New retrieval approach to tropospheric NO_2 by synergistic inversion of satellite nadir DOAS soundings and ground-based FTIR measurements

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Motivation - why another approach to tropospheric NO_2?

- **Bovensmann et al., 1999; Richter and Burrows, 2002; Boersma et al., 2004:** diff. approaches to separate strat. background: use limb measurements, use reference sector, use CTM assimilaton
- **Heland et al., 2002; Petritoli et al., 2004; Boersma et al., 2004:** significant errors in trop. NO_2 retrievals
- **Lambert et al., 2004:** lack of direct measurements of trop. NO_2 column for validation purposes
- **Richter; Lambert; Sussmann (ACVE-2, 2004):** ground-based DOAS and FTIR disagree as to total NO_2
- **Rinsland et al., 2003:** possible strong a priori contribution to FTIR retrieval of total NO_2
- **Rodgers and Connor, 2003:** intercomparison of remote sounding instruments
- **Eskes and Boersma, 2003:** averaging kernels for DOAS

⇒ **Sussmann, Stremme, Burrows, Richter, Seiler, Rettinger, ACP, 5, 2657-2677, 2005:** Tropospheric NO_2 from combined SCIAMACHY and Zugspitze FTIR retrieval

Definitions: Total column averaging kernels (row vectors $\boldsymbol{a}^T$)

$$\boxed{col_{ret} = col_a + \boldsymbol{a}^T \cdot (\boldsymbol{x}_{true} - \boldsymbol{x}_a)}$$

$$with \quad col_a = (111...1)\,\boldsymbol{x}_a \;,$$

$$and \;\; neglecting \;\; measurement \; errors$$

$$and \;\; forward \;\; model \; errors$$

$$\textbf{Case (i).} \quad \boldsymbol{a}^T = (111...1)$$

$$\Rightarrow \quad col_{ret} \quad = \quad col_{true}$$

$$\textbf{Case (ii).} \quad a_i = 0 \quad \forall \quad x_{true,i} \quad \neq \quad x_{a,i} \;\; with \; i : layer \; index$$

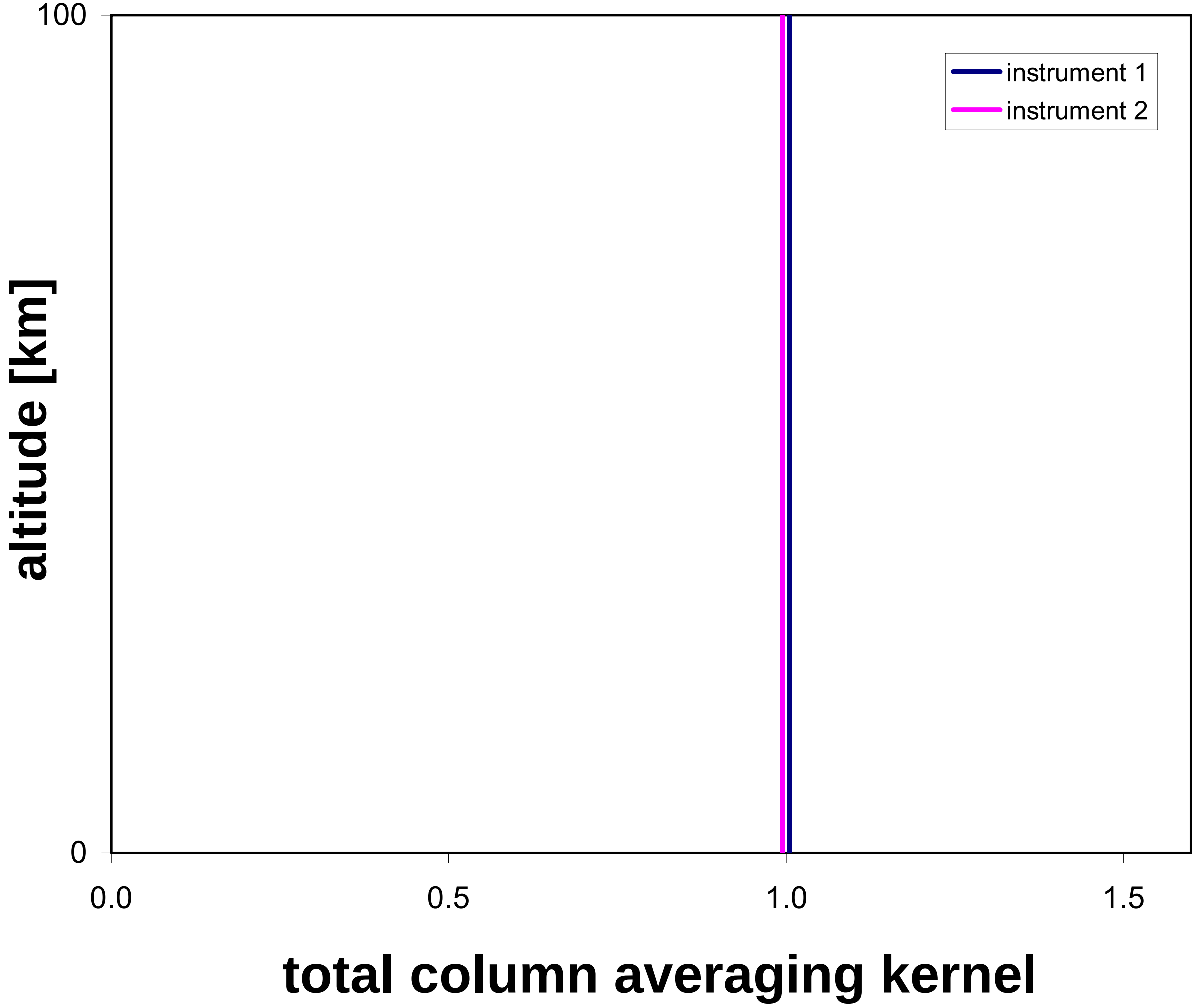
$$\Rightarrow \quad col_{ret} \quad = \quad col_a \quad \neq \quad col_{true}$$

$$\textbf{Case (iii).} \quad \boldsymbol{x}_{true} \quad = \quad \boldsymbol{x}_a$$

$$\Rightarrow \quad col_{ret} \quad = \quad col_{true} \quad = \quad col_a$$

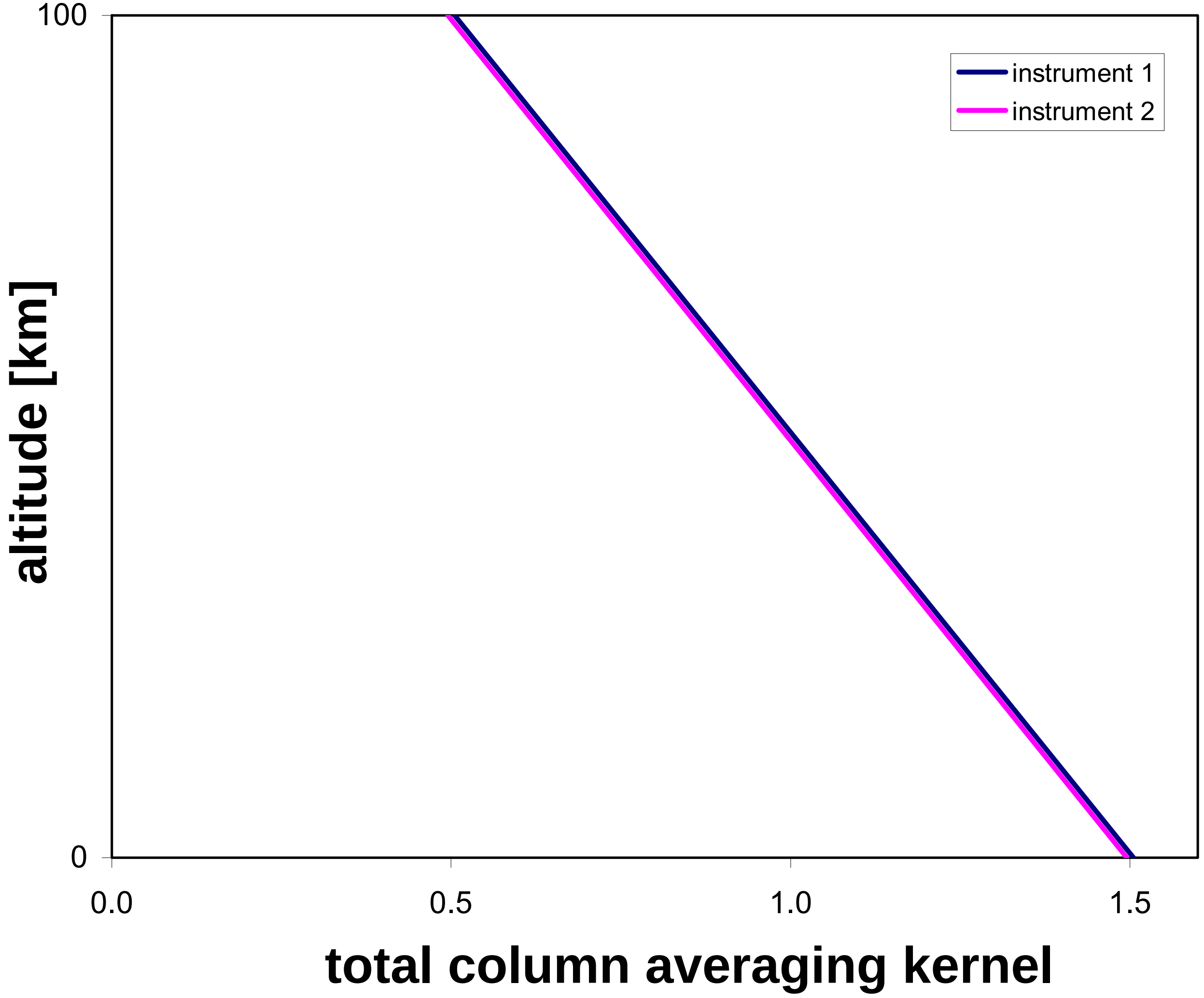
taken from:
Sussmann, R.: Ground-based Fourier transform spectrometry at the NDSC site Zugspitze: Geophysical products for satellite validation, in *Proceedings of the European Symposium on Atmospheric Measurements from Space*, ESTEC, Noordwijk, The Netherlands, 18-22 Jan 1999, WPP-161, Vol. 2, pp. 661-664, 1999.

