

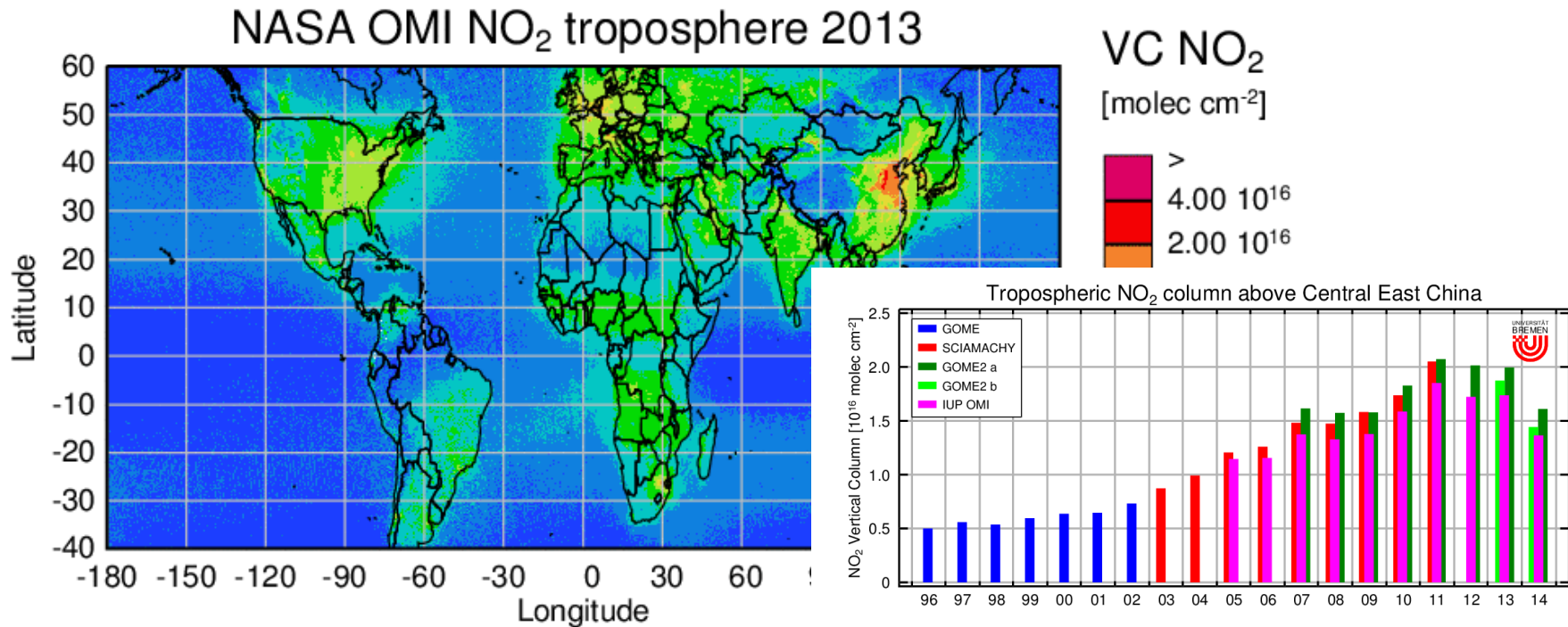
Improving S5P NO₂ retrievals

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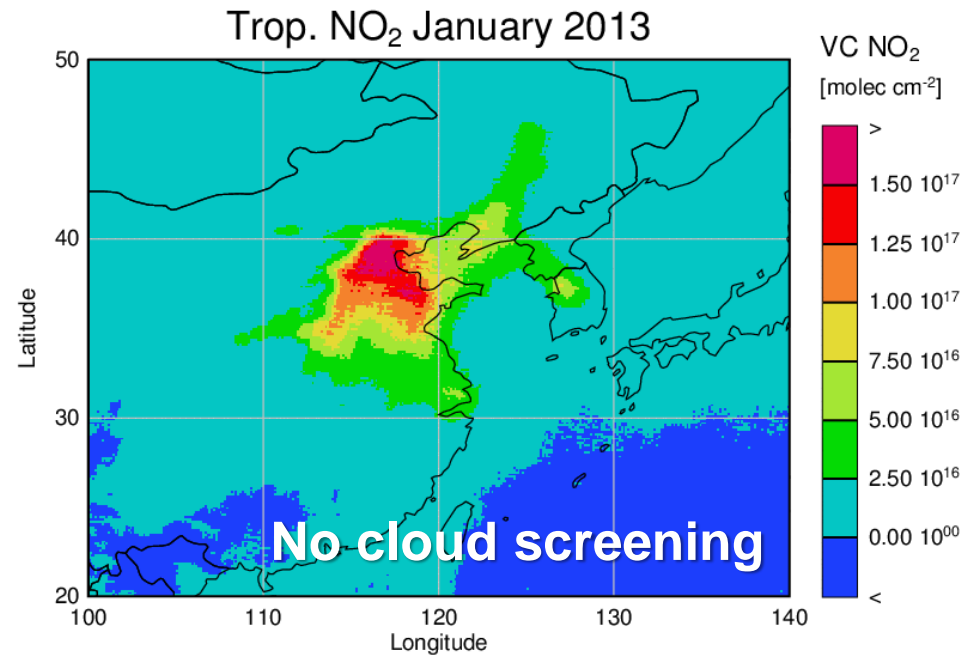
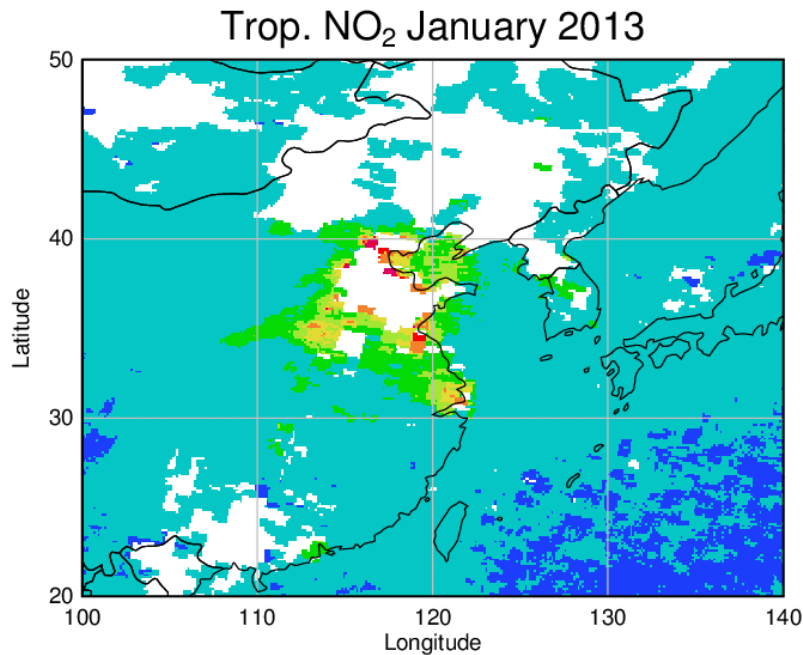
Introduction



- Current instruments provide excellent coverage and spatial detail for NO₂
- They will soon be followed by the TROPOMI / S5P instrument which promises even better performance and spatial resolution

