Supplementary material for:

Structural uncertainty in AMF calculation for NO_2 and HCHO satellite retrievals.

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Supplementary material

Equation S1. Effective temperature at altitude z:

$$T_{eff} = \frac{\int_z^{\infty} T(z) \cdot m(z) \cdot n(z) dz}{\int_z^{\infty} m(z) \cdot n(z) dz} \quad \text{(S1)}$$

Where T(z) is the temperature profile, m(z) is the altitude-dependent air mass factor, and n(z) is the NO_2 number-density profile.

Equation S2. Temperature correction factor from Boersma et al. (2002).

$$c_l = \frac{T_o - 11.4}{T_l - 11.4} \quad \text{(S2)}$$

T_o : Cross section temperature used in the DOAS fit (220 K in this study).

T_l : Temperature in layer l

Equation S3. Temperature correction factor from Bucsela et al. (2013).

$$c_l = 1 - 0.003 \bullet (T_l - T_o) \quad \text{(S3)}$$

T_o : Cross section temperature used in the DOAS fit (220 K in this study).

T_l : Temperature in layer l

Equation S4. Cloud radiance fraction (Boersma et al., 2004).

$$w = \frac{f_{cl} I_{cl}}{f_{cl} I_{cl} + (1 - f_{cl}) I_{cr}} \quad \text{(S4)}$$

f_{cl} is the cloud fraction, and I_{cr} and I_{cl} the fit-window averaged radiances for 100% clear and cloudy scenes, respectively.

Table S1. Model settings for top-of-the-atmosphere reflectance calculation with different RTMs, as described in Section 3.1.

Input Parameter	**Number of reference points**	**Values of reference points**
Wavelength	7	340, 360, 380, 400, 420, 440, 460 nm
Atmospheric profile	N.A.	mid-latitude summer atmosphere including O_3 (335 DU)
μ_0 (cosine solar zenith angle)	10	1.00, 0.80, 0.60, 0.50, 0.30, 0.25, 0.15, 0.05, 0.03, 0.00
Solar zenith angles	10	0°, 36.9°, 53.1°, 60°, 72.5°, 75.5°, 81.4°, 87.1°, 88.3°, 90°
μ (cosine viewing zenith angle)	2	1.00, 0.30
Viewing zenith angles	2	0°,72.5°
$180-\lvert\varphi-\varphi_0\rvert$ (relative azimuth angle)	5	0°, 60°, 90°, 120°, 180°
Surface albedo	1	0.00
Surface pressure	1	1013 hPa

Table S2. Model settings for altitude dependent (box-) AMFs calculation in Section 3.2.

Input Parameter	**Number of reference points**	**Values of reference points**
Atmospheric profile	N.A.	mid-latitude summer atmosphere including O3 (335 DU)
Layering	170	0, 0.1, 0.2, …. 10 km 10, 11, 12, …. 60 km 60, 62, 64, …. 100 km
μ_0 (cosine solar zenith angle)	12	1.00, 0.90, 0.80, 0.70, 0.60, 0.50, 0.30, 0.25, 0.15, 0.05, 0.03, 0.00
Solar zenith angle	12	0°, 25.8°, 36.9°, 45.6°, 53.1°, 60°, 72.5°, 75.5°, 81.4°, 87.1°, 88.3°, 90°

μ (cosine viewing zenith angle)	6	1.00, 0.90, 0.80, 0.70, 0.50, 0.30
Viewing zenith angle	6	0°, 25.8°, 36.9°, 45.6°, 60°, 72.5°
$180-\|\varphi-\varphi_0\|$ (relative azimuth angle)	13	0°, 15°, 30°, 45°, 60°, 75°, 90°, 105°, 120°, 135°, 150°, 165°, 180°
Surface albedo	7	0.00, 0.05, 0.1, 0.2, 0.5, 0.8, 1.0
Surface height pressure (hPa)	5	1013, 902, 802, 554, 281