Two remote sounding instruments: ideal for validation, no synergistic use



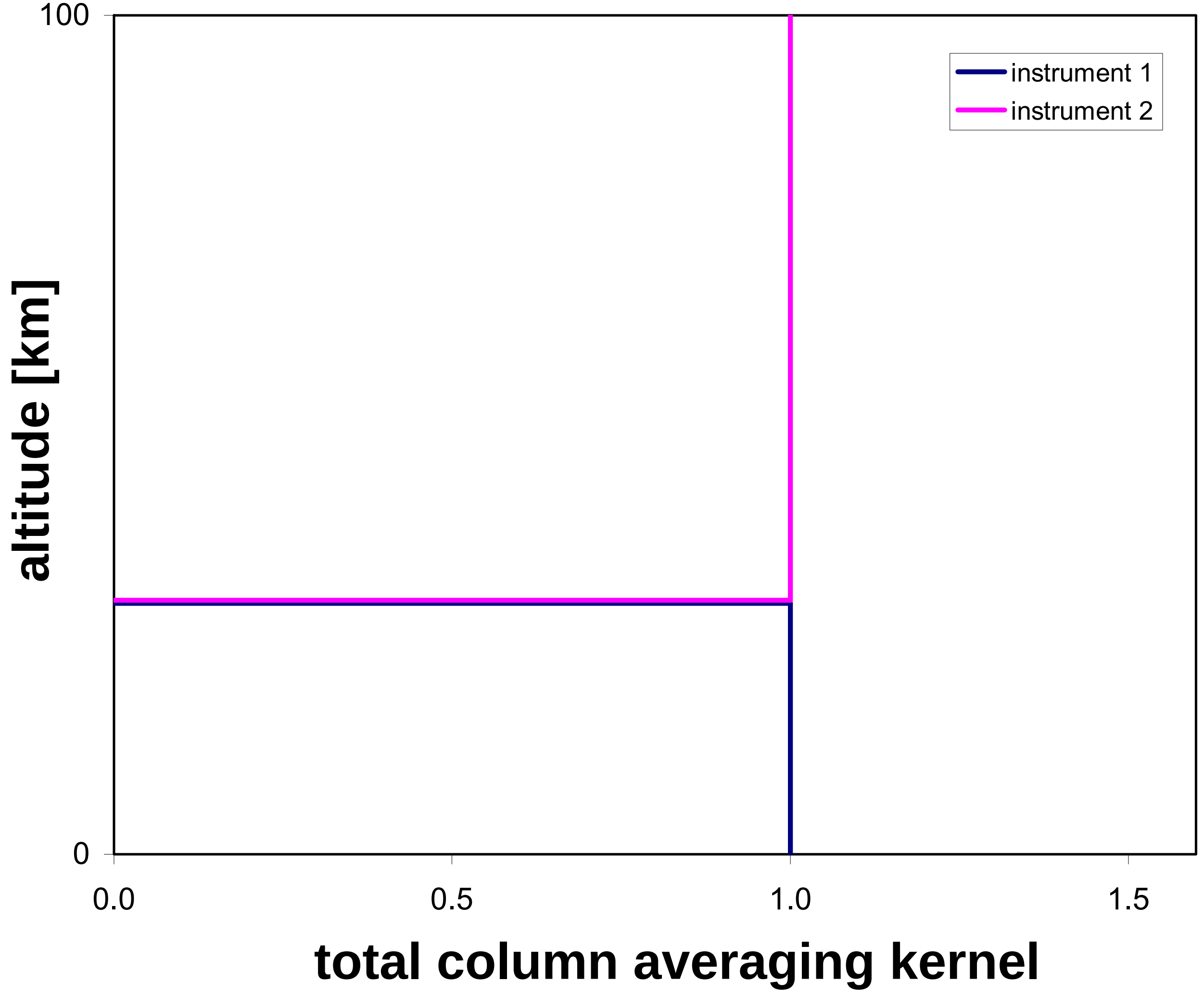
Example 1:
ideal instruments

Two remote sounding instruments: ideal for validation, no synergistic use



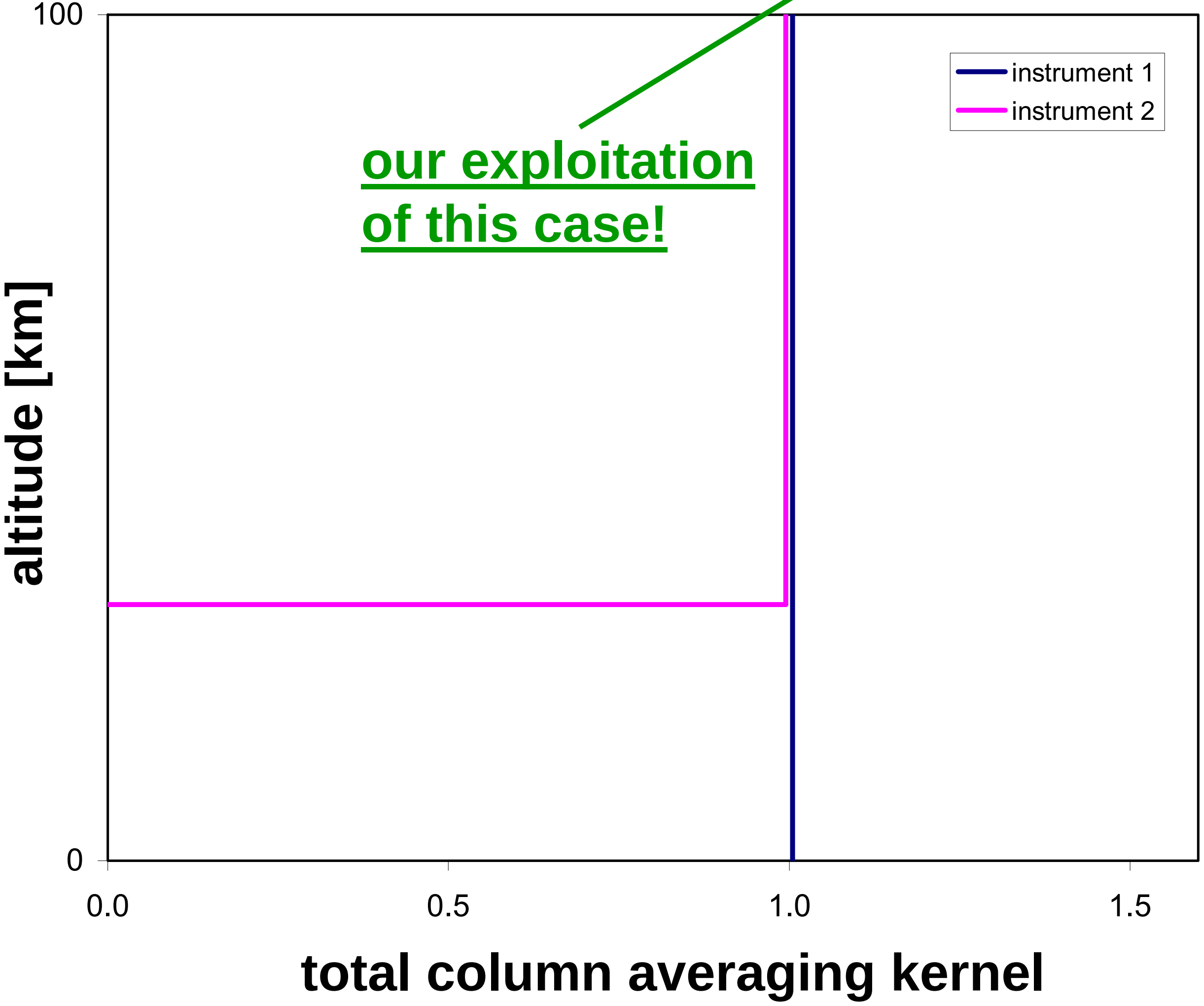
Example 2:
non-ideal instruments

Two remote sounding instruments: ideal for synergistic use, no validation



Example 3

Two remote sounding instruments: ideal for synergistic use, validation limited to sub-set of states



our exploitation of this case!

Rodgers, Connor, 2003

Example 4

Total column averaging kernels: DOAS

DOAS sensitivity usually characterized by Air Mass Factor.

First DOAS-Averaging Kernel: Eskes and Boersma, ACP, 3, 2003:

$$\boldsymbol{a}_i = \frac{AMF(\boldsymbol{x}_{ref,i}, \boldsymbol{b})}{AMF(\boldsymbol{x}_{ref}, \boldsymbol{b})}$$

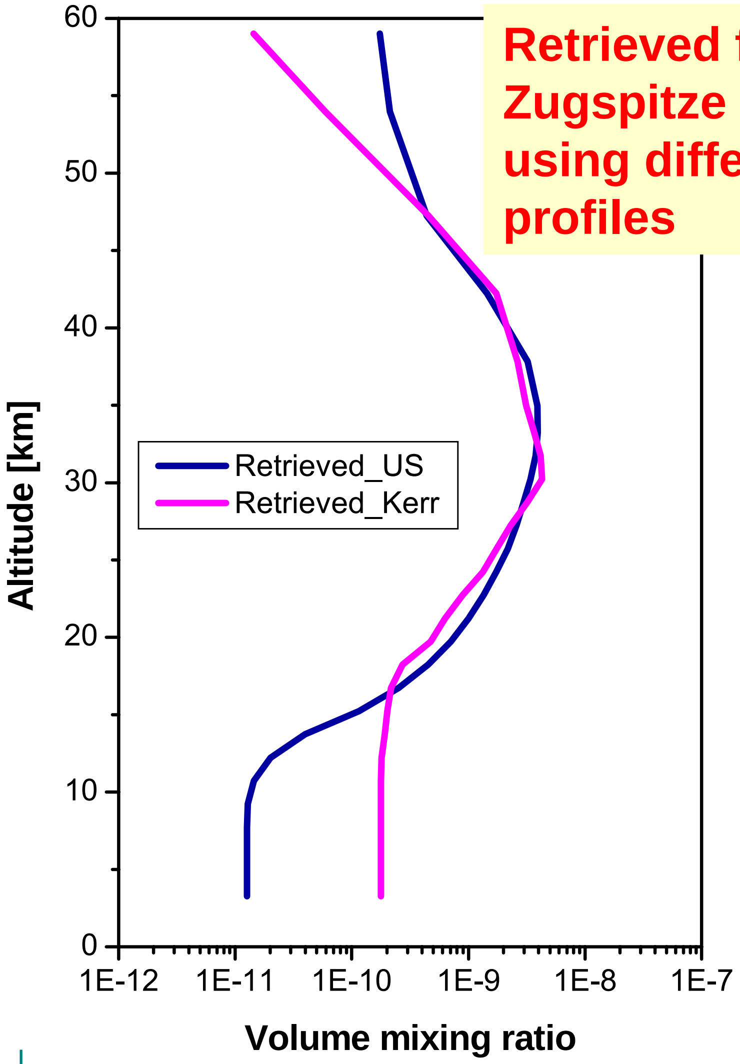
i: layer index
$\boldsymbol{x}_{ref}$: reference state (e.g., a priori NO_2 profile)
$\boldsymbol{b}$: SZA, viewing angle, clouds, albedo,
aerosols, …

Have to decide upon NO_2 a priori profile:

DOAS AMF *strongly* depends on a priori NO_2 profile shape in the troposphere, DOAS AMF does nearly *not depend* on a priori NO_2 profile in the stratosphere

⇒ DOAS a priori := US Standard NO_2 profile with VMR = 0 up to 10 km (A. Richter: SCIAMACHY UB1.5 total NO_2 retrieval)

Total column averaging kernels: NO_2 a priori profile for FTIR?