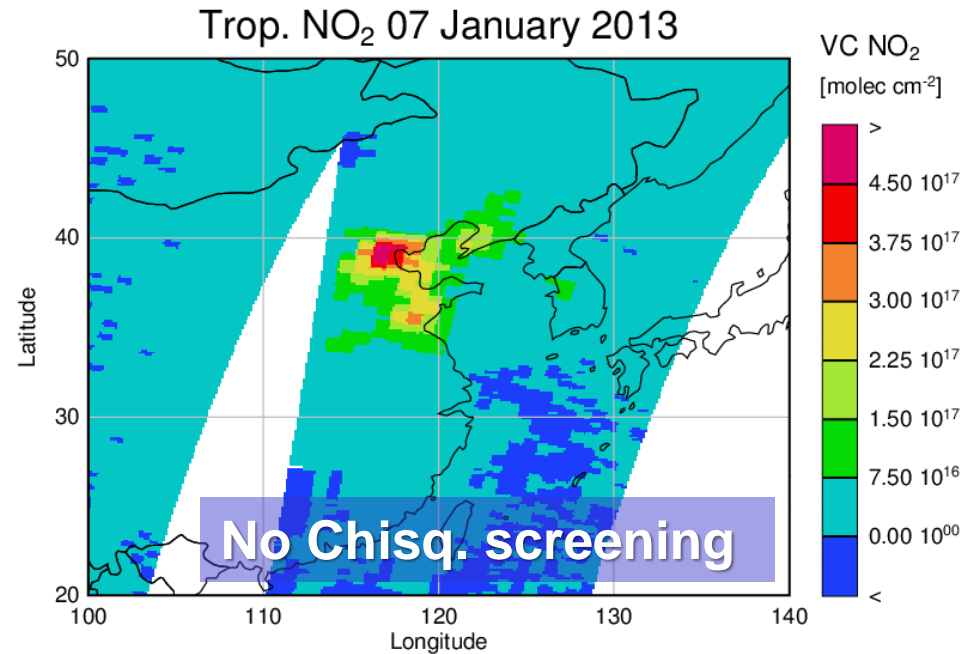
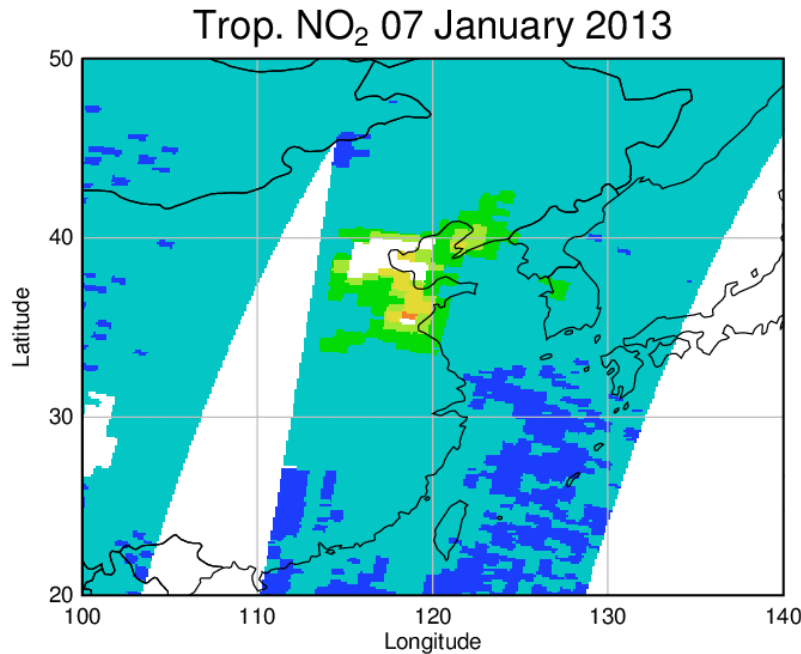
S5P tropospheric NO₂ retrieval is state of the art – so what's left to improve?

GOME-2A NO₂ above China



- Monthly GOME-2 tropospheric NO₂ data are missing most of the large values
- These were removed by cloud filtering as aerosol was so thick that data were classified as partially cloudy

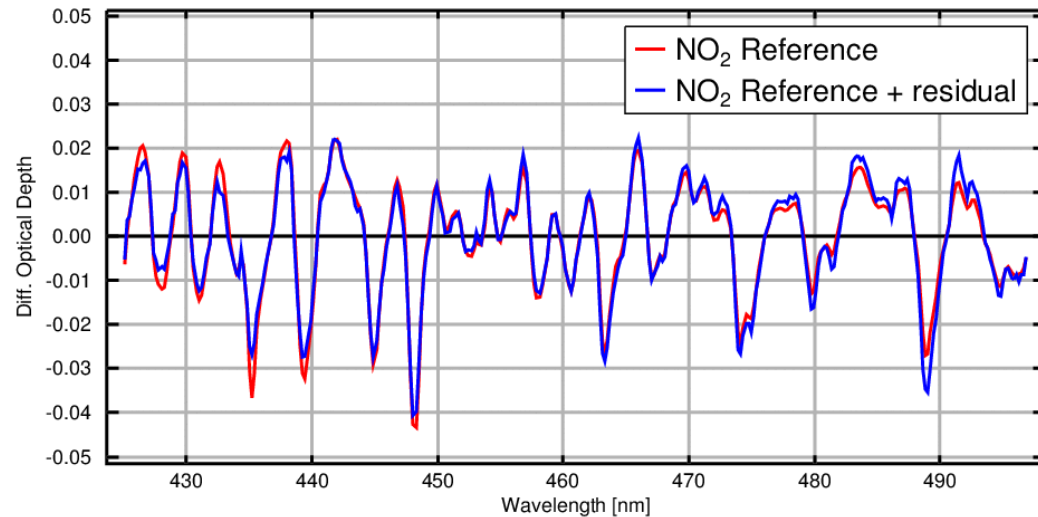
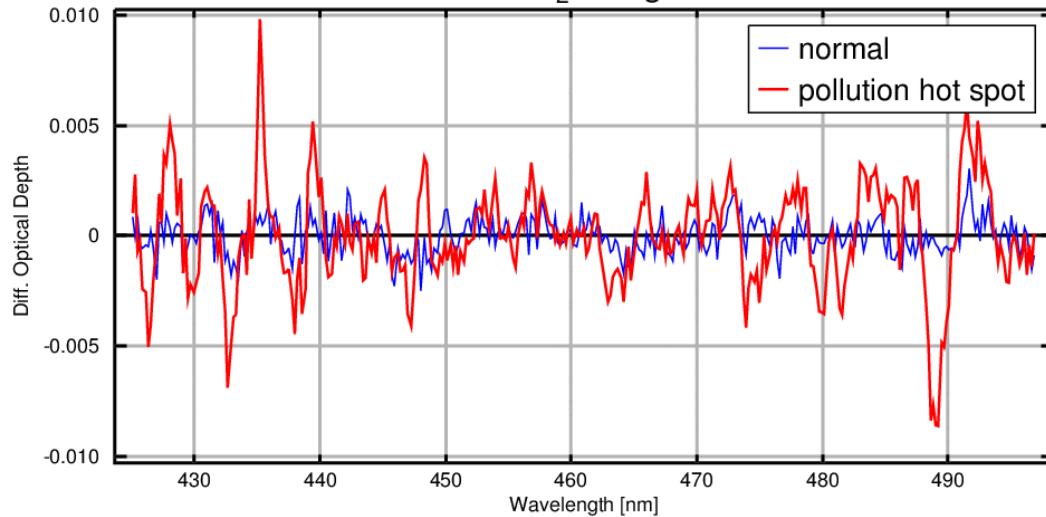
Is it only Aerosols?



- Even without cloud screening, there are data gaps over pollution hot spots on some days
- This is due to quality checking as these fits are poor

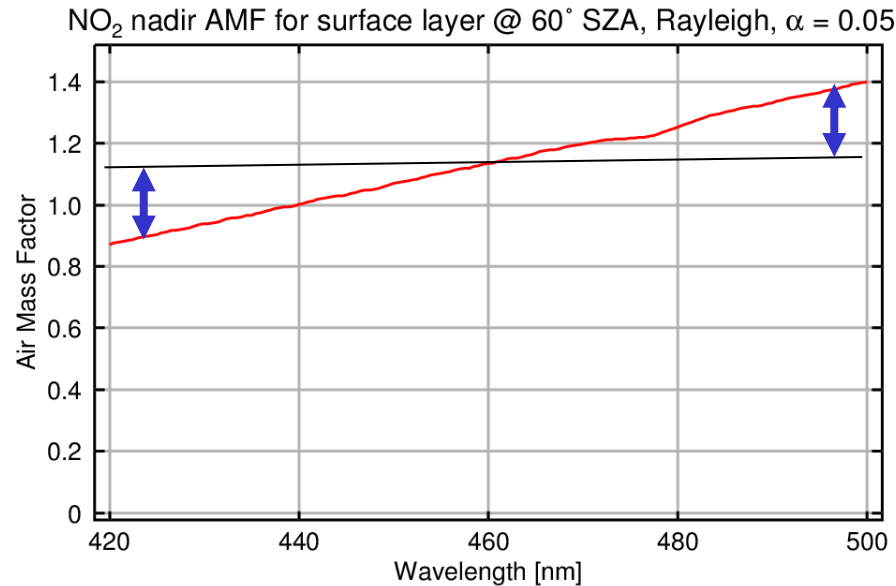
Why are the fits poorer at strong pollution?

