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Supplement of

Spatial and temporal representativeness of point measurements for nitrogen dioxide pollution levels in cities

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In order to verify whether the LP DOAS measurements are representative for the whole city, NO₂ data from the Ozone Monitoring Instrument (OMI) on-board the NASA Aura satellite was used. Satellite measurements are commonly used for global scale long-term observation of aerosols and trace gases (Silvern et al., 2019; Zara et al., 2018; Laughner and Cohen, 2017; Inness et al., 2015). Validation studies revealed that satellite retrievals generally underestimate urban areas but also
5 found good correlations between satellite and ground based observations (Chan et al., 2018; Lin et al., 2012; Lamsal et al., 2008; Wenig et al., 2008; Petritoli et al., 2004). Section 2 presents the comparison of LP DOAS NO₂ measurements with OMI satellite data, and analyzes the characteristics of seasonal NO₂ variation.

S1 The Ozone Monitoring Instrument (OMI) satellite observations

The Ozone Monitoring Instrument (OMI) is an imaging spectrometer on board the NASA Aura satellite. It measures earthshine
10 radiances with two grating spectrometers which cover the wavelength range from 264 nm to 504 nm. OMI provides the daily measurements of NO₂, BrO, SO₂, O₃, HCHO, OCIO, BrO and aerosols in a global coverage. It is able to detect the cloud radiance fraction, cloud pressure and albedo.

NASA's OMI standard product version 3 (SPv3) (Krotkov et al., 2017; Marchenko et al., 2015) is used in this study. The OMI NO₂ data is publicly available at the Goddard Earth Sciences Data and Information Services Center (GES DISC)
15 (<https://disc.gsfc.nasa.gov/>). For our comparison we gridded the OMI VCDs onto a high resolution grid with $0.02^\circ \times 0.02^\circ$ following the approach described in previous studies (Chan et al., 2015; Kuhlmann et al., 2014).

S1.1 Converting OMI vertical column densities to ground mixing ratio using modeled NO₂ profiles

In order to examine how representative the LP DOAS data is for the temporal pattern observed by OMI, which covers the entire city, OMI's vertical column densities (VCDs) are converted into ground concentrations. For the conversion, vertical
20 profile information is needed. We utilized NO₂ vertical profile information simulated by the chemistry transport model (CTM) GEOS-Chem (Bey et al., 2001). The horizontal resolution of the simulation is 2.0° (latitude) \times 2.5° . Vertical profiles of NO₂ are spatially interpolated within the 4 closest grid cell to the measurement location. Detailed description of the GEOS-Chem simulation can be found in previous studies (Chan, 2017a, b).

Since we use the LP DOAS data for the diurnal correction, we tested the correlation between LP DOAS measurement values
25 and OMI data. The OMI satellite measurements cover a larger area of Munich with the instrument's ground pixel footprint of $\sim 320 - 6400 \text{ km}^2$ than the 2 km path length of the LP DOAS instrument. This correlation refers to the spatial coverage of course, and not necessarily also to the diurnal cycle, since OMI measures only once per day.

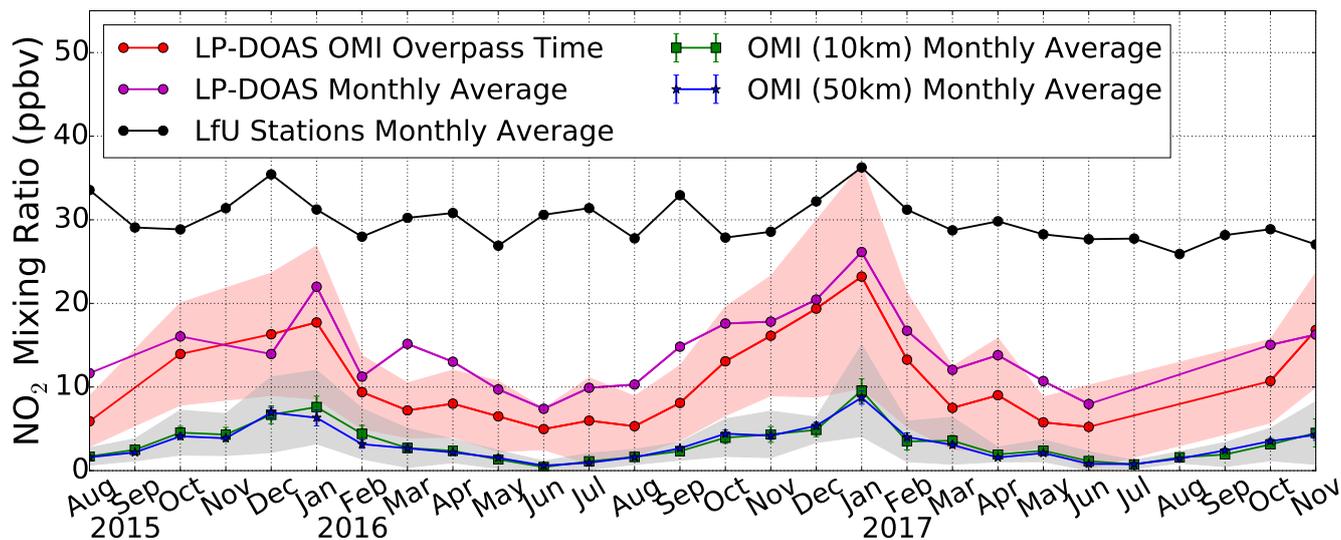


Figure S1. Monthly average ground-level NO₂ mixing ratio. The monthly average NO₂ of the governmental monitoring stations (Landshuter Allee, Lothstraße, Stachus) is shown. LP DOAS data are temporal averages around OMI overpass time (12:30 – 14:30 UTC). OMI data are spatial averages over pixels within a 10 km and 50 km radius of our institute. The red shadow indicate the variability of 1 σ (standard deviation) of averaged LP DOAS measurements of OMI overpass time. The light gray regions indicate 1 σ variability of OMI data within

S2 Comparison between ground measurement and converted OMI observation

Satellite measurements are strongly affected by clouds, as clouds shield ground level NO₂. Hence, to compare with the LP DOAS data, OMI data with cloud fractions larger than 50 %, which were significantly influenced by clouds, were filtered out. LP DOAS data from 12:00 - 15:00 UTC, same as the OMI overpass time for Munich, were used for comparing with the average

5 OMI data sets within 10 km and 50 km from the measurement site (Figure 1). Since OMI only has one measurement per day pass over Germany, and it is able to detect cloud radiance fraction, cloud pressure and albedo, too. Based on different weather condition, the viewing pixel may be missing and has large uncertainty. Each day is gridded separately and then averaged for one month. To reduce the impact of clouds and local spatial variations, we use monthly average data to compare. The uncertainty of the LP DOAS measurements, which was smaller than 1 ppb for a single measurement, is too small to be shown for monthly

10 averages so the standard deviation within each month is shown in Figure 1. Observation of LP DOAS and OMI both showed a similar annual trends, with higher NO₂ levels in winter and lower NO₂ levels in summer.

Comparing monthly means of LP DOAS overpass time measurements with OMI retrieved ground mixing ratios of NO₂ and VCDs within 10 km show correlation coefficients of 0.85 and 0.72, respectively (Figure 2). OMI and the corresponding overpass time measurements of LP DOAS correlate well, indicating OMI measured reliable tendency of ground level NO₂.

15 The discrepancy of correlation coefficients were mainly caused by the vertical profile used for the OMI retrieval and the

