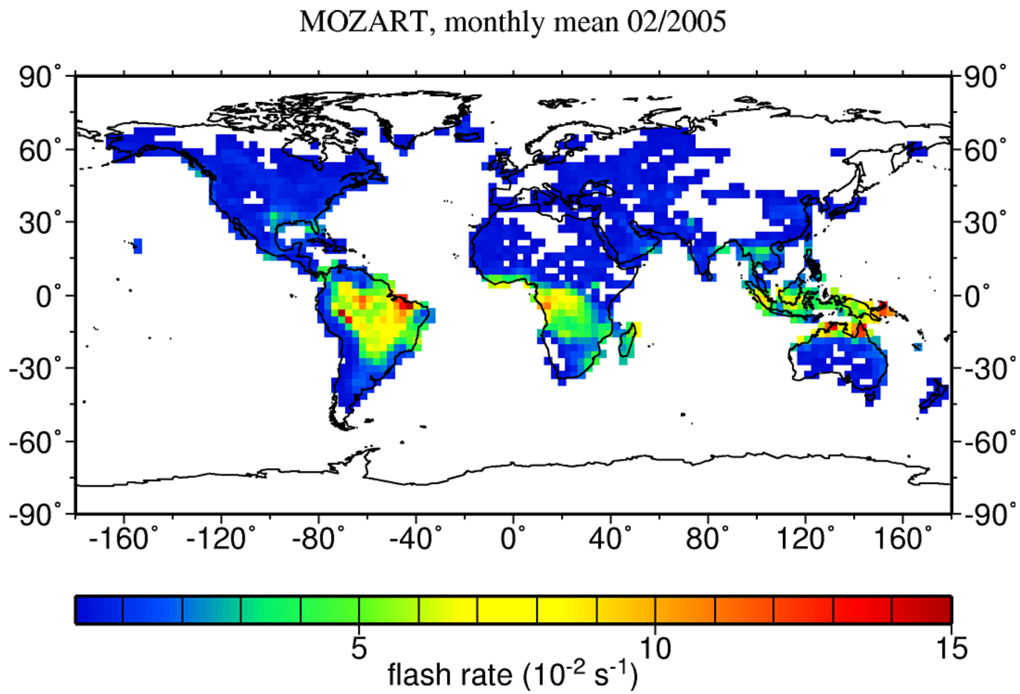
+ MPIC-<u>MATCH</u> (M. Lawrence & L. Labrador) for LNOX with S = 2 Tg/a
+ preliminary version <u>E5M0</u> (C. Kurz)

Modelled and observed Lightning flash frequency

E5M1: Updraft velocity -> (Grewe et al., 2001), same model for land & ocean







MZ4: Cloud top height -> (Price et al., 1997), different for land & ocean

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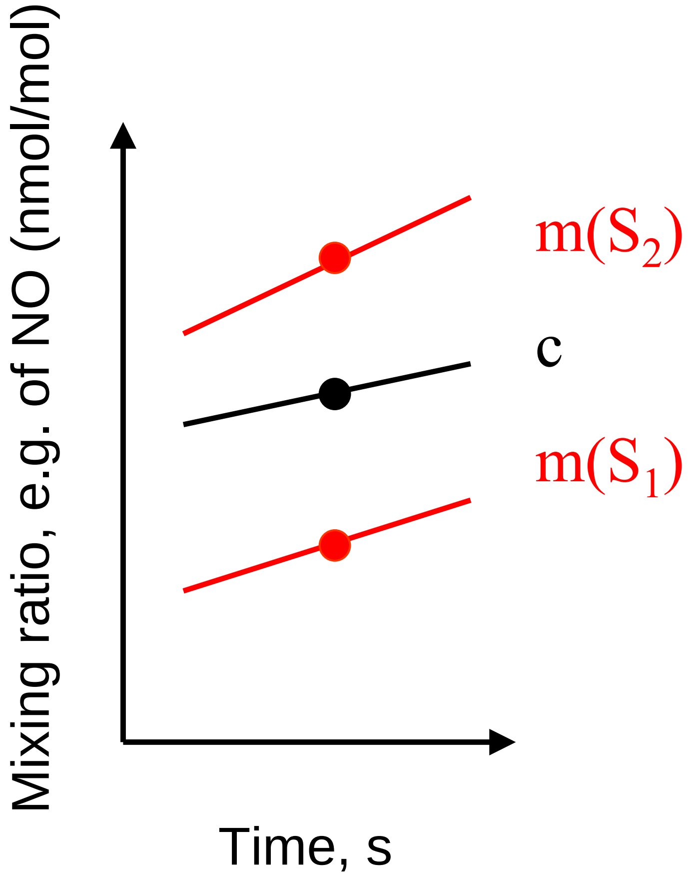
Determination of the global LNOx source S (Tg/a), direct for one observation



$$S = S_1 + (S_2 - S_1) \times \frac{c - m(S_1)}{m(S_2) - m(S_1)}$$

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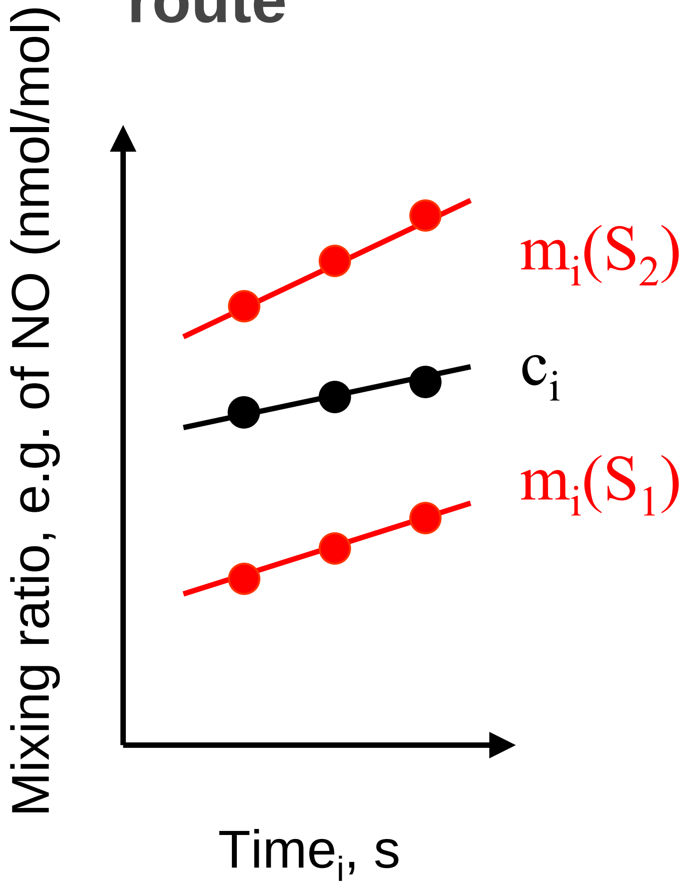
Determination of the global LNOx source S (Tg/a), by least square fit for many observations along flight route



$$S = S_1 + (S_2 - S_1) \times \frac{c - m(S_1)}{m(S_2) - m(S_1)}$$

$$\sigma^2(S) = \sum w_i^2 \, [c_i - m_i(S)]^2 = \min!$$

$$m_i(S) = m_i(S_1) + b_i\,(S - S1),$$

$$b_i = \partial m_i / \partial S \approx [m_i(S_2) - m_i(S_1)] / (S_2 - S_1)$$

$$S = S_1 + [\sum (b_i\, w_i)^2]^{-1} \sum b_i\, w_i^2 [c_i - m_i(S_1)]$$

w_i = suitable weight for observation i

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Random and Systematic Error Estimates:
$S \pm \Delta S_{ran} \pm \Delta S_{sys}$

$\varepsilon_i = c_i - m_i$, the differences between measured and modelled data, cause errors or uncertainties in S according to linear error propagation:

<u>random (ran)</u> error of the result S:

$\Delta S_{ran}{}^2 = (\sum (b_i w_i)^2)^{-2} \sum\{ b_i w^2{}_i \, \varepsilon_i \}^2$, valid for independent random errors.

(Decreases with $n^{½}$)

<u>systematic (sys)</u> error of the result S:

$\Delta S_{sys} \leq (\sum (b_i w_i)^2)^{-1} \sum | b_i w^2{}_i \, \varepsilon_i |$, valid always.

(Minimum when $w_i = \delta_{il}$ selects the most accurate model/data combination l)

Selection of weights w_i to minimize ΔS_{sys}

The weights w_i are selected as constant versus time during a flight, but different for each flight/parameter (NO, O3, CO, NOy) combination i:

$$w_i^2 = 1/ [\sigma_{c,i}^2 \times \Delta S_{sys,\ i}/\max(0, S_i)]$$

$\sigma_{c,i}$ = (error-) variance of measured data,

$\Delta S_{sys,\ i}/S_i$= relative accuracy for individual set of data

Sensitivity of relative error $\Delta S_{sys}/S$ on data c_i and model results m_i (S)

