Increase of tropospheric ozone over Arabian Sea during pre-monsoon season

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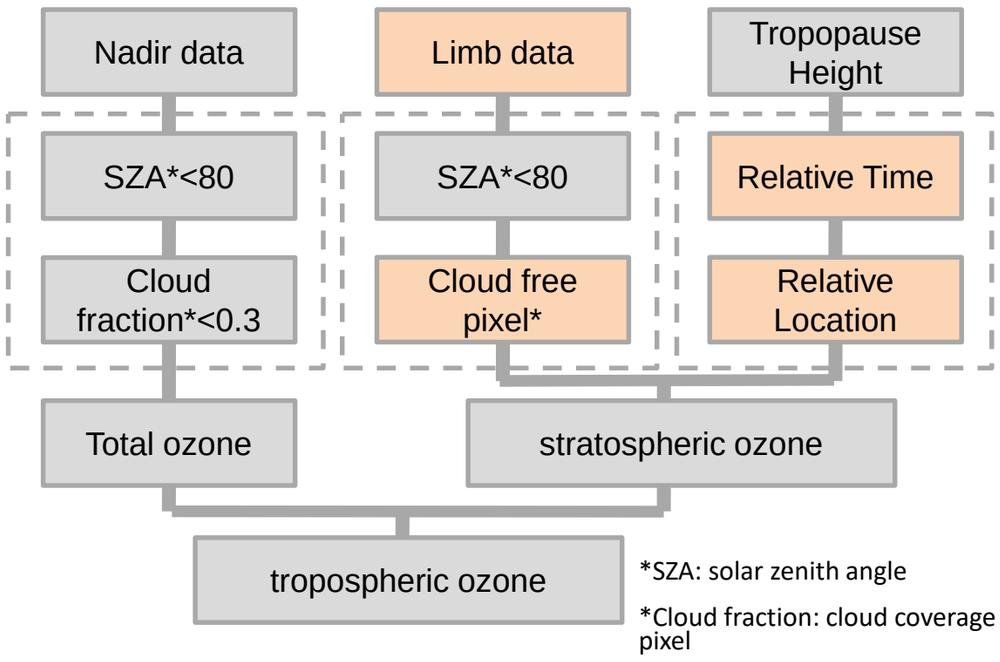
1. Introduction - SCIAMACHY LNM method

Tropospheric ozone (O_3) , one of the most important greenhouse gases, is a photochemically produced pollutant and a main component of summer smog. Measurements from the space borne spectrometer SCIAMACHY are well suitable to investigate sources and transport mechanisms of tropospheric O_3 in a global view. Exploiting alternating observations in limb and nadir modes, the Limb-Nadir Matching technique (LNM) is used to retrieve global distributions of the tropospheric O_3 for the entire duration of the SCIAMACHY mission: Aug.2002-Apr.2012 (Bovensmann et al., 1999; Sierk et al., 2006, Eboje et al., 2014) (Fig. 1).



*SZA: solar zenith angle

*Cloud fraction: cloud coverage ratio in a pixel

*Cloud free: the relative cloud top height is lower than the tropopause height

2. Arabian Sea (AS) Trop. O_3 and its variation

With the benefit of the improved tropospheric O_3 column product from SCIAMACHY LNM retrieval, the spring O_3 maxima over the Arabian Sea (AS) during the pre-monsoon can be seen (Fig. 2) (Jia et al., 2016). The AS is a remote area with few local pollutions. However, a spring enhancement of ~42 DU in monthly average (Fig. 3) is identified from our study, which is similar in magnitude to the well-known biomass burning plume in the southern hemisphere. With the help of MACC reanalysis data, our results showed that ¾ of the enhanced O_3 is contributed in the 0-8 km height range. The monthly averaged O_3 and CO have very similar spring variation in the year 2008-2011 (Fig. 3).