GOME-2 NO₂ fitting residuals



- There are large and clearly structured residuals in fits over pollution hot spots
- This is not random noise!
- Comparison to NO₂ cross-sections shows that scaling of NO₂ should change over fitting window

Wavelength dependence of Air Mass Factor

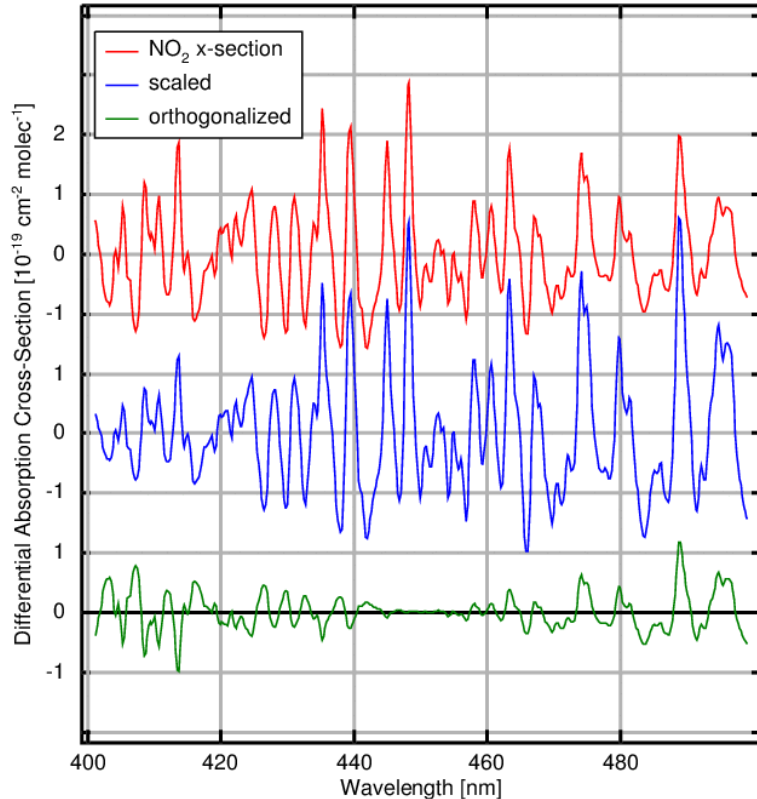


**About
+/- 20%**

- For constant albedo, AMF of NO₂ layer close to the surface increases with wavelength in a Rayleigh atmosphere
- For a surface layer, this can be a significant effect
- With radiative transfer modelling and a formal inversion, this should provide information on the altitude of the NO₂

Empirical Approach for $\text{NO}_2(\lambda)$

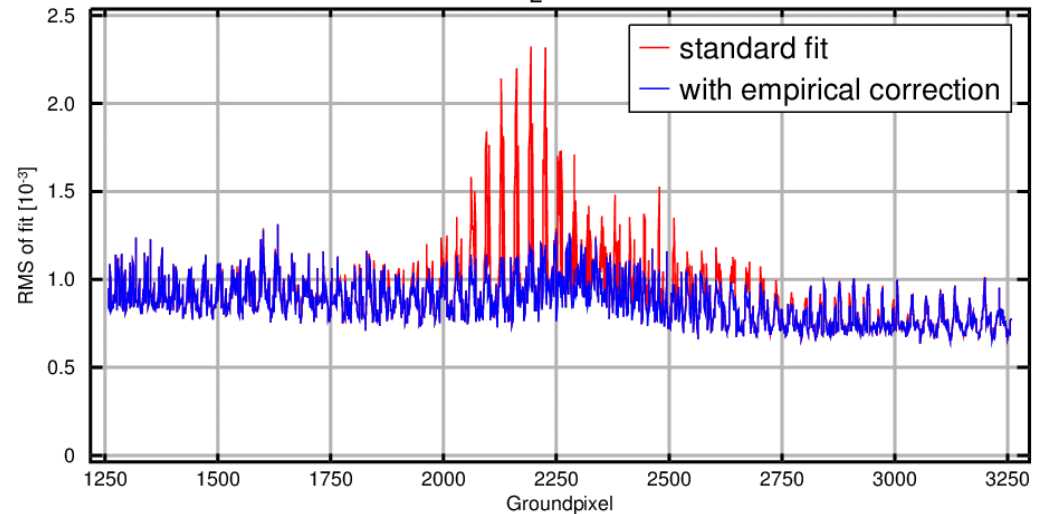
Empirical lower troposphere NO_2 x-section



- Take standard NO_2 x-section
- Scale to increase amplitude with wavelength
- Orthogonalise to leave NO_2 columns unchanged

When introduced in the fit,
large residuals are fixed

GOME-2 Residual of NO_2 fit above China 07-01-2013

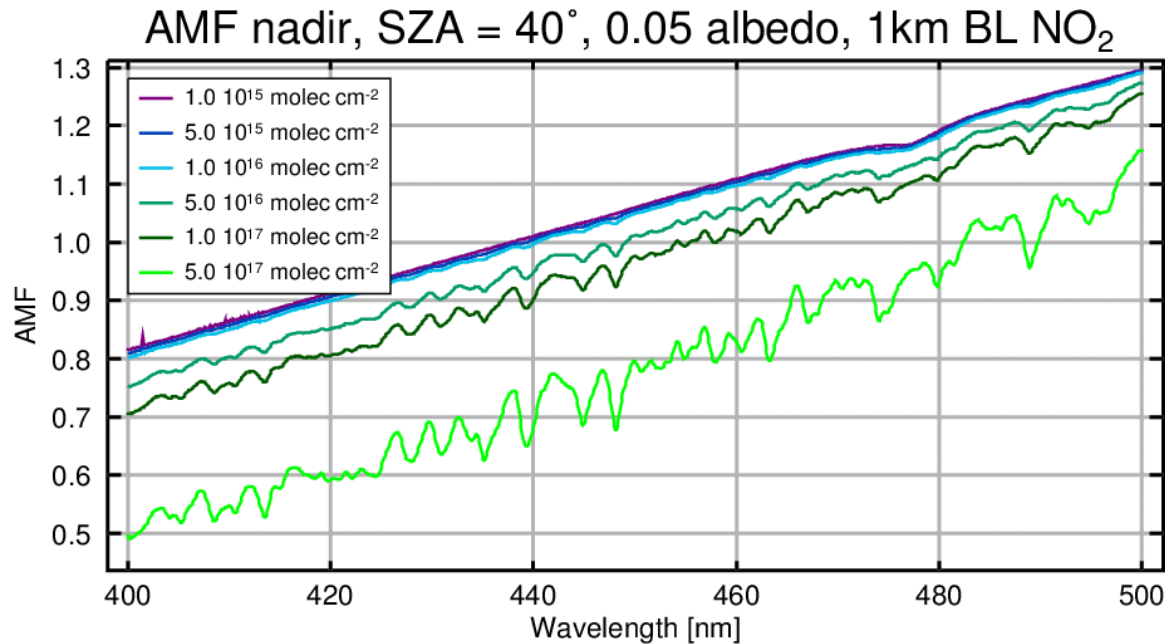


Is this the only problem at large NO₂ columns?

- One of the main DOAS assumptions is, that the light path enhancement (AMF) for a trace gas is independent of its column amount
- For strong absorbers (O₃, SO₂, IR gases) this approximation is not good enough and the change of sensitivity with wavelength needs to be accounted in the fit ("modified" DOAS)
- NO₂ is generally considered to be a weak absorber, but is that still true for very polluted scenarios?

