Using GEOS-Chem for retrieval of vertical profiles of atmospheric composition over Central London

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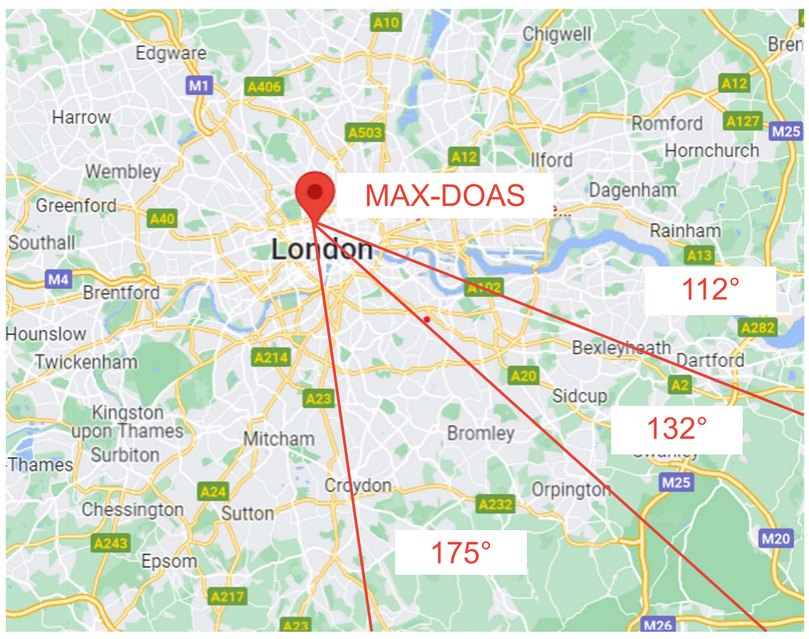
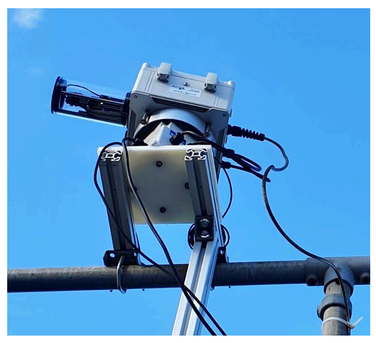
1. Motivation

- There is a lack of vertical profiles of trace gases over Central London.
- The UCL Multi Axis Differential Absorption Spectroscopy (MAX-DOAS) instrument is addresses this with vertical profiles from ~60 m to 8 km..
- It provides invaluable data to assess best understanding of the distribution of air pollution in Central London.

2. The UCL MAX-DOAS

- The UCL MAX-DOAS was installed mid-June 2022 on a 60 m altitude rooftop on the UCL Bloomsbury campus.
- The instrument measures spectra over elevation angles -1°, 1°, 2°, 3°, 5°, 10°, 20°, 40°, and 90°.
- Three azimuth angles (112°, 132° and 175°) scan southeast of London between buildings. Objects on the roof obscure the northerly region.
- The instrument detects UV-Vis active atmospheric species.
- Vertical profiles of HCHO and NO_2 obtained with the RAPDOSI retrieval algorithm have been used to characterize ozone production in London for summer 2022. [1]