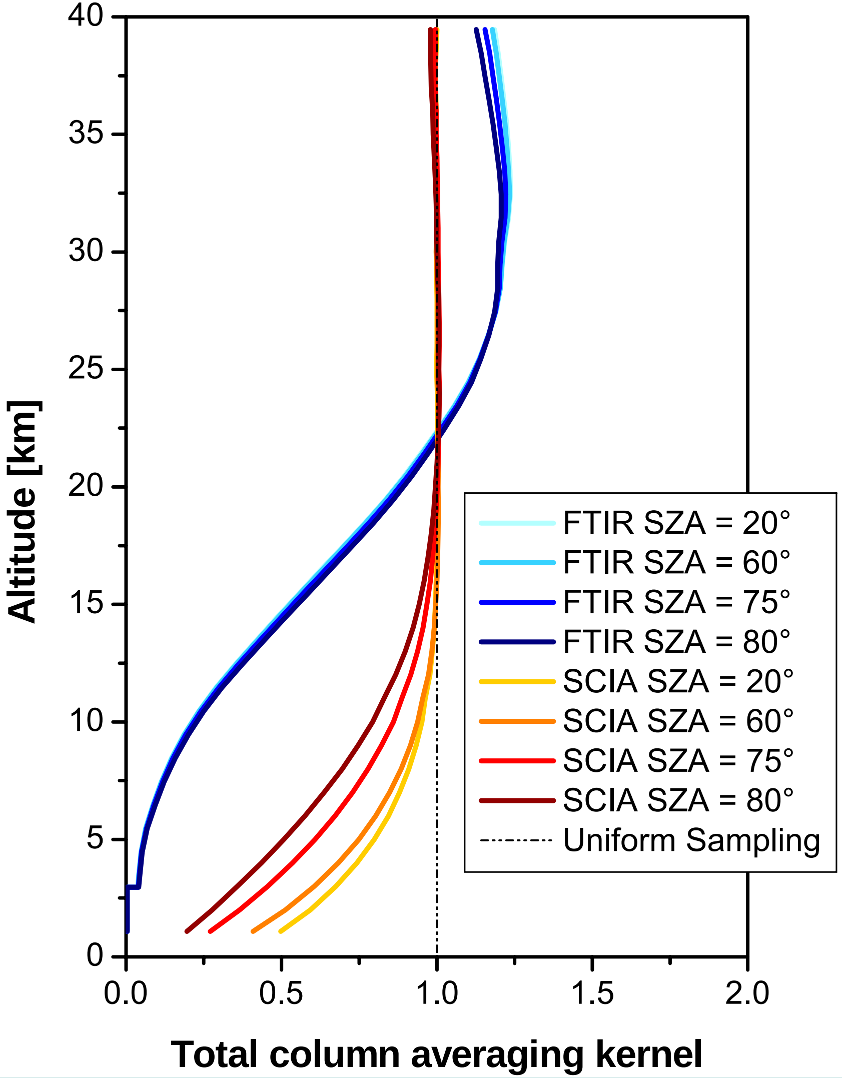


Retrieved from the same Zugspitze FTIR spectrum using differing a priori profiles

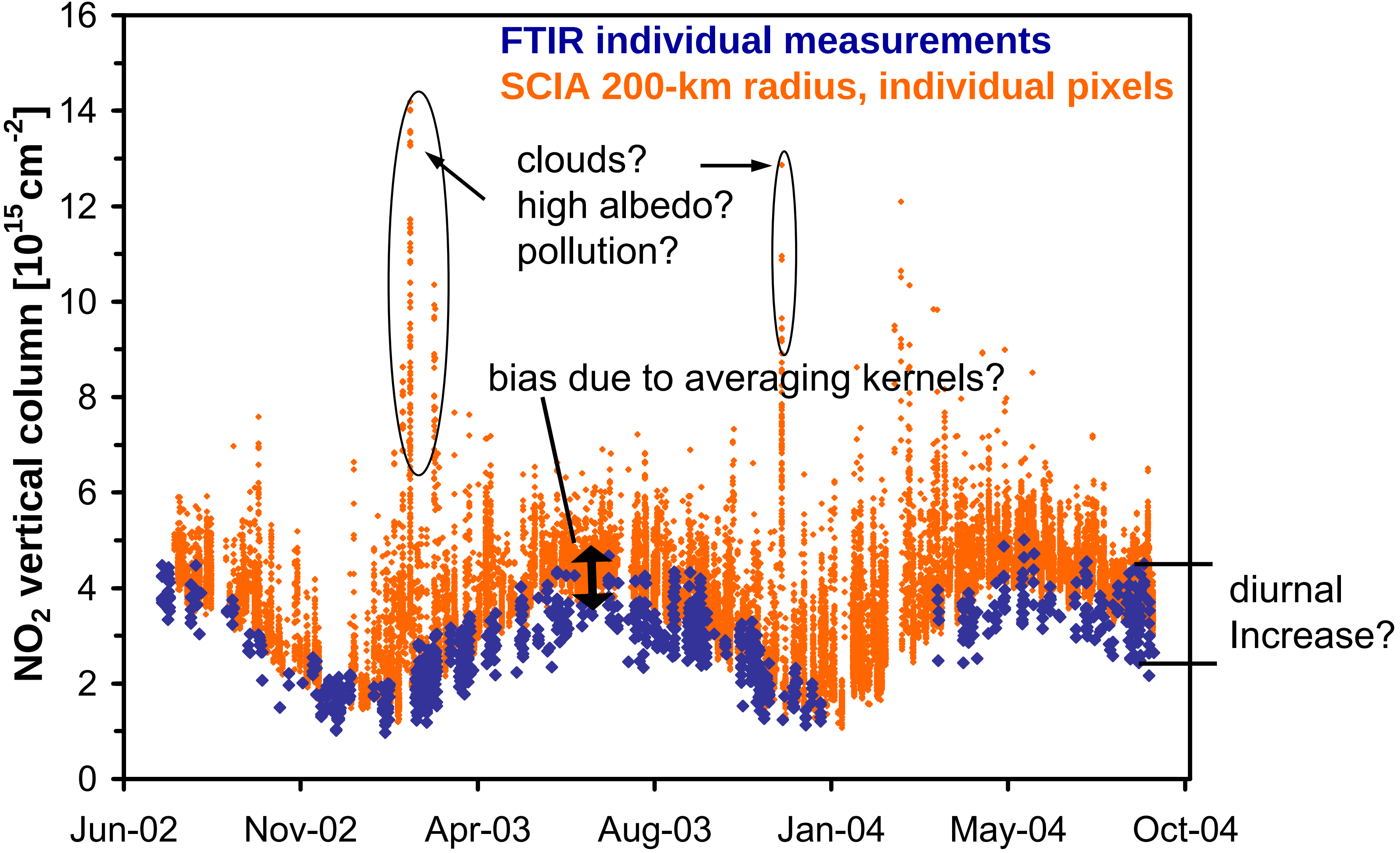
Solar FTIR retrieval not sensitive in the troposphere,
thus strongly depends on a priori NO_2 profile shape in the troposphere

⇒ use FTIR a priori = DOAS a priori :=
US Standard profile with VMR = 0 up to 10 km altitude

Total column averaging kernels: FTIR versus SCIAMACHY



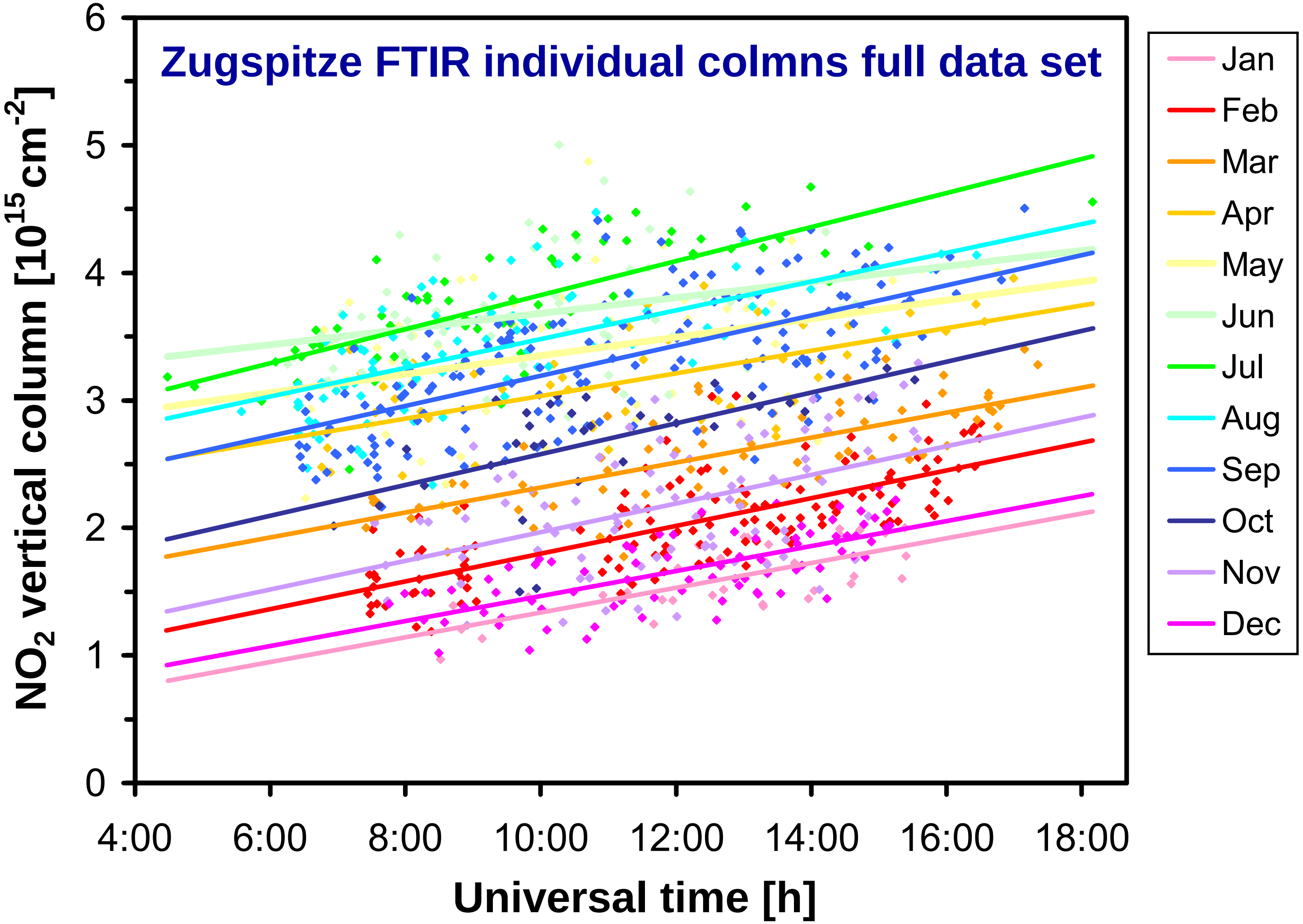
SCIA UB1.5 versus Zugspitze FTIR total columns: Direct comparison