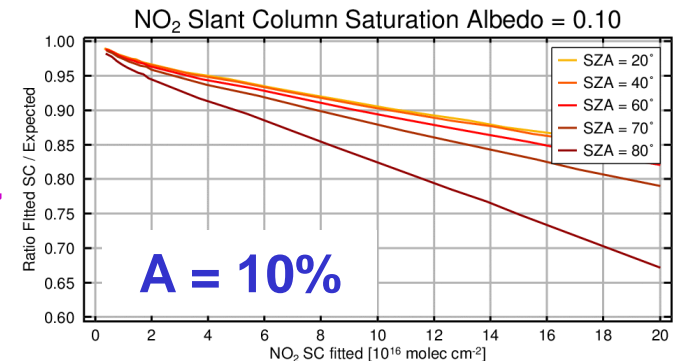
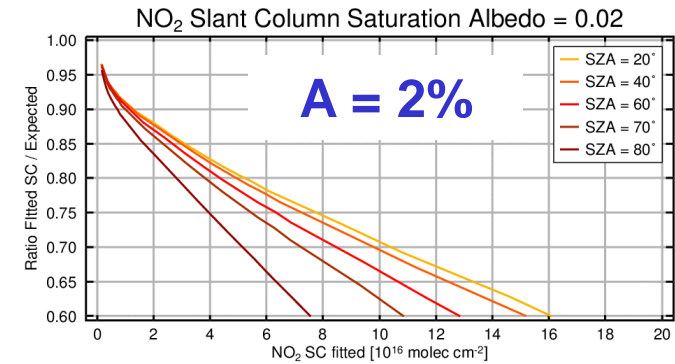
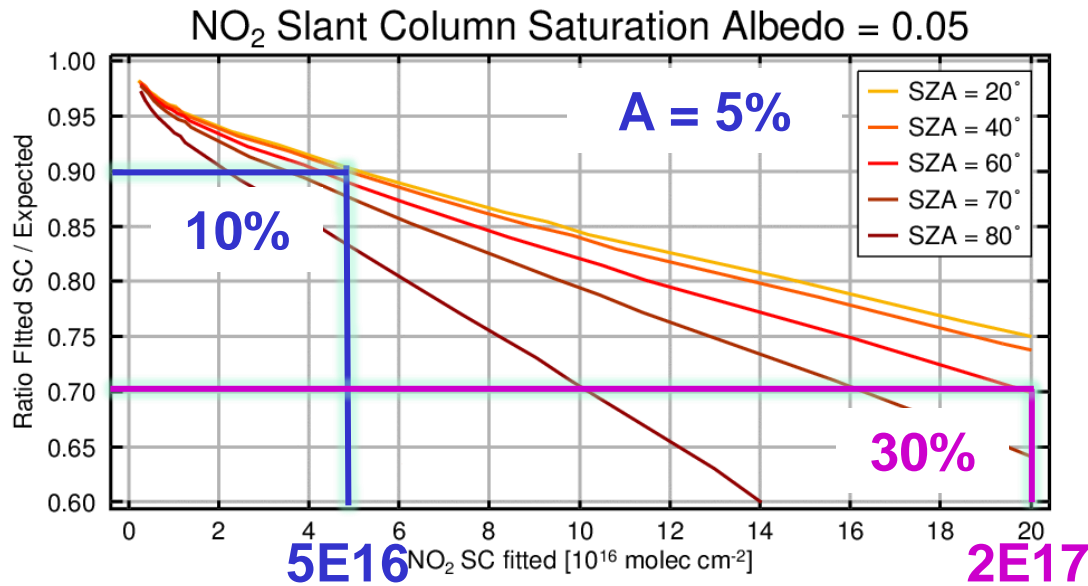


AMF dependence on NO₂ column



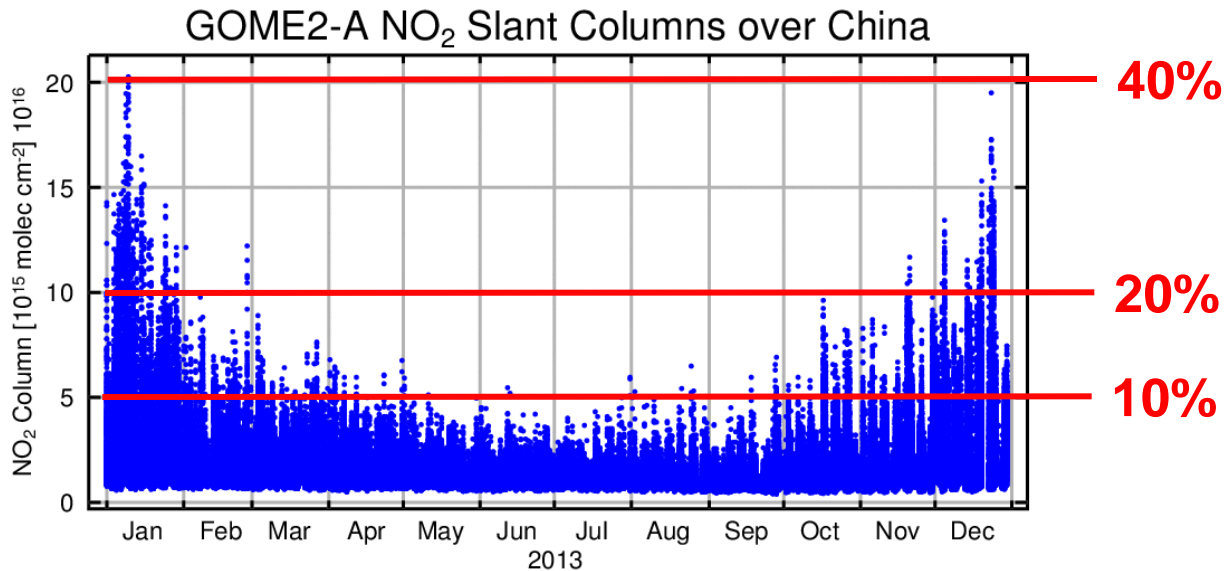
- Up to a vertical column of about 1×10^{16} molec cm⁻², small dependence of AMF on NO₂ amount
- For larger columns, the AMF decreases and NO₂ absorption structures appear

AMF dependence on NO₂ column



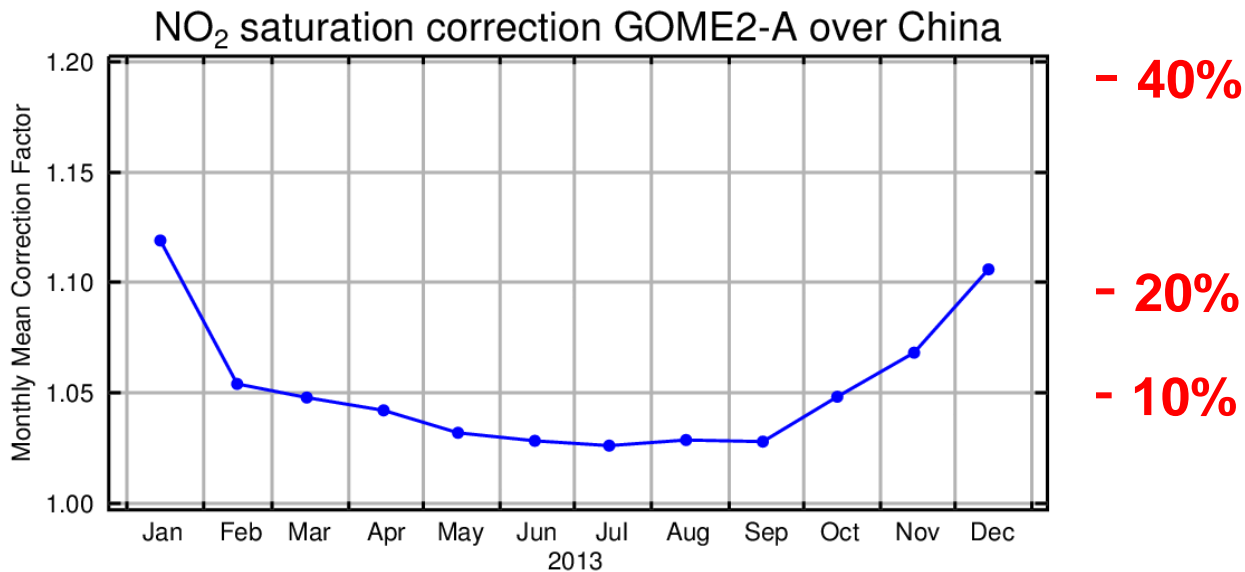
- Effect on DOAS fit on synthetic data is larger than on AMF alone as both smaller AMF and spectral structures reduce NO₂ columns
- Effect increases with SZA
- Even at a column of 5×10^{16} molec cm², the error is > 10%
- Effect decreases with increasing albedo

