

New retrieval approach to tropospheric NO₂ by synergistic inversion of satellite nadir DOAS soundings and ground-based FTIR measurements

R. Sussmann¹, W. Stremme¹, J.P. Burrows², A. Richter², W. Seiler¹, and M. Rettinger¹

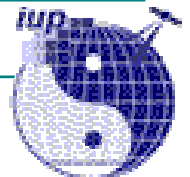
¹*Forschungszentrum Karlsruhe, IMK-IFU, Garmisch-Partenkirchen, Germany*

²*Universität Bremen, Institut für Umweltphysik*

+49 8821 183 159, Ralf.Sussmann@imk.fzk.de

Motivation - why another approach to tropospheric NO₂?

- **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 assimilation
 - **Heland et al., 2002; Petritoli et al., 2004; Boersma et al., 2004:** significant errors in trop. NO₂ retrievals
 - **Lambert et al., 2004:** lack of direct measurements of trop. NO₂ column for validation purposes
 - **Richter; Lambert; Sussmann (ACVE-2, 2004):** ground-based DOAS and FTIR disagree as to total NO₂
 - **Rinsland et al., 2003:** possible strong a priori contribution to FTIR retrieval of total NO₂
 - **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 NO₂ from combined SCIAMACHY and Zugspitze FTIR retrieval



Definitions: Total column averaging kernels (row vectors a^T)

$$col_{ret} = col_a + a^T \cdot (x_{true} - x_a)$$

with $col_a = (111\dots 1) x_a$,
and neglecting measurement errors
and forward model errors

Case (i). $a^T = (111\dots 1)$

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

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

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

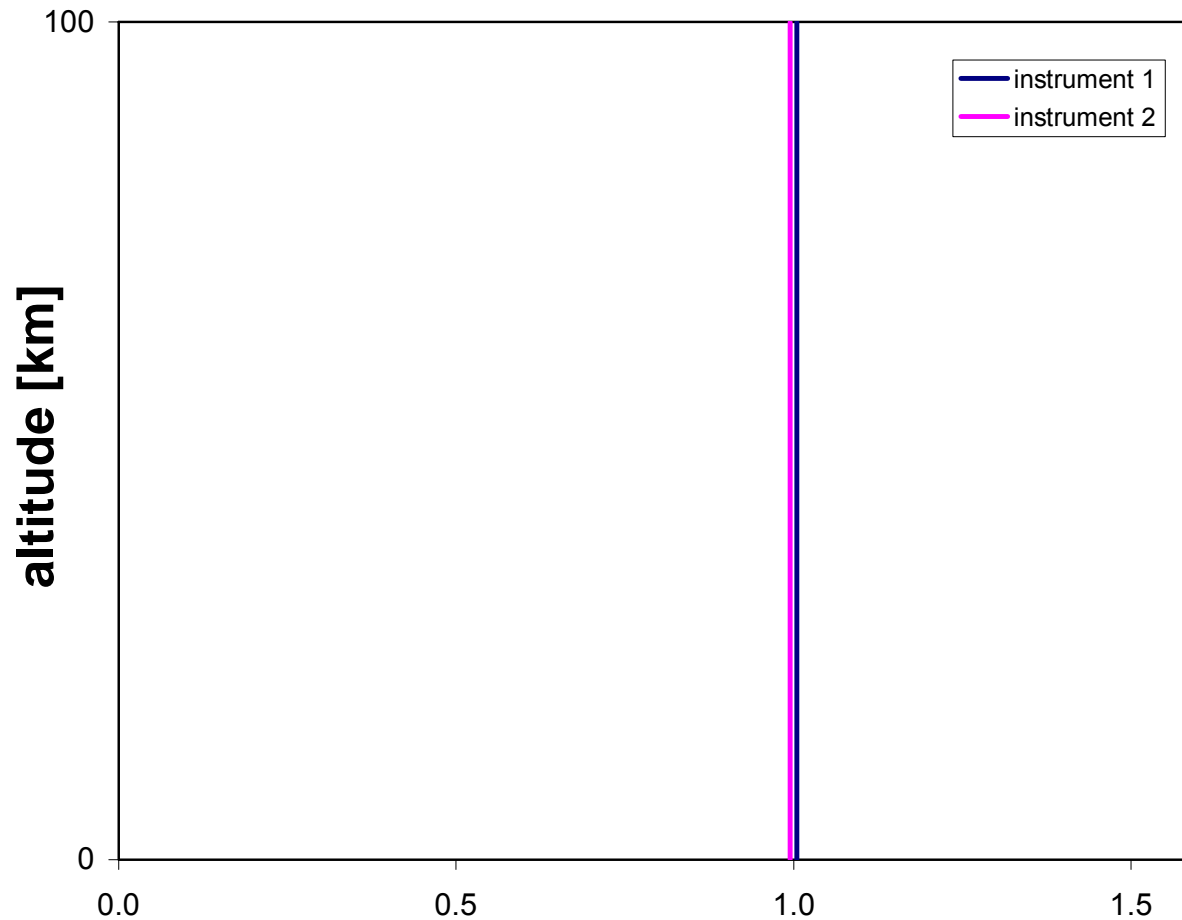
Case (iii). $x_{true} = x_a$

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

taken from:

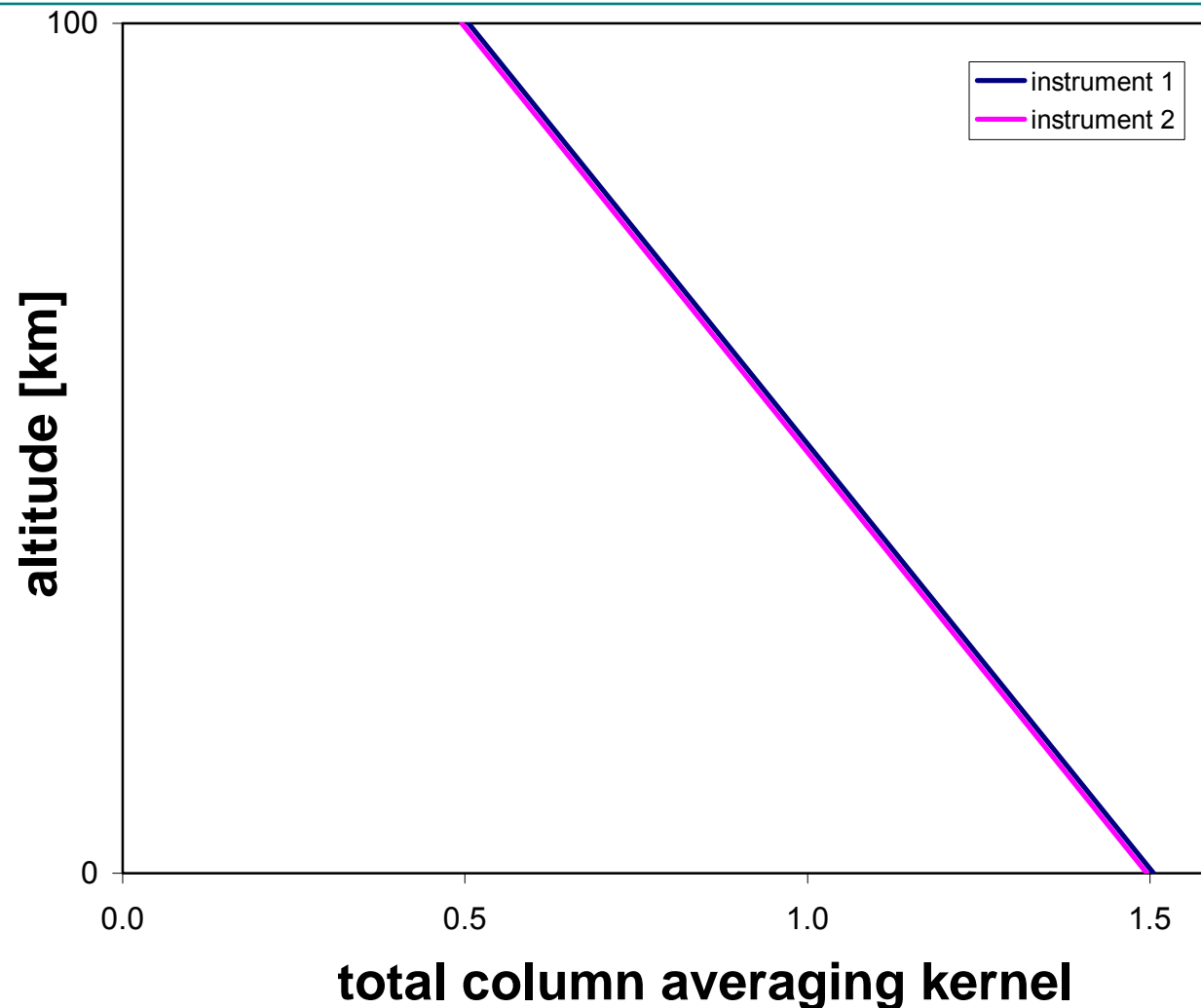
Sussmann, R.: Ground-based Fourier transform spectrometry at the NDSC site Zugspitze: Geophysical products for satellite validation, in *Proceedings of the European Symposium on Atmospheric Measurements from Space*, ESTEC, Noordwijk, The Netherlands, 18-22 Jan 1999, WPP-161, Vol. 2, pp. 661-664, 1999.