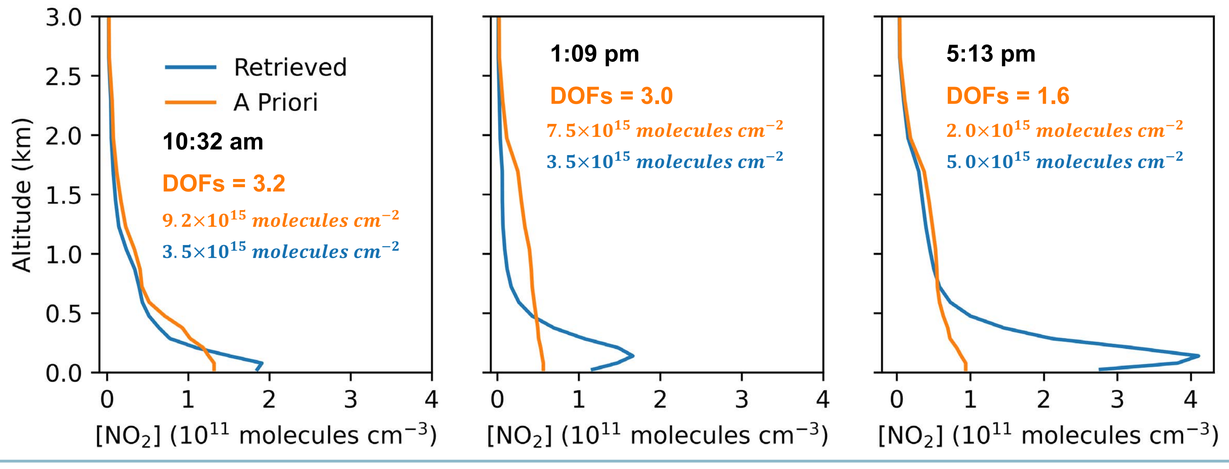




3. GEOS-Chem for MAX-DOAS Retrievals

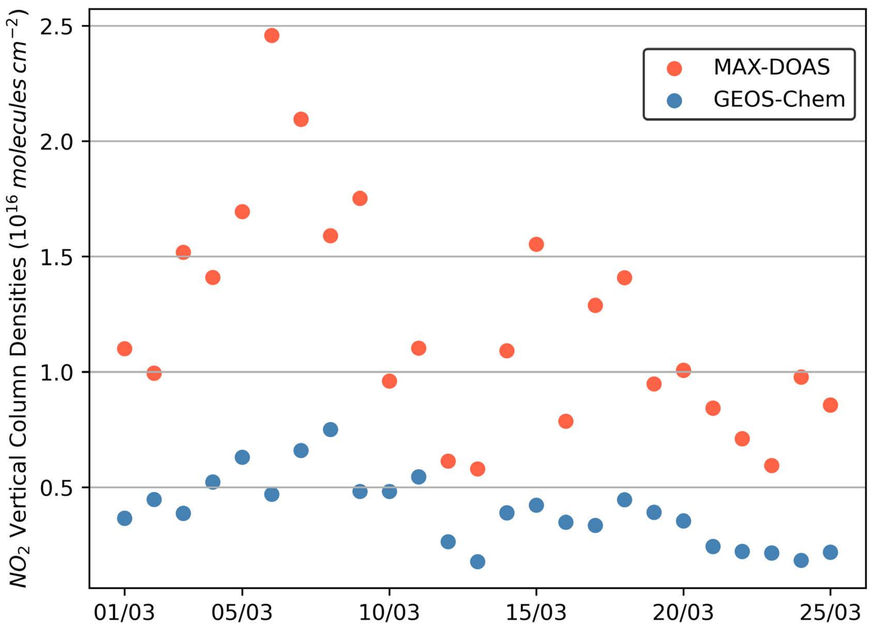
- We use GEOS-Chem nested grid simulations $(0.25°\times0.3125°)$ centered over Greater London $(49.25°N - 59.5°N, 9.375°W - 3.75°E)$ as the a priori for the retrieval of MAX-DOAS vertical profiles.[1]
- Model inputs are NASA GEOS-FP assimilated meteorology and anthropogenic emissions from the UK National Atmospheric Emission Inventory.
- Sensitivity tests show that the retrievals for HCHO and NO_2 are insensitive to the a priori
- MAX-DOAS vertical profiles can be used to assess equivalent GEOS-Chem profiles.
- GEOS-Chem underestimates NO_2 in the ten lowest layers of the MAX-DOAS retrieval (0 – 800 m).
- GEOS-Chem does not capture the diurnal variability in NO_2.

Retrieved MAX-DOAS profiles at 112° and GEOS-Chem a priori profiles on 1 March 2023