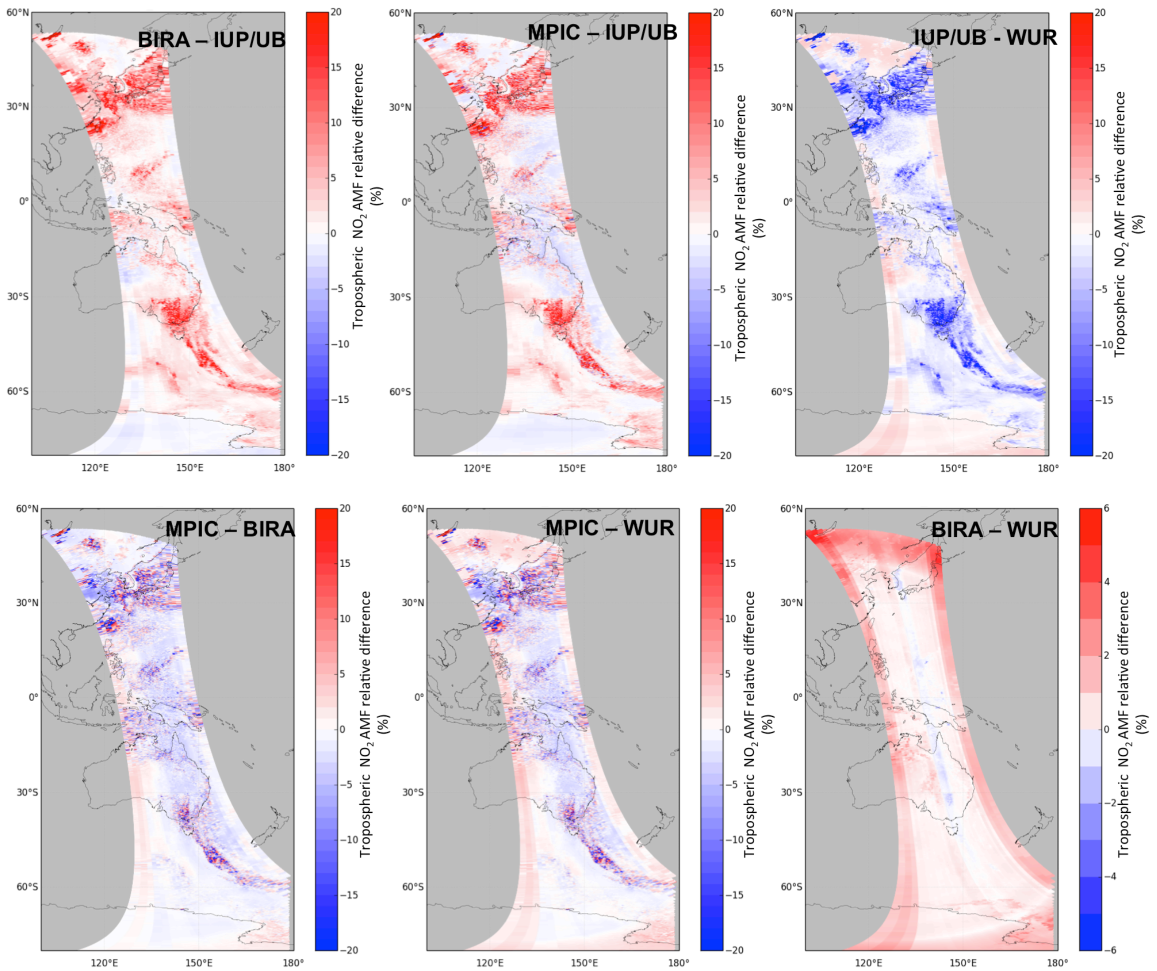


Figure S1: Relative differences of tropospheric NO_2 AMFs between each research group using harmonized settings. Only pixels with SZA < 70 ° are shown. The selected OMI orbit is from 02 February 2005 (2005m0202-o02940_v003). Different scale was used for the differences between BIRA – WUR (lower right panel).

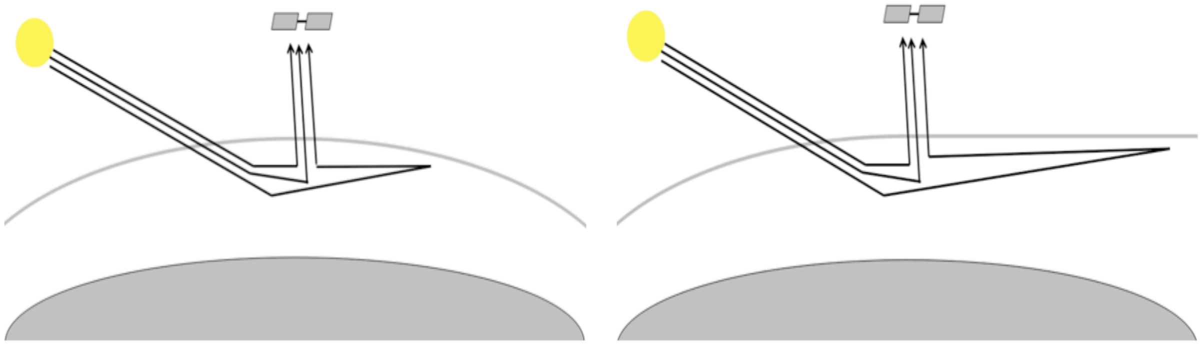


Figure S2. Schematic representation of differences in model design between McArtim (left) and DAK, VLIDORT and SCIATRAN (right) for the direct solar beam (left side of the individual figures) and the multiple scattered photons (left side of the individual figures). The grey line indicates the atmosphere's confinement (either spherical or plane parallel).

