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Can tropospheric NO₂ be directly retrieved from space using only an earthshine reference spectrum?

1. Motivation

Could a compact high-resolution nadir-viewing instrument accurately retrieve tropospheric NO₂ without a solar reference?

- Despite efforts to control urban air quality, NO₂ emissions from traffic and industry are still a widespread issue, with emissions being linked to increased respiratory problems.
- Satellite measurements of tropospheric NO₂ provide vital mapping and trend information on a global scale.
- However, current satellite instruments are limited in detecting NO₂ at sub-city scales due to poor spatial and temporal resolution, and are reliant on direct solar calibration.
- An ideal solution would be to have a fleet of nadir-viewing, compact, high-resolution satellite instruments to provide high temporal and spatial resolution data.
- But orbits are constrained by viewing the sun to provide a reference spectrum. Could a purely nadir-viewing instrument retrieve tropospheric NO₂ without requiring a solar reference?

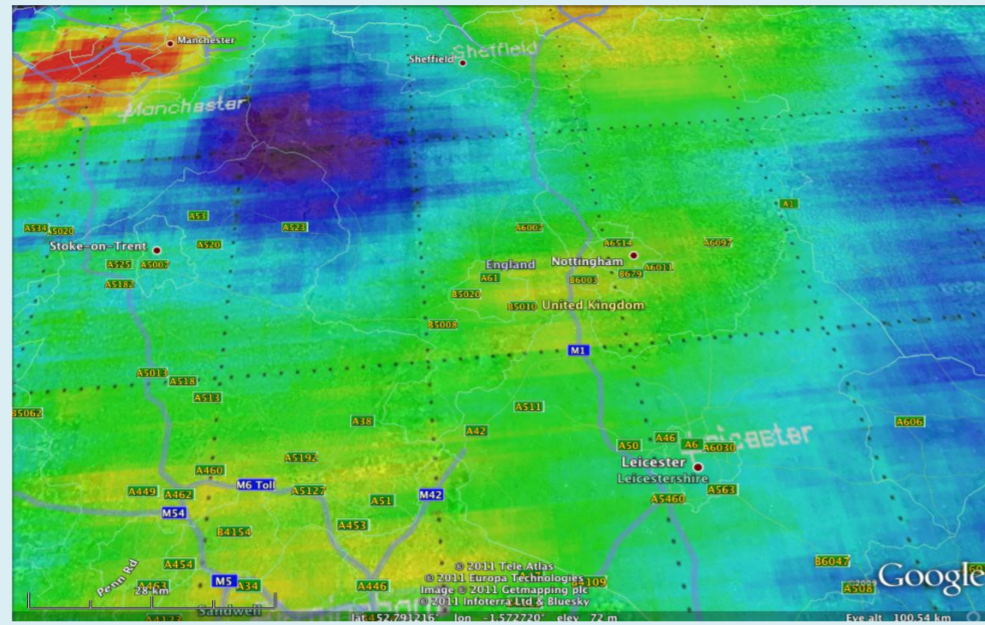


Figure 1: Averaged tropospheric NO₂ vertical column density (in molcm⁻²) over the Midlands during August 2008, measured by OMI (0.05° x 0.05° regrided). While individual cities' pollution can be resolved, there is little sub-city information that can be analysed, even on monthly timescales.

2. Algorithm Design

- Spectral and retrieval data are taken from the NASA/KNMI AURA Ozone Monitoring Instrument (OMI, Levelt et al, 2006). OMI is a push-broom spectrometer (VIS channel: 350-500 nm) that provides global coverage every 24 hrs (overpass time ~ 1340). Nadir ground pixel size: 13 x 24 km² (60 cross-track ground pixels). OMI radiance spectra and L2 NO₂ data are obtained from the OMI1BRVG, OMNO2 and DOMINO data products (NASA/TEMIS). Data from June 2005 was used to avoid coverage issues due to row anomalies.
- As the Pacific is known to have little tropospheric NO₂, earthshine spectra from this region could be used as a reference to directly retrieve tropospheric NO₂.
- Earthshine spectra collected over a clean region of the Pacific Ocean (Fig. 2) under cloud-free conditions (CF < 25%) are binned for each day in 1° latitude bands for each cross-track pixel
- Differential Optical Absorption Spectroscopy (DOAS) fitting performed using QDOAS software package (Fayt et al, 2012) developed at BIRA-IASB. Retrieval settings closely approximate the interpolation methods and cross-sections used in the DOMINO algorithm (Boersma et al, 2011).
- A stratospheric (T = 220 K) NO₂ absorption cross section was used. Liquid H₂O and sand absorption cross sections (Richter et al, 2011) were also included to further reduce residuals over deserts and oceans.