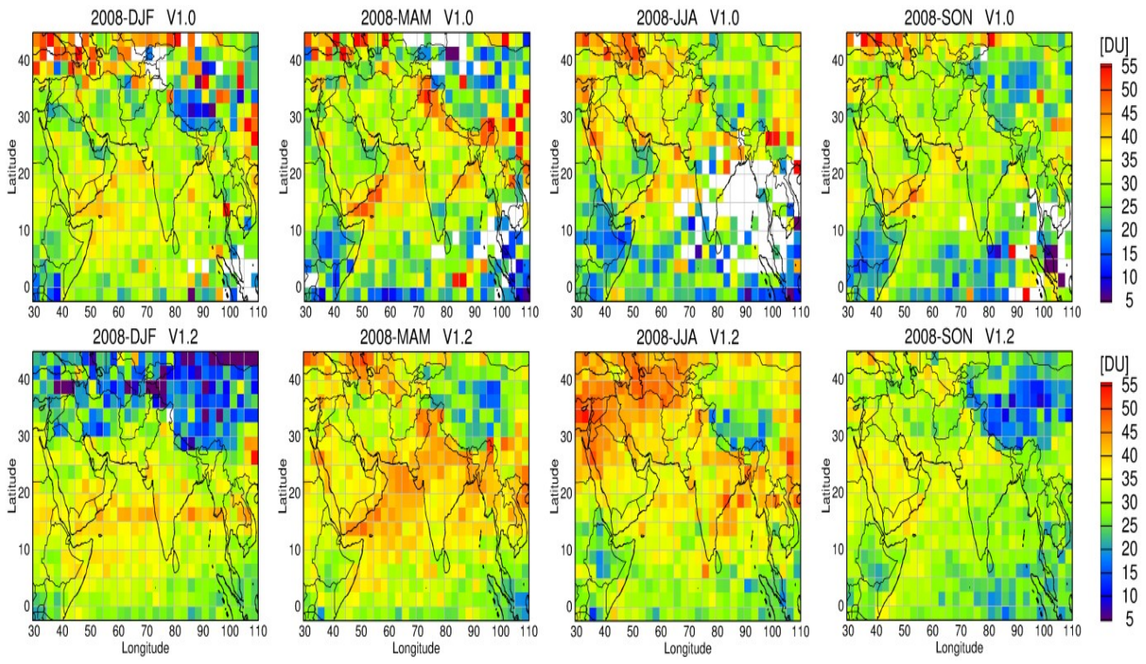


Fig. 2 SCIAMACHY LNM tropospheric ozone columns in the year 2008. From left to right: DJF, MAM, JJA, SON. Upper panels are V1.0 (old) products, lower panels are V1.2 (new) products.

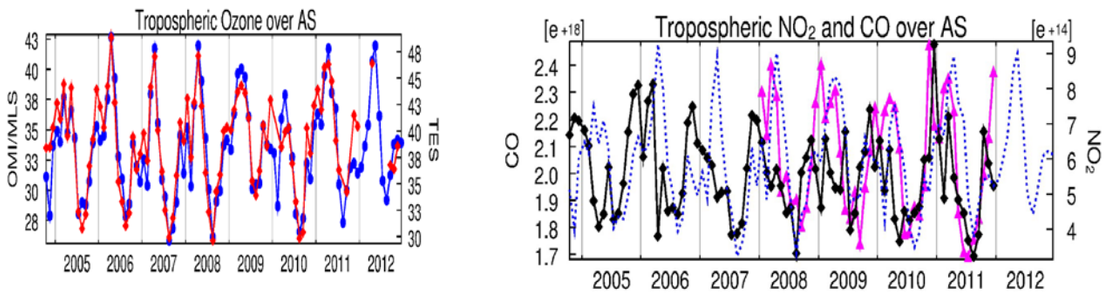


Fig. 3 Trace gas time series over AS (10-20° N, 60-70° E) from 2004 to 2012. The blue (solid and dot) curve represents OMI/MLS ozone, red is TES O_3, magenta is IASI CO and black stands for SCIAMACHY NO_2. The vertical columns are given in DU for O_3 and molec/cm^2 for NO_2 and CO.

3. Potential sources for Trop. O_3

1) LRT: The similar seasonal variation between O_3 and CO and the previous studies both suggest that LRT of O_3 plays an important role in the AS pre-monsoon O_3 pool. Our simulation using MOZART-4 CTM identified that the sources are mainly Middle East, India, Africa and North America at lower troposphere (0-8km). The transporting source region distribution varies in different altitude range (Fig. 4).