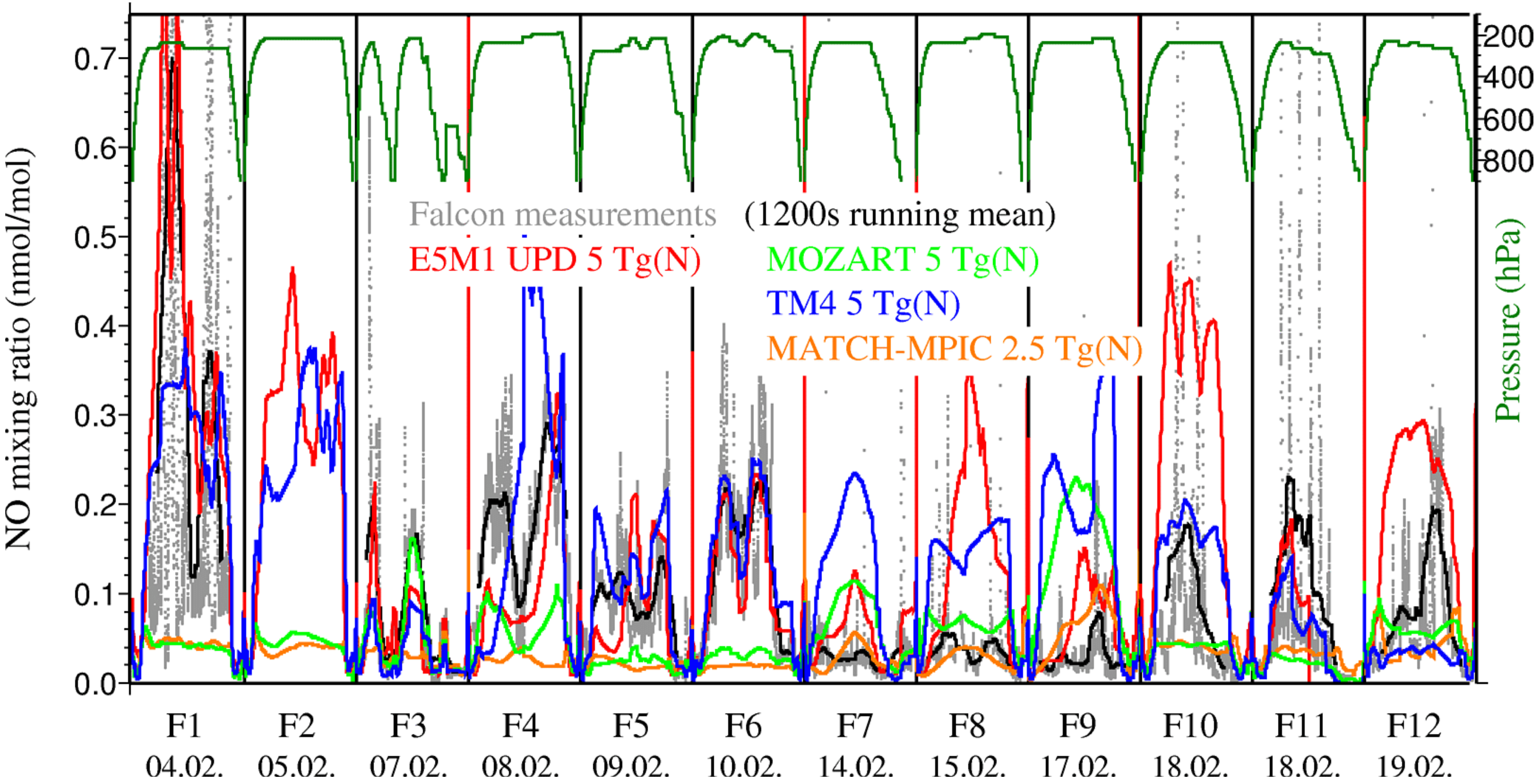
$$\frac{\Delta S_{sys}}{S} = \frac{\sum \left| b_i\, w^2_i[c_i-m_i(S)] \right|}{\sum \left| b_i w^2_i\, (c_i-m_i(0)) \right|} \approx \frac{\sum \left| c_i-m_i(S) \right|}{\sum \left| (m_i(S) - m_i(0)) \right|}$$

Hence, S most accurate for

small difference c-m between data and model

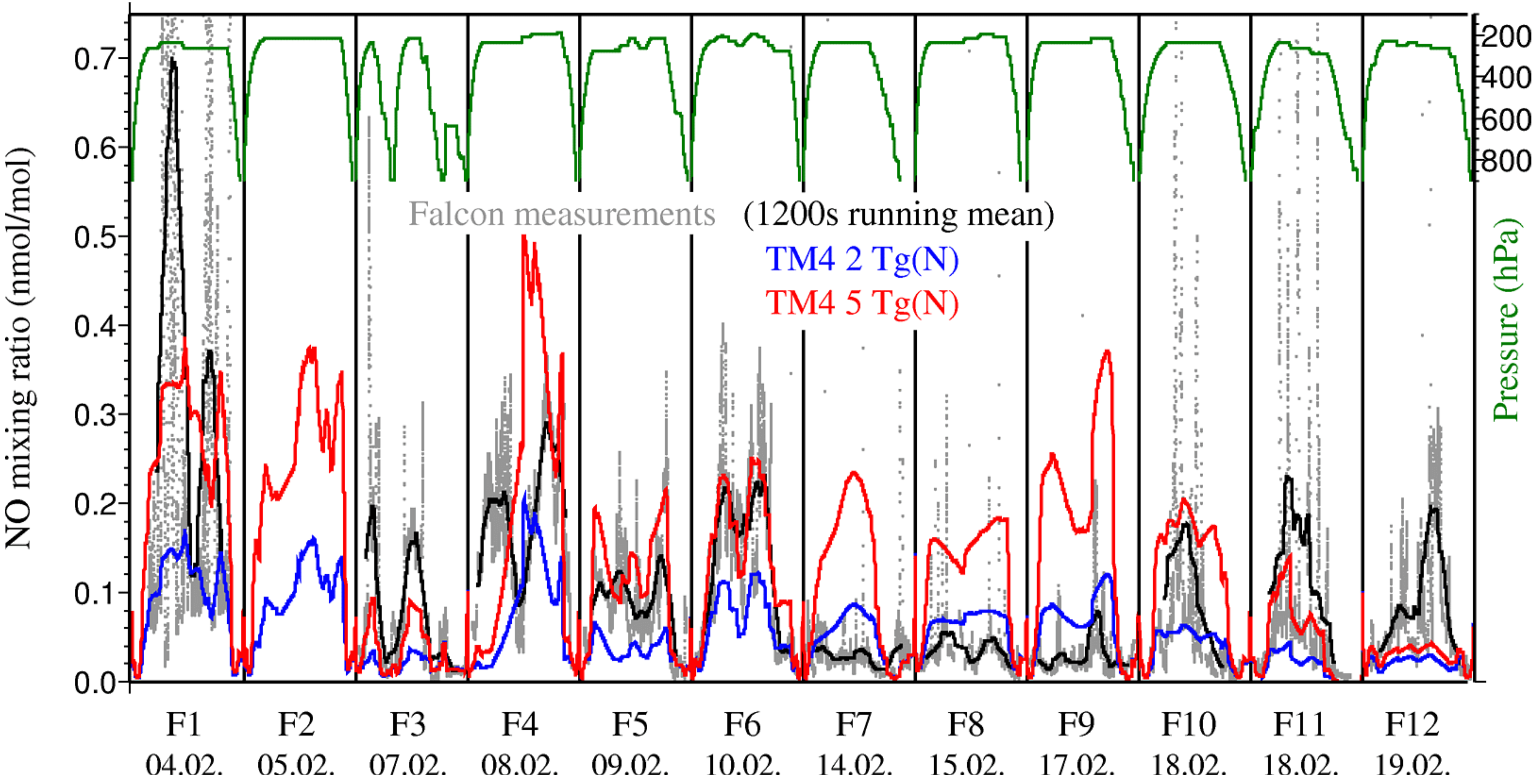
and large sensitivity of m to LNOX .

NO Falcon 2005, all models



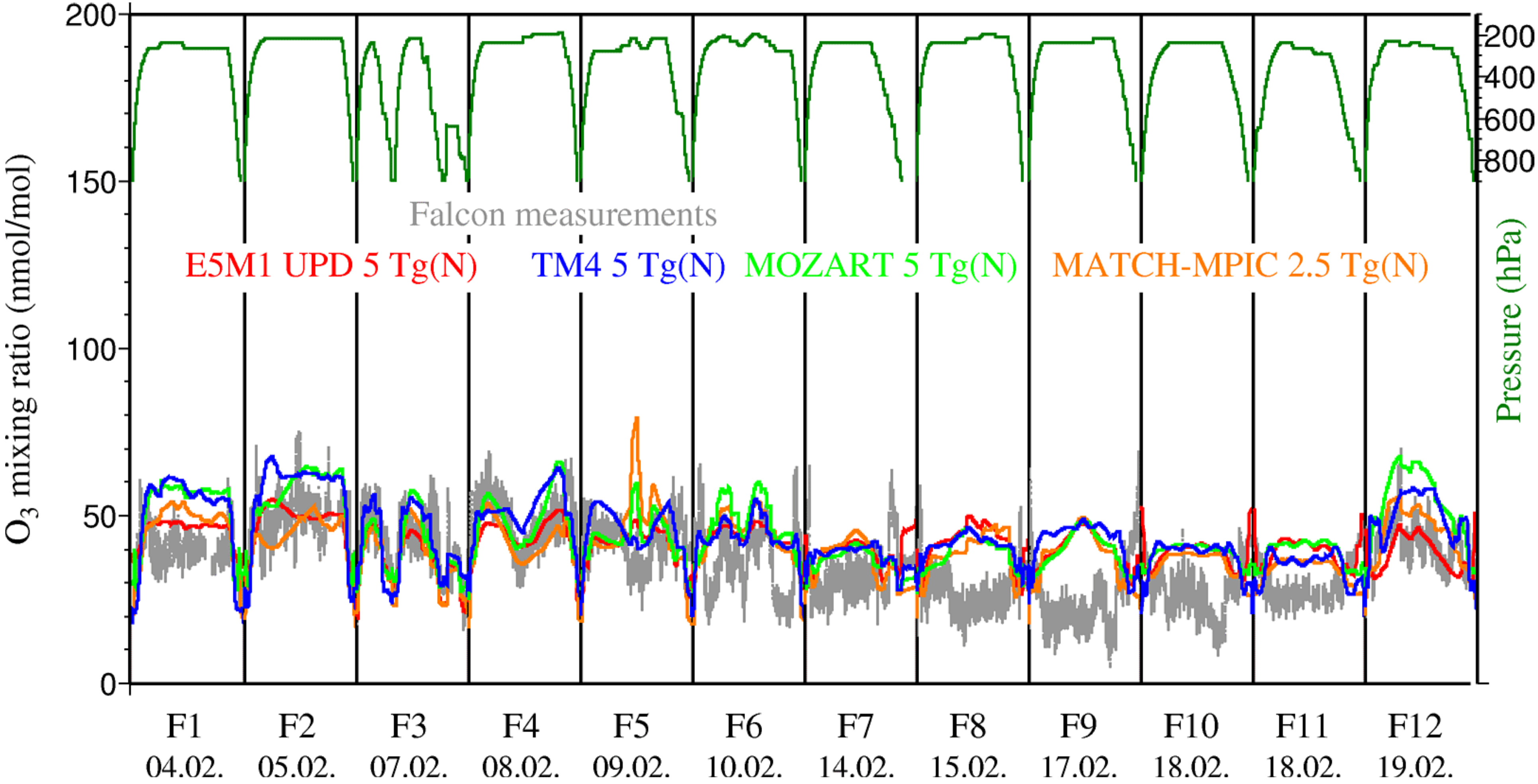
Highly variable conditions
E5M1 and TM4 show similar variability
CTH parameterisation as in MZ4 and MATCH shows too little variability

NO Falcon 2005, TM4 for 2 LNOx values



Precipitation based model better than CTH; less variability than for UPD-Model in E5M1

O_3 Falcon 2005, all models



All models similar, do not follow the minimum in the second part.