NO₂ saturation on real **GOME-2A** data



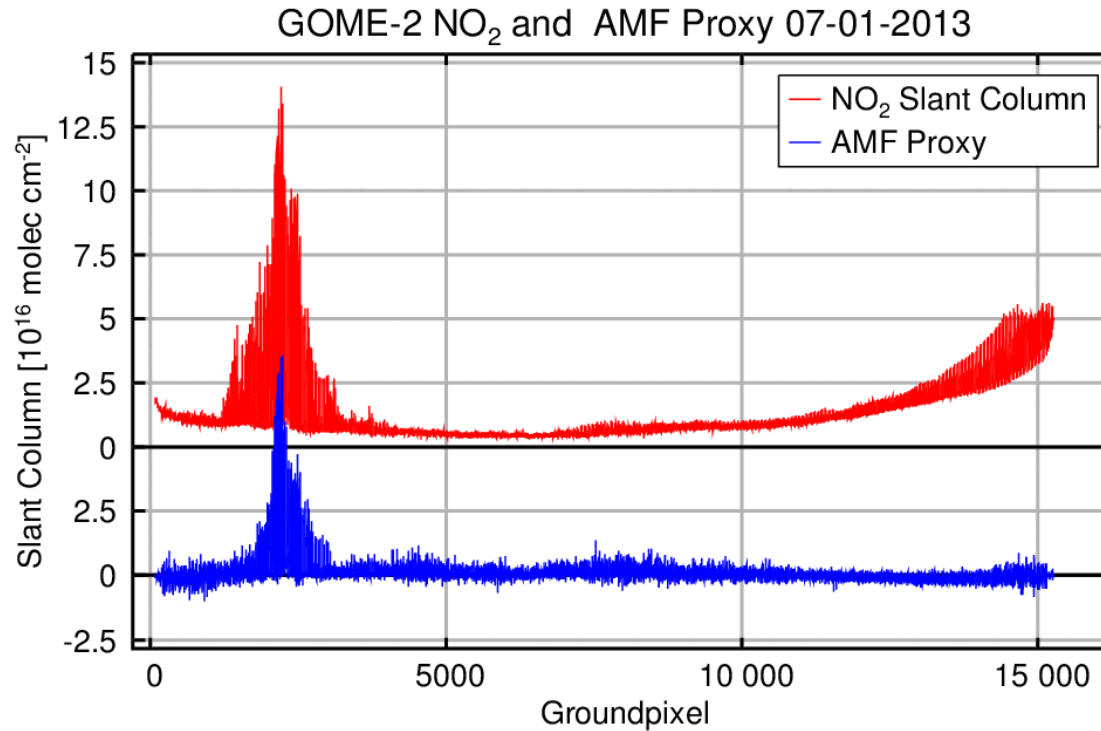
- Many measurements above China are in the saturation range
- Corrections of 5% - 40% need to be applied to individual pixels
- In monthly averages, the effect is still $> 10\%$ in January and December
- Real effect is even larger as cloud effects reduce columns but not saturation effects

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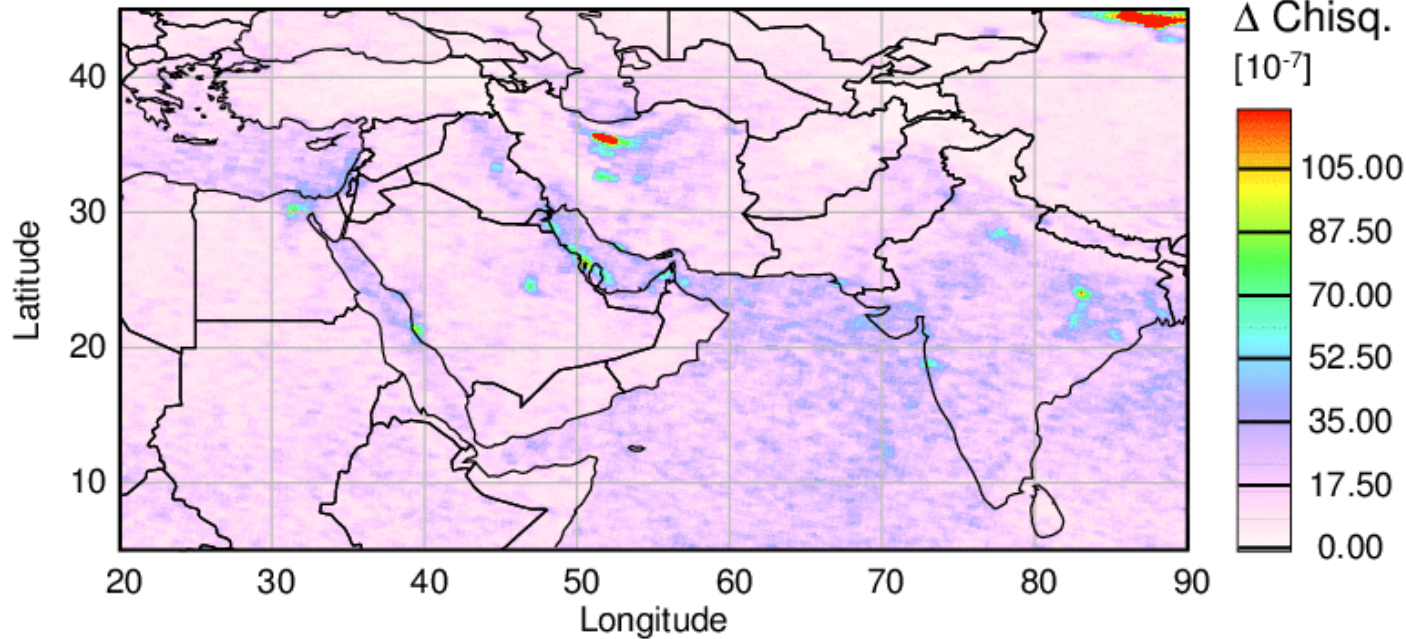
Results Empirical Approach $\text{NO}_2(\lambda)$: **GOME-2A**



- The empirical NO_2 AMF proxy is found over the pollution hotspot in China
- It is not found at other locations where the NO_2 slant column is large
- There is some noise in the retrieval of the proxy

Is there more than China? **GOME-2**

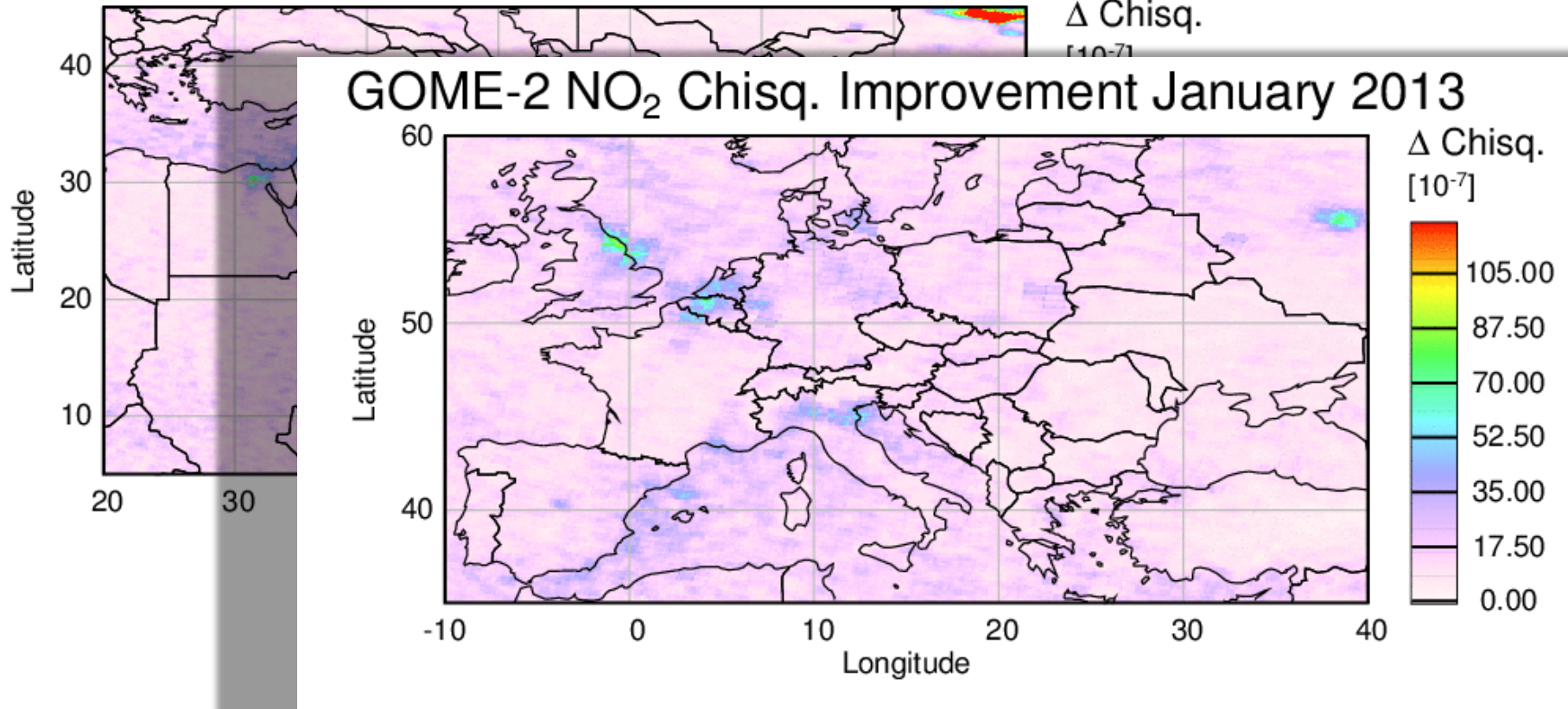
GOME-2 NO₂ Chisq. Improvement January 2013