S1. Preferred settings for NO_2 tropospheric AMF calculation

In this section we give a summary of the preferred settings for AMF calculation from the groups that do not have a published reference.

S1.1 BIRA – IASB

For the radiative transfer modelling and box-AMF calculation, BIRA uses the VLIDORT radiative transfer model (see Sect. 2.2).

The surface reflectivity is a combination of the MODIS black sky albedo (BSA) gap filled product (MCD43GF) and the OMI minimum LER from Kleipool et al. (2008) at 440 nm. The MODIS BSA values are averaged over 10 years of measurements and the OMI min LER dataset is used to fill the gaps and for scenes over water.

Surface pressure is from the Global Multi-resolution Terrain Elevation Data 2010 with 30 x 30 km resolution, corrected following the approach by Zhou et al. (2009).

The cloud parameters (cloud fraction and cloud pressure) are taken from the OMI 02-02 cloud retrieval (OMCLDO2, Acarreta et al., 2004).

For the cloud correction they apply IPA for cloud fractions higher than 0.2 and cloud masking for cloud fractions lower than 0.2. They apply an implicit aerosol correction.

The NO_2 a priori profiles are daily profiles from the TM5 chemistry transport model at a resolution of 1x1 degrees.

S1.2 IUP-UB

For the radiative transfer modelling and box-AMF calculation, IUP-UB uses the SCIATRAN radiative transfer model (see Sect. 2.2).

Surface albedo is from Kleipool et al. (2008) updated dataset (version 3), which uses 5 years of OMI measurements. The monthly minimum LER at 442 nm is used.

Surface pressure is from the Global Multi-resolution Terrain Elevation Data 2010. They are gridded to 0.25 x 0.25 degrees and corrected following the approach by Zhou et al. (2009).

For the cloud correction they apply IPA for cloud fractions higher than 0.1 and cloud masking for cloud fractions lower than 0.1. They use modelled reflectances for the current albedo and a cloud albedo of 0.8 to convert O2-O2 cloud fraction to radiance fraction. The cloud fraction threshold is cloud radiance fraction of 50%. They apply an implicit aerosol correction.

The NO_2 and temperature profiles come are daily MACC-II reanalysis profiles with a resolution of 1.25 x 1.25°

S1.3 MPI-C

For the radiative transfer modelling and box-AMF calculation, MPI-C uses the McArtim radiative transfer model (see Sect. 2.2).

Surface albedo is from Kleipool et al. (2008), version 002, which uses 3.5 years of OMI measurements. The monthly minimum LER at 440 nm is used.

Surface pressure is from TM4 chemistry transport model and corrected following the Zhou et al. (2009) approach using the high resolution DEM_3km Earth Science Data type database.

The NO_2 and temperature profiles are daily TM4 model at a resolution of 3 x 2 degrees.

In the preferred settings, MPI-C accounts for the possibility of cloud aerosol mixtures or layer of other different types of aerosol. For this purpose they differentiate three different cases:

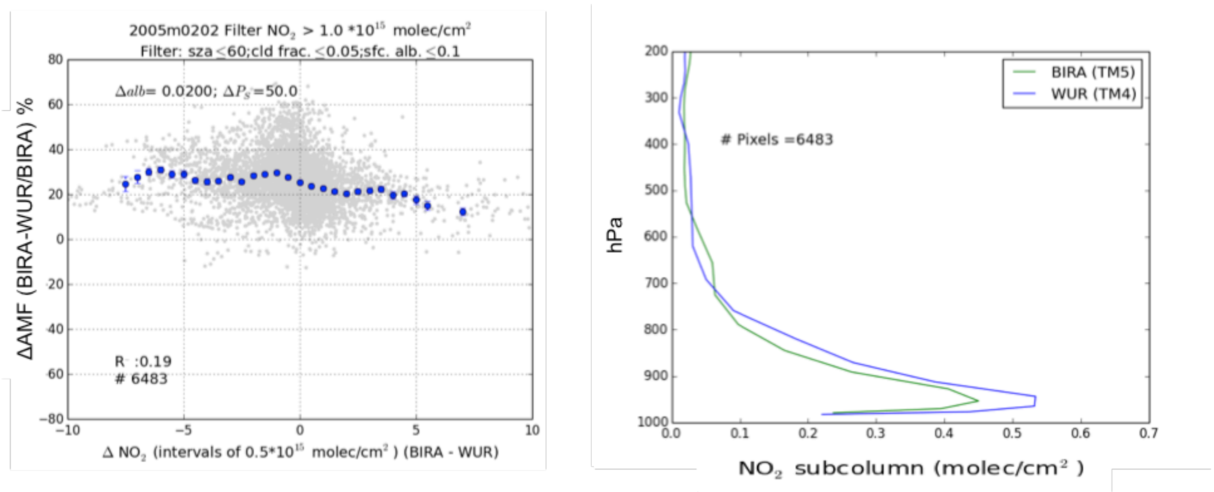
A. Clouds higher than 3 km. The independent pixel approximation is applied to calculate the AMF.