TM4 and MZ4 a bit high.

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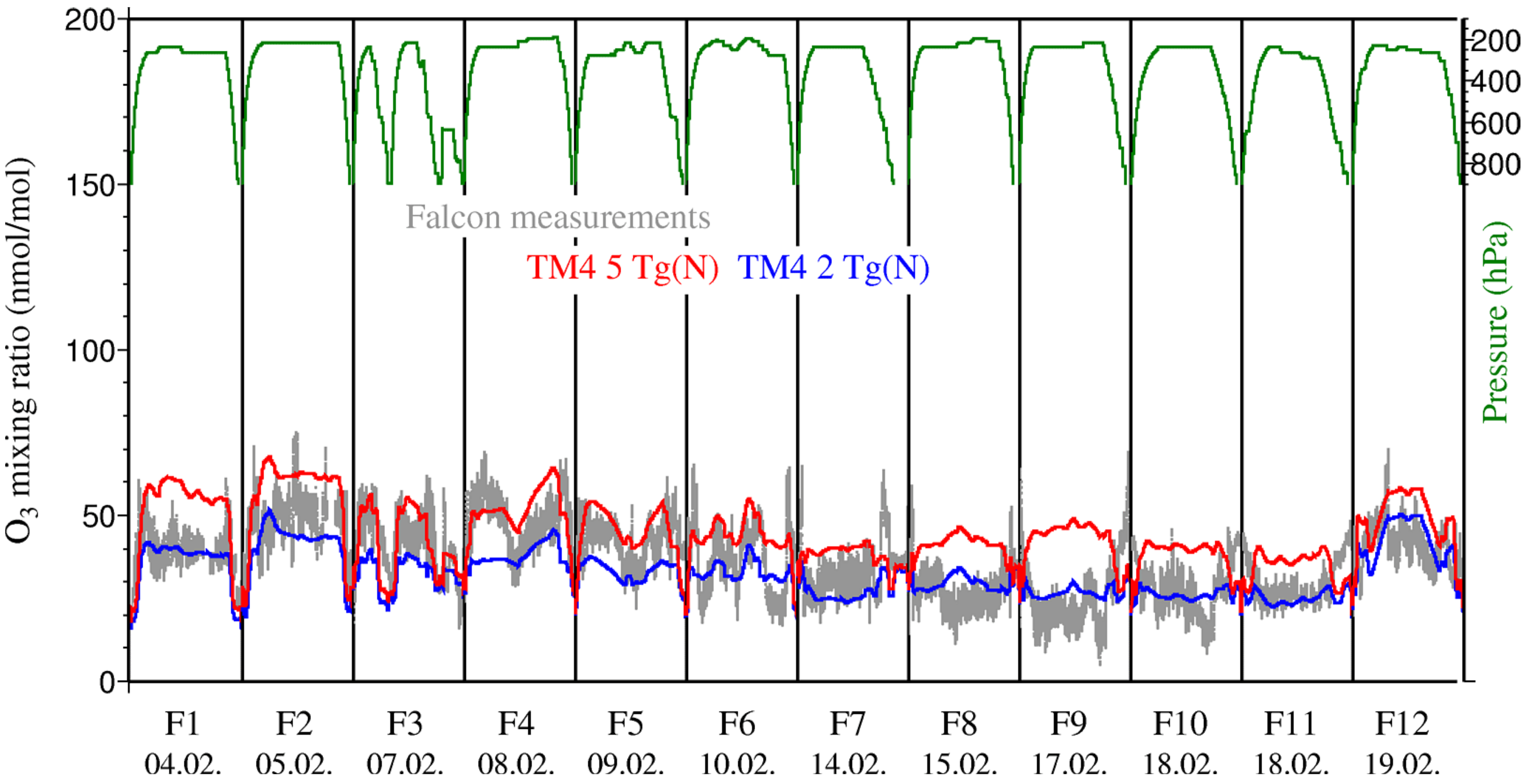
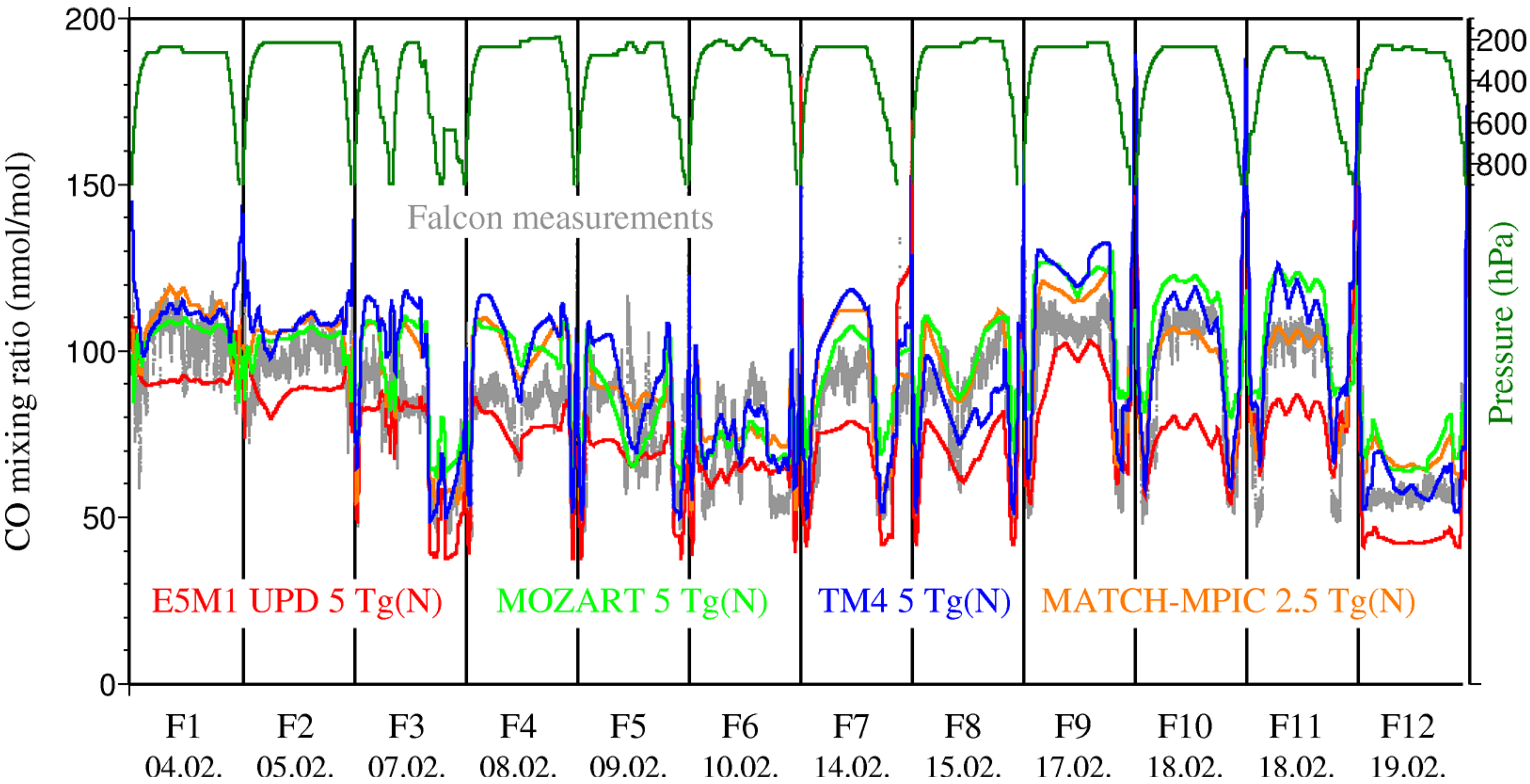
O_3 Falcon 2005, TM4 for 2 LNOx values