2) Pollutant accumulation: As seen in Fig. 5, The air masses at lower troposphere subsides locally with anti-circulation in 10 days. Furthermore, the air masses over AS at 4-8 km are rather dry compared to the surroundings (not shown here), which reduced the impact of HO_x removal on O_3. The accumulation of O_3 pollutant is considered to also contribute to the O_3 pool (Fig. 6).

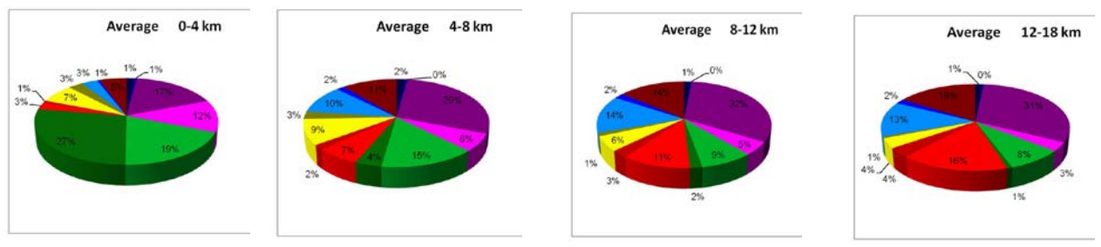


Fig. 4 Upper panel: Regional separation for tracer tagging with distributions of the spring mean emission rate (µg/m2/s) of NO (including anthropogenic, biomass burning, and soil emissions) at the surface used in the model simulations during 1997–2007. Lower panel: Averaged LRT contributions from different source regions to the 4 atmospheric layers over AS in April 1997-2007. PBL (planetary boundary layer) is defined as the region from surface to the top of the boundary layer. FT (free troposphere) is defined as extending to the tropopause above the BL.

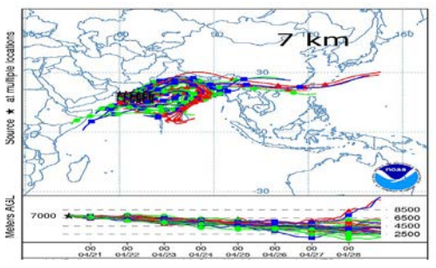


Fig. 5 HYSPLIT trajectory model (240 hr forward) results for air masses at AS with source location at 7km in 20 April, 2008.

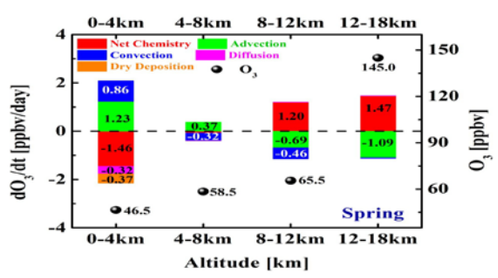


Fig. 6 Averaged O_3 budget in pre-monsoon from MOZART-4 at four layers (0-4, 4-8, 8-12, 12-18 km) over AS region.

3) Local chemistry: One question is that has more O_3 been photochemically produced during the long accumulating time in the middle (4-8 km) or lower (0-4 km) troposphere? Our O_3 budget study shown: in the 0-4 km layer, the destruction of O by OH radicals and reactive halogens in the marine boundary layer domains. In the 4-8 km layer, the chemistry budget is rather small. In the higher layers, photochemical production becomes a major source of O_3. However, these products are quickly transported horizontally in the entire latitude bin by strong advection.

3. Summary

1) The tropospheric O_3 column produced by SCIAMACHY LNM retrieval is significantly improved. The comparison with O_3-sonde show yearly mean differences of less than 5 DU globally. More details can be observed on global distribution.

2) A tropospheric O_3 enhancement of ~ 42 DU over AS in pre-monsoon was observed from satellite observations and MACC model, 3/4 of which is contributed by the 0-8 km height range.

3) Our study suggested the sources of tropospheric O_3 to be mainly LRT from 'Euro_FT' (including Middle East and Africa) with 30% contribution in average, followed by 'India' region with over 20%. The source regions and contributions are identified by analysing the MOZART-4 CTM model simulations.

4) Local chemical production mainly happens at >8 km altitudes and the produced O_3 are removed from AS by advection. The photochemical production of O_3 is negligible in the 4-8 km range.

4. Selected references

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