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



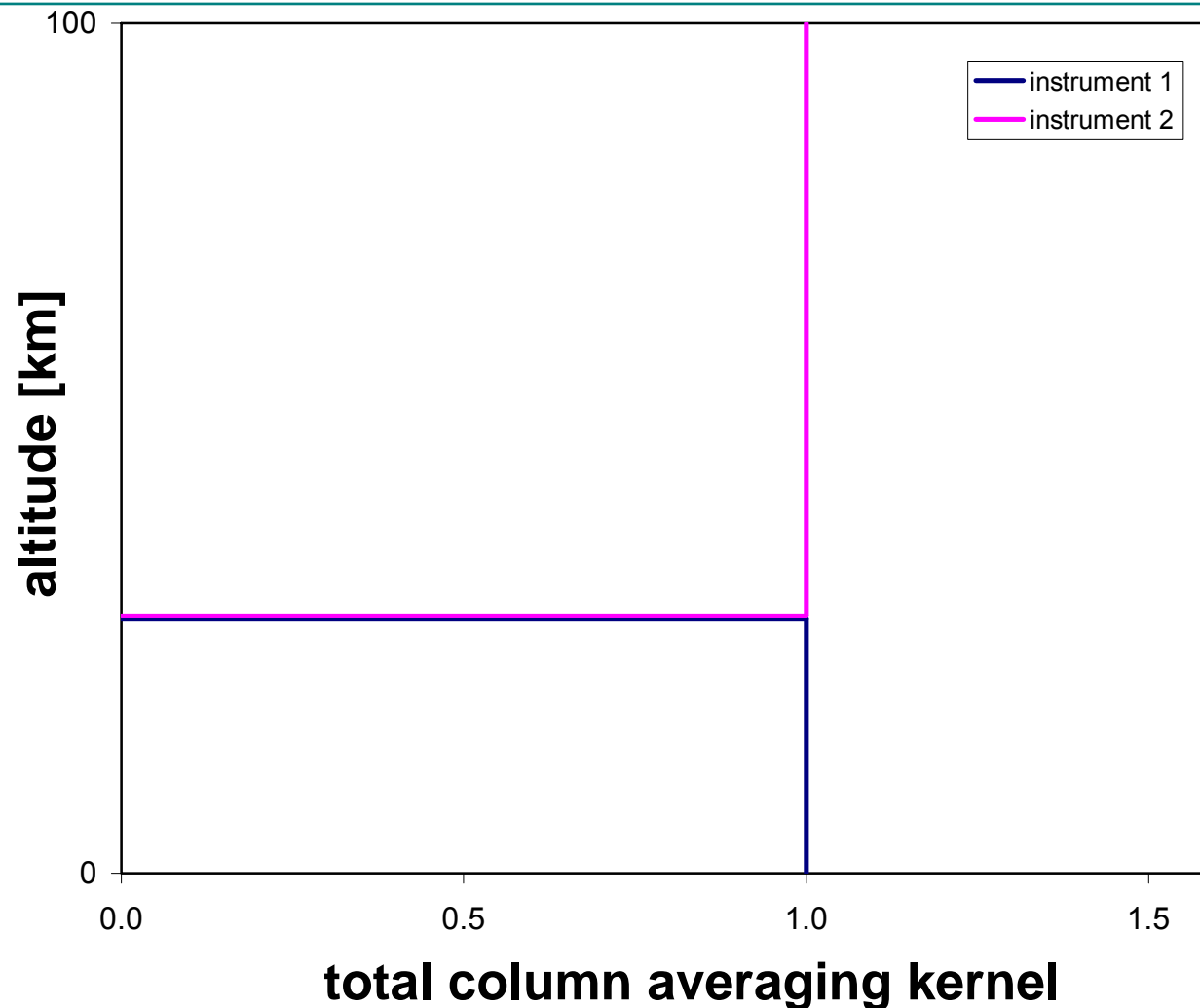
Example 1:
ideal instruments

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



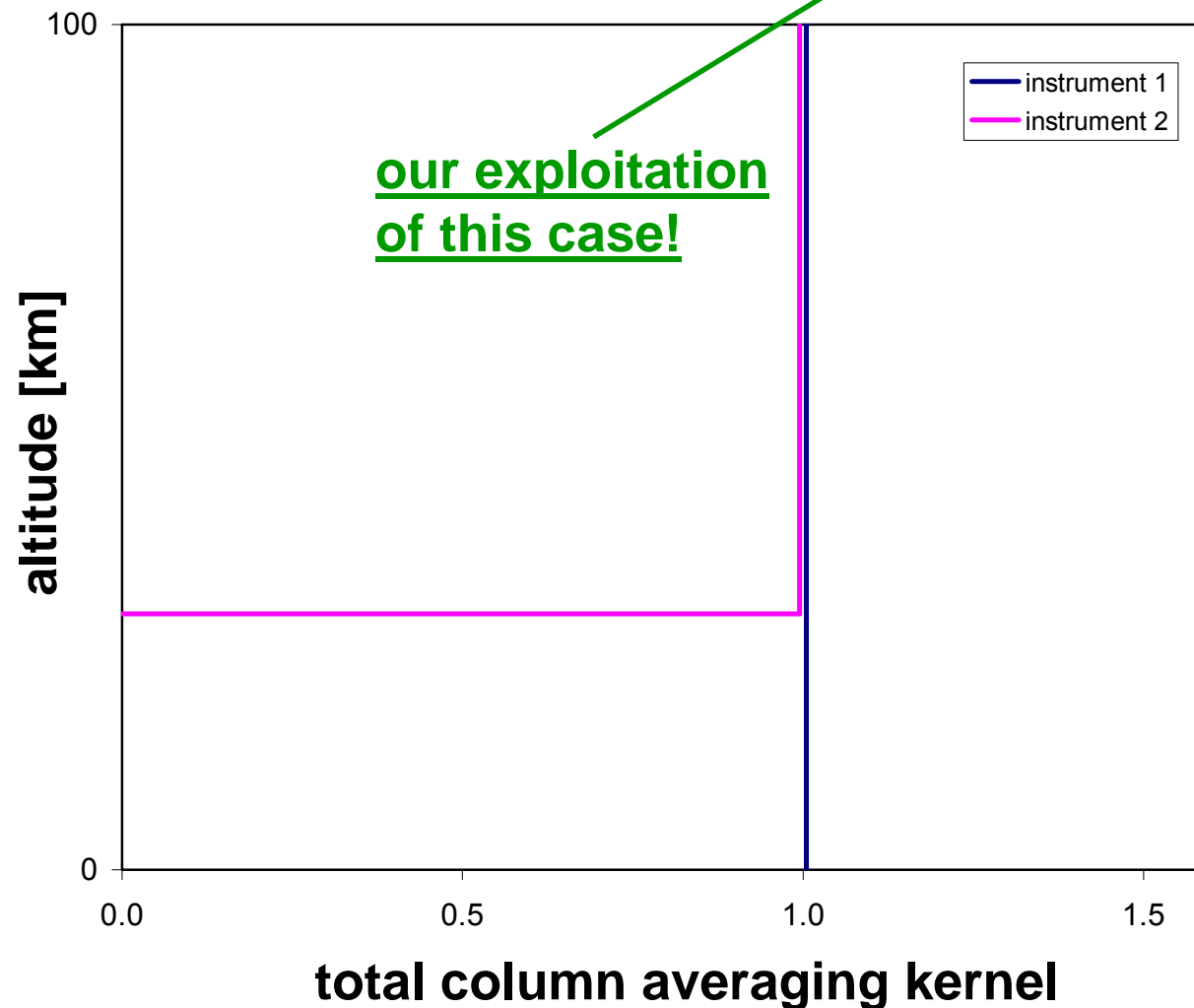
Example 2:
non-ideal instruments

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



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:

$$a_i = \frac{AMF(x_{ref,i}, \mathbf{b})}{AMF(x_{ref}, \mathbf{b})}$$

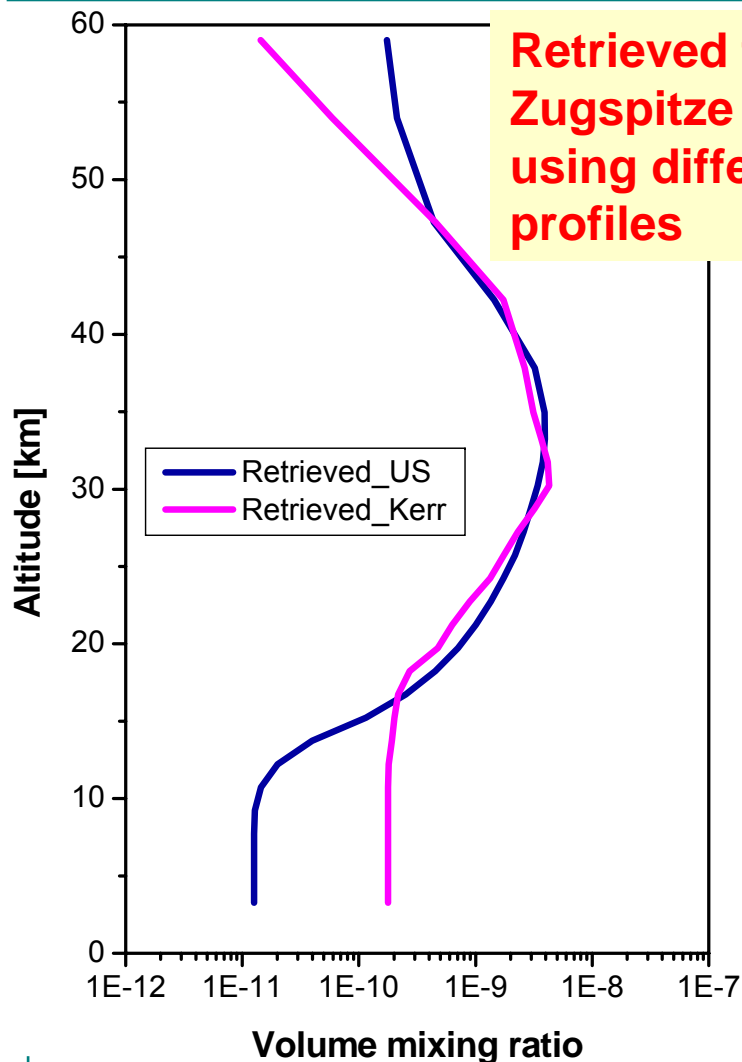
i : layer index
 x_{ref} : reference state (e.g., a priori NO₂ profile)
 \mathbf{b} : SZA, viewing angle, clouds, albedo, aerosols, ...