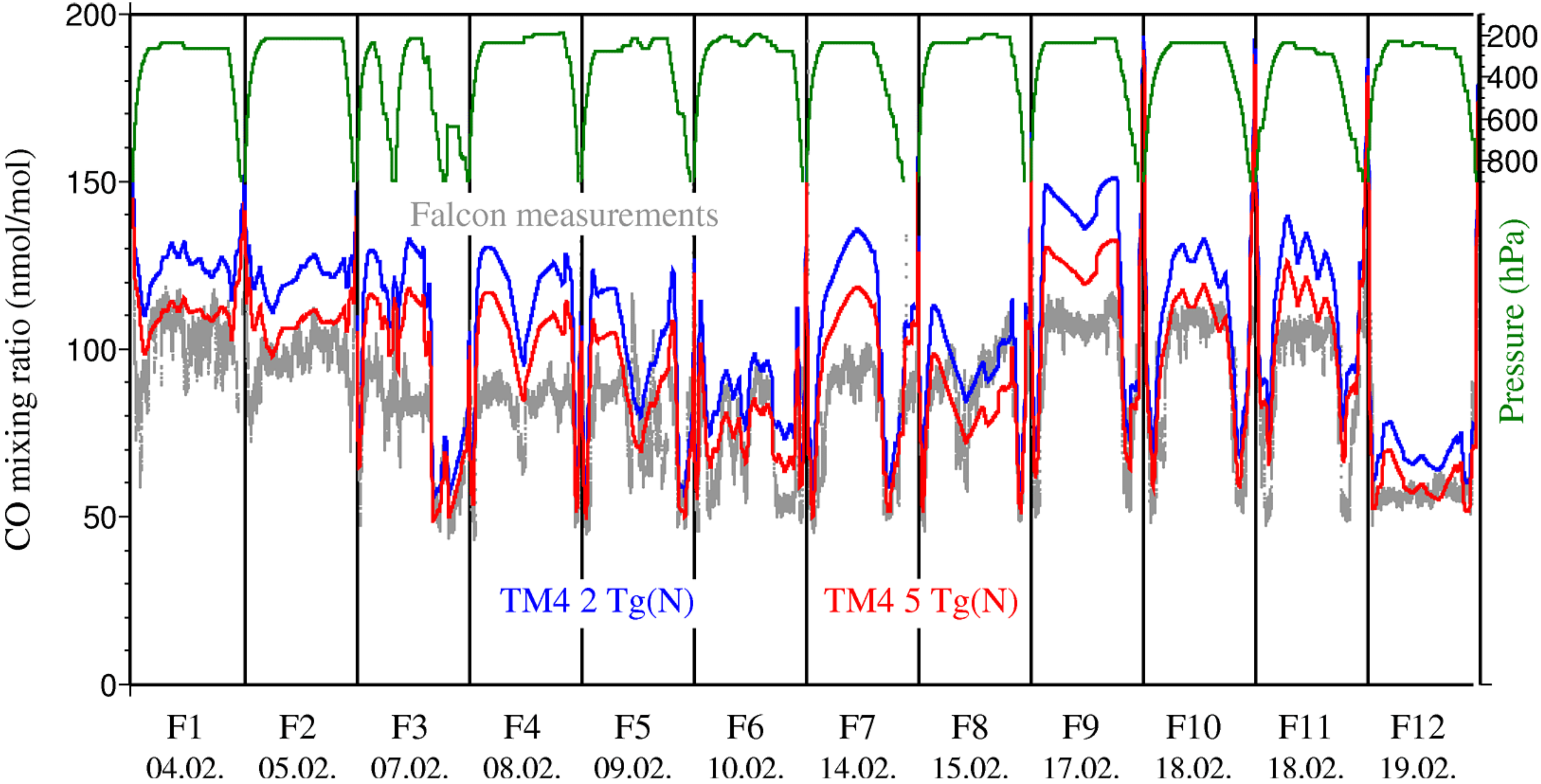


Large difference between 2 and 5 Tg/a, suitable for LNOX determination

CO Falcon 2005, all models



CO Falcon 2005, TM4 for 2 LNOx values



Large difference between 2 and 5 Tg/a, but not enclosing data

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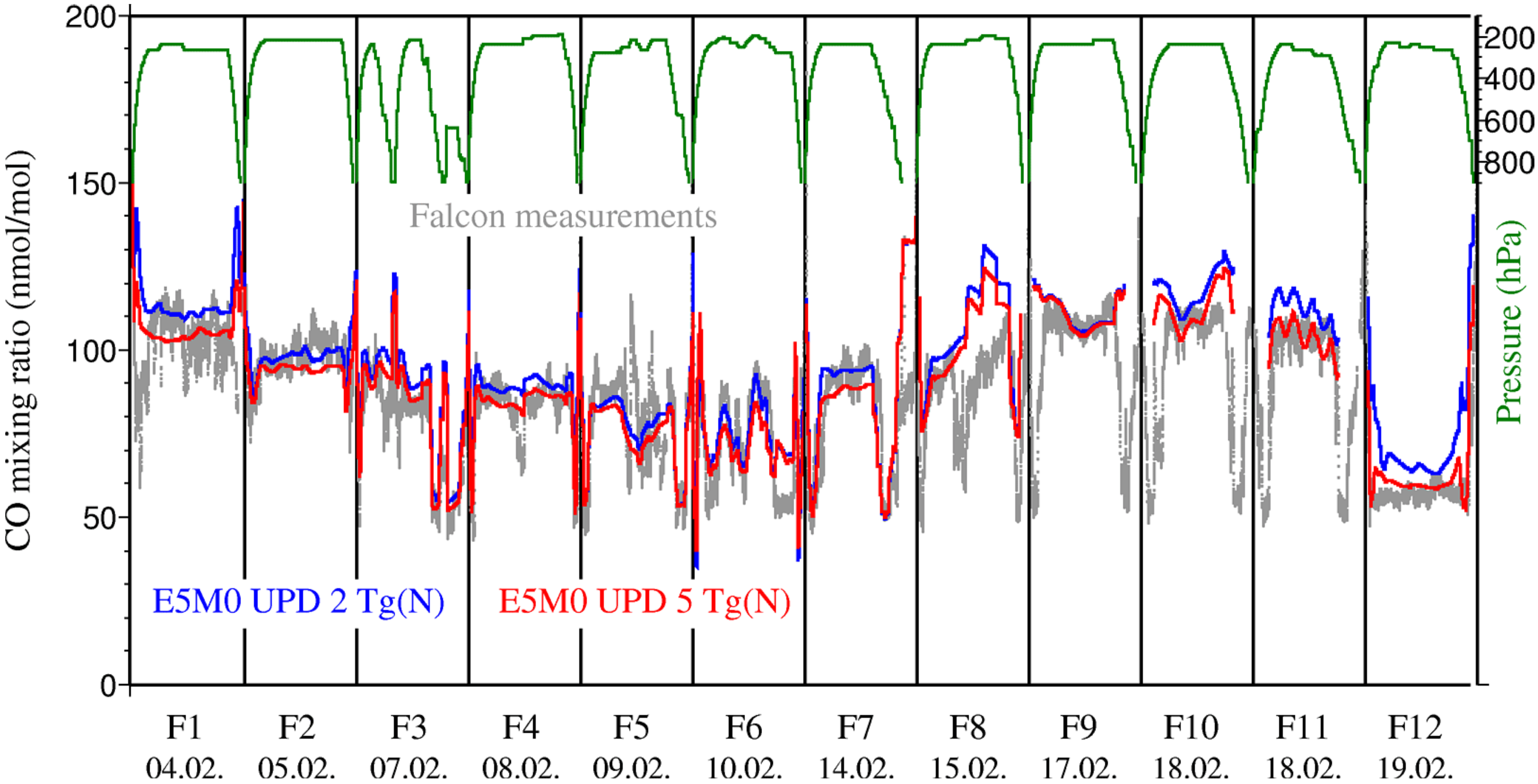
CO Falcon 2005, E5M0 for 2 LNOx values



Best fit with preliminary ECHAM5/MESSy version E5M0

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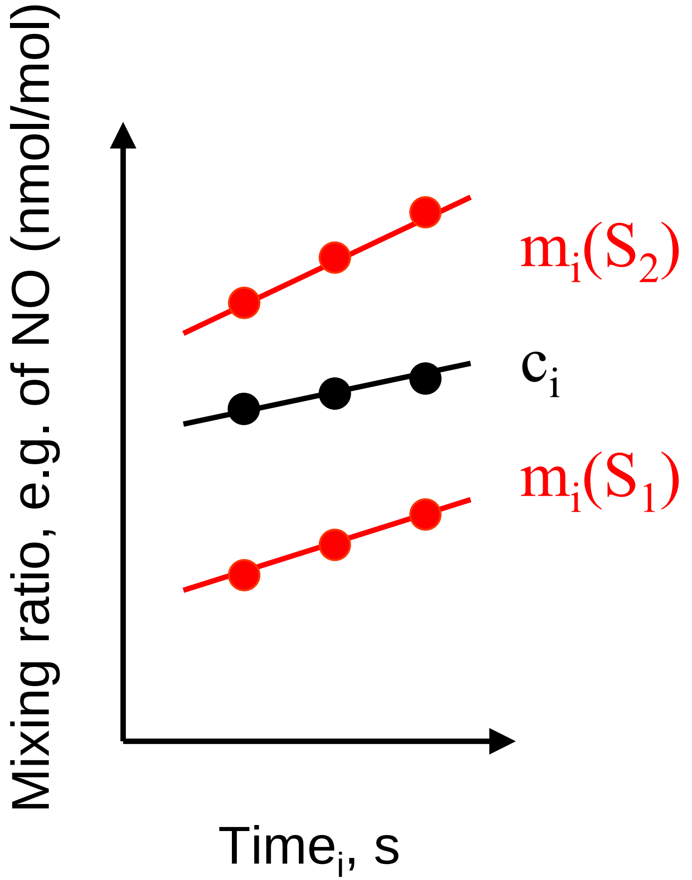
Determination of the global LNOx source S (Tg/a), by least square fit for many observations



$$\sigma^2(S) = \sum w^2_i\,[c_i - m_i(S)]^2 = \text{min!}$$

$$m_i(S) = m_i(S_1) + b_i\,(S - S1),$$

$$b_i = \partial m_i/\partial S \approx [m_i(S_2) - m_i(S_1)]/\,(S_2 - S_1)$$

$$S = S_1 + [\sum (b_i\, w_i)^2]^{-1} \sum b_i\, w^2_i [c_i - m_i(S_1)]$$

$\Delta S = ?$

$w_i = ?$

$\sigma^2(S)$ for various models?