4. MAX-DOAS vs GEOS-Chem Columns

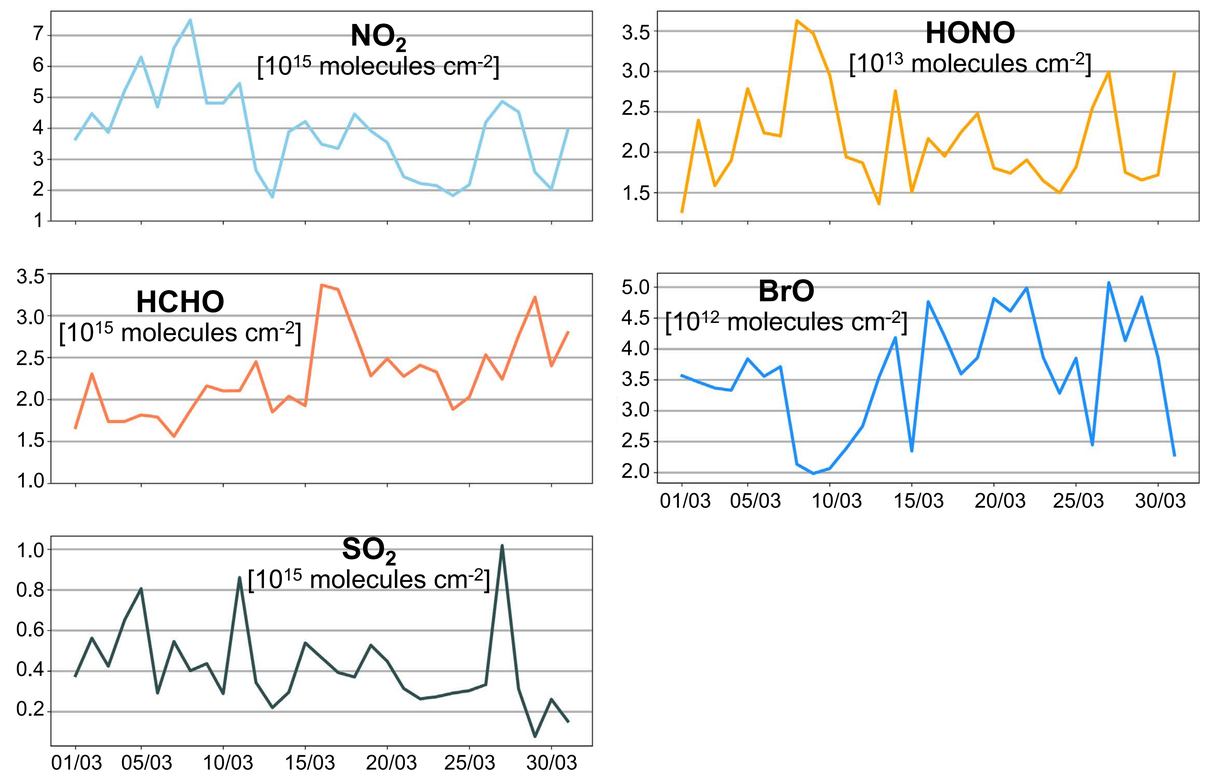
- Preliminary comparison of GEOS-Chem and MAX-DOAS daytime means for March 2023 shows that the model is underestimating NO_2 by 66 %.
- However, the comparison doesn't yet adjust GEOS-Chem by the vertical sensitivity of MAX-DOAS using the retrieved averaging kernels.



- GEOS-Chem is underestimating NO_2 by 66 %
- March in Central London was the wettest March in 40 years. With below average temperatures at the beginning of the month.

5. GEOS-Chem Trace Gas Columns

Daytime means of GEOS-Chem tropospheric columns of trace gases



- For MAX-DOAS observations to be reliable, the concentration of the species must be over the detection limit of the instrument.
- Short lived species such as BrO have been observed in polar regions and the marine boundary layer, but not in a city environment.[2,3]
- GEOS-Chem provides a preliminary assessment of concentrations of UV-vis active species to evaluate whether these could be detected with MAX-DOAS.
- Determination of detection limits for MAX-DOAS is not standardized, so values vary widely. This aspect of the work is underway.

Future Work

- Ongoing retrieval of MAX-DOAS trace gas profiles using GEOS-Chem as a priori.
- Use MAX-DOAS to characterise vertical profiles of pollutants over Central London
- Extend GEOS-Chem vertical column densities to that of MAX-DOAS beyond March 2023 to assess best understanding of pollution over Central London.
- Define the detection limits for trace gases of interest to assess which species can be reliably measured by the UCL MAX-DOAS.

References

1. R. G. Ryan, E. A. Marais, E. Gershenson-Smith, R. Ramsay, J.-P. Muller, J.-L. Tirpitz and U. Frieß, *Atmos. Chem. Phys.*, 2023, **23**, 7121–7139.
2. H. Leser, G. Hönninger and U. Platt, *Geophysical Research Letters*, , DOI:10.1029/2002GL015811.
3. U. Frieß, K. Kreher, R. Querel, H. Schmithüsen, D. Smale, R. Weller and U. Platt, *Atmos. Chem. Phys.*, 2023, **23**, 3207–3232.