Have to decide upon NO₂ a priori profile:

**DOAS AMF strongly depends on a priori NO₂ profile shape in the troposphere,
DOAS AMF does nearly not depend on a priori NO₂ profile in the stratosphere**

**⇒ DOAS a priori := US Standard NO₂ profile with VMR = 0 up to 10 km
(A. Richter: SCIAMACHY UB1.5 total NO₂ retrieval)**

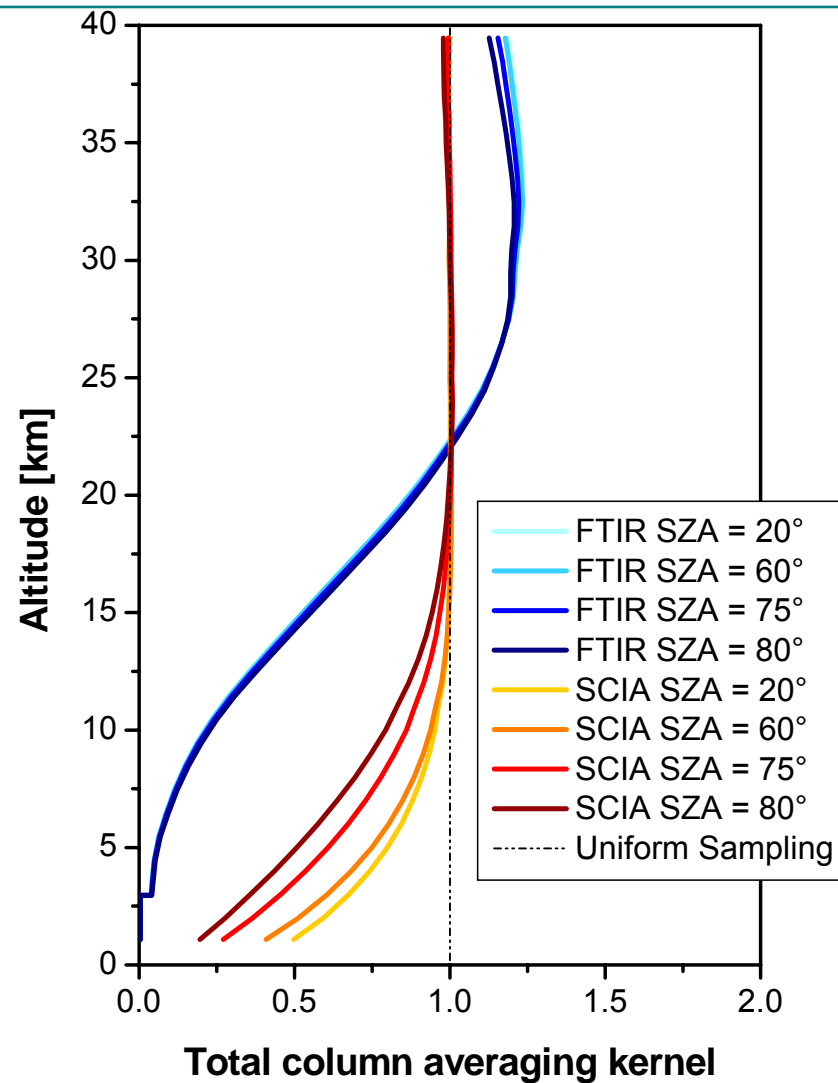
Total column averaging kernels: NO_2 a priori profile for FTIR?



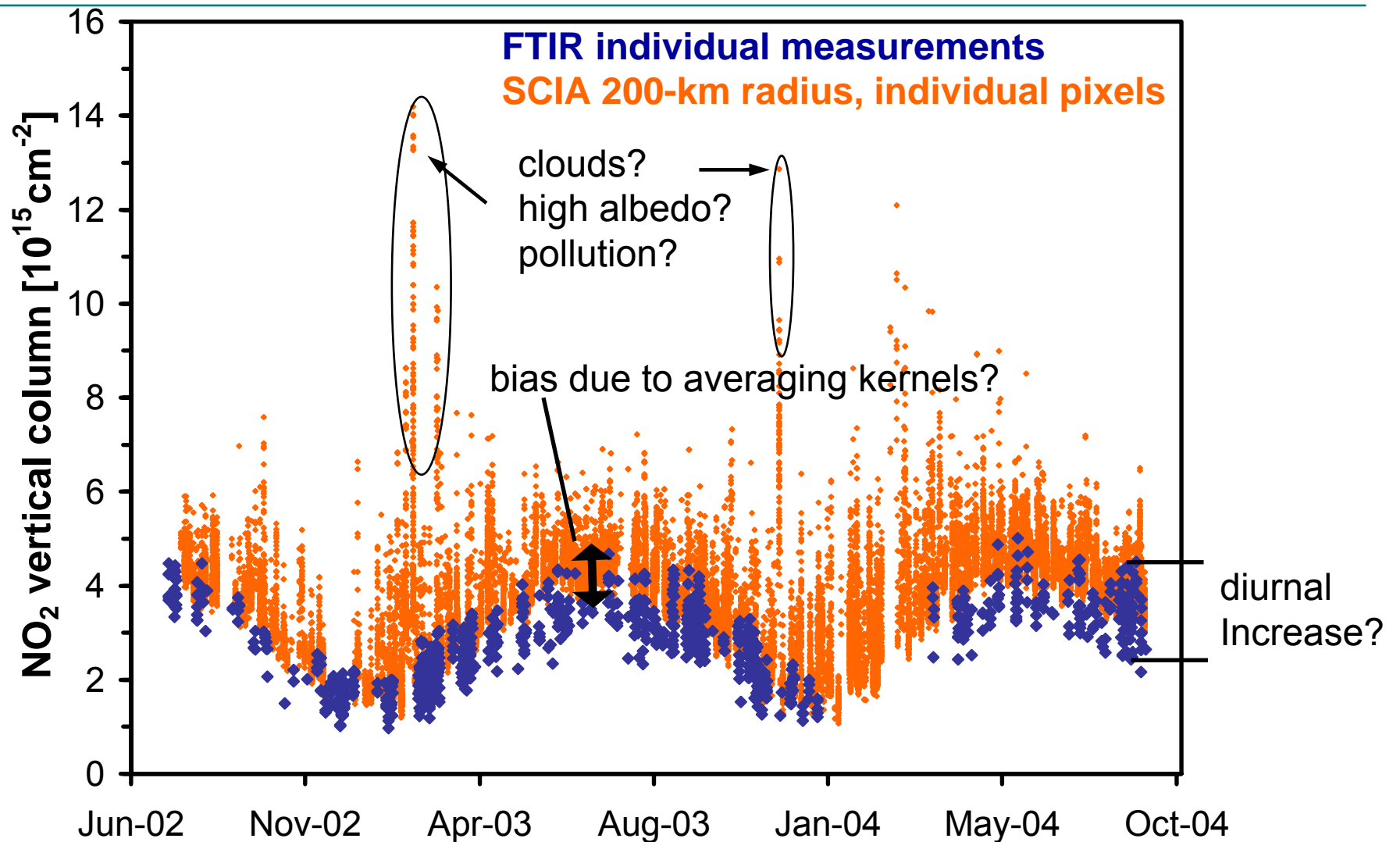
Solar FTIR retrieval not sensitive in the troposphere, thus strongly depends on a priori NO_2 profile shape in the troposphere

⇒ 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



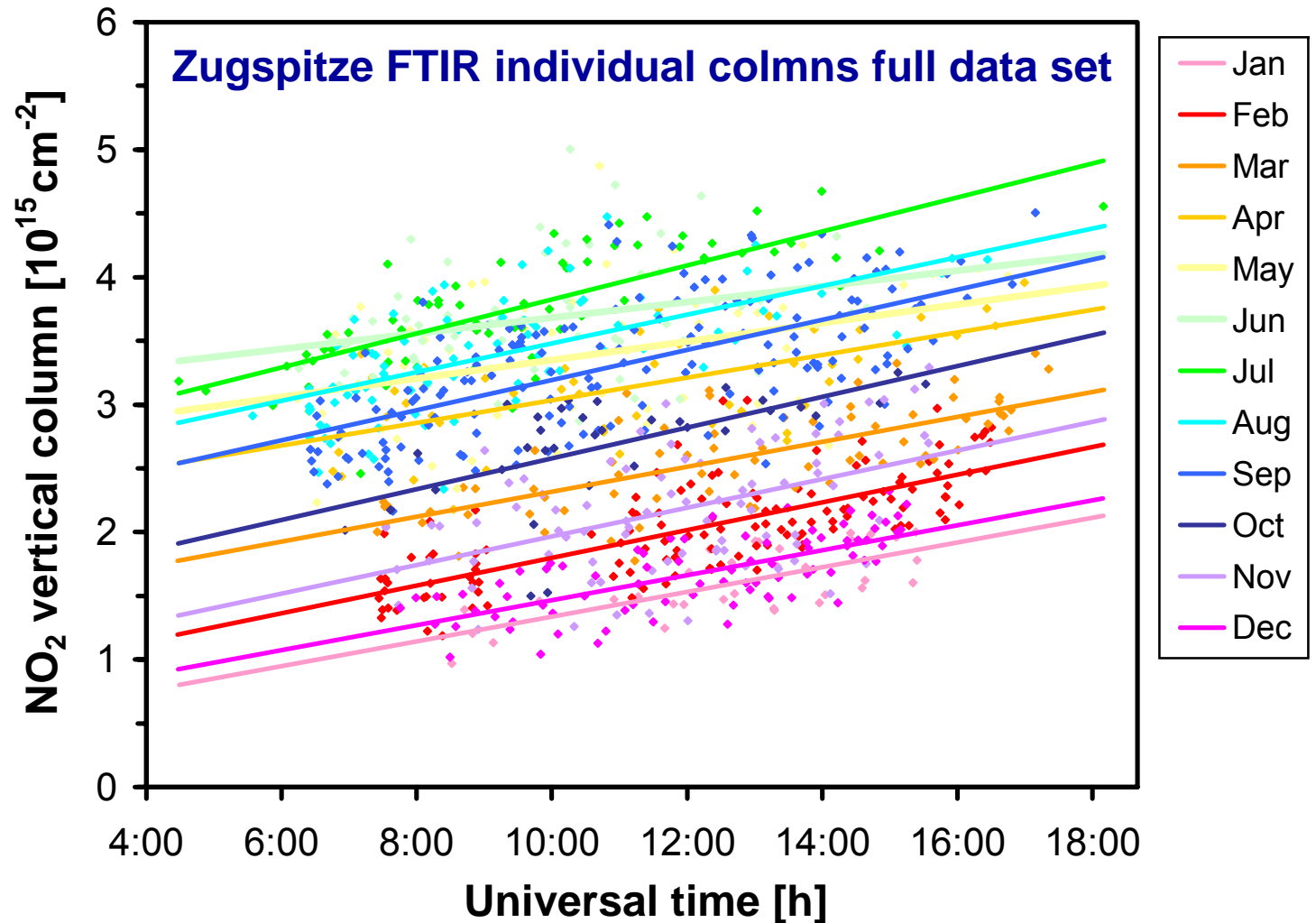
SCIA UB1.5 versus Zugspitze FTIR total columns: Direct comparison