B. Low clouds and aerosols. For cloud altitudes below 2 km, a parameterized aerosol cloud layer is included between 0 and 1 km above the surface. This parameterization only represents a coarse cloud/aerosol model that assumes small cloud fractions to be pure aerosols and high cloud fractions to be pure clouds both with a fixed layer thickness of 1 km. They determine the relation between optical depth of an aerosol/cloud layer and the cloud radiance fraction using McArtim simulations. For this purpose they expand the LUT by the optical depth (OD), single scattering albedo and the Henyey Greenstein asymmetry parameter. Depending on the optical depth, they assume typical optical parameters of aerosols for OD <= 1, aerosols/cloud particle mixture for 1 < OD < 3 and cloud particles for OD >3.

C. Low cloud fraction. For clouds between 2 and 3 km with cloud radiance fraction below 10%, they use the clear sky AMF.

D. High cloud fraction. For clouds between 2 and 3 km with cloud radiance fraction higher than 10%, they flag the pixel as invalid as it cannot be differentiate between white Lambertian clouds and mixtures of clouds and aerosols.

02 February 2005



16 August 2005

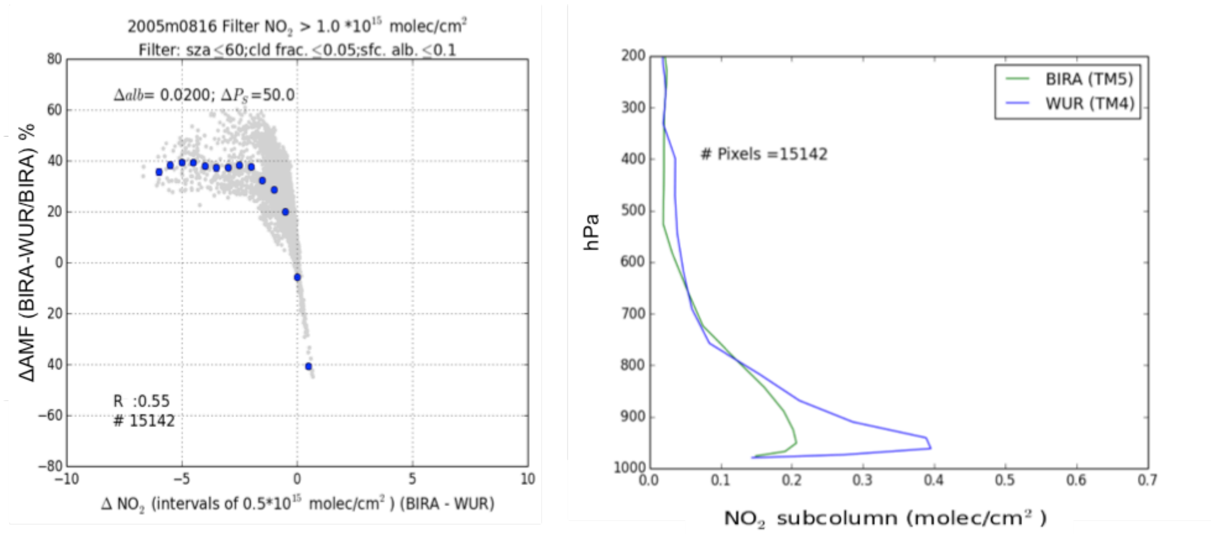


Figure S3: Example of correlation between AMF differences by BIRA and WUR (ΔAMF) and differences in NO_2 vertical columns (ΔNO_2) for 02 February 2005 (upper panels) and 16 August 2005 (lower panels). The panels on the right show the average NO_2 vertical profiles for the scenarios shown in the left panels (green, TM5 by BIRA and blue, TM4 by WUR).