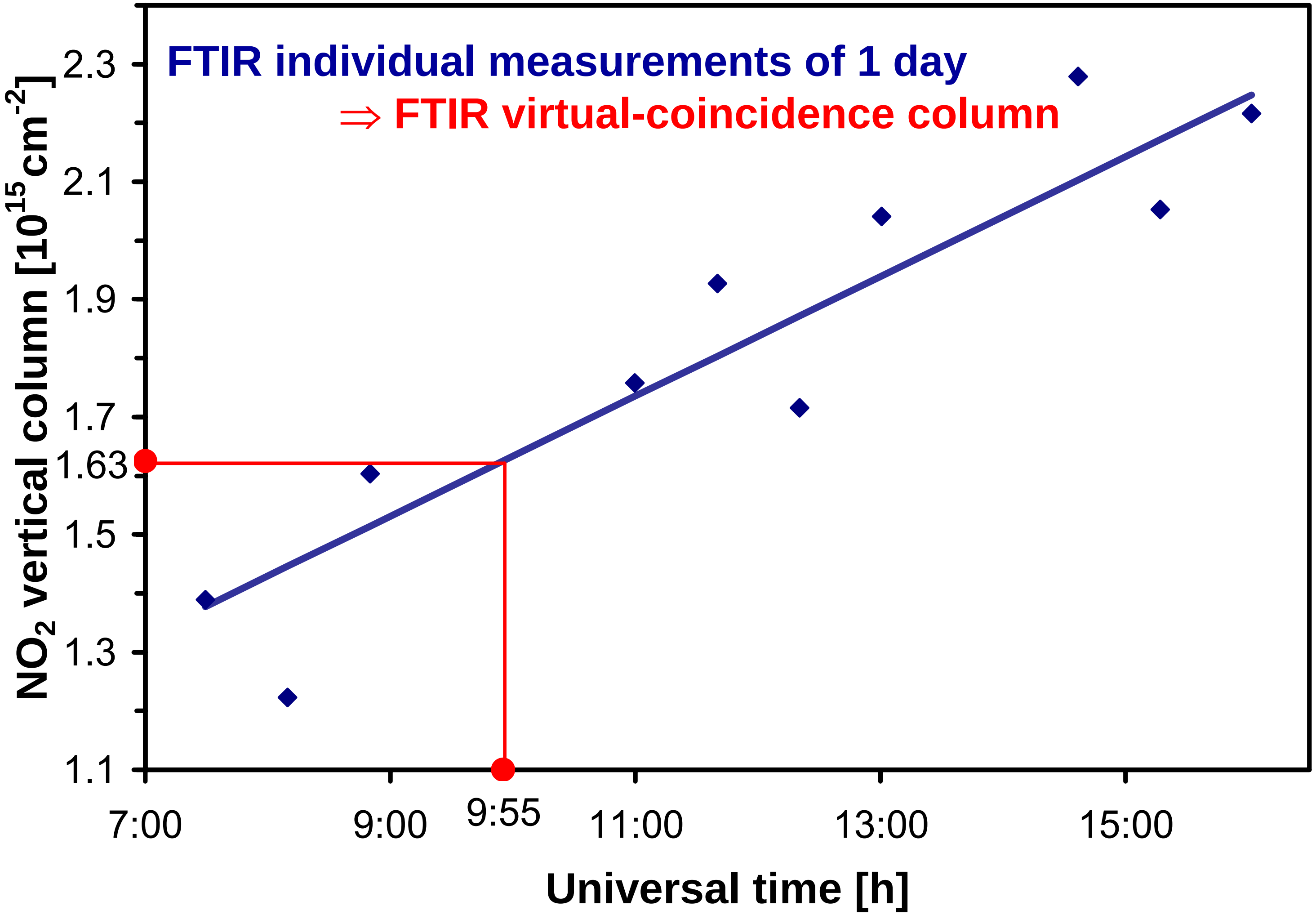
Total NO_2 diurnal increasing rate: Does it change with season?

„No annual cycle of diurnal increasing rate found“

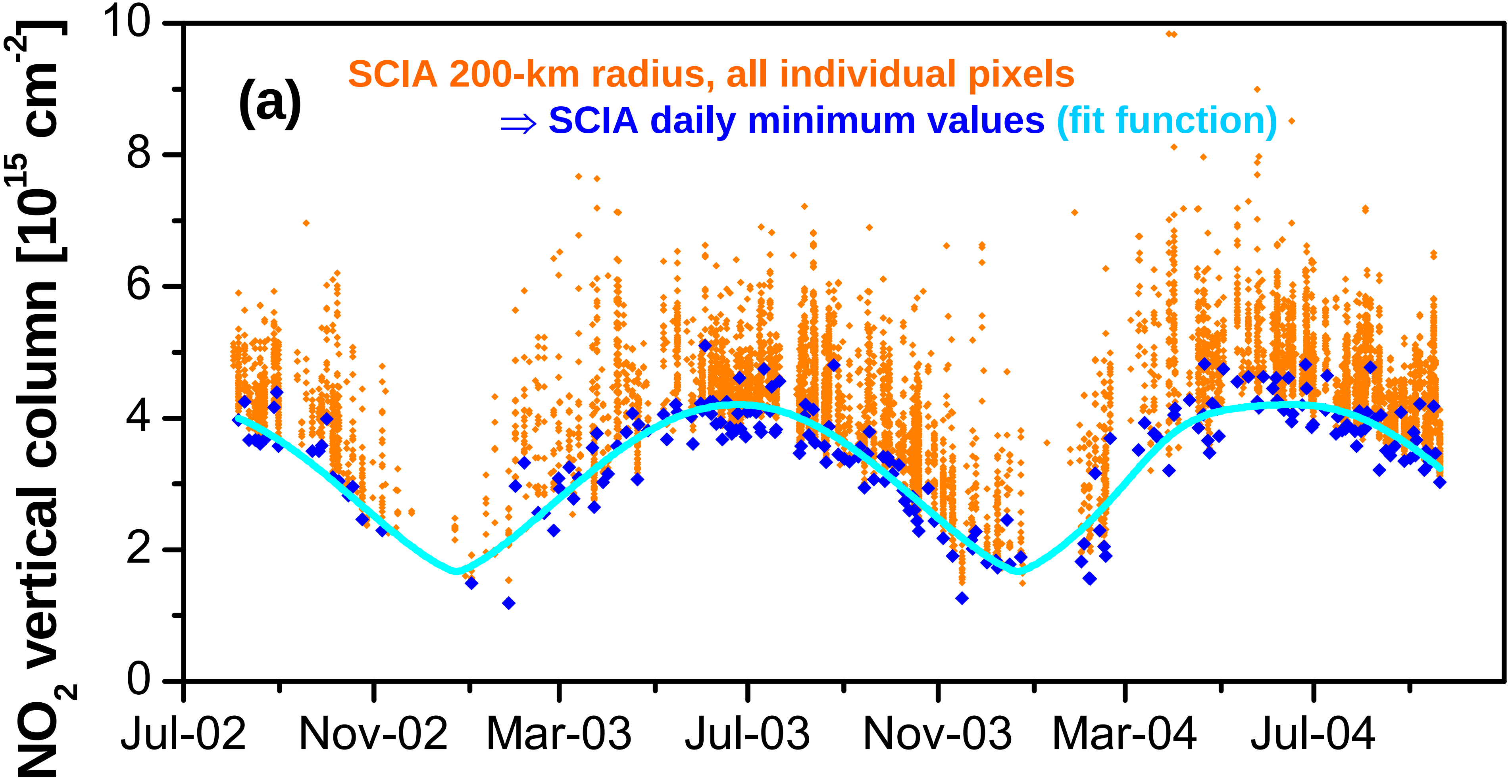
⇒ **Mean diurnal increasing rate for mid-latitudes = + 1.02(6)E+14 cm^{-2}/h**



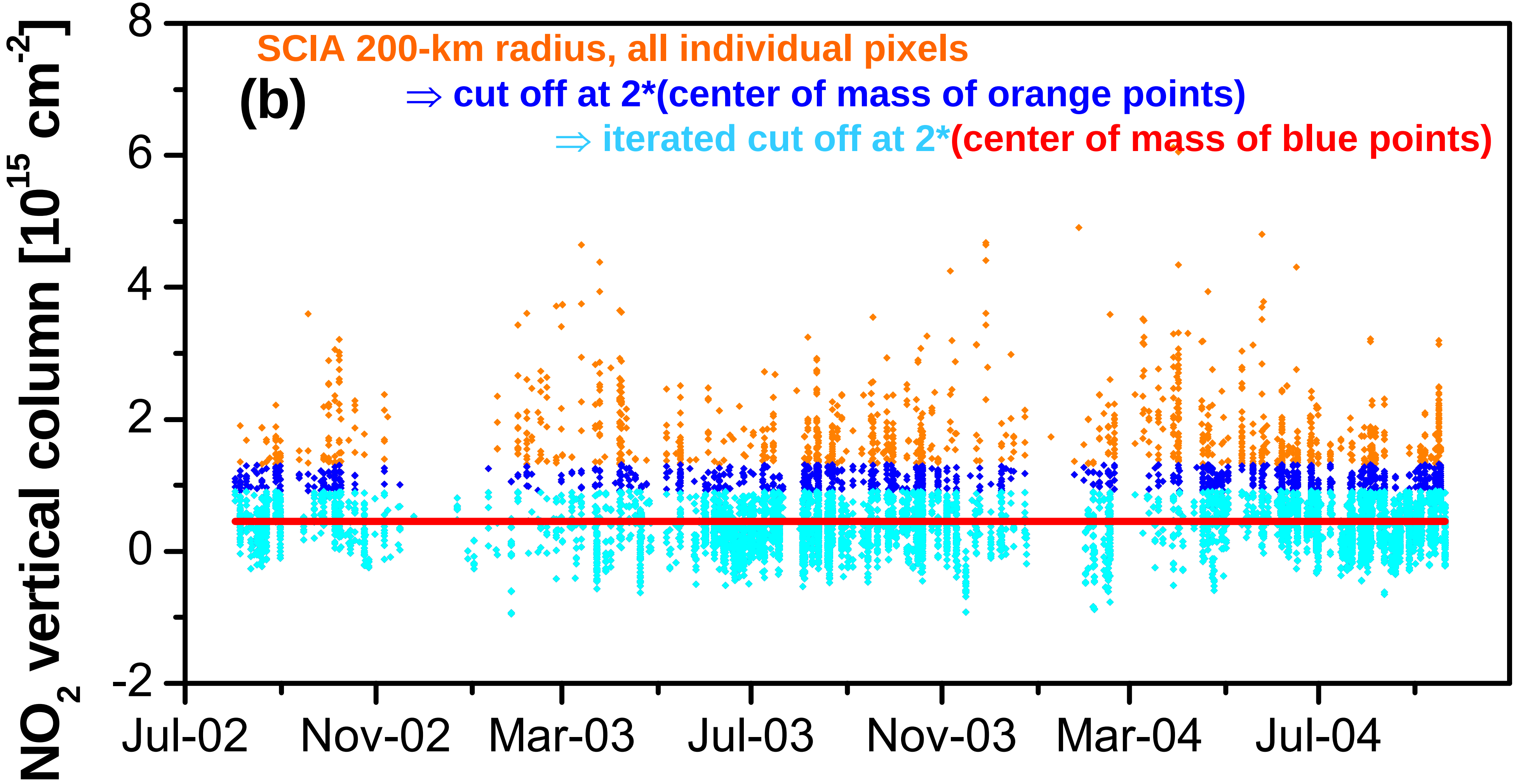
Matching FTIR to satellite overpasses: Concept of „virtual coincidences“