- Fit is improved by AMF proxy everywhere over pollution hotspots

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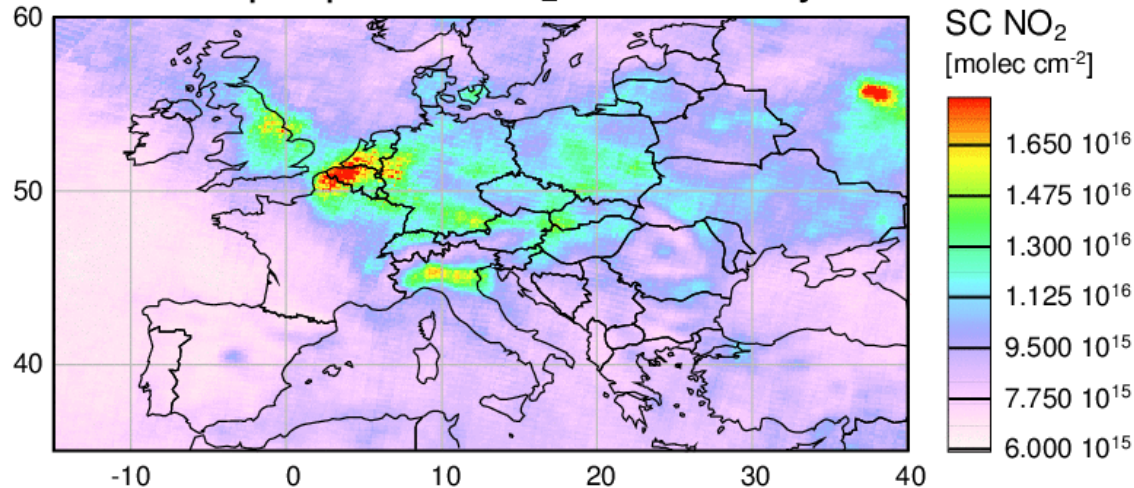
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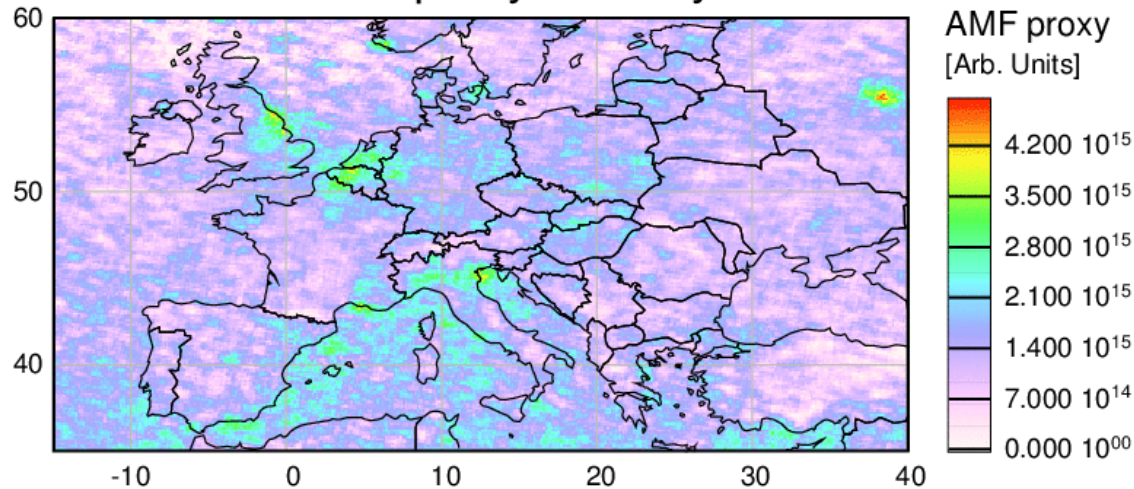
Comparison to NO₂ columns: **GOME-2**

GOME2 tropospheric NO₂ SC January 2013



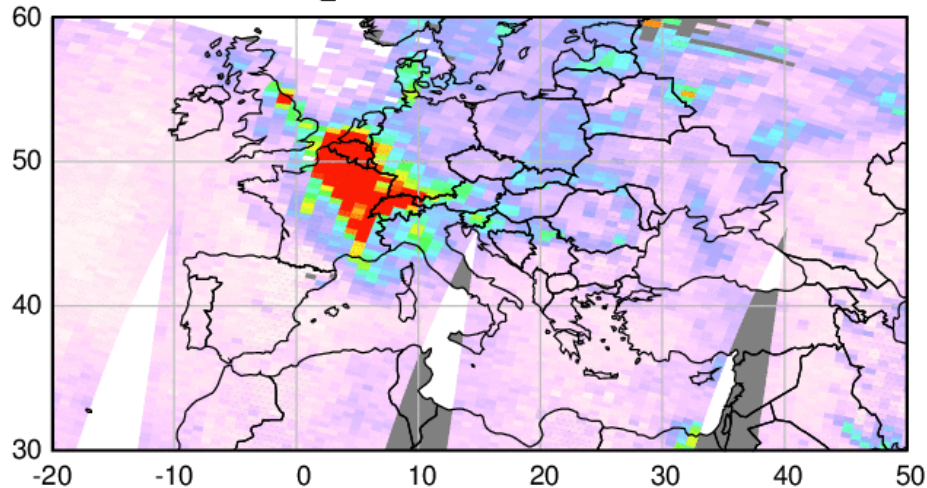
- Overall pattern similar to NO₂ map
- Differences in distributions of maxima
- Artefacts over water
- noise

GOME2 AMF proxy January 2013

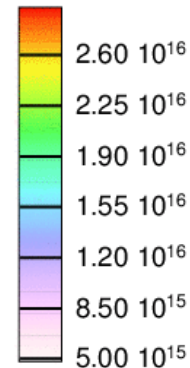


Impact of Clouds: GOME-2

GOME-2 NO₂ Slant Columns 2013/01/17

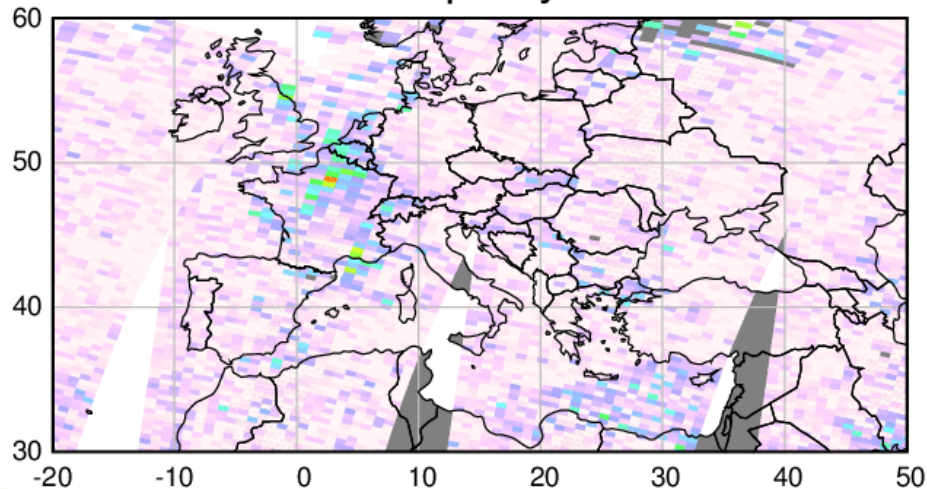


SC NO₂
[molec cm⁻²]