Estimate S ±ΔS for various models, years and species

Flights	Species	TM4		E5M1		MZ4		E5M0	
	Tg/a	S	ΔS	S	ΔS	S	ΔS	S	ΔS
F05	NO	3.6	2.0	4.0	1.6	6.8	6.3	5.6	1.7
	NO+NOy	2.3	1.8	4.3	2.7	11.9	8.6		
	NO+O3	3.2	1.2	6.3	3.6	3.4	1.5	4.8	1.9
	NO+CO	11.4	2.5	6.2	1.0	11.7	4.3	6.6	2.8
F05+G05	NO	3.7	2.1	3.5	1.9	7.7	6.8	4.8	2.4
	NO+NOy	3.9	3.6	3.3	2.9	13.9	10.1		
	NO+O3	3.2	1.2	6.5	4.3	3.5	1.6	6.4	1.7
	NO+CO	10.1	3.9	6.2	1.0	11.5	4.4		
F04+F05+G05	NO	4.1	2.7	3.5	1.9	10.1	9.1	3.9	1.5
	NO+NOy	4.3	3.7	3.3	2.9	17.1	11.5		
	NO+O3	4.8	2.5	6.5	4.3	6.3	3.8	6.3	1.8
	NO+CO	10.3	4.2	6.2	1.0	15.0	6.5		
	all-CO	4.8	2.5	6.5	4.3	6.3	3.8	5.7	1.8

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Estimate S ±ΔS for various models, years and species

Flights	Species	TM4							
	Tg/a	S	ΔS						
F05	NO	3.6	2.0						
	NO+NOy								
	NO+O3								
	NO+CO								
F05+G05	NO								
	NO+NOy								
	NO+O3								
	NO+CO								
F04+F05+G05	NO								
	NO+NOy								
	NO+O3								
	NO+CO								
	all-CO								

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Estimate S ±ΔS for various models, years and species

Flights	Species	TM4								
	Tg/a	S	ΔS							
F05	NO	3.6	2.0							
	NO+NOy	2.3	1.8							
	NO+O3									
	NO+CO									
F05+G05	NO									
	NO+NOy									
	NO+O3									
	NO+CO									
F04+F05+G05	NO									
	NO+NOy									
	NO+O3									
	NO+CO									
	all-CO									

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Estimate S ±ΔS for various models, years and species

Flights	Species	TM4							
	Tg/a	S	ΔS						
F05	NO	3.6	2.0						
	NO+NOy	2.3	1.8						
	NO+O3	3.2	1.2						
	NO+CO								
F05+G05	NO								
	NO+NOy								
	NO+O3								
	NO+CO								
F04+F05+G05	NO								
	NO+NOy								
	NO+O3								
	NO+CO								
	all-CO								

Estimate S ±ΔS for various models, years and species

Flights	Species	TM4							
	Tg/a	S	ΔS						
F05	NO	3.6	2.0						
	NO+NOy	2.3	1.8						
	NO+O3	3.2	1.2						
	NO+CO	11.4	2.5						
F05+G05	NO								
	NO+NOy								
	NO+O3								
	NO+CO								
F04+F05+G05	NO								
	NO+NOy								
	NO+O3								
	NO+CO								
	all-CO								

Estimate S ±ΔS for various models, years and species

Flights	Species	TM4							
	Tg/a	S	ΔS						
F05	NO	**3.6**	**2.0**						
	NO+NOy	**2.3**	**1.8**						
	NO+O3	**3.2**	**1.2**						
	NO+CO	**11.4**	**2.5**						
F05+G05	NO	**3.7**	**2.1**						
	NO+NOy	3.9	3.6						
	NO+O3	**3.2**	**1.2**						
	NO+CO	10.1	3.9						
F04+F05+G05	NO	**4.1**	**2.7**						
	NO+NOy	4.3	3.7						
	NO+O3	**4.8**	**2.5**						
	NO+CO	10.3	4.2						
	all-CO	**4.8**	**2.5**						

Estimate S ±ΔS for various models, years and species

Flights	Species	TM4		E5M1		MZ4		E5M0	
	Tg/a	S	ΔS	S	ΔS	S	ΔS	S	ΔS
F05	NO	**3.6**	**2.0**	**4.0**	**1.6**	6.8	6.3	**5.6**	**1.7**
	NO+NOy	**2.3**	**1.8**	**4.3**	**2.7**	11.9	8.6		
	NO+O3	**3.2**	**1.2**	6.3	3.6	**3.4**	**1.5**	**4.8**	**1.9**
	NO+CO	**11.4**	**2.5**	**6.2**	**1.0**	11.7	4.3	**6.6**	**2.8**
F05+G05	NO	**3.7**	**2.1**	**3.5**	**1.9**	7.7	6.8	**4.8**	**2.4**
	NO+NOy	3.9	3.6	**3.3**	**2.9**	13.9	10.1		
	NO+O3	**3.2**	**1.2**	6.5	4.3	**3.5**	**1.6**	**6.4**	**1.7**
	NO+CO	10.1	3.9	**6.2**	**1.0**	11.5	4.4		
F04+F05+G05	NO	**4.1**	**2.7**	**3.5**	**1.9**	10.1	9.1	**3.9**	**1.5**
	NO+NOy	4.3	3.7	**3.3**	**2.9**	17.1	11.5		
	NO+O3	**4.8**	**2.5**	6.5	4.3	6.3	3.8	**6.3**	**1.8**
	NO+CO	10.3	4.2	**6.2**	**1.0**	15.0	6.5		
	all-CO	**4.8**	**2.5**	6.5	4.3	6.3	3.8	**5.7**	**1.8**

bold: ΔS < 3 Tg/a

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Estimate S ±ΔS for various models, years and species

Flights	Species	TM4		E5M1		MZ4		E5M0	
	Tg/a	S	ΔS	S	ΔS	S	ΔS	S	ΔS
F05	NO	**3.6**	**2.0**	**4.0**	**1.6**	6.8	6.3	5.6	1.7
	NO+NOy	**2.3**	**1.8**	**4.3**	**2.7**	11.9	8.6		
	NO+O3	**3.2**	**1.2**	6.3	3.6	**3.4**	**1.5**	4.8	1.9
	NO+CO	**11.4**	**2.5**	**6.2**	**1.0**	11.7	4.3	6.6	2.8
F05+G05	NO	**3.7**	**2.1**	**3.5**	**1.9**	7.7	6.8	4.8	2.4
	NO+NOy	3.9	3.6	**3.3**	**2.9**	13.9	10.1		

LNOx = 4.8 ±2.5 (2.3 to 7.3) Tg/a

from TROCCINOX & TM4 with NO, NOy, and O_3

Flights	Species	TM4 S	TM4 ΔS	E5M1 S	E5M1 ΔS	MZ4 S	MZ4 ΔS	E5M0 S	E5M0 ΔS
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
	NO+NOy	4.3	3.7	**3.3**	**2.9**	17.1	11.5		
	NO+O3	**4.8**	**2.5**	6.5	4.3	6.3	3.8	6.3	1.8
	NO+CO	10.3	4.2	**6.2**	**1.0**	15.0	6.5		
	all-CO	**4.8**	**2.5**	6.5	4.3	6.3	3.8	5.7	1.8

LNOx contribution to NO_2 column from TM4 for comparisons with SCIAMACHY

TM4 provides global 3d NO_2 field at 10:30 LT

for LNOx nitrogen mass source of 2 and 5 Tg/a

Total and tropospheric columns (subtraction of mean between 180 and 220°E)

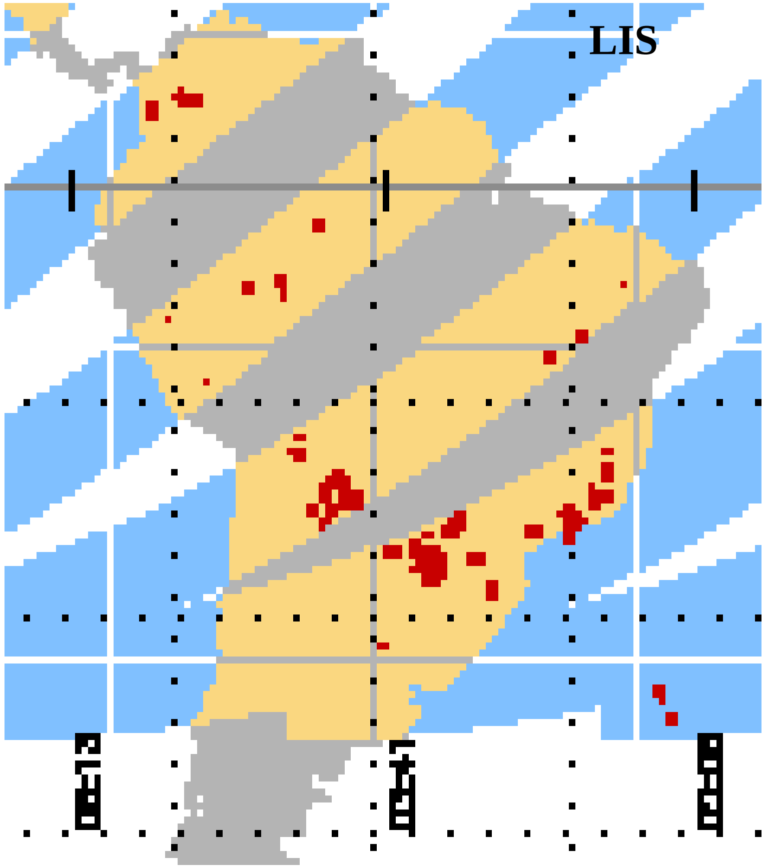
R = {m(5 Tg) - m(2 Tg)} / m(5Tg) shows LNOx contributions

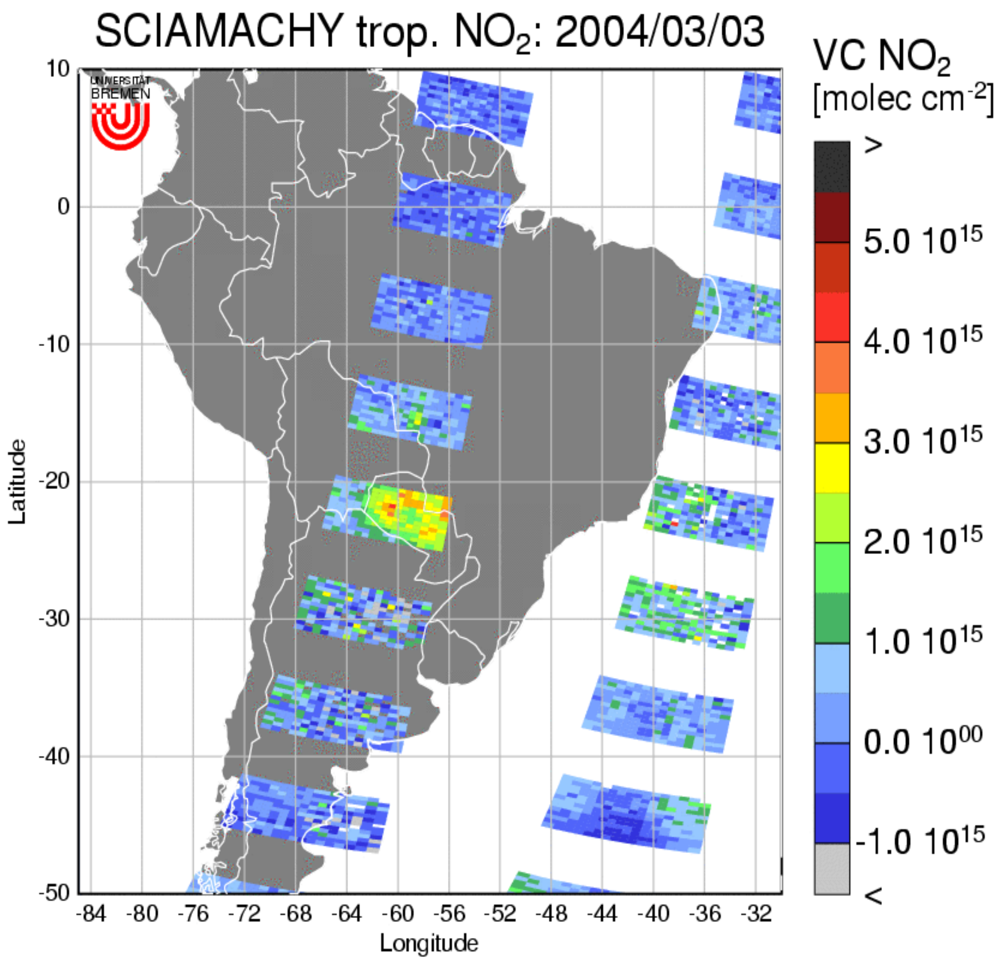
daily values and monthly mean values February/March 2004