2005m0202	ΔAMF vs ΔNO_2	ΔAMF vs ΔA_s	ΔAMF vs ΔP_s
# Pixels	6483	1876	1303
R	-0.19	0.50	-0.04
2005m0816	**ΔAMF vs ΔNO_2**	**ΔAMF vs $\Delta\ A_s$**	**ΔAMF vs ΔP_s**
# Pixels	15142	5382	2736
R	-0.55	0.21	-0.01

Table S3: Number (#) of pixels and correlation coefficient (R) for the correlation between air mass factor differences between WUR and BIRA (ΔAMF) with differences in modelled NO_2 vertical column (ΔNO_2)**,** surface albedo (ΔA_s) and surface pressure (ΔP_s) for 02 February 2005 and 16 August 2005. The first column corresponds to the correlation shown in left panels in Fig. S3.

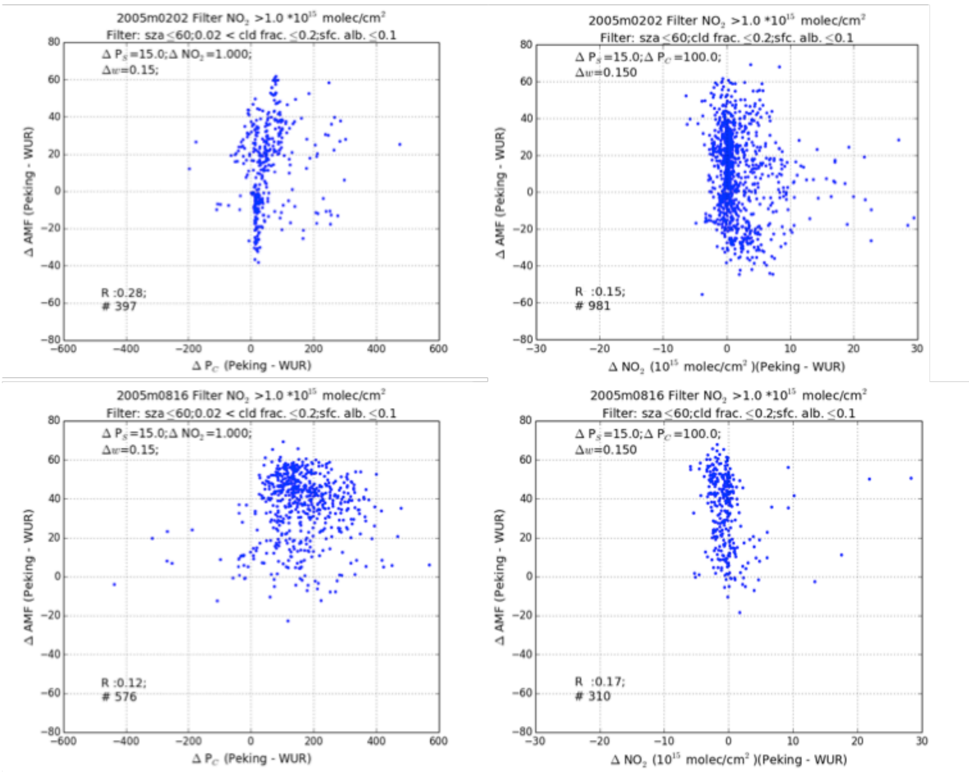


Figure S4: Correlation between AMF differences by Peking University and WUR (ΔAMF) and differences in cloud pressure (ΔP_c) and NO_2 vertical columns (ΔNO_2) for the 02 February 2005 (upper panels) and 16 August 2005 (lower panels).

2005m0202	**ΔAMF vs ΔP_c**	**ΔAMF vs ΔNO_2**
# Pixels	397	981
R	0.28	0.15
2005m0816	**ΔAMF vs ΔP_c**	**ΔAMF vs ΔNO_2**
# Pixels	576	310
R	0.12	0.17

Table S4: Number (#) of pixels and correlation coefficient (R) for the correlation between air mass factor differences between Peking Uni. and WUR (ΔAMF) with differences in cloud pressure (ΔP_c) and modelled NO_2 vertical column (ΔNO_2) and for the 02 February 2005 and 16 August 2005.