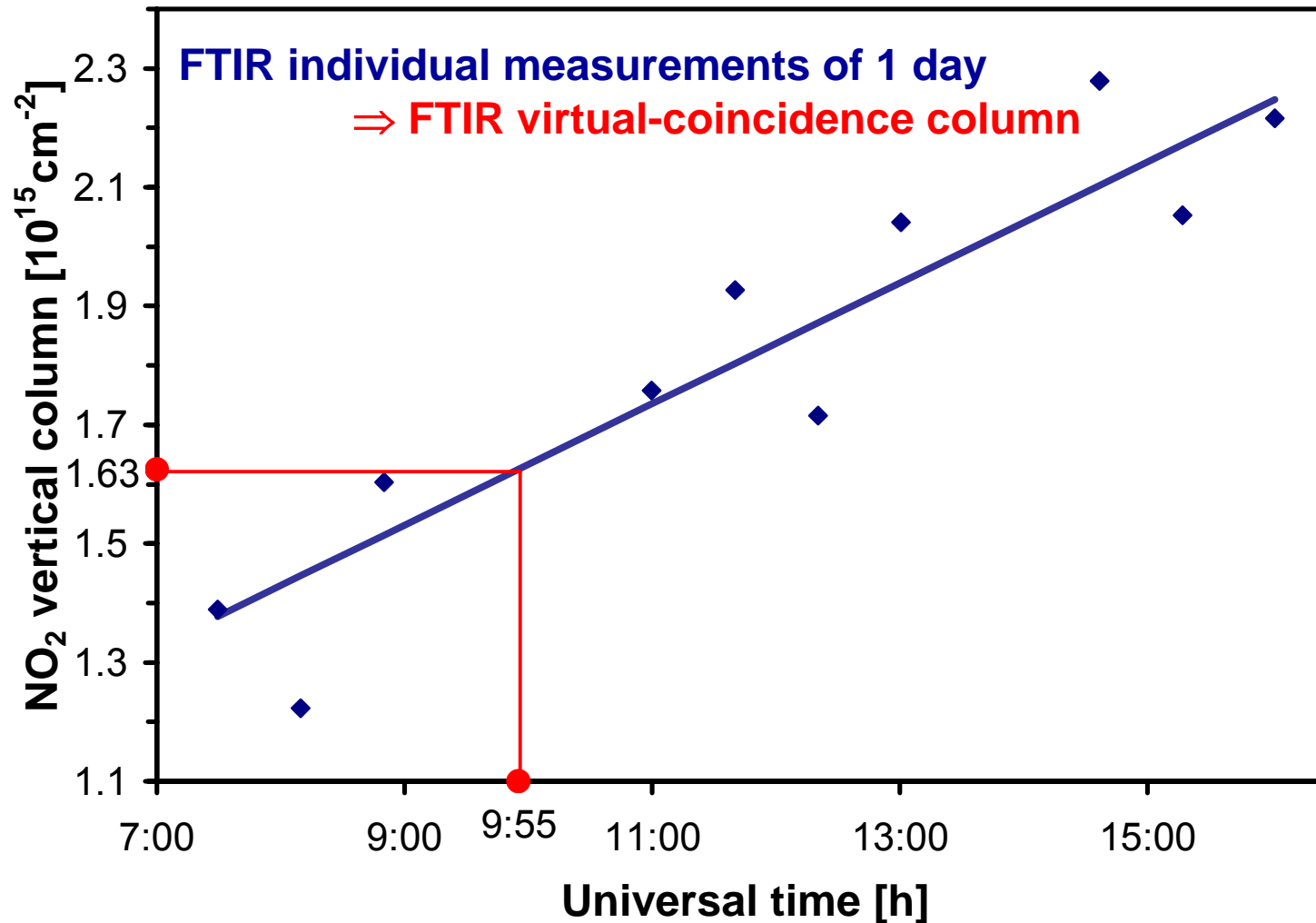
Total NO₂ diurnal increasing rate: Does it change with season?

„No annual cycle of diurnal increasing rate found“

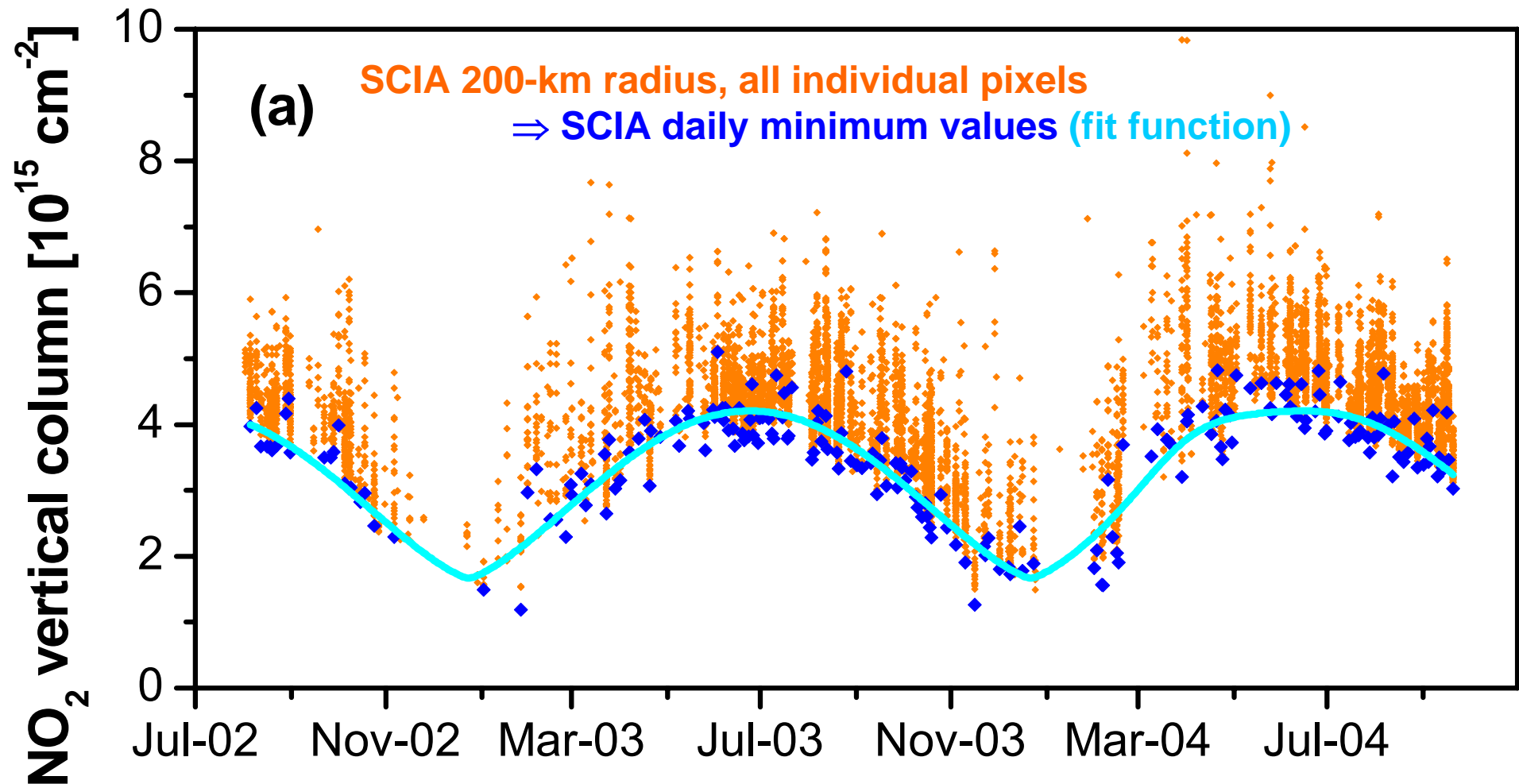
⇒ Mean diurnal increasing rate for mid-latitudes = + 1.02(6)E+14 cm⁻²/h



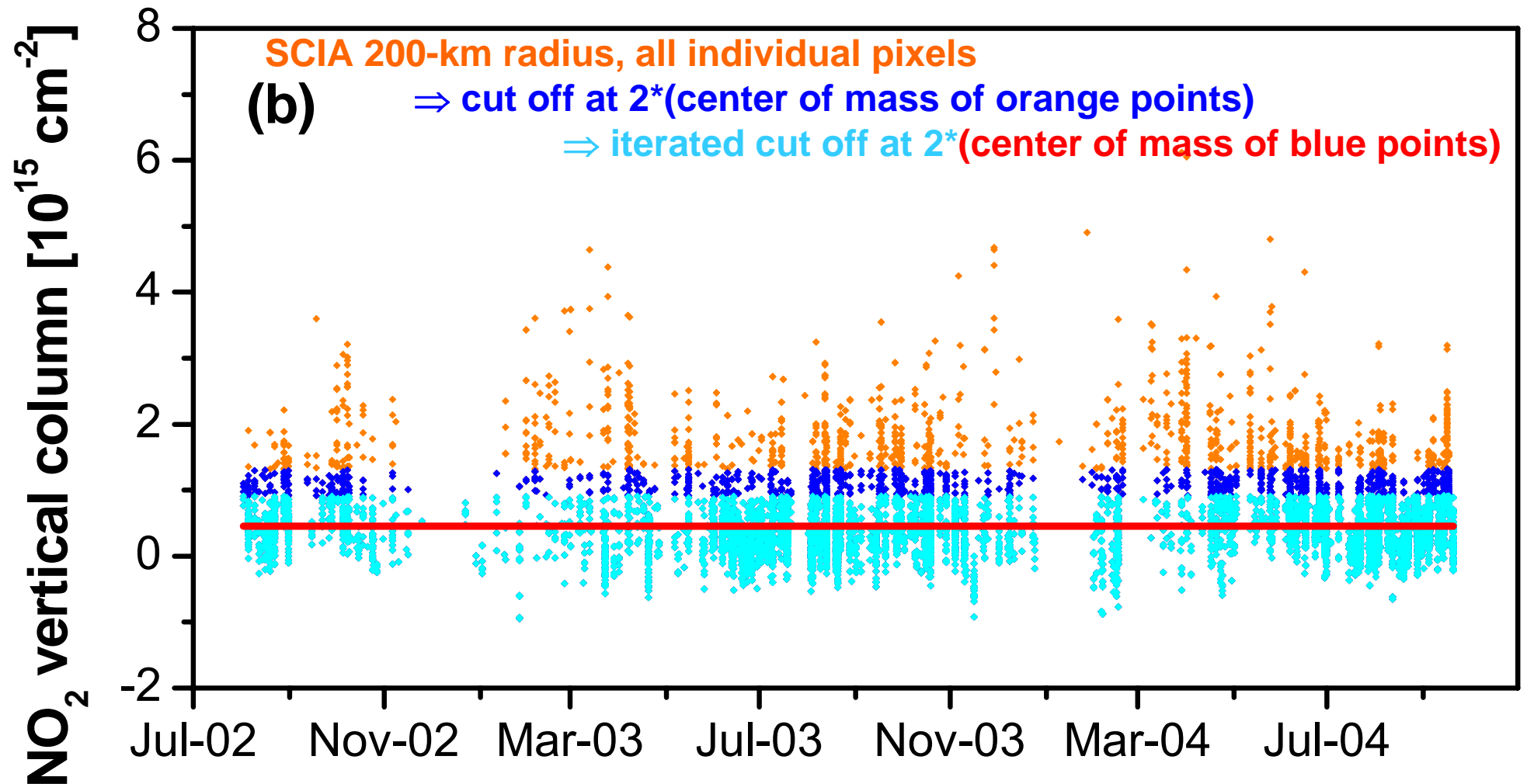
Matching FTIR to satellite overpasses: Concept of „virtual coincidences“