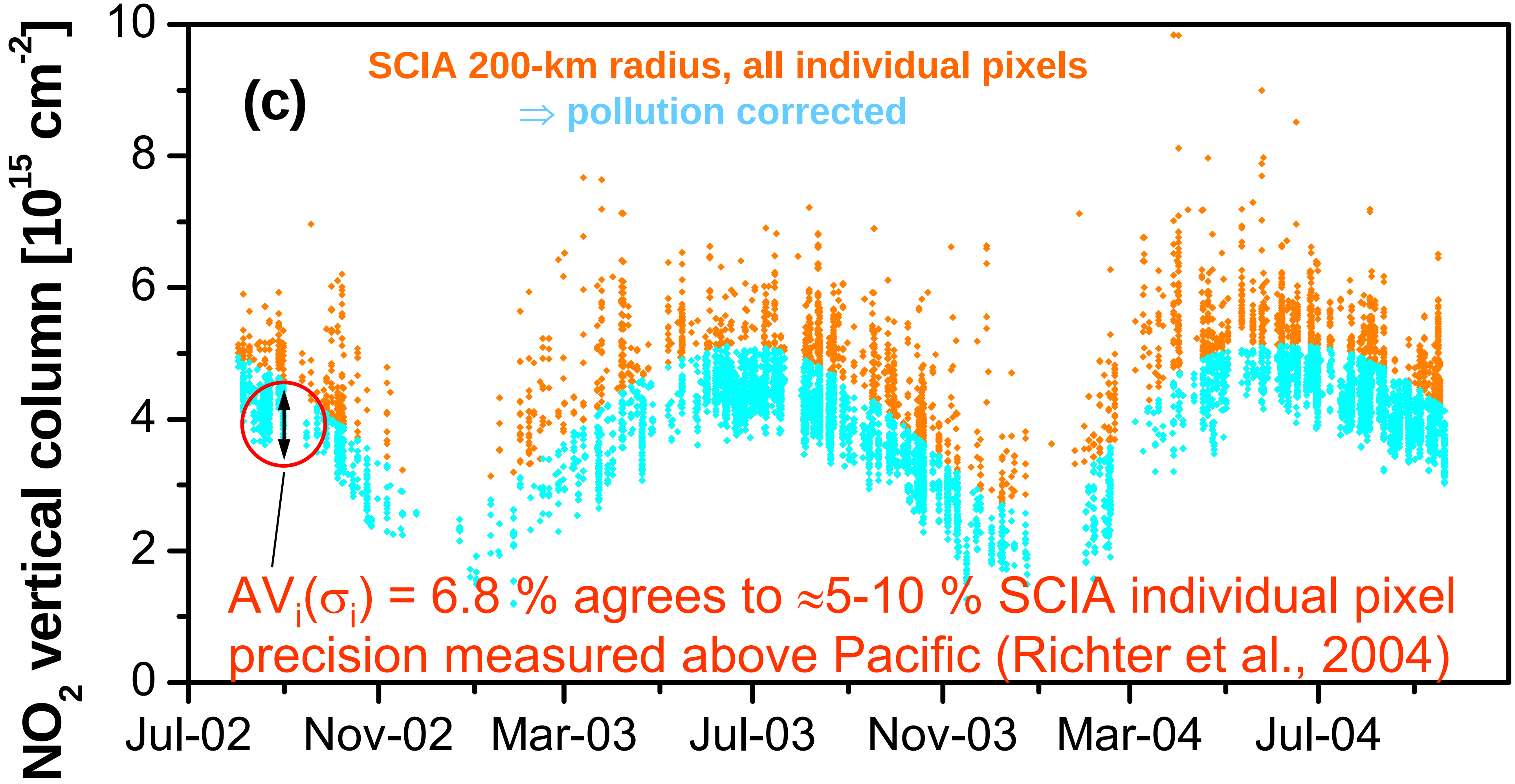
Satellite Pollution Correction Scheme: (a) Substract fit to daily minima



Sat. Pollution Correction Scheme: (b) Cut off at 2*(center of mass) and iterate

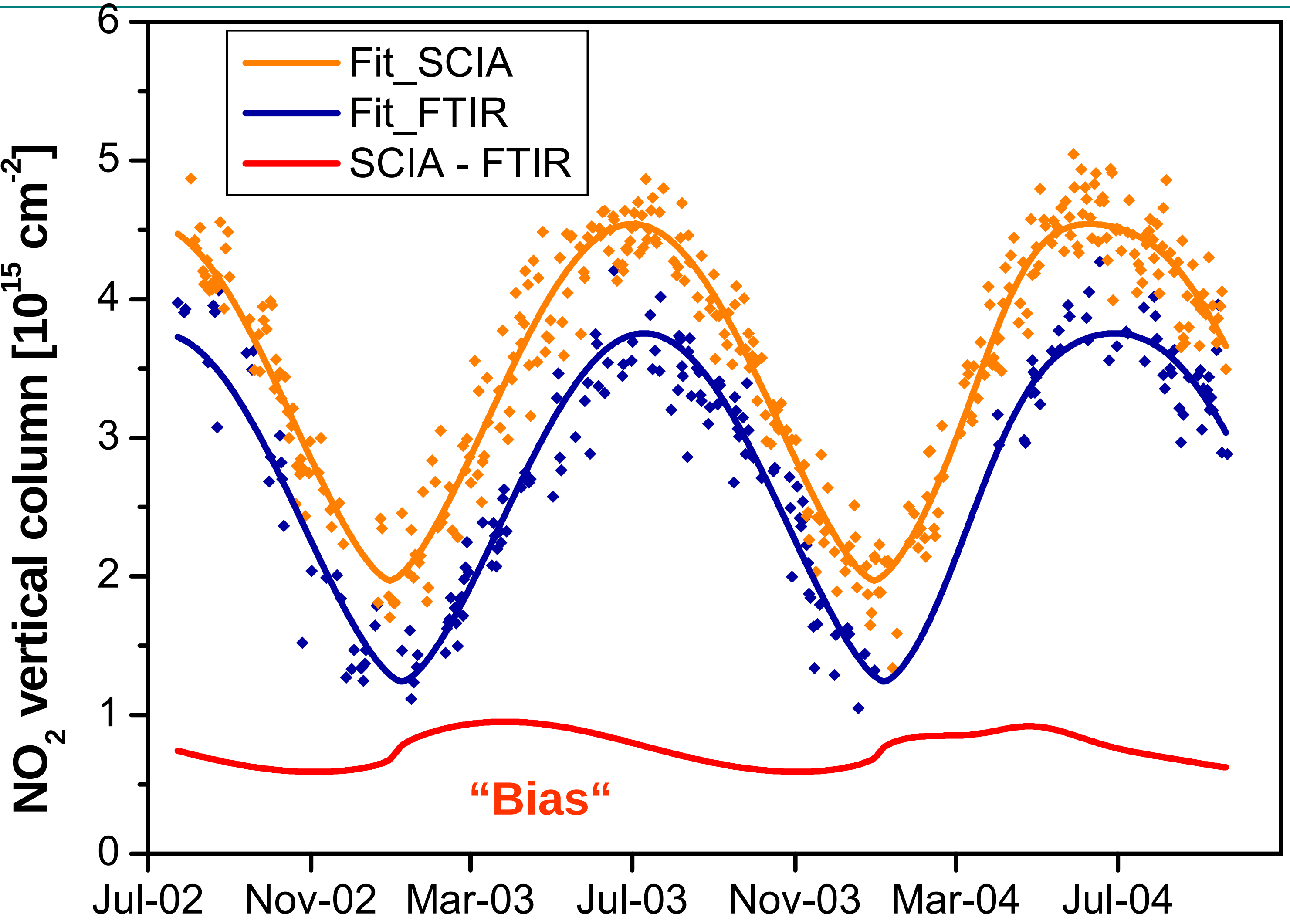


Sat. Pollution Correction Scheme: (c) Re-add fit-function (ann. cycle of minima)

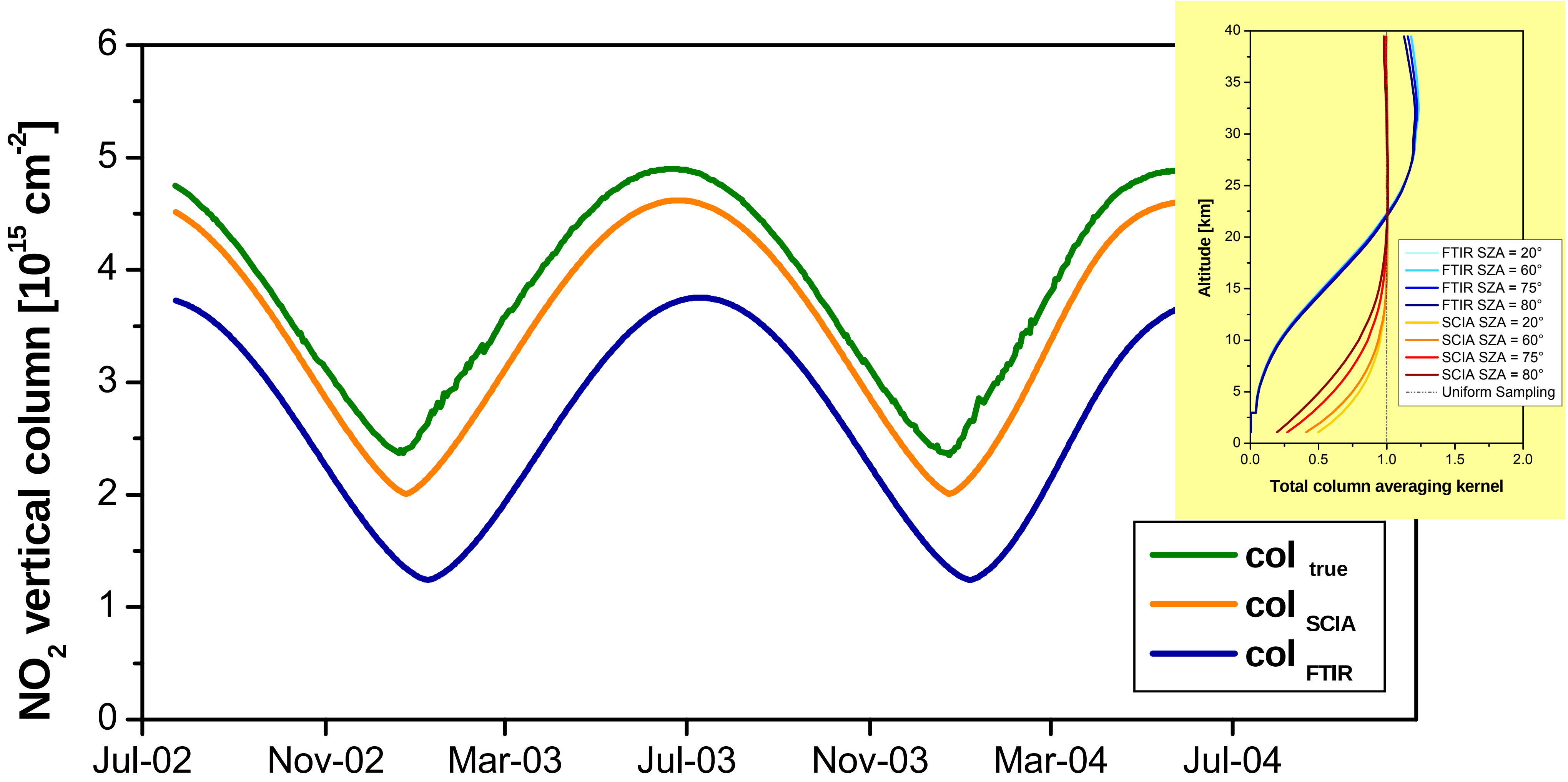


$AV_i(\sigma_i)$ = 6.8 % agrees to ≈5-10 % SCIA individual pixel precision measured above Pacific (Richter et al., 2004)