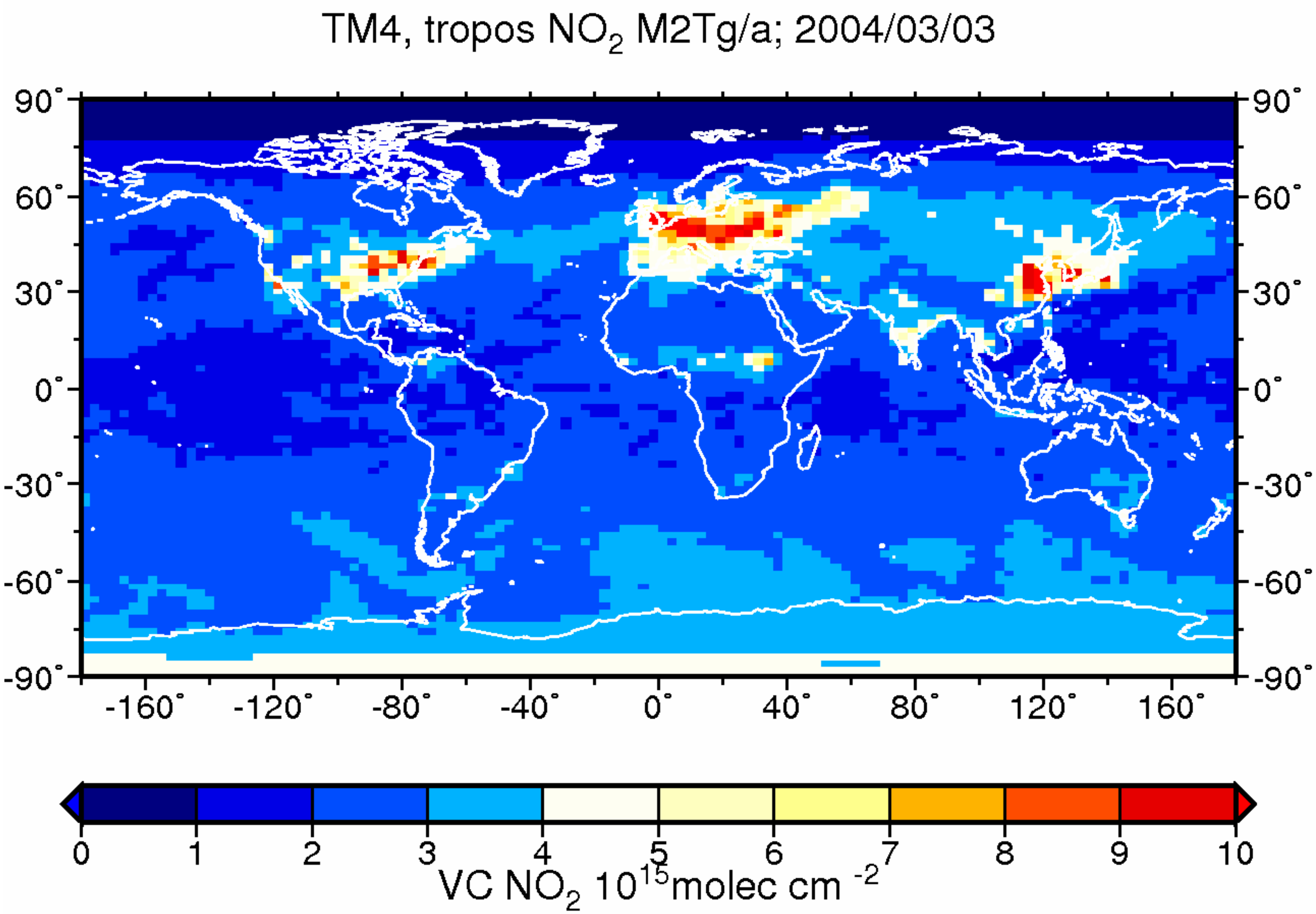
globally

South Atlantic Anomaly (SAA) and Cloud-dependent air mass factors (AMF) have still to be taken into account

Event with maximum LNOx during TROCCINOX: 03.03.2004







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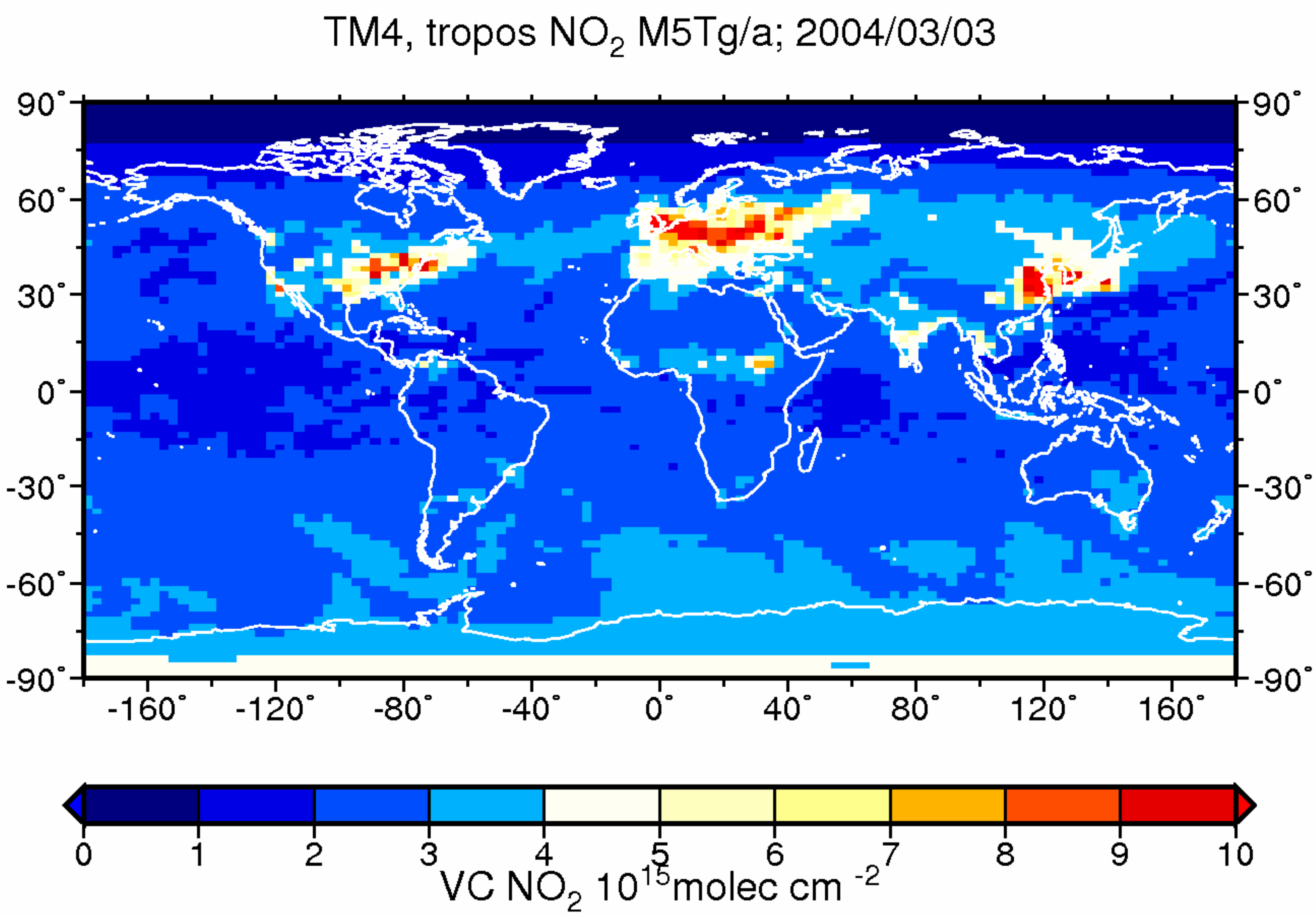