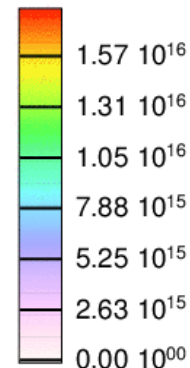


- On many days in winter, very large NO₂ slant columns are observed over Europe and the US
- The NO₂ AMF proxy picks up only very few of these signals

GOME2 AMF proxy 2013/01/17

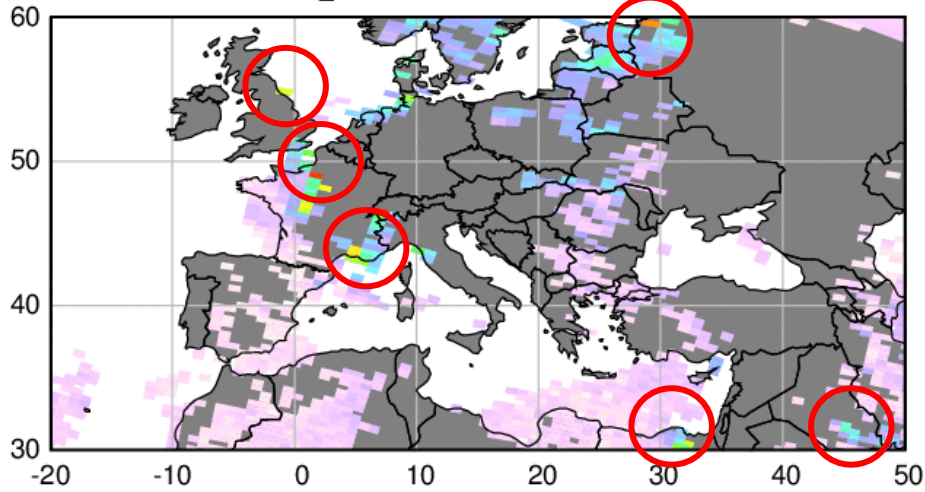


AMF proxy
[Arb. Units]

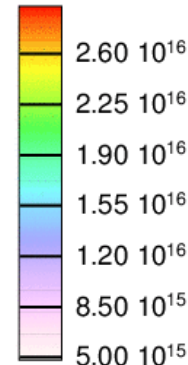


Impact of Clouds: GOME-2

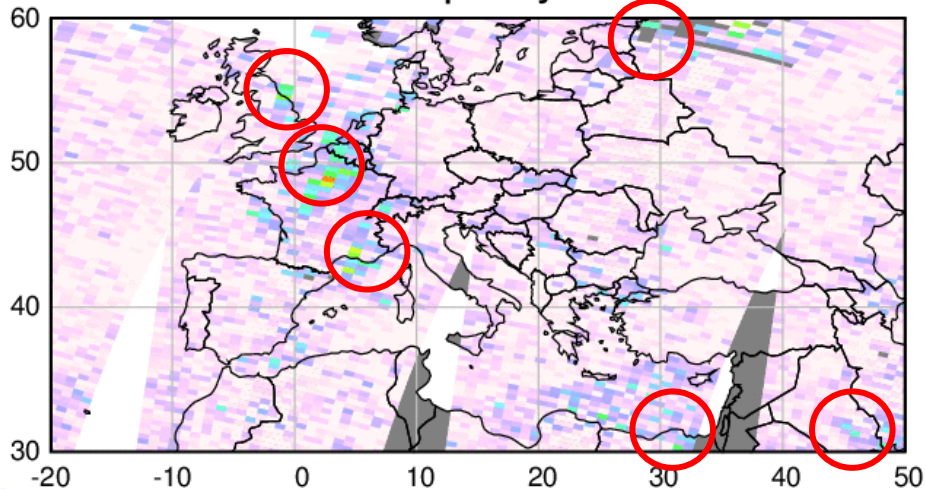
GOME-2 NO₂ Slant Columns 2013/01/17



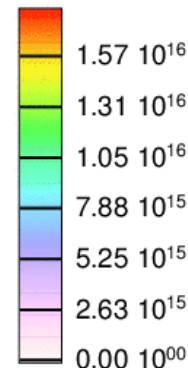
SC NO₂
[molec cm⁻²]



GOME2 AMF proxy 2013/01/17

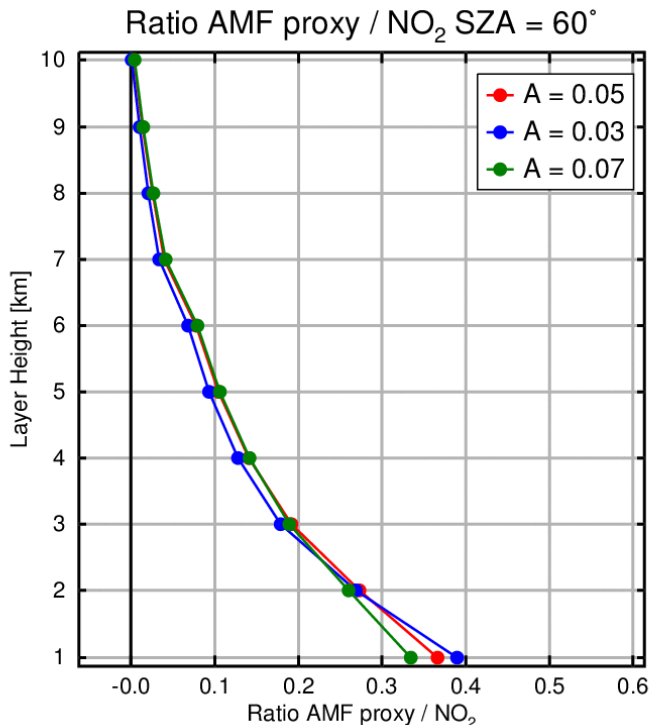


AMF proxy
[Arb. Units]



- On many days in winter, very large NO₂ slant columns are observed over Europe and the US
- The NO₂ AMF proxy picks up only very few of these signals
- This is linked to the fact that most of the events are related to cloudy scenes or snow on the surface, resulting in small wavelength dependence