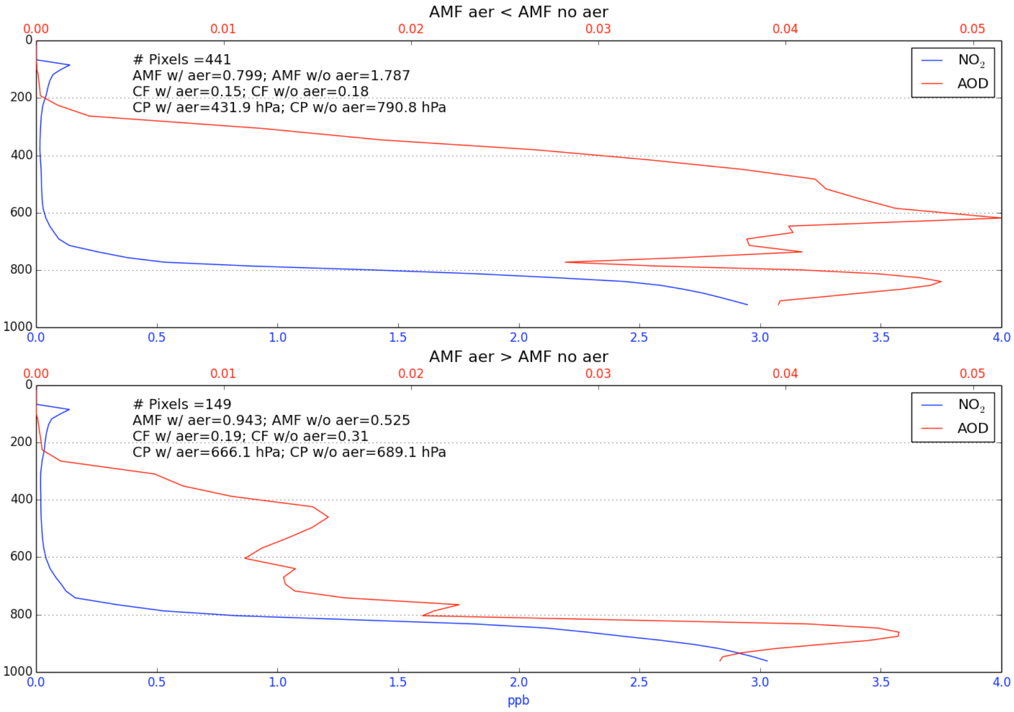


Figure S5: Aerosol optical depth (red line) and a priori NO_2 (blue line) vertical profiles for the 02 February 2005. Upper panel shows pixels where AMF_{aer} (with explicit aerosol correction) are lower than AMF (without explicit aerosol correction), due to the screening effect of the aerosols layer above the NO_2 layer. Lower panel shows pixels where AMF_{aer} (with explicit aerosol correction) are higher than AMF (without explicit aerosol correction), due to the increased scattering probability within the NO_2 + aerosol layer. Only pixels where AMF relative differences are higher than 25% are shown, as well as surface reflectance < 0.3, effective cloud fraction < 0.5 and AOD > 0.5.

	AMF_{aer} < AMF		AMF_{aer} > AMF	
# Pixels	441		149	
AOT	1.1		0.7	
SSA	0.90		0.88	
	Without correction	**With correction**	**Without correction**	**With correction**
Cloud fraction	0.18	0.15	0.31	0.19
Cloud Pressure	791 hPa	432 hPa	689 hPa	666 hPa
AMF	1.78	0.80	0.53	0.94

Table S5. Tropospheric NO_2 AMFs calculated by Peking University with and without an explicit aerosol correction over China on the 02 February 2005. Pixels with AOT > 0.5, albedo < 0.3 and effective cloud fraction < 0.5 were selected. The average AOT and single scattering albedo originate from the GEOS-Chem aerosol simulations for the location and time of the pixels. The average cloud fraction and cloud pressure are the result from Peking University's cloud retrieval.