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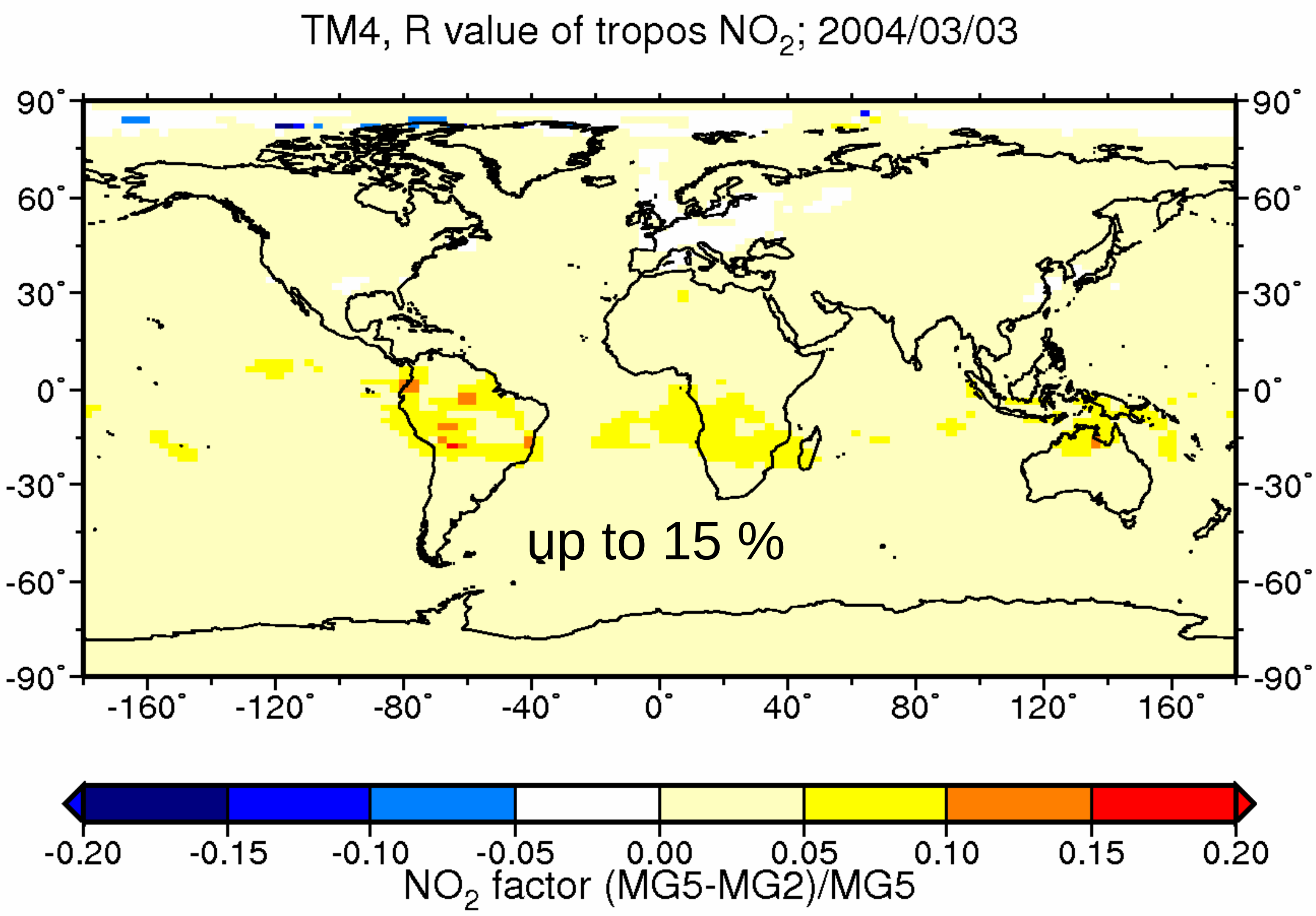

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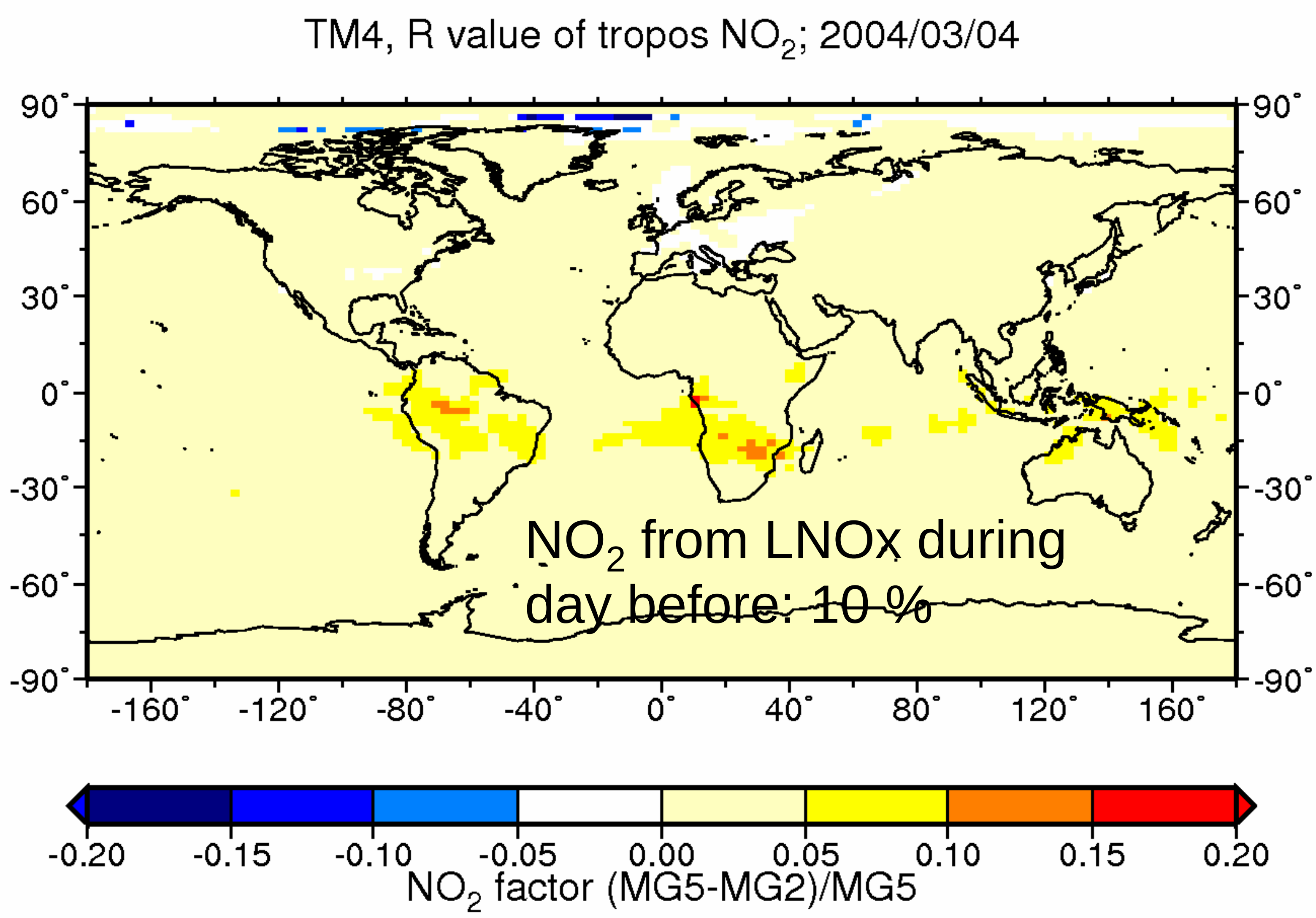
next day