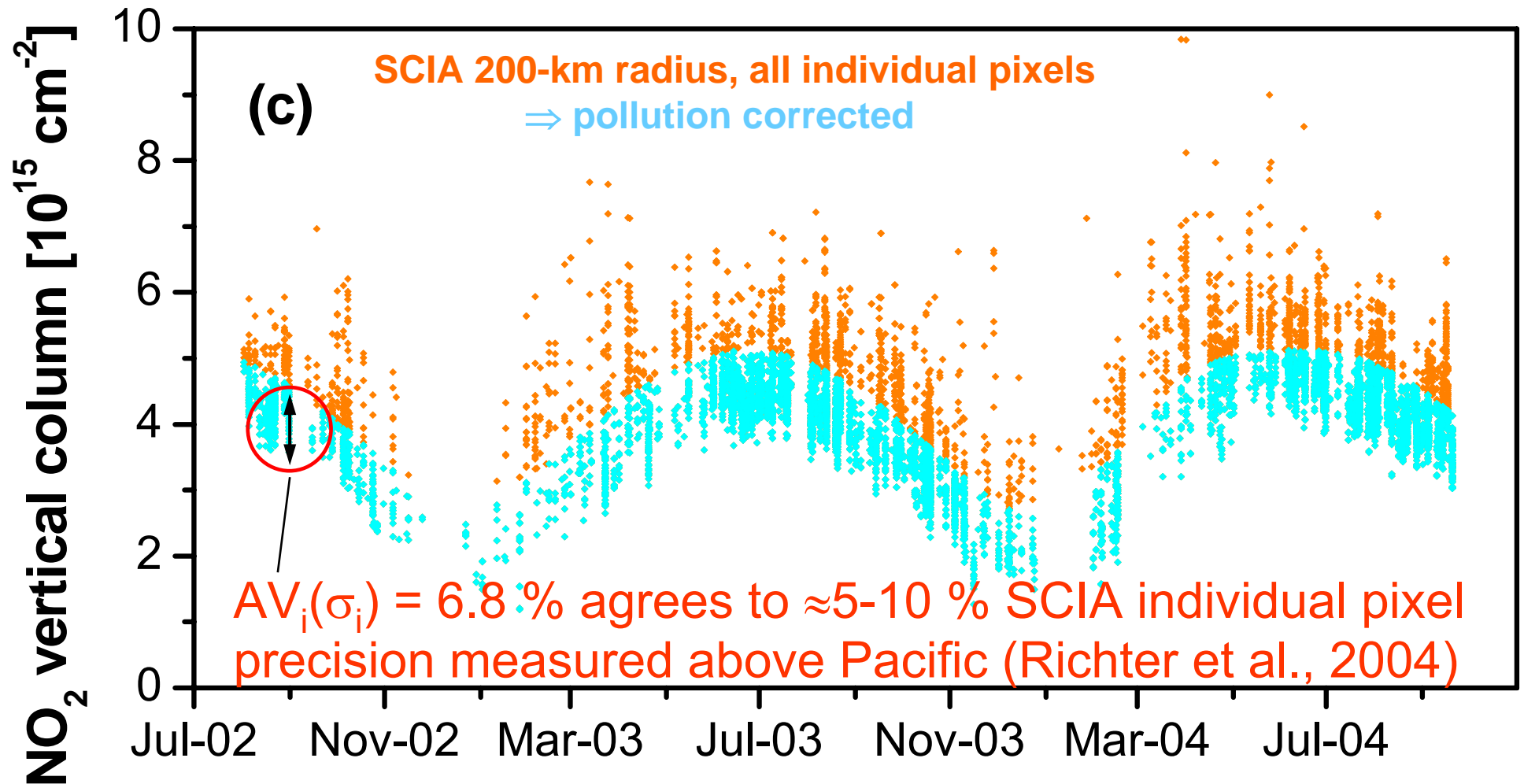
Satellite Pollution Correction Scheme: (a) Subtract fit to daily minima



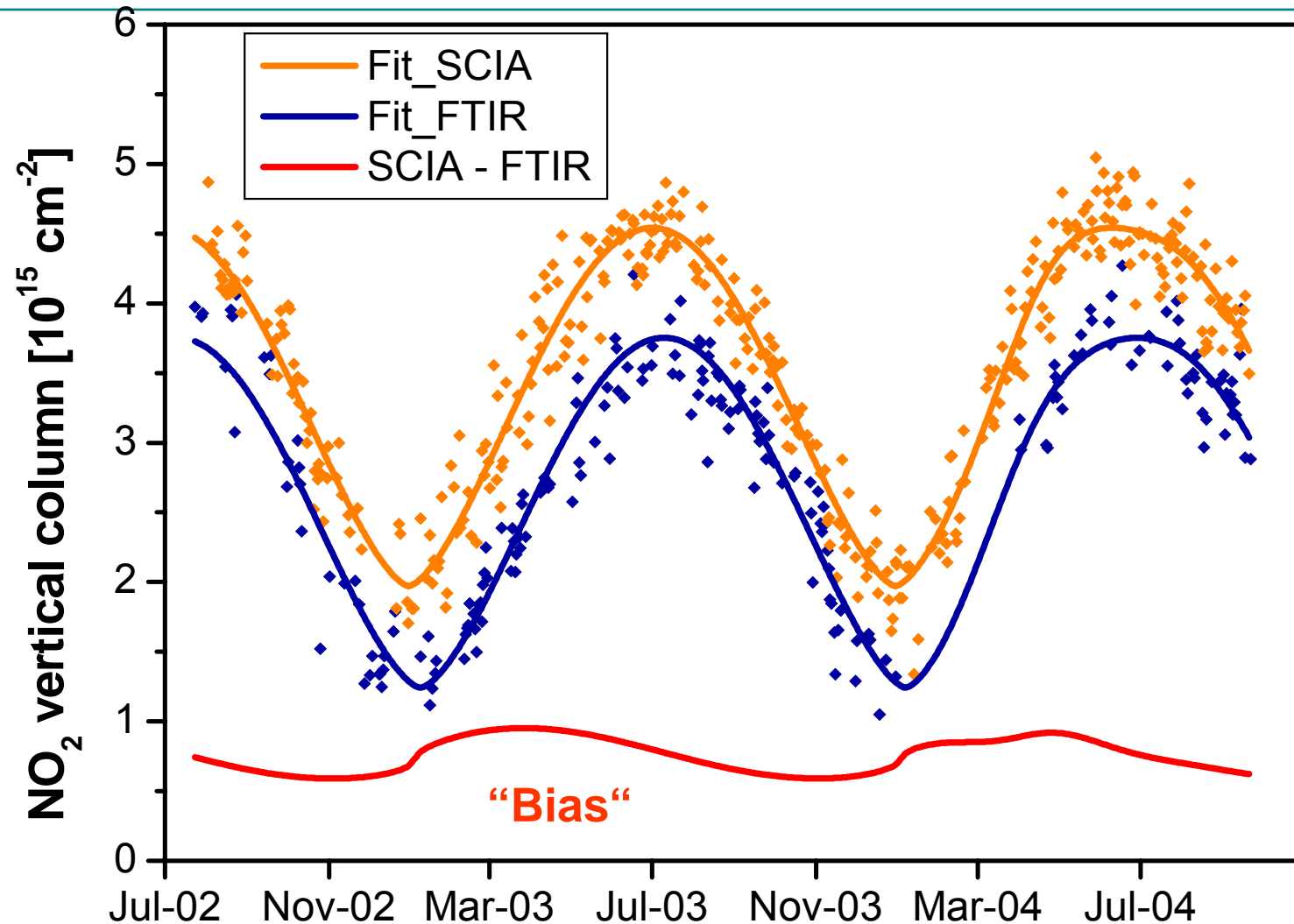
Sat. Pollution Correction Scheme: (b) Cut off at 2*(center of mass) and iterate



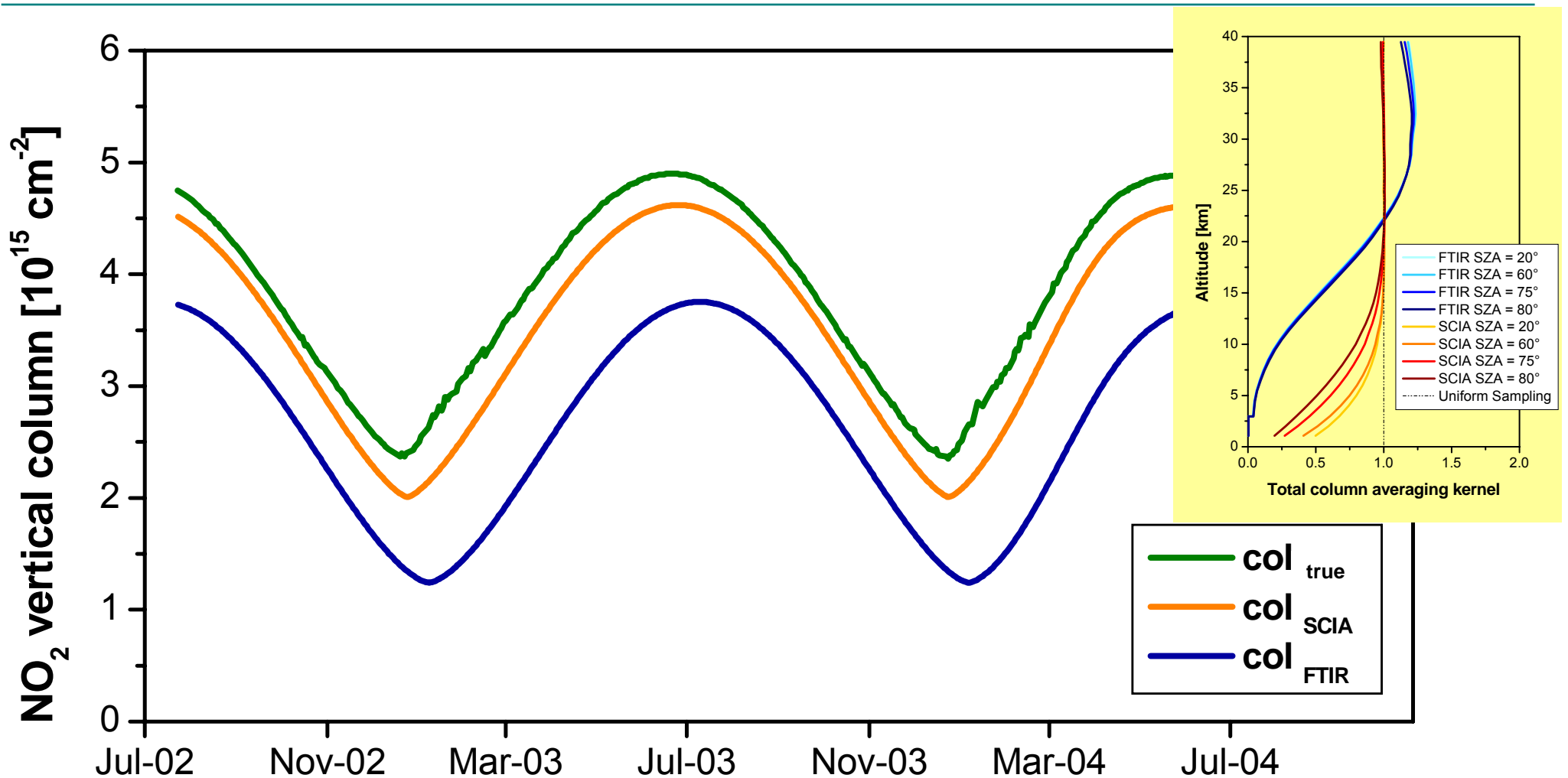
Sat. Pollution Correction Scheme: (c) Re-add fit-function (ann. cycle of minima)