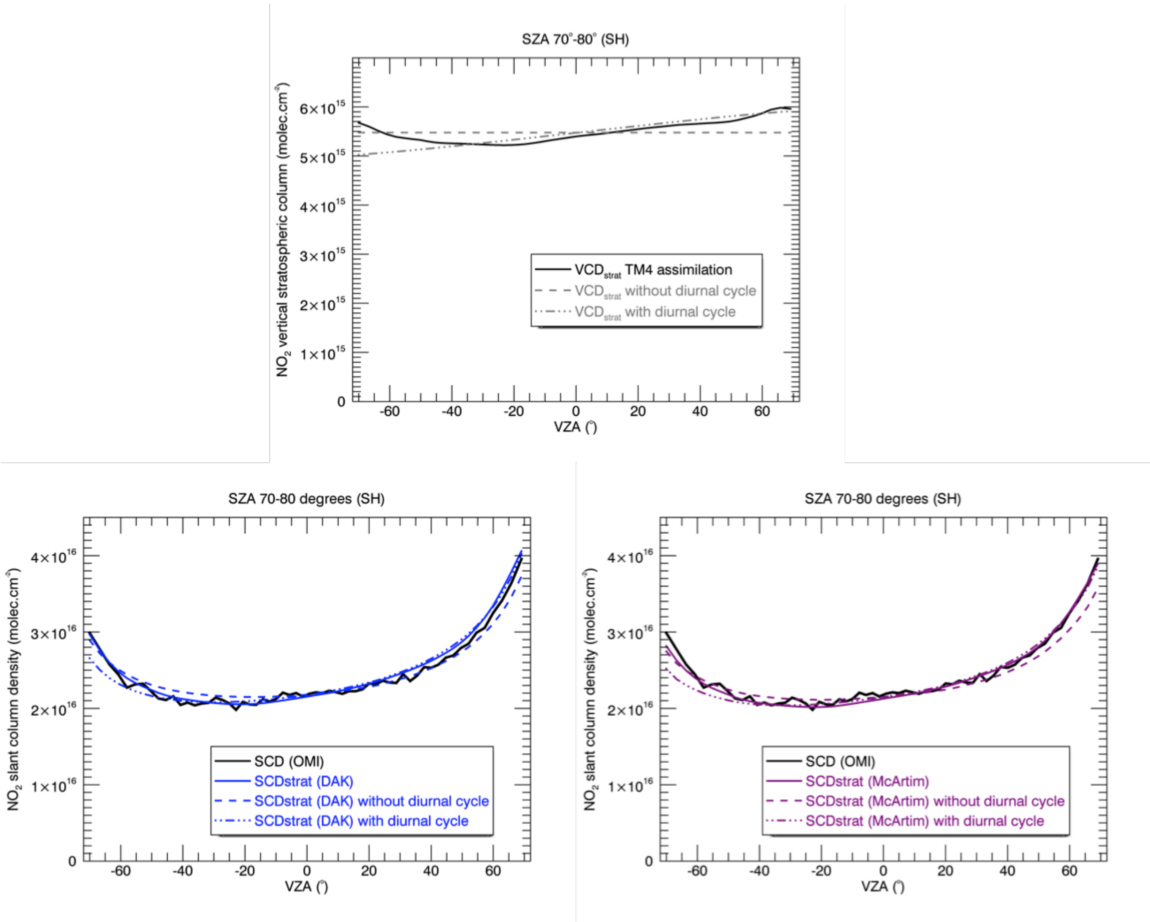


Figure S6: (Upper panel) Stratospheric vertical NO_2 columns as a function of VZA from assimilation of OMI NO_2 SCDs in TM4 (DOMINOV2 product). The dashed line indicates a scenario without any diurnal variation in stratospheric NO_2 (local solar time differences are up to 6 hours at these latitudes), and dashed-dotted line indicates a scenario with a strong, consistent stratospheric NO_2 increase rate (of 0.15 10^{15} molec/cm^2/h). The lower panels compare the three corresponding simulated stratospheric slant columns (from TM4 assimilated VCD, without diurnal cycle and with diurnal cycle) from DAK (left panel) and McArtim (right panel) to the observed OMI total SCD (black solid line) as a function of OMI VZA.

Table S6. Statistical parameters of the comparison with the model mean ($((\overline{AMF} - AMF_x)/\overline{AMF})*100$, in %) of total tropospheric NO_2 AMFs calculated by each group over the globe for polluted and unpolluted pixels (pixels with model NO_2 vertical column higher or lower than $1•10^{15}$ molec/cm^2 respectively). Upper panels correspond to OMI measurements for the 02 February 2005 and the lower panels for the 16 August 2005. Only pixels with cloud fraction ≤ 0.2 and SZA < 60° are considered in the comparison.