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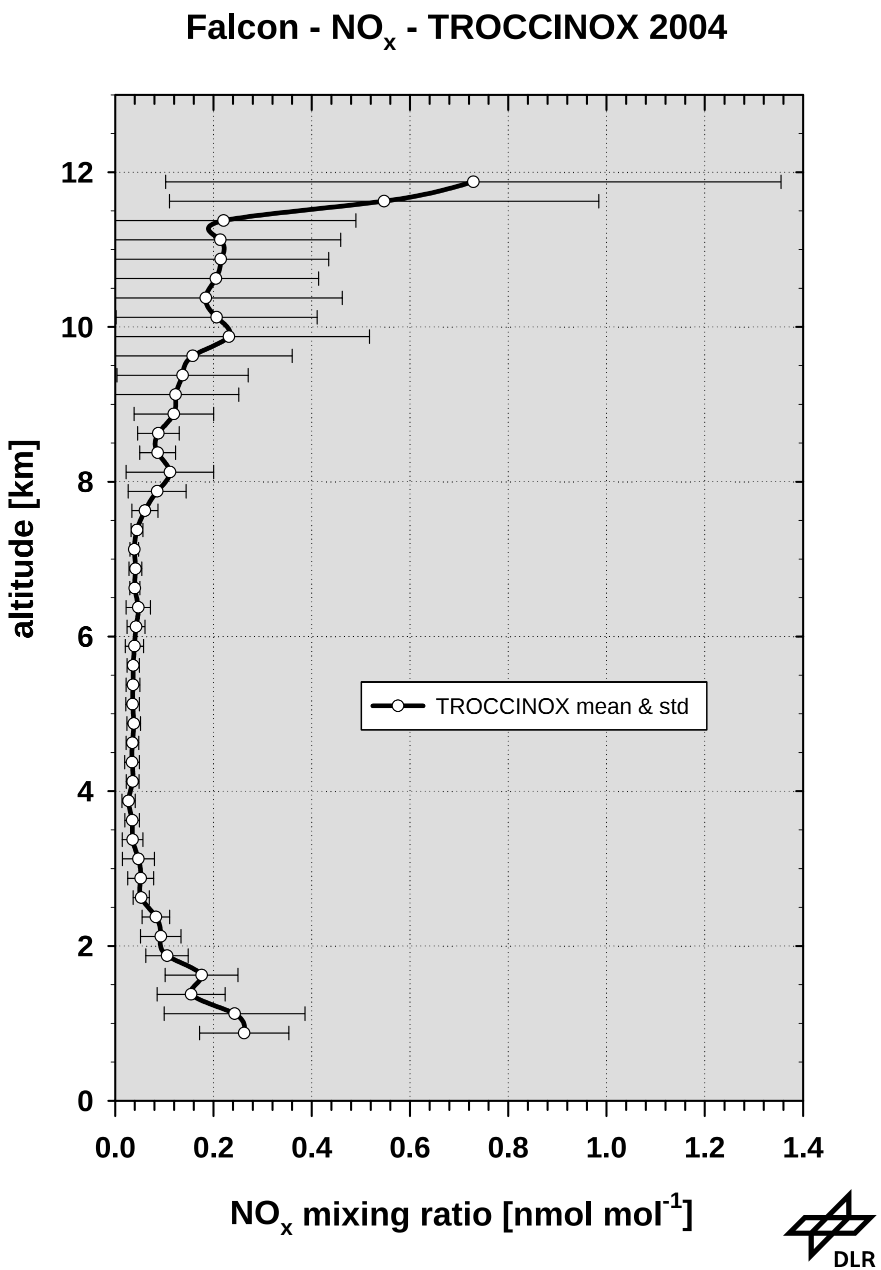
Falcon - NOx - TROCCINOX 2004
altitude [km]
NOx mixing ratio [nmol mol-1]
TROCCINOX mean & std
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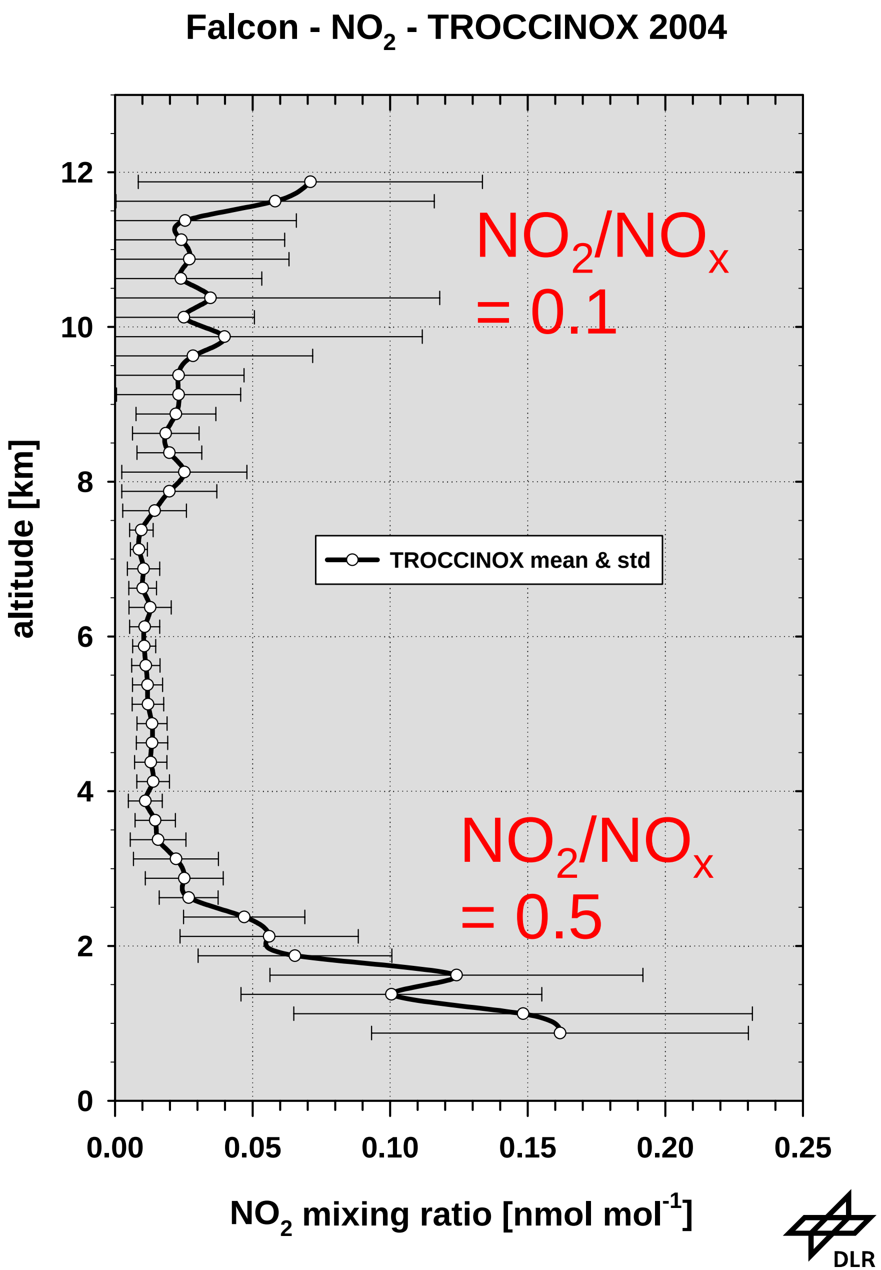
Falcon - NO2 - TROCCINOX 2004
altitude [km]
NO2 mixing ratio [nmol mol-1]
TROCCINOX mean & std
NO2/NOx = 0.1
NO2/NOx = 0.5
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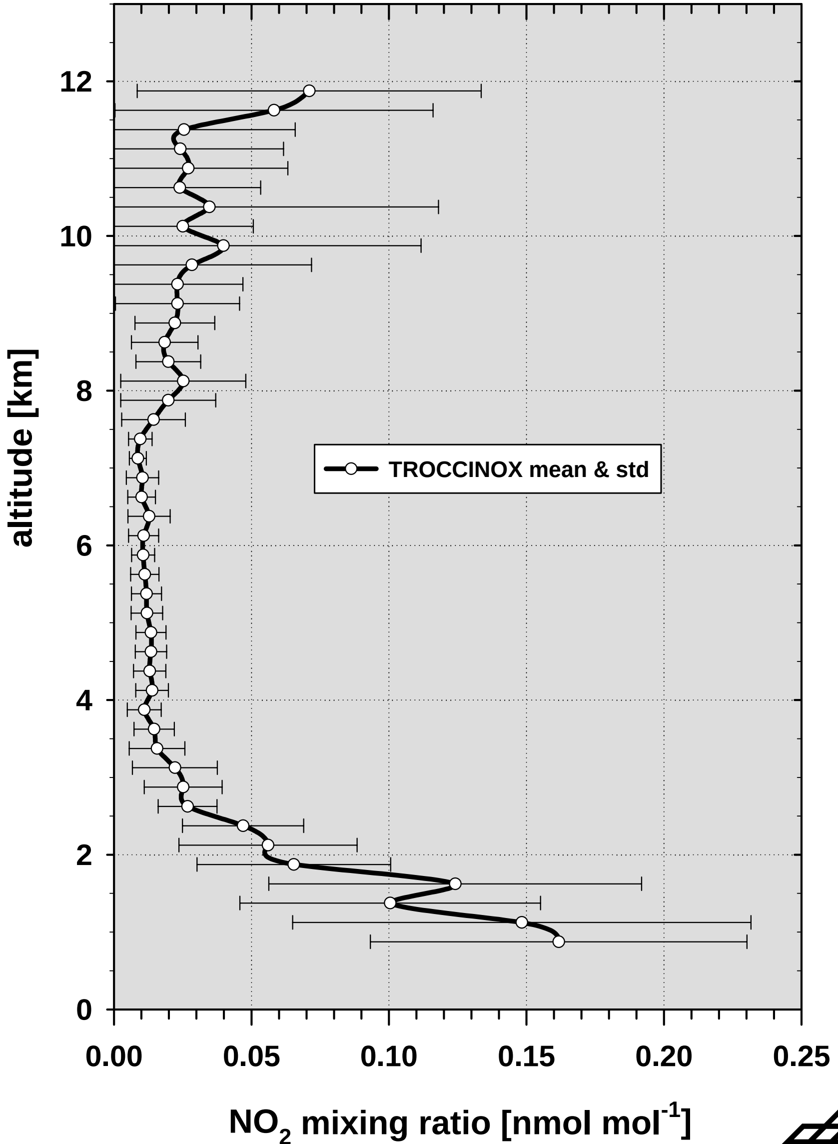
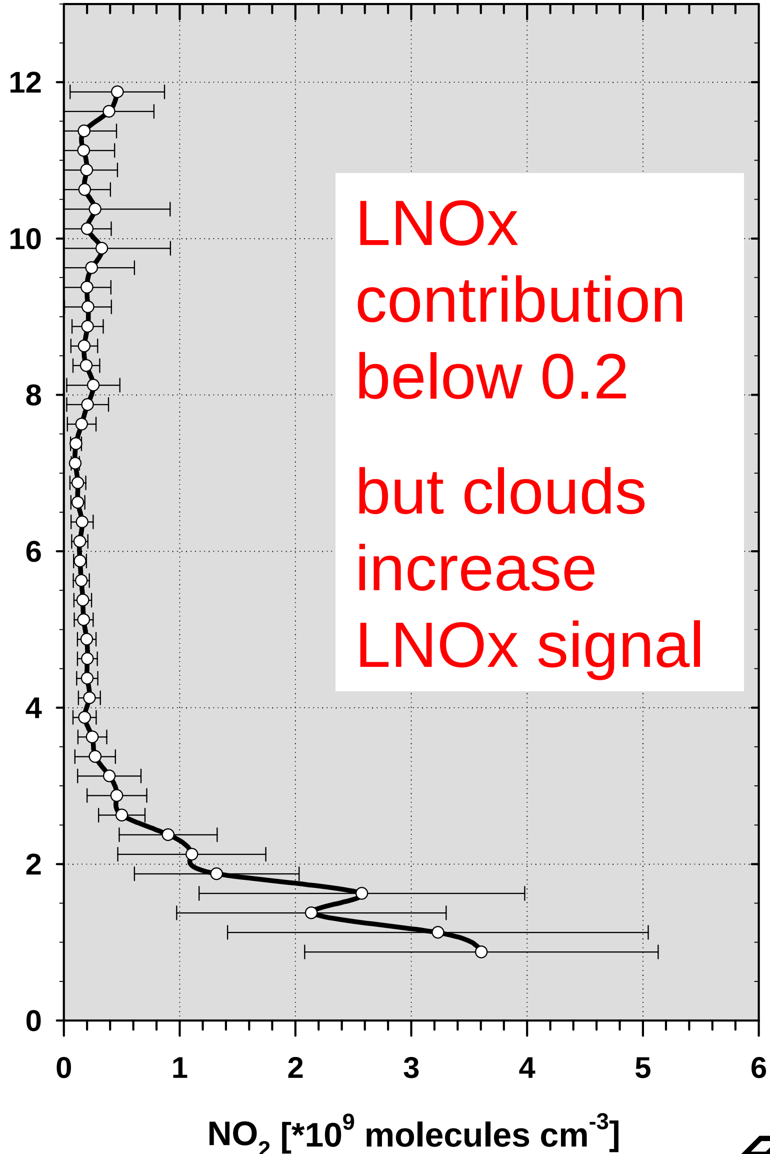
NO_2 mixing ratio - NO_2 molecule concentration





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Conclusion

LNOx value derived by best fit of models and TROCCINOX insitu data S = 4.8±2.5 Tg/a :

Least Square Fit gives S value with error estimates ΔS .

S-value computable from NO, NO_y, and O_3, and CO.

S-value from best fit of models and TROCCINOX insitu data: S = 4.8±2.5 Tg/a.

Sampling error due to observations in two months in the tropics should be reduced by inclusion of further data, including data from SCIAMACHY, GOME, OMI, etc.

LNOX contribution to SCIAMACHY NO2 column up to 15 % over tropical continents

Evaluation of LNOx from SCIAMACHY and OMI appears possible