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



Combine FTIR and SCIAMACHY to retrieve tropospheric NO₂: The Idea



Combined a posteriori retrieval of tropospheric NO₂: Constraint

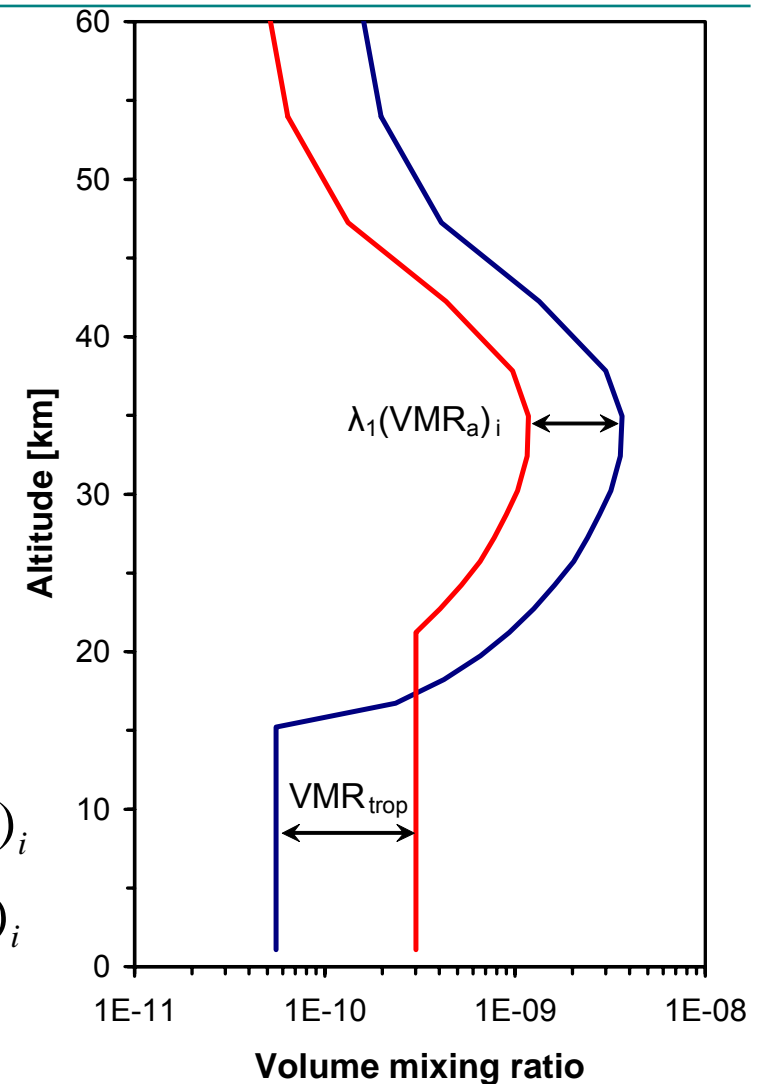
Our a posteriori retrieval constraint is set up as follows

$$\mathbf{x}_{true} = \lambda_1 \cdot \mathbf{x}_a + \mathbf{x}_{trop} \quad \text{with}$$

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

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 NO₂: 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.,

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

and

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

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

Combined a posteriori retrieval of tropospheric NO₂: Retrieval equation

We derive a starting value for λ_1 by using

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

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

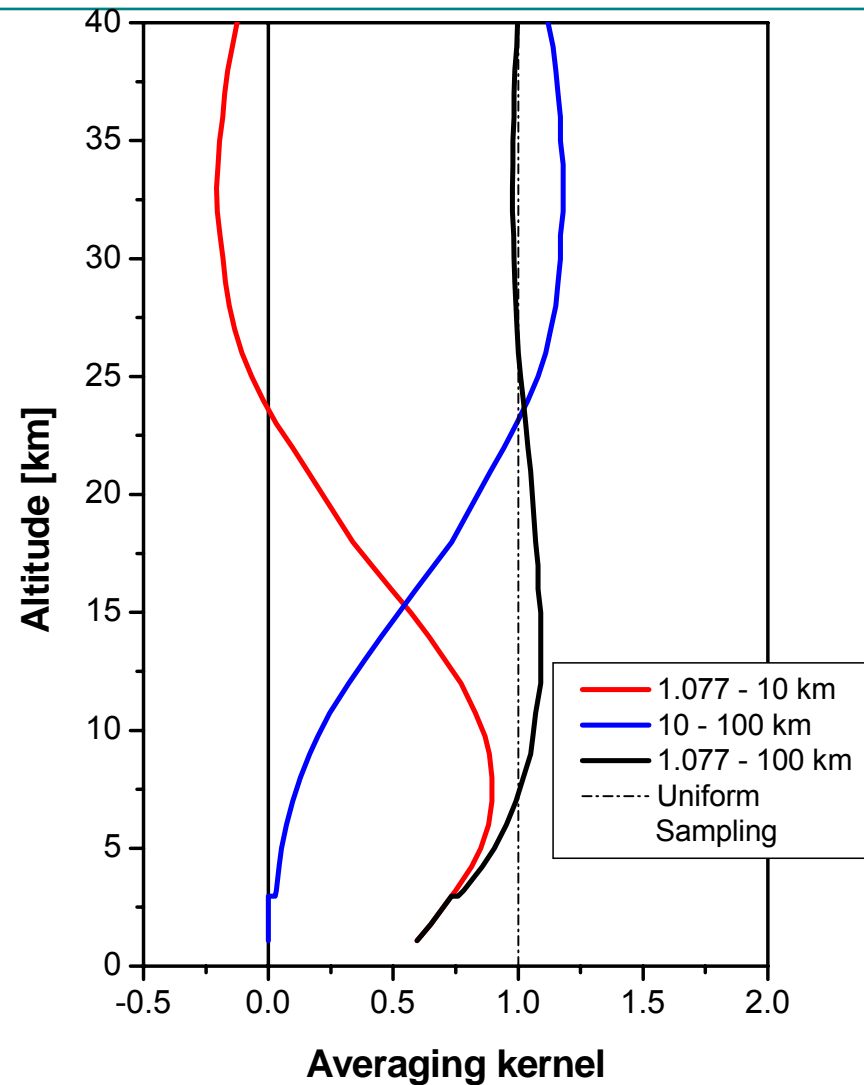
$$\lambda_1 = \frac{col_{FTIR}}{\mathbf{a}_{ideal}^T \cdot \mathbf{x}_a}$$

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

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

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

Characterization: Averaging kernels of the combined a-posteriori NO₂ retrieval

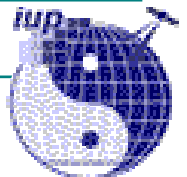


Forschungszentrum Karlsruhe, IMK-IFU, Garmisch-Partenkirchen

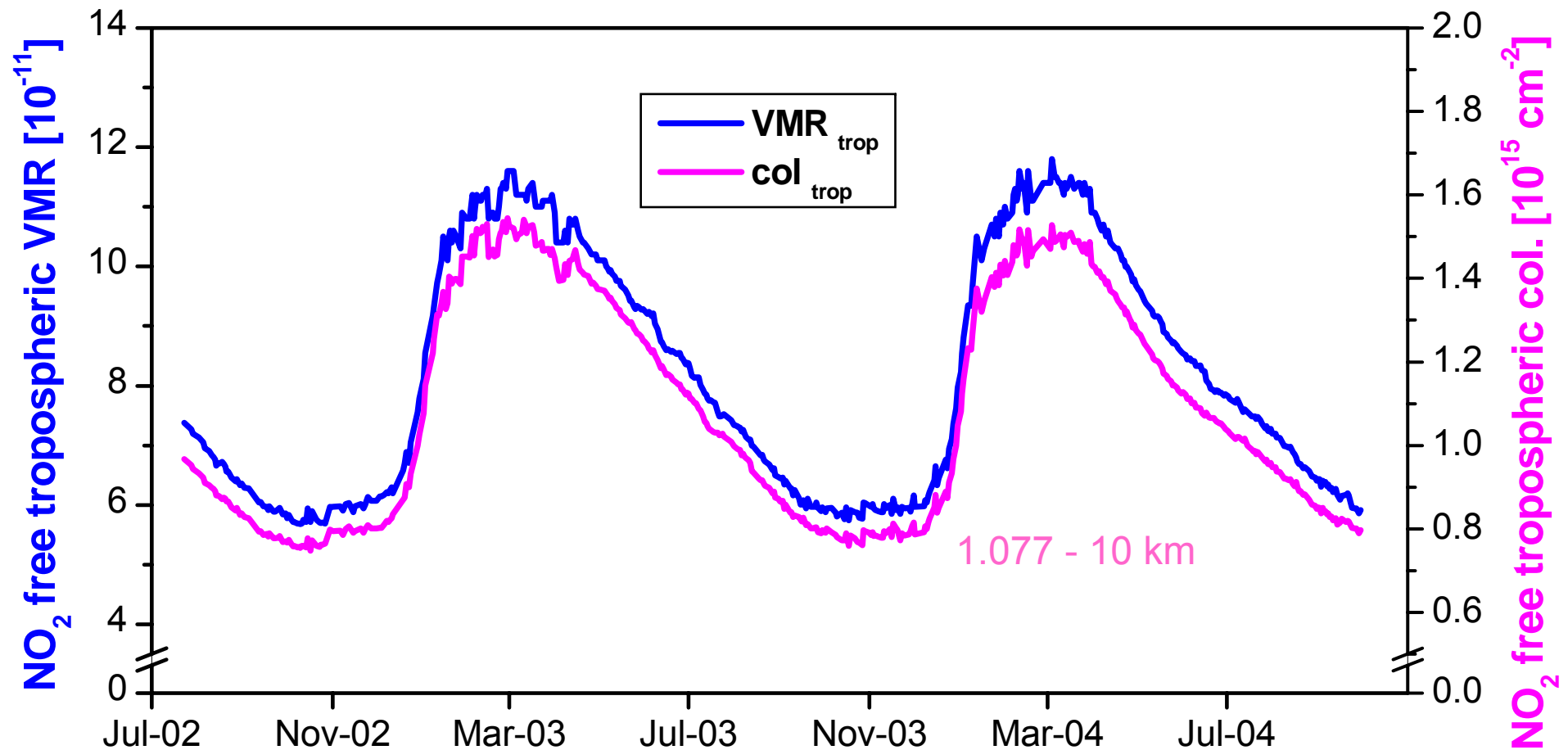
Sussmann et al.: *Trop. NO₂ from combined satellite and FTIR*