	Polluted pixels						**Unpolluted pixels**					
	Mean	**Median**	**σ**	**R^2**	**Slope**	**Offset**	**Mean**	**Median**	**σ**	**R^2**	**Slope**	**Offset**
BIRA	-14.0	-15.8	15.6	0.840	1.40	-0.33	-5.9	-6.1	10.4	0.897	1.08	-0.05
IUP-UB	2.0	2.0	20.2	0.606	1.10	-0.17	1.7	1.2	11.6	0.861	1.05	-0.12
Leicester Uni.	3.7	3.8	14.6	0.788	1.07	-0.14	7.4	7.7	8.4	0.926	1.04	-0.2
MPIC	-8.0	7.1	42.1	0.699	2.60	-1.85	-2.6	-2.4	17.9	0.827	1.45	-0.75
NASA	-1.6	-1.2	11.7	0.847	1.05	-0.05	-2.5	-1.5	9.5	0.940	1.16	-0.31
WUR	18.0	18	12.5	0.814	1.06	-0.30	1.8	1.5	8.8	0.938	1.13	-0.26

	Polluted pixels						**Unpolluted pixels**					
	Mean	**Median**	**σ**	**R^2**	**Slope**	**Offset**	**Mean**	**Median**	**σ**	**R^2**	**Slope**	**Offset**
BIRA	-9.7	-13.2	15.5	0.916	1.39	-0.36	-5.2	-5.9	9.3	0.929	1.1	-0.09
IUP-UB	-7.3	-6.2	15.3	0.859	1.04	0.02	-0.8	-0.7	11.3	0.875	1.01	-0.01
Leicester Uni.	1.3	1.9	10.6	0.921	0.97	0.01	6.0	6.7	8.5	0.923	0.99	-0.1
MPIC	1.8	10.2	31.1	0.643	1.54	-0.71	-2.0	-1.4	15.9	0.871	1.36	-0.61
NASA	-1.7	-1.5	11.5	0.918	1.08	-0.08	-1.6	-0.9	9.6	0.915	1.03	-0.03
WUR	15.7	13.9	10.3	0.926	1.03	-0.23	3.7	3.6	9.3	0.932	1.13	-0.29

Table S7. Statistical parameters of the comparison with the model mean ($((\overline{AMF} - AMF_x)/\overline{AMF})*100$, in %) of total tropospheric NO_2 AMFs calculated by each group over China (20°-53°N / 80°-130°W) for polluted and unpolluted pixels (pixels with model NO_2 vertical column higher or lower than $1 \cdot 10^{15}$ molec/cm^2 respectively). Upper panels correspond to OMI measurements for 02 February 2005 and the lower panels for 16 August 2005. Only pixels with cloud fraction ≤ 0.2 and SZA < 60° are considered in the comparison.

	Polluted pixels						**Unpolluted pixels**					
	Mean	**Median**	**σ**	**R^2**	**Slope**	**Offset**	**Mean**	**Median**	**σ**	**R^2**	**Slope**	**Offset**
BIRA	-10	-7.5	18.2	0.769	1.42	-0.37	-15	-15.1	15.2	0.860	1.27	-0.18
IUP-UB	5.1	9.0	14.9	0.728	0.70	0.27	8.3	12.6	16.2	0.745	1.08	-0.24
Leicester Uni.	-8.8	-3.3	20.4	0.649	0.96	0.13	1.2	2.1	10.2	0.905	0.98	0.01
MPIC	7.1	8.2	37.1	0.781	2.46	-1.72	-5.1	-3.2	27.5	0.728	1.64	-0.88
NASA	-0.2	1.5	11.9	0.843	0.94	0.06	-2.5	-2.9	11.4	0.910	1.13	-0.15
Peking Uni.	-3.3	-4.8	18.0	0.774	1.27	-0.28	2.9	3.9	20.7	0.762	1.33	-0.54
WUR	10.7	9.9	13.0	0.880	1.22	-0.37	10.1	10.2	12.3	0.882	1.11	-0.31

	Polluted pixels						**Unpolluted pixels**					
	Mean	**Median**	**σ**	**R^2**	**Slope**	**Offset**	**Mean**	**Median**	**σ**	**R^2**	**Slope**	**Offset**
BIRA	-10.5	-10.9	13.4	0.855	1.41	-0.33	-10.3	-10.2	8.6	0.960	1.27	-0.26
IUP-UB	-20.0	-20.8	14.0	0.767	1.13	0.06	-0.9	0.1	9.6	0.899	0.87	0.20
Leicester Uni.	7.2	8.1	9.7	0.871	1.06	-0.14	8.0	7.6	7.8	0.931	0.97	-0.09
MPIC	26.0	27.0	12.7	0.708	1.06	-0.34	-1.0	-0.9	10.6	0.929	1.27	-0.40
NASA	-0.9	0.7	17	0.778	1.43	-0.46	4.7	4.3	11.2	0.874	1.05	-0.16
Peking Uni.	-24.6	-25.3	20.0	0.775	1.81	-0.60	-3.2	-0.9	15.3	0.822	1.18	-0.24
WUR	22.8	23.3	12.5	0.701	1.04	-0.29	2.8	3.3	8.6	0.934	1.10	-0.19