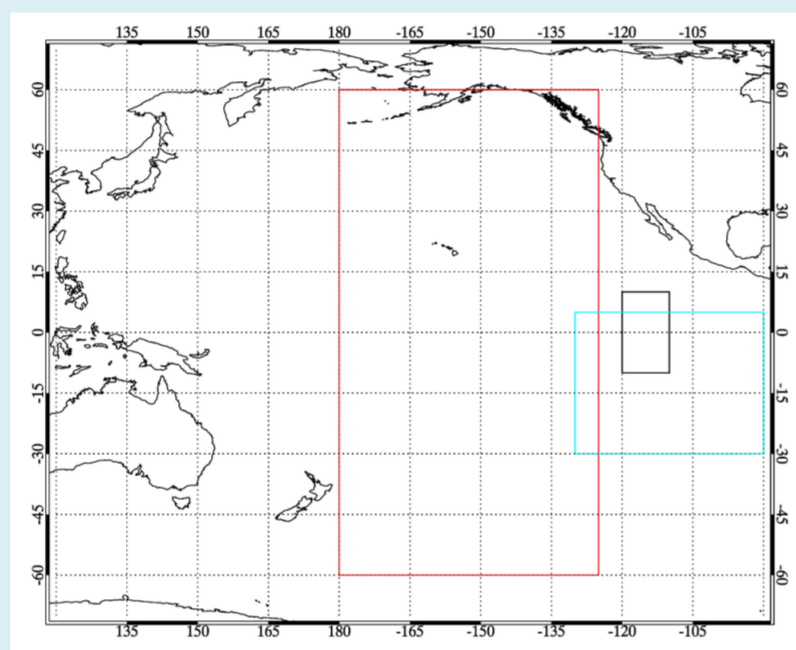


Figure 2: Regions Pacific regions used for earthshine reference spectra collection (red), determination of retrieval uncertainty (black) and cross-track biases (blue)

3. Tropospheric NO₂ SCD Retrieval Results

- Retrieval appears to largely resolve the spatial variability and magnitude of tropospheric NO₂ slant column densities (SCDs) over urban regions.
- Potential sources of residual bias could be from transient longitudinally varying stratospheric NO₂ features outside of the reference sector, or due to absorption from warmer, boundary layer NO₂.
- Global distribution of sand and liquid water SCD appears to be similar to that retrieved by Richter et al (2011). This technique could potentially be used to retrieve aerosol properties over these regions.