Sensitivity Study for $\text{NO}_2(\lambda) / \text{NO}_2$

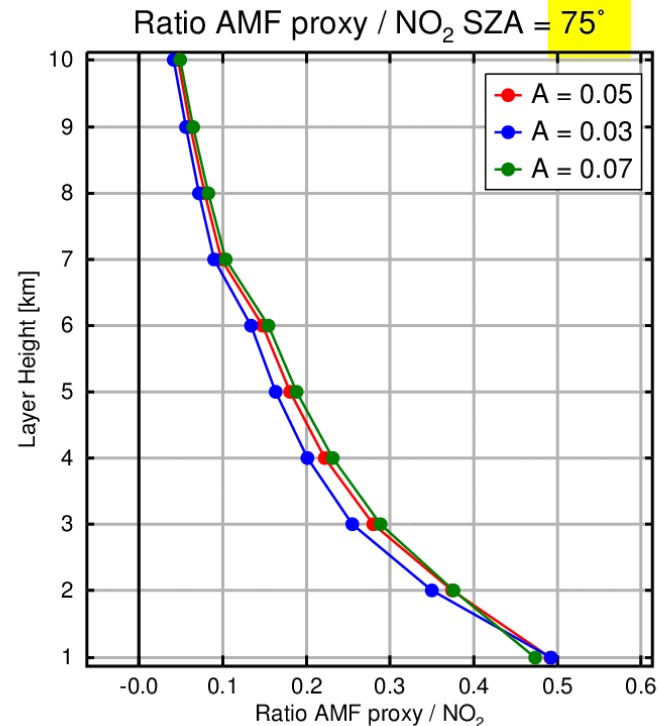
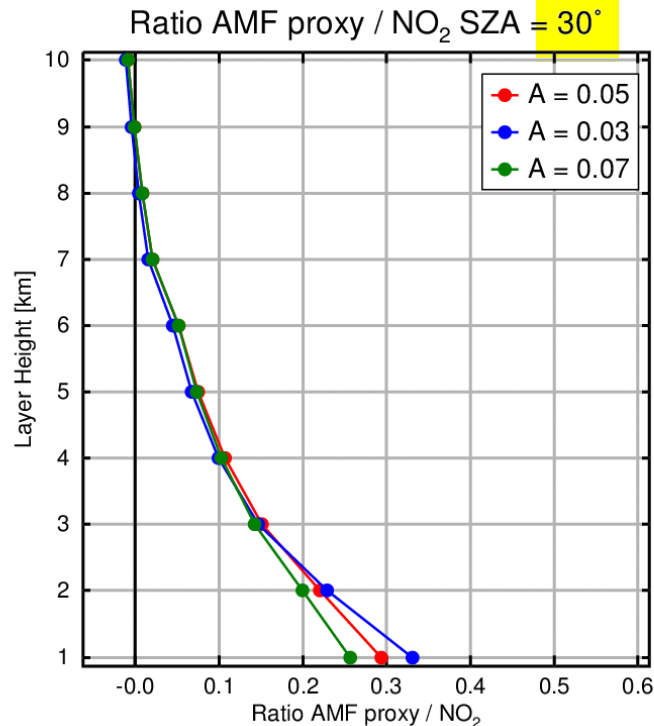


Synthetic data:

- Rayleigh atmosphere
- Constant albedo
- NO_2 layer in different altitudes
- DOAS fit on spectra
- NO_2 temperature dependence corrected by using 2 NO_2 x-sections
- AMF proxy included
- Ratio of AMF proxy / NO_2 to normalise signal

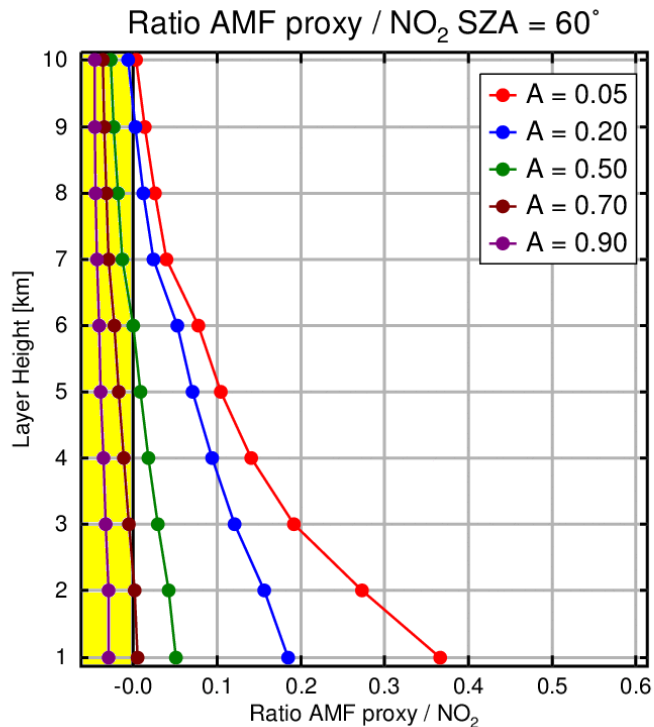
- Ratio of AMF proxy and NO_2 has strong dependence on NO_2 layer height
- Dependence on albedo is small between 3% and 7%

Sensitivity Study $\text{NO}_2(\lambda) / \text{NO}_2$: SZA



- Effect varies with SZA; larger effect at larger SZA
- At large SZA, AMF proxy also found for high NO_2
- Dependence on albedo is small between 3% and 7%

Sensitivity Study $\text{NO}_2(\lambda) / \text{NO}_2$: Bright Surfaces

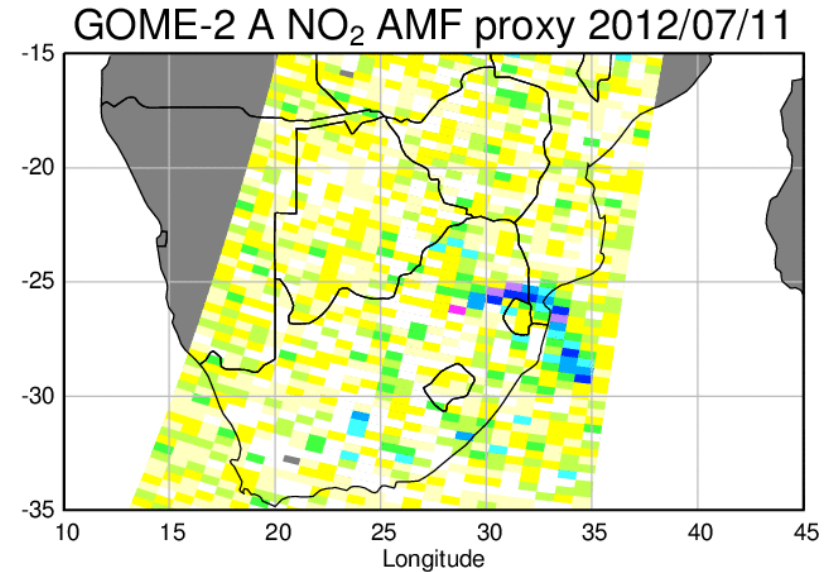
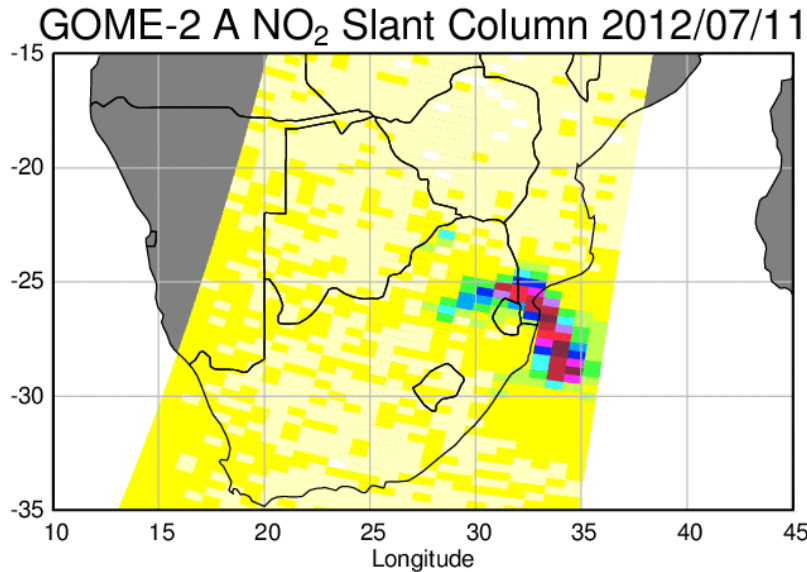


- Increasing albedo reduces effect as expected for reduced importance of Rayleigh scattering
- For large albedo ($> 50\%$), negative fit factors are found for AMF proxy \Rightarrow wavelength dependence is inverted and only weakly dependent on altitude

\Rightarrow multiple scattering over bright surfaces is stronger at shorter wavelengths

\Rightarrow wavelength dependence of AMF is inverted

NO₂(λ) / NO₂: Case Study Highveld: GOME-2



- NO₂ plume from Highveld power plants can be tracked onto the ocean
- NO₂ SC values increase downwind of the source
- AMF Proxy also has higher values within the plume, but
 - Is more narrow
 - Has largest values at beginning of plume, not at the end of it

Summary

- At large NO₂ values ($> 5 \times 10^{16}$ molec cm⁻²), AMF becomes a clear function of NO₂ column which needs to be corrected ($> 10\%$ effect)
- The effect increases with large SZA and low surface albedo
- A simple correction can be applied using tabulated factors

- At large BL NO₂ values, the wavelength dependence of the NO₂ AMF becomes relevant in the fit
- A simple empirical correction can be used to account for the AMF change
- At large NO₂ columns, the effect can be used to derive some information on NO₂ layer height

- All effects of large NO₂ values are expected to increase in frequency for instruments having better spatial resolution (S5P, S4)