“Validation“: FTIR virt. coincidence col´s versus SCIA poll. corr. 200-km means



Combine FTIR and SCIAMACHY to retrieve tropospheric NO_2: The Idea



Combined a posteriori retrieval of tropospheric NO_2: Constraint

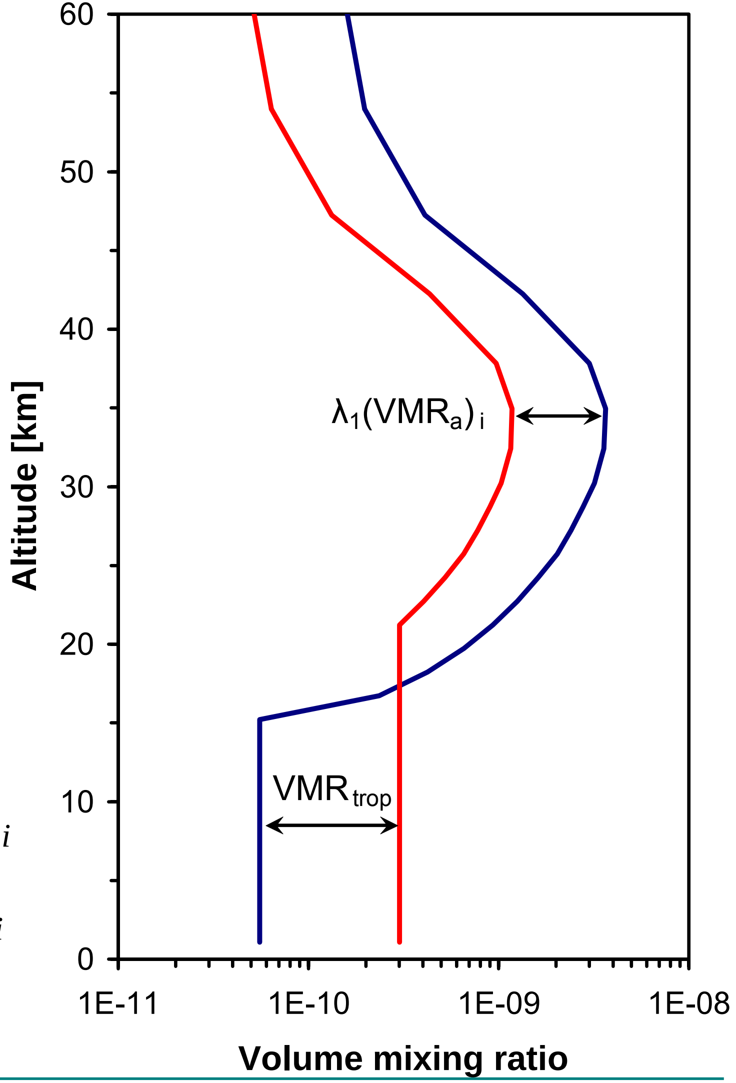
Our a posteriori retrieval constraint is set up as follows

$$\boldsymbol{x}_{true} = \lambda_1 \cdot \boldsymbol{x}_a + \boldsymbol{x}_{trop} \qquad \text{with}$$

$$\boldsymbol{x}_{trop} = \begin{pmatrix} VMR_{29} \cdot AMF_{29} \\ \vdots \\ VMR_1 \cdot AMF_1 \end{pmatrix}$$

where

$$VMR_i = \begin{cases} 0 & \text{if } VMR_{trop} < \lambda_1 (VMR_a)_i \\ VMR_{trop} - \lambda_1 (VMR_a)_i & \text{if } VMR_{trop} \geq \lambda_1 (VMR_a)_i \end{cases}$$



Combined a posteriori retrieval of tropospheric NO_2: The set up

We write two relations for the true column col_{true}, the column retrieved by FTIR col_{FTIR}, and the column retrieved by SCIAMACHY col_{SCIA}, *i.e.*,

$$col_{FTIR} - col_{true} = col_{FTIR} - \boldsymbol{a}_{ideal}^{T} \cdot \boldsymbol{x}_{true} = \boldsymbol{a}_{FTIR}^{T} \cdot (\boldsymbol{x}_{true} - \boldsymbol{x}_{a}) + \boldsymbol{a}_{ideal}^{T} \cdot \boldsymbol{x}_{a} - \boldsymbol{a}_{ideal}^{T} \cdot \boldsymbol{x}_{true}$$

and

$$col_{SCIA} - col_{true} = col_{SCIA} - \boldsymbol{a}_{ideal}^{T} \cdot \boldsymbol{x}_{true} = \boldsymbol{a}_{SCIA}^{T} \cdot (\boldsymbol{x}_{true} - \boldsymbol{x}_{a}) + \boldsymbol{a}_{ideal}^{T} \cdot \boldsymbol{x}_{a} - \boldsymbol{a}_{ideal}^{T} \cdot \boldsymbol{x}_{true}$$

where vectors $\boldsymbol{x}$ are partial columns profiles, and $\boldsymbol{x_a}$ is the a priori profile common to FTIR and SCIA.

Combined a posteriori retrieval of tropospheric NO_2: Retrieval equation

We derive a starting value for λ_1 *by using*

$$col_{FTIR} - col_{true}(10 - 100\ km) = 0$$

i.e., we assume that FTIR is a good measure for the true pure stratospheric column.
It follows

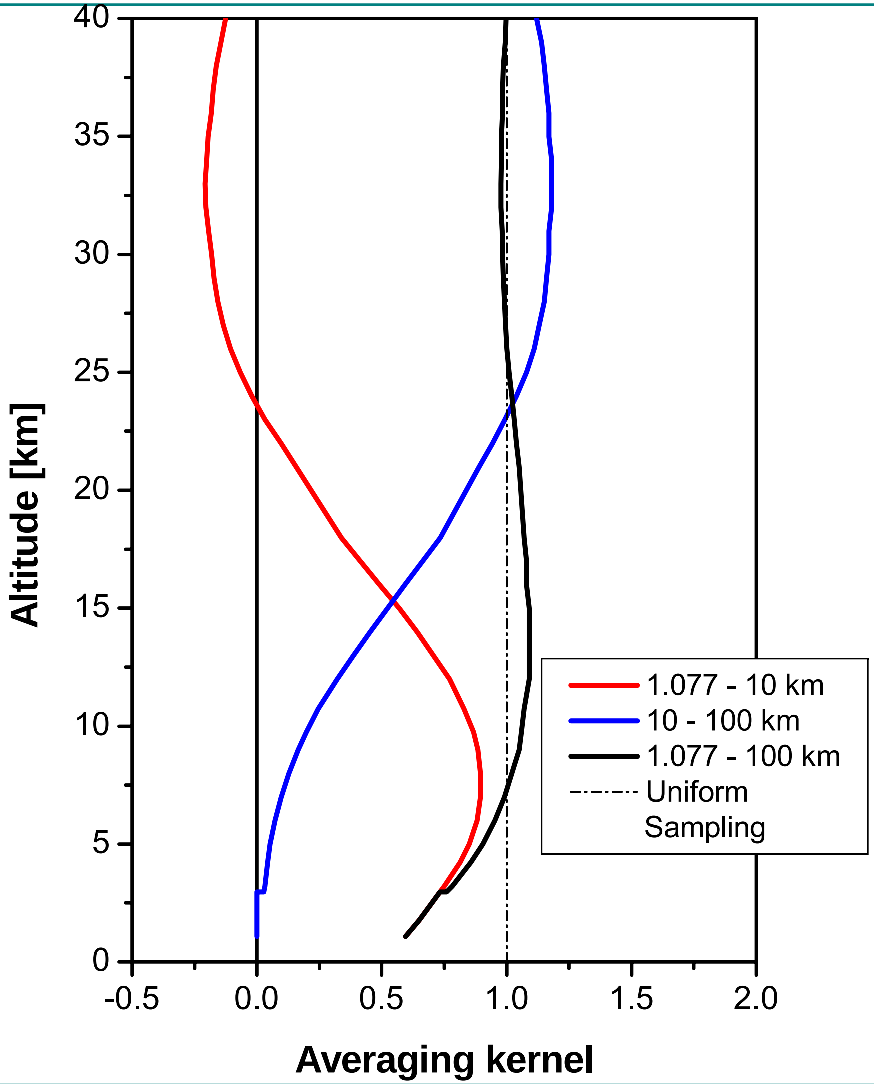
$$\lambda_1 = \frac{col_{FTIR}}{\boldsymbol{a}_{ideal}^{T} \cdot \boldsymbol{x}_a}$$

For the given λ_1 we subsequently apply the a posteriori retrieval equation

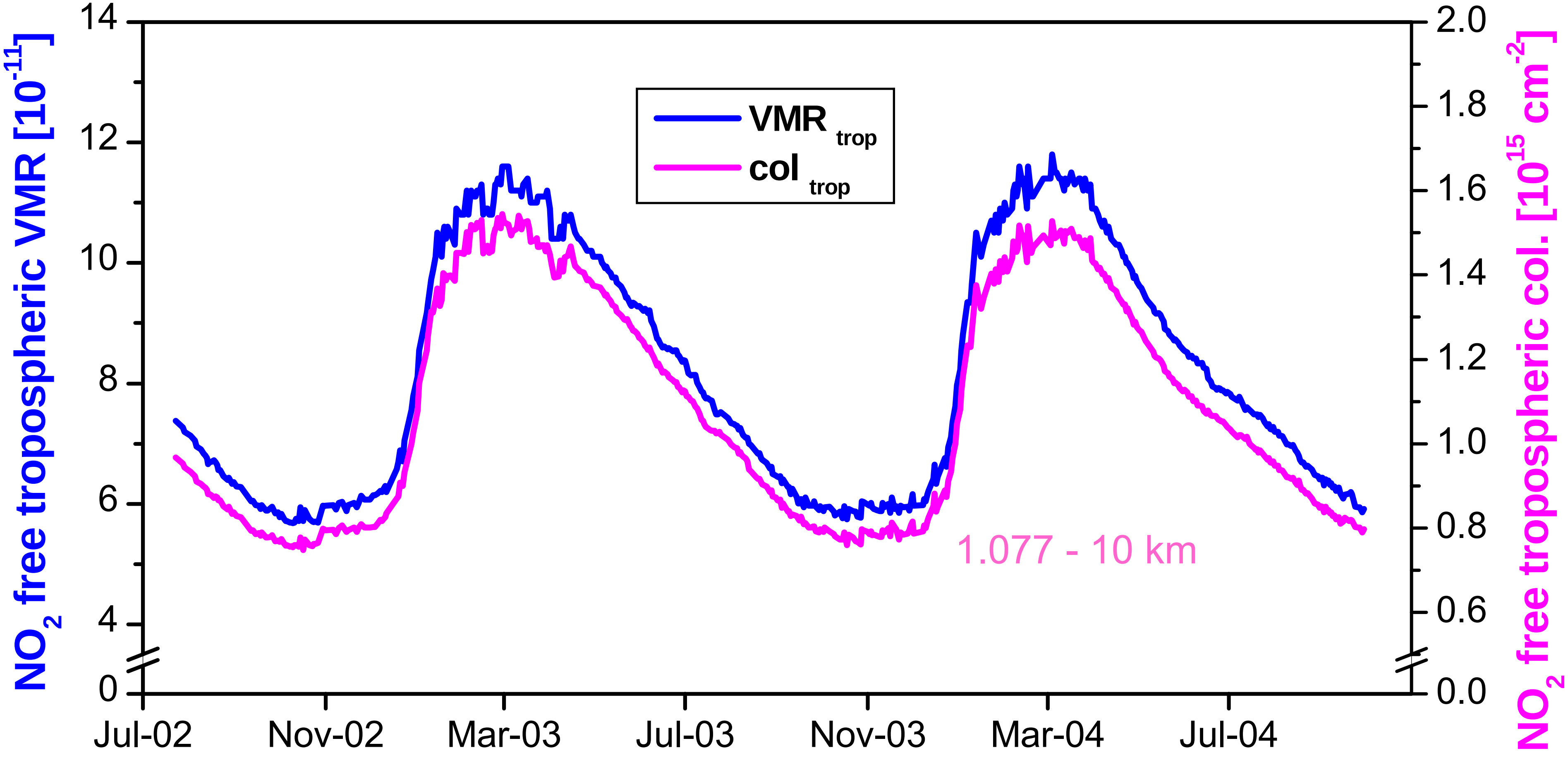
$$col_{FTIR} - col_{SCIA} = (\boldsymbol{a}_{FTIR} - \boldsymbol{a}_{SCIA})^{T}[(\lambda_1 - 1)\boldsymbol{x}_a + \boldsymbol{x}_{trop}]$$

which describes the iteration of VMR_{trop} *via* $\boldsymbol{x}_{trop}$ to match the measured columns difference $col_{FTIR} - col_{SCIA}$.