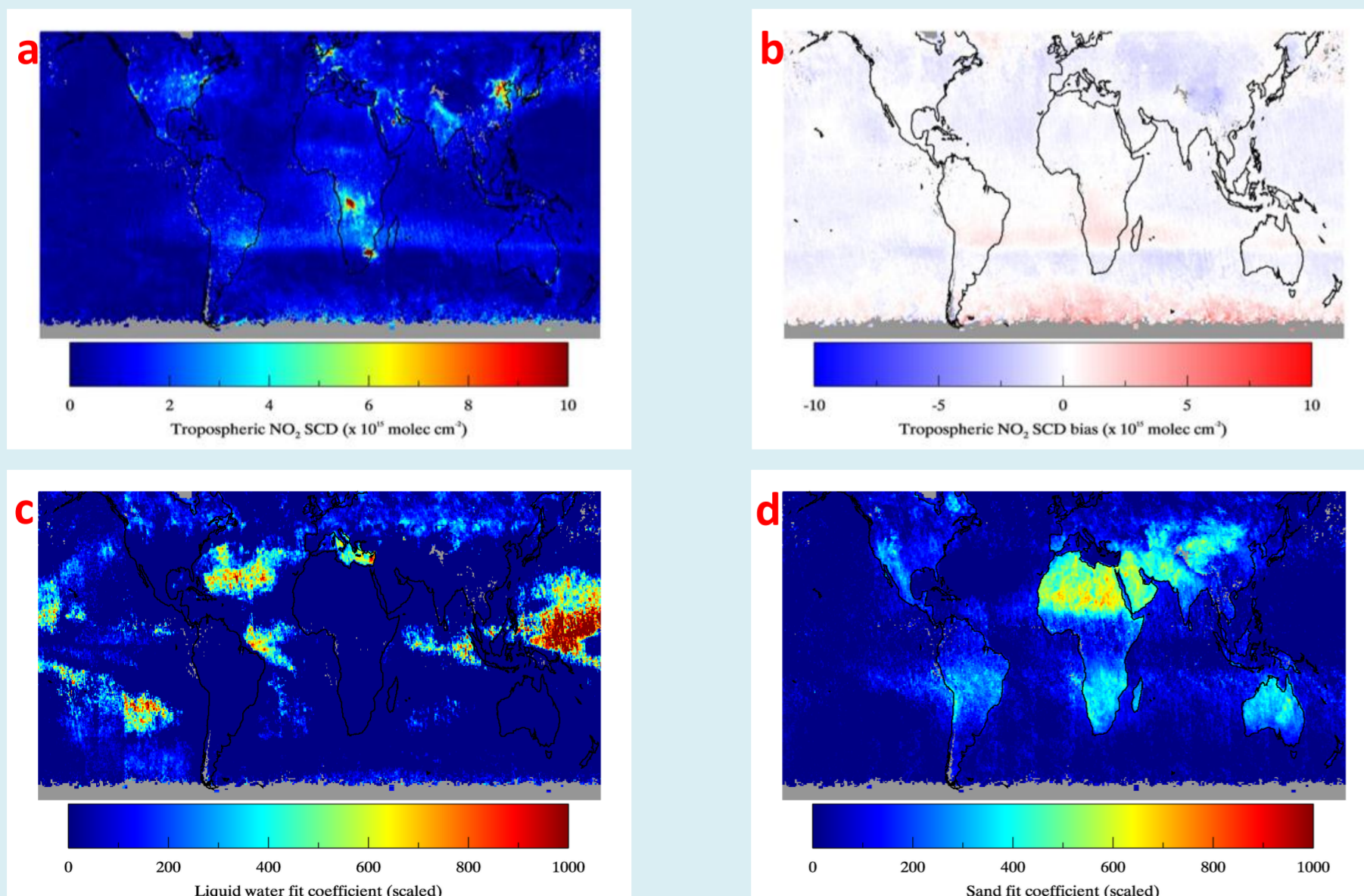


Figure 3: (a) tropospheric SCD retrieved using an earthshine reference, (b) The average bias between the tropospheric SCDs retrieved by this technique and DOMINO, (c) the average liquid H₂O SCD retrieved using an earthshine reference, (d) the average sand SCD retrieved using an earthshine reference.

4. Quantification of Retrieval Improvements

- Cross-track biases arising from diffusor features can also be reduced with this technique. Over the Pacific region in Fig. 2, the average deviation from the along-track mean SCD for each cross-track pixel is calculated and compared with the original and manually destriped mean SCD deviation from the DOMINO and OMNO2 algorithms. As shown in Fig. 4, using an earthshine reference produces less cross-track biases, and the resulting deviations are comparable to existing post-hoc destriping techniques.