Universität Bremen



Combined a posteriori retrieval: Result for free tropospheric NO₂ column



Summary and Outlook

- stratospheric NO₂ diurnal increasing rate does not depend on season
- concept of FTIR “virtual-coincidence column”
- pollution clearing scheme for satellite NO₂ 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 NO₂ is between the phase of boundary layer NO₂, and stratospheric NO₂

Vision: integrated global NO₂ observing system based on synergistic use of nadir satellites and a set of ground-based FTIRs

Acknowledgments

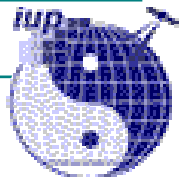
BMBF/DLR-GCVOS, EC-UFTIR, EC-ACCENT-AT2, ESA-TASTE

Forschungszentrum Karlsruhe, IMK-IFU, Garmisch-Partenkirchen

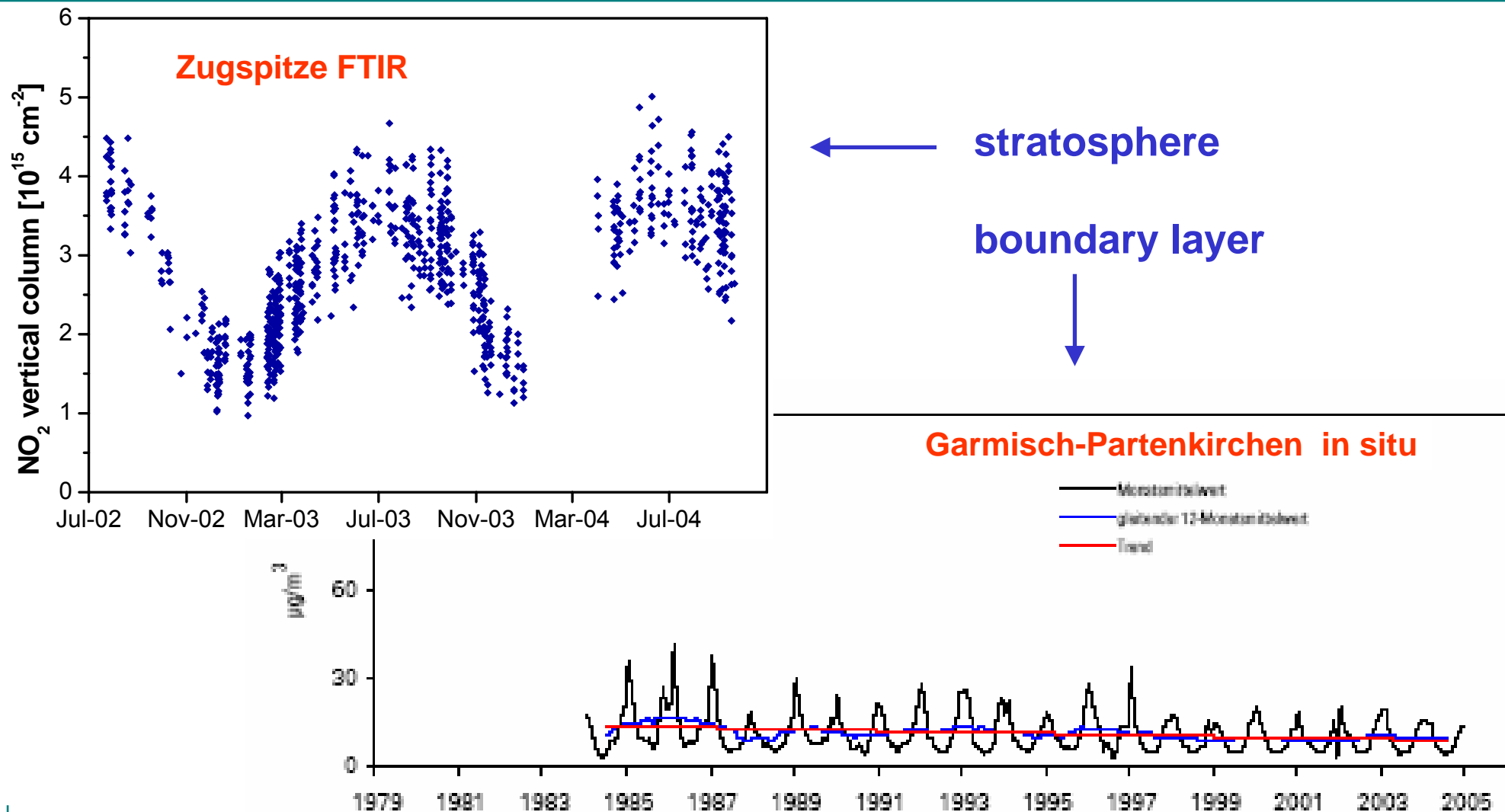
Sussmann et al.: *Trop. NO₂ from combined satellite and FTIR*



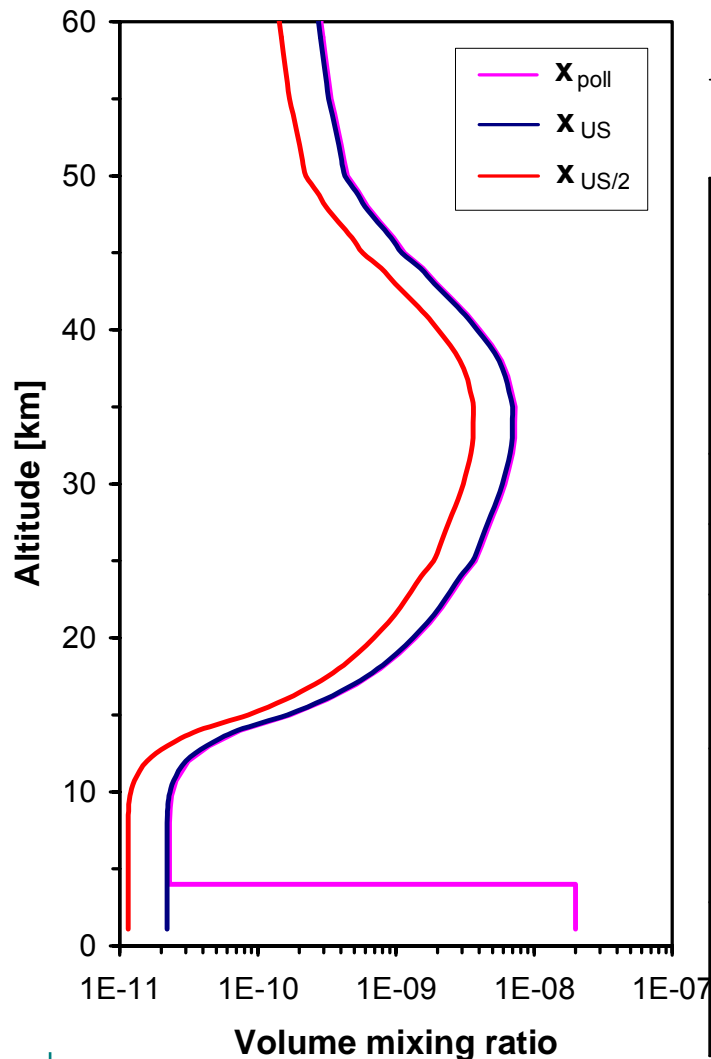
Universität Bremen



Combined a-posteriori retrieval: Annual cycle of free tropospheric NO₂ column?



Different sensitivity to tropospheric pollution: FTIR versus SCIAMACHY



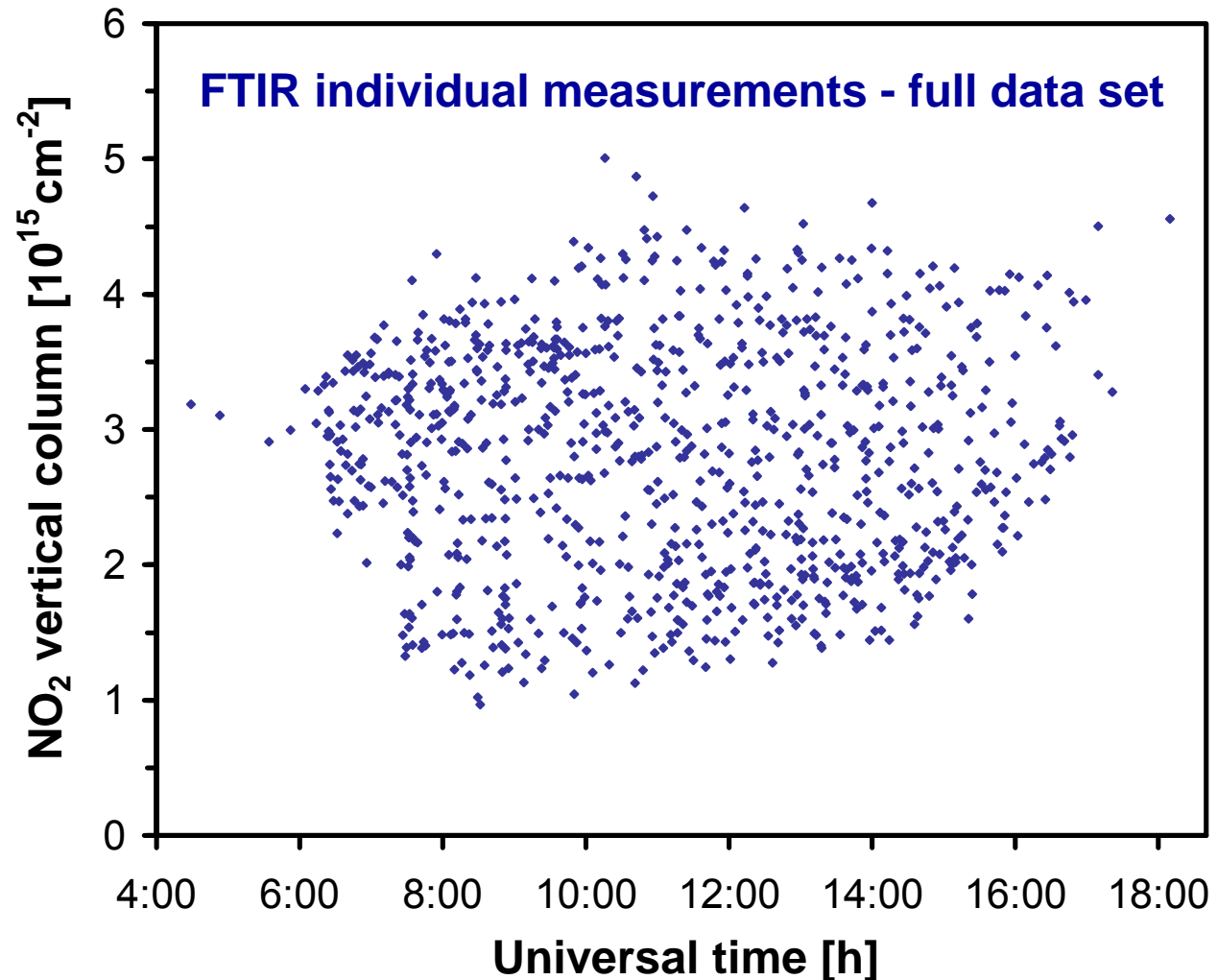
$$\frac{col_{ret}}{col_{true}} = \frac{\mathbf{a}^T (\mathbf{x}_{true} - \mathbf{x}_a) + \mathbf{a}_{ideal}^T \mathbf{x}_a}{col_{true}} \quad \text{with} \quad col_{true} = (111\dots 1) \mathbf{x}_{true} = \mathbf{a}_{ideal}^T \mathbf{x}_{true}$$