Characterization: Averaging kernels of the combined a-posteriori NO_2 retrieval



Combined a posteriori retrieval: Result for free tropospheric NO_2 column



Summary and Outlook

- stratospheric NO_2 diurnal increasing rate does not depend on season
- concept of FTIR “virtual-coincidence column”
- pollution clearing scheme for satellite NO_2 nadir DOAS
- mountain-site FTIR with a priori set to zero in the troposphere is a good measure for the pure stratospheric column
- combined FTIR-satellite DOAS a posteriori retrieval with dofs = 2
- phase of annual cycle of free tropospheric NO_2 is between the phase of boundary layer NO_2, and stratospheric NO_2

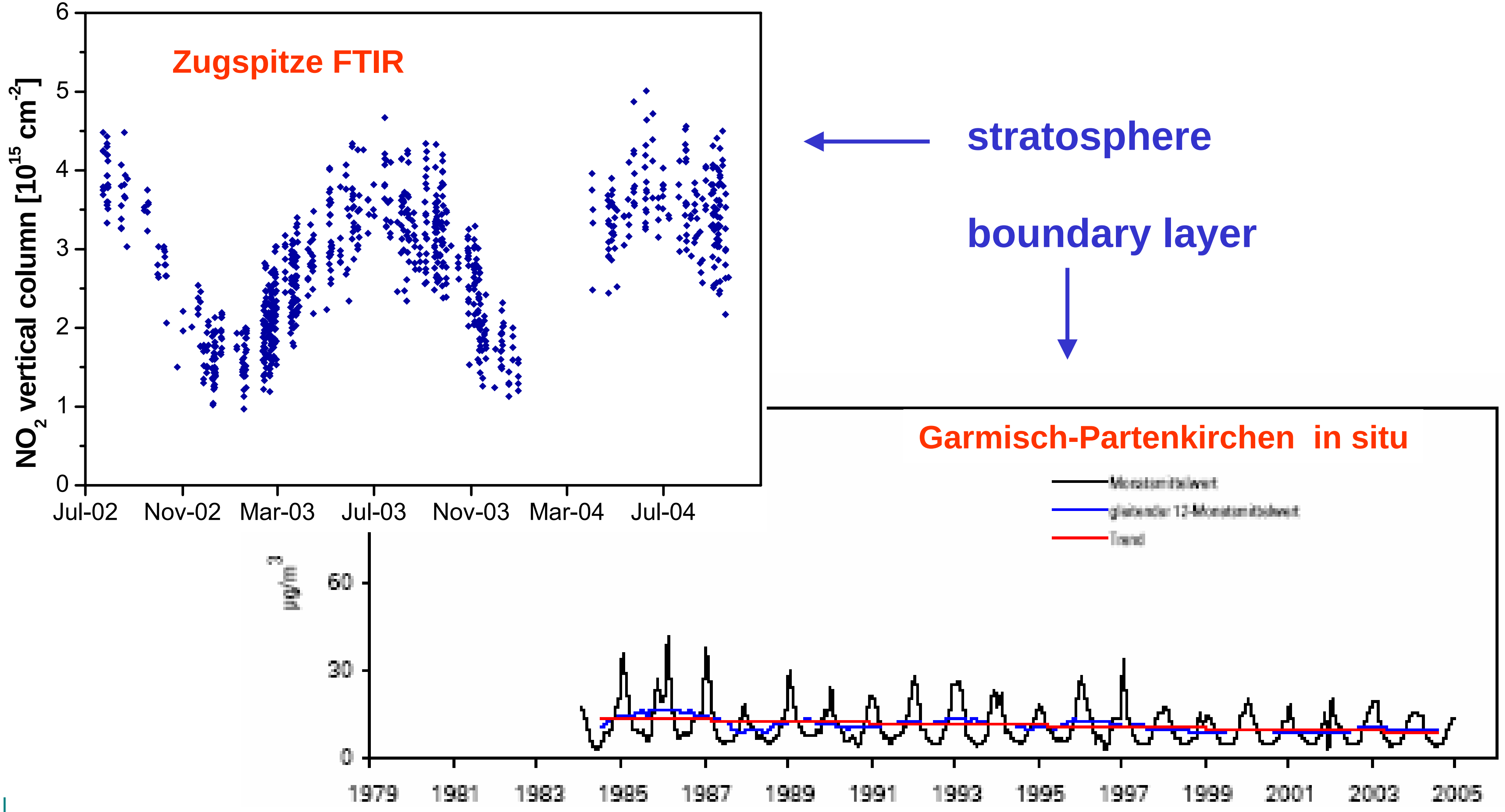
Vision: integrated global NO_2 observing system based on synergistic use of nadir satellites and a set of ground-based FTIRs

Acknowledgments

BMBF/DLR-GCVOS, EC-UFTIR, EC-ACCENT-AT2, ESA-TASTE

Appendix 1:

Combined a-posteriori retrieval: Annual cycle of free tropospheric NO_2 column?

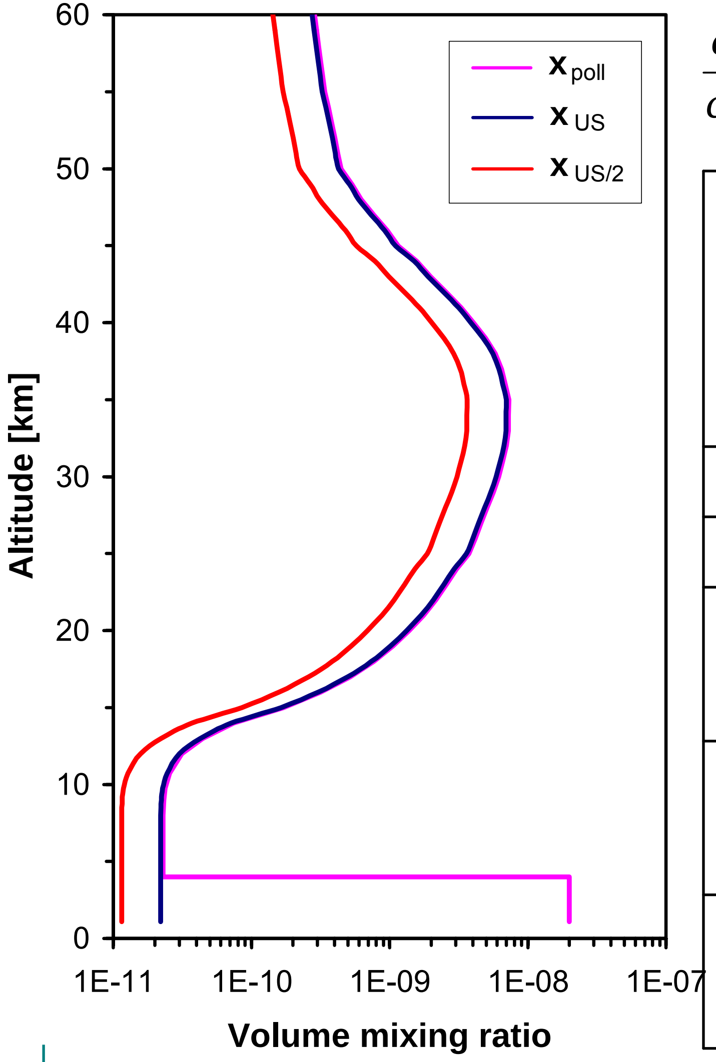


stratosphere

boundary layer

Appendix 2:

Different sensitivity to tropospheric pollution: FTIR versus SCIAMACHY



$$\frac{col_{ret}}{col_{true}} = \frac{\mathbf{a}^T(\mathbf{x}_{true}-\mathbf{x}_a)+\mathbf{a}^T_{ideal}\mathbf{x}_a}{col_{true}}$$

with $col_{true} = (111...1)\mathbf{x}_{true} = \mathbf{a}^T_{ideal}\mathbf{x}_{true}$