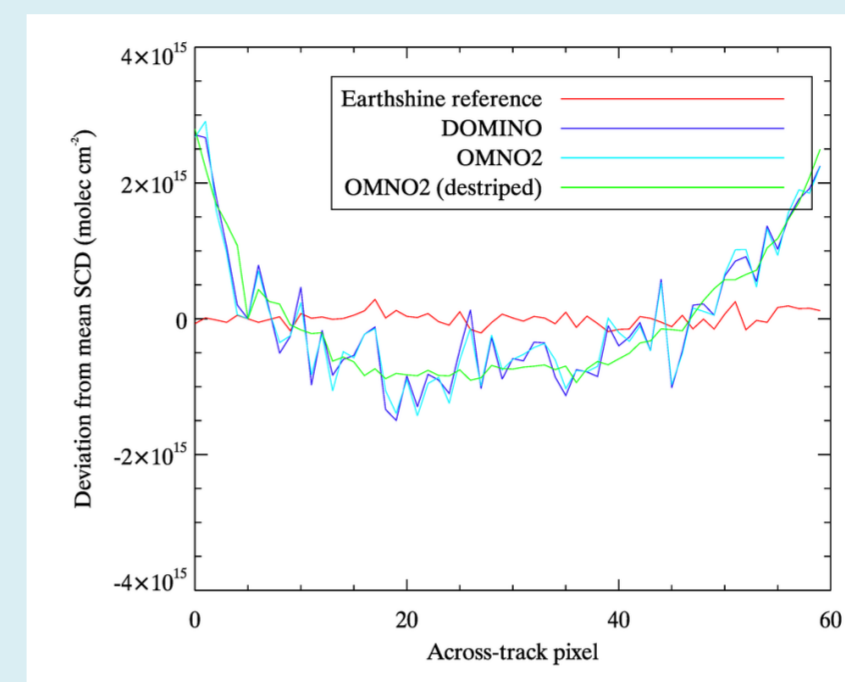


Figure 4: Comparison of cross-track biases in the tropospheric NO₂ SCD retrievals over the region shown in Fig. 2. The earthshine retrieval considerably reduces the across-track biases present in the solar reference retrievals. Note that the earthshine retrieval also reduces the path-length enhancement seen at the swath edge pixels.

- The transect over China in Fig. 5 shows that the variability of the retrieved tropospheric NO₂ SCD largely appears to be similar to those reported by the DOMINO and OMNO2 products over urban areas. While this indicates that the retrieval can observe the spatial variability of NO₂ over urban areas, the warmer boundary layer temperature causes a net offset in the earthshine retrieval.

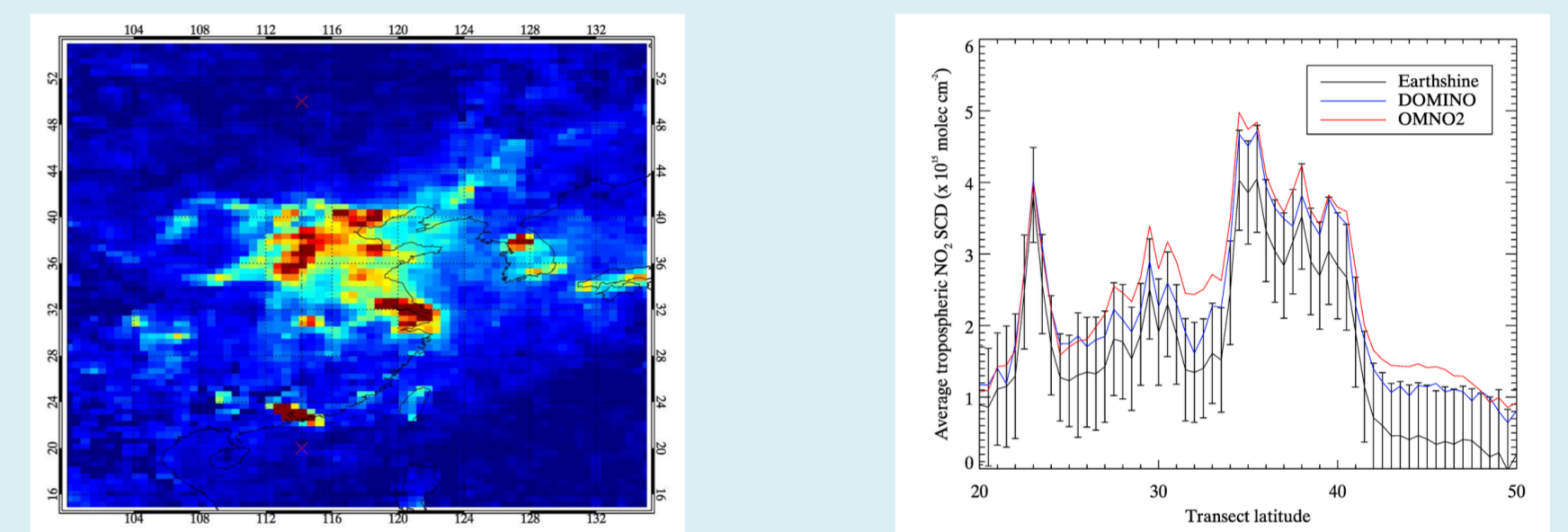


Figure 5: The average tropospheric NO₂ SCD over China as retrieved by DOMINO (left) and the average SCD over the indicated transect retrieved by the earthshine reference, DOMINO and OMNO2 algorithms(right). Error bars are the average DOAS fit error along the transect