	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}
$\frac{col_{true}(1.077-10\text{ km})}{col_{true}(1.077-100\text{ km})}$	0.053	0.053	0.955	0.955	0.053	0.053	0.955
$\frac{col_{ret}(1.077-100\text{ km})}{col_{true}(1.077-100\text{ km})}$	0.950	0.949	0.058	0.078	0.984	0.984	0.570
$\frac{col_{ret}(1.077-100\text{ km})}{col_{true}(10-100\text{ km})}$	1.003	1.002	1.239	1.656	1.039	1.039	12.54

Total NO₂ 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 N₂O₅ depends on the length of the night and because its rate of photo dissociation varies with solar elevation.”



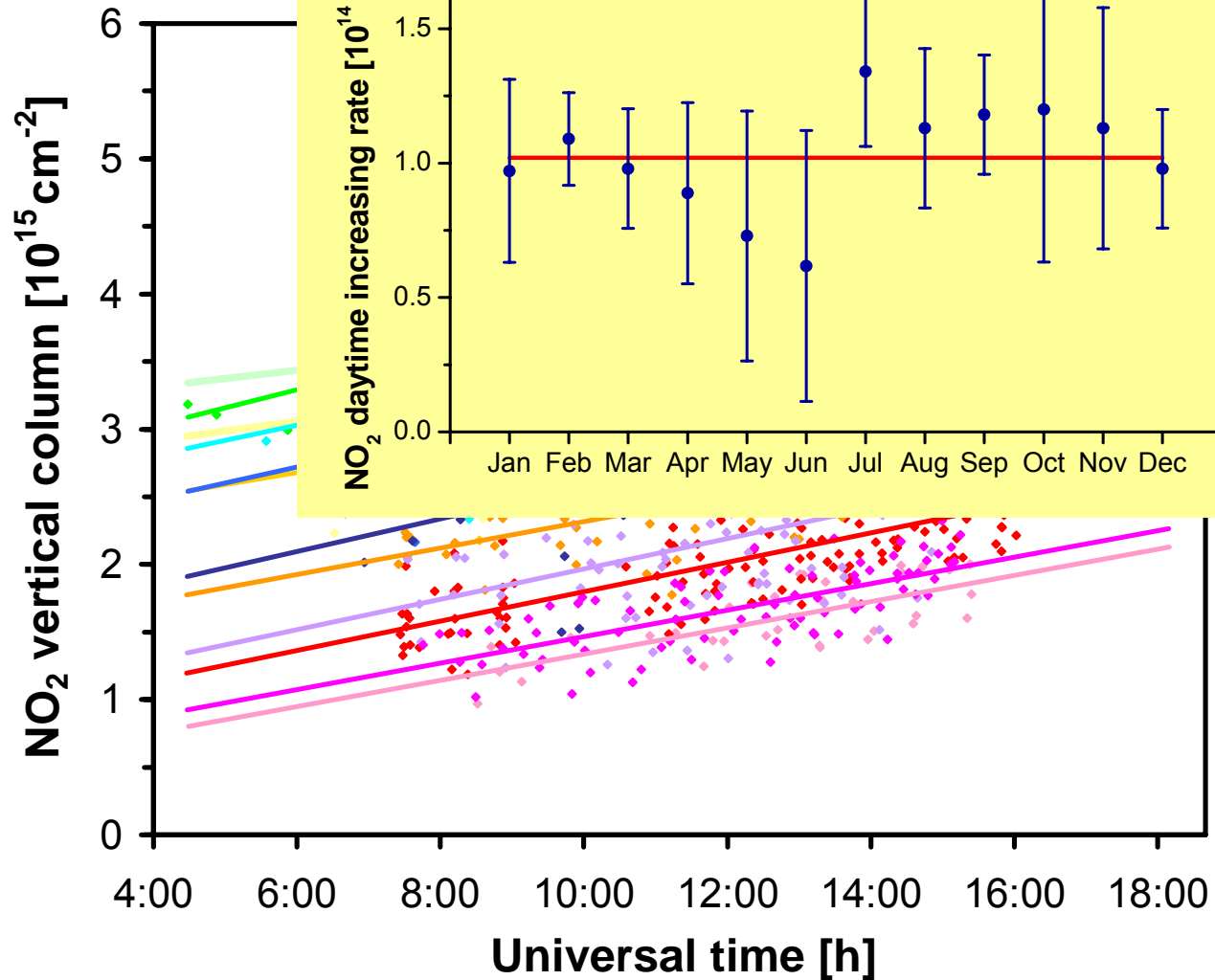
Appendix 4:

Total NO₂ diurnal increasing rate:

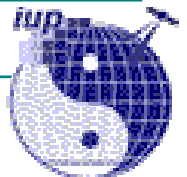
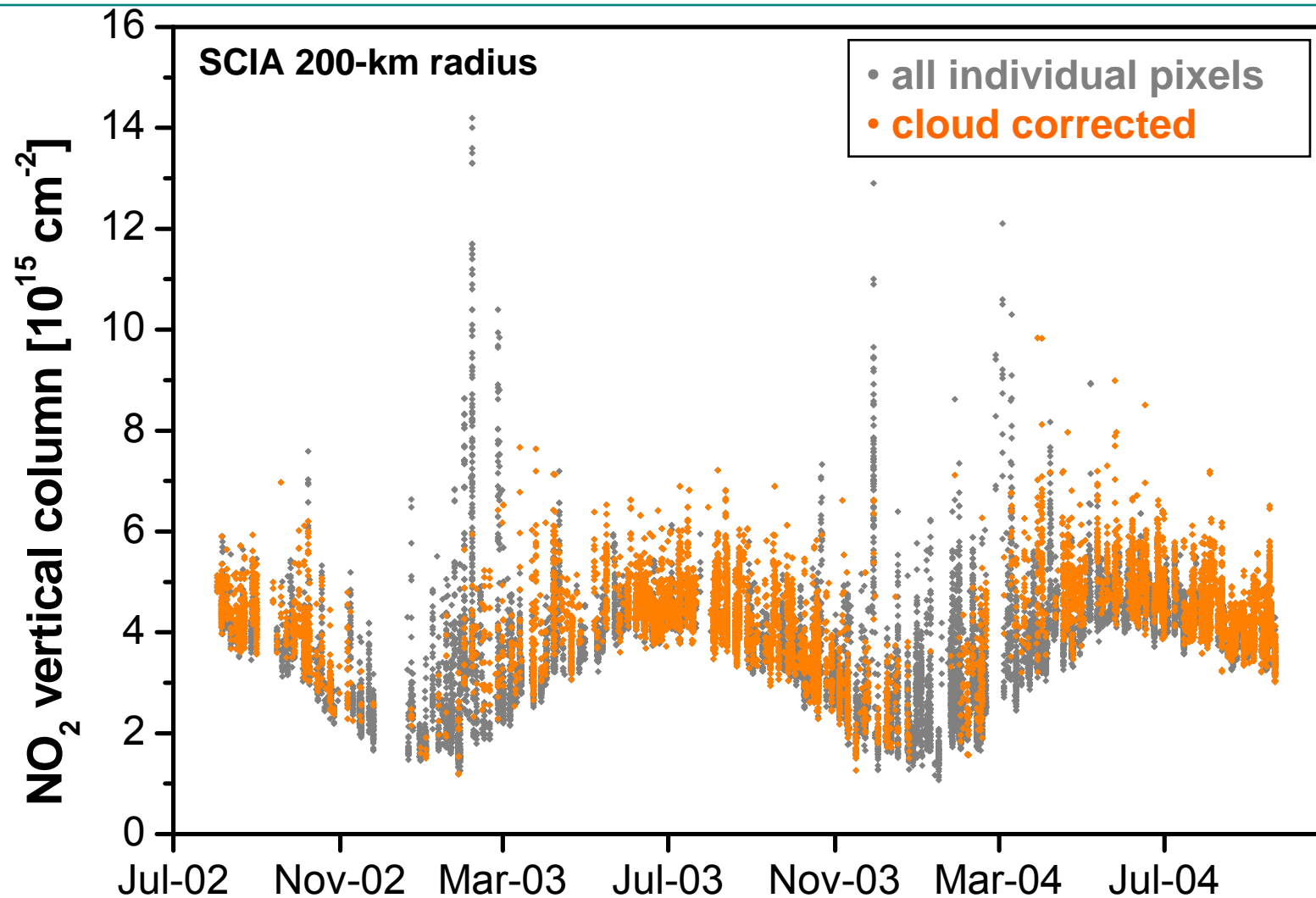
Susmann et al.,
ACP, 5, 2005:

„No annual cycle of
diurnal increasing
rate found“

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



SCIA Cloud Correction Scheme:



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

	$AV_i(n_i)$	$AV_i(\sigma_i)$	$AV_i(\sigma_i/\sqrt{n_i})$	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 %



agrees with $\approx 5-10$ % individual pixel precision above Pacific (Richter et al., 2004)

Appendix 7:

Analytic function to fit NO₂ annual cycles