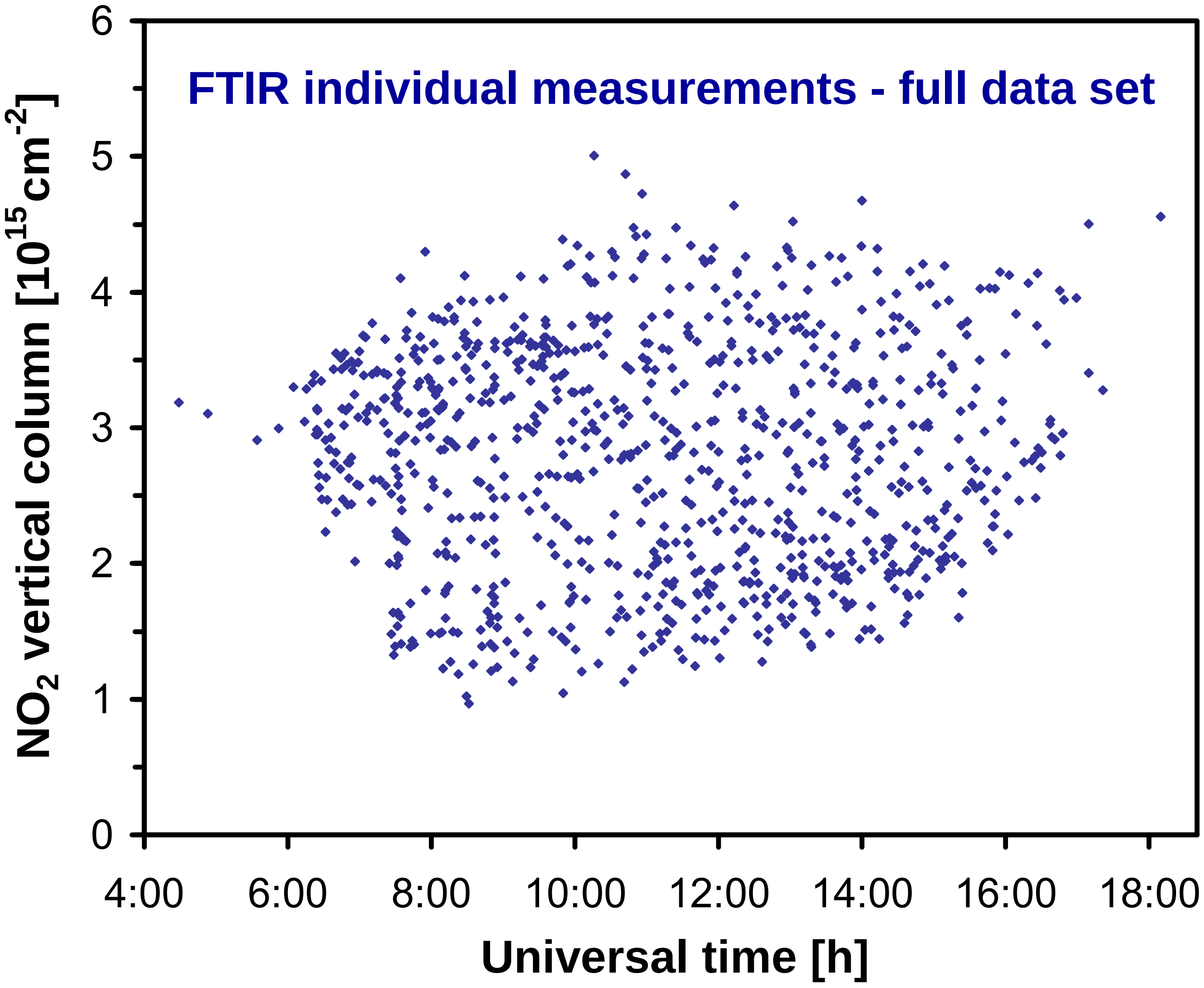
	Zugspitze FTIR @ 2.964 km asl.			FTIR @ 1.077 km asl.	SCIAMACHY		
Scenario	1	2	3	3	1	2	3
x_{true}	$\mathbf{x}_{US}$	$\mathbf{x}_{US/2}$	$\mathbf{x}_{poll}$	$\mathbf{x}_{poll}$	$\mathbf{x}_{US}$	$\mathbf{x}_{US/2}$	$\mathbf{x}_{poll}$
$\frac{col_{true}(1.077-10\,km)}{col_{true}(1.077-100\,km)}$	0.053	0.053	0.955	0.955	0.053	0.053	0.955
$\frac{col_{ret}(1.077-100\,km)}{col_{true}(1.077-100\,km)}$	0.950	0.949	0.058	0.078	0.984	0.984	0.570
$\frac{col_{ret}(1.077-100\,km)}{col_{true}(10-100\,km)}$	1.003	1.002	1.239	1.656	1.039	1.039	12.54

Appendix 3:

Total NO_2 diurnal increasing rate: Does it change with season?

Flaud et al., GRL, 10, 1983 (strat. photochem. model):

"**diurnal increasing rate should be a function of the season** since the build up of N_2O_5 depends on the length of the night and because its rate of photo dissociation varies with solar elevation."



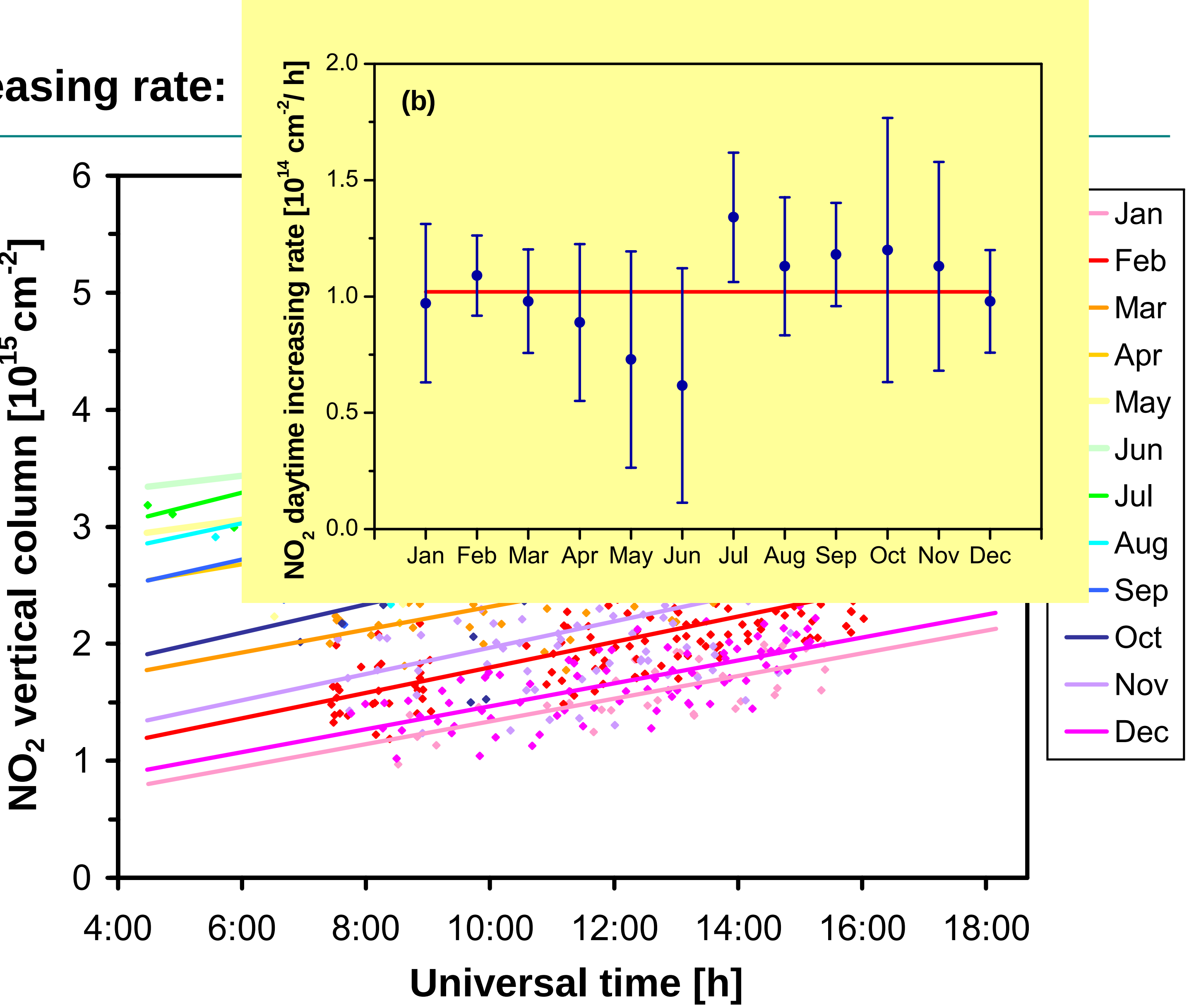
Appendix 4:

Total NO_2 diurnal increasing rate:

Susmann et al., ACP, 5, 2005:

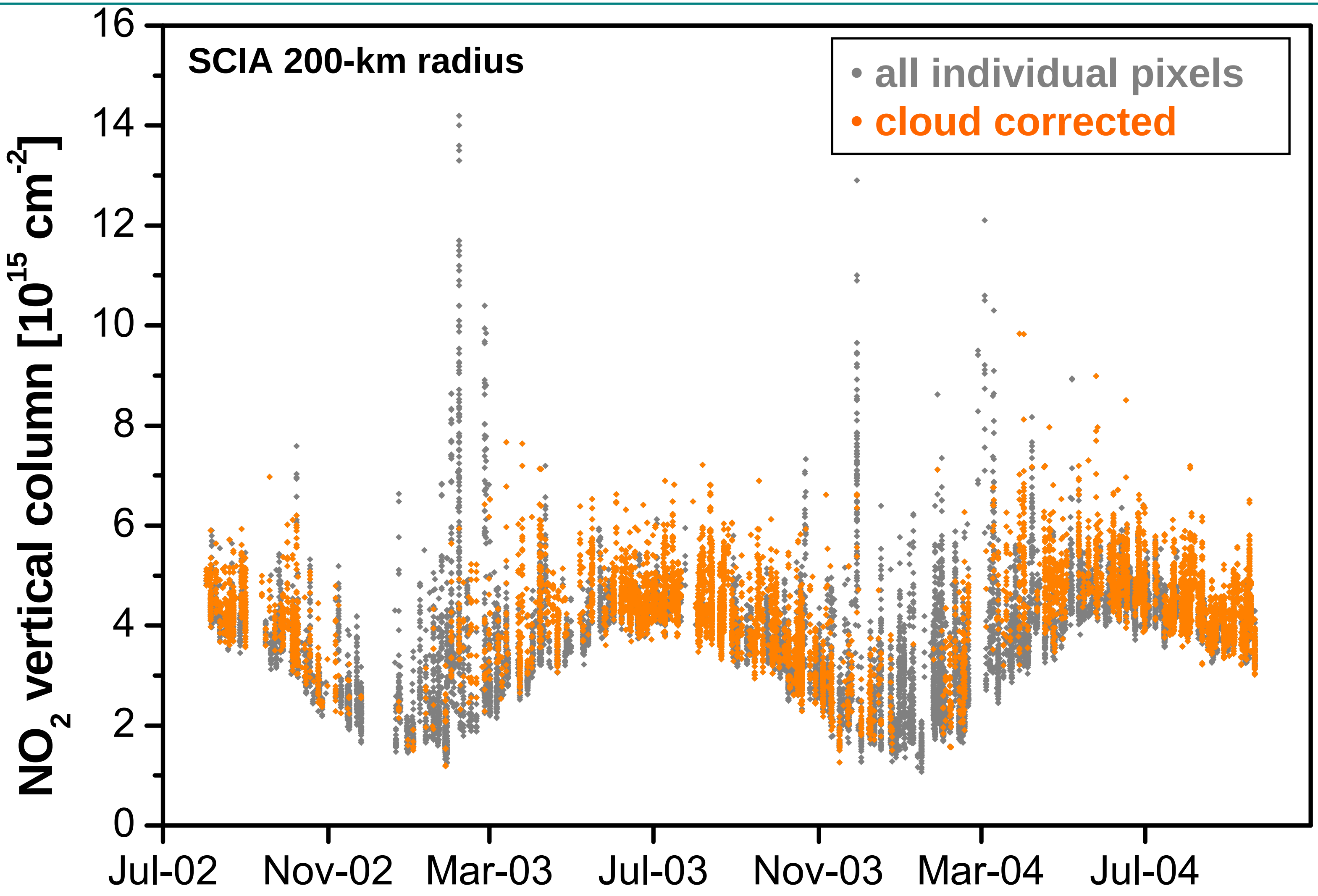
„No annual cycle of diurnal increasing rate found“

⇒ Mean diurnal increasing rate = + 1.02E+14 cm^{-2}/h for mid-latitudes



Appendix 5:

SCIA Cloud Correction Scheme:



Appendix 6:

FTIR versus SCIAMACHY: Statistical erros and day-to-day scatter

	$AV_i(n_i)$	$AV_i(\sigma_i)$	$AV_i(\sigma_i/\mathrm{sqrt}(n_i))$	σ of daily means corrected for annual cycle
Zugspitze FTIR	4.6	8.8 %	4.3 %	9.2 %
SCIAMACHY (200 km, poll. corr.)	22	6.8 %	1.9 %	6.5 %

agres with ≈5-10 % individual pixel precision above Pacific (Richter et al., 2004)

Appendix 7:

Analytic function to fit NO_2 annual cycles