- A statistical technique (Valks et al, 2011) can be used to determine the retrieval uncertainty due to noise. Over the region indicated in Fig. 2 the deviations from the local mean SCDs are binned into histograms. These are then fitted with Gaussian functions, where the FWHM represents the retrieval uncertainty. Fig. 6 shows that using the earthshine reference algorithm results in an error of 8.0 x 10¹⁴ molcm⁻² – 27% less than DOMINO. This increase in precision could be due to the removal of noise that would be otherwise present in the irradiance spectra used by both products.

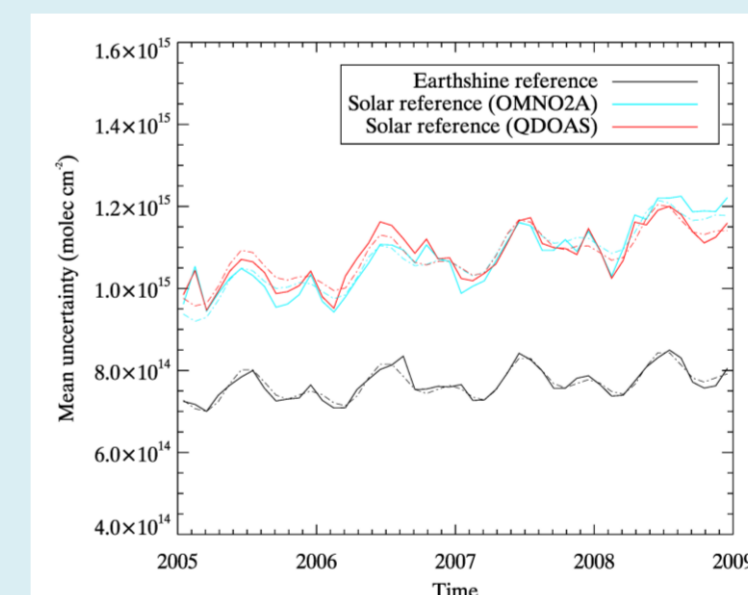


Figure 6: The monthly mean fit uncertainty of retrieved SCDs over the region indicated in Fig. 2, for DOAS retrievals using an earthshine and solar reference (from DOMINO data and DOAS retrievals performed using QDOAS). An annual oscillation was fitted to demonstrate the dependence on the annual irradiation variation.

5. Discussion and Conclusions

Preliminary results suggest that a trop. NO₂ DOAS retrieval using only earthshine reference spectra is viable and results in greater retrieval accuracy

- Retrieval accuracy hindered by cloud cover at northern latitudes in reference sector; may need to decrease temporal resolution of reference spectra.
- Sand SCD spatial variability retrieved over desert regions closely match that found by Richter et al (2011), this could be used to retrieve aerosol properties over these regions.
- Retrieval sensitivity to noise, instrument design, etc is unknown. Similarly, converting SCDs to usable vertical column densities (VCD) will require knowledge of appropriate air mass factors (AMF)
- Future instruments using this technique would have much simpler optical designs and overall have a smaller, less costly payload.
- This work has been submitted to Atmospheric Measurement Techniques (AMT) for publication

6. Future Work

- Determine the effects of stratospheric NO₂ seasonality and longitudinal variation on the reference spectra taken to quantify and remove residual offsets – perhaps multiple reference sectors could be used?
- Incorporate an appropriate AMF calculation into the algorithm, and research the impact of clouds, aerosols and scene inhomogeneity on retrieval accuracy.
- Validate retrieval accuracy using data from past ground-based intercomparison campaigns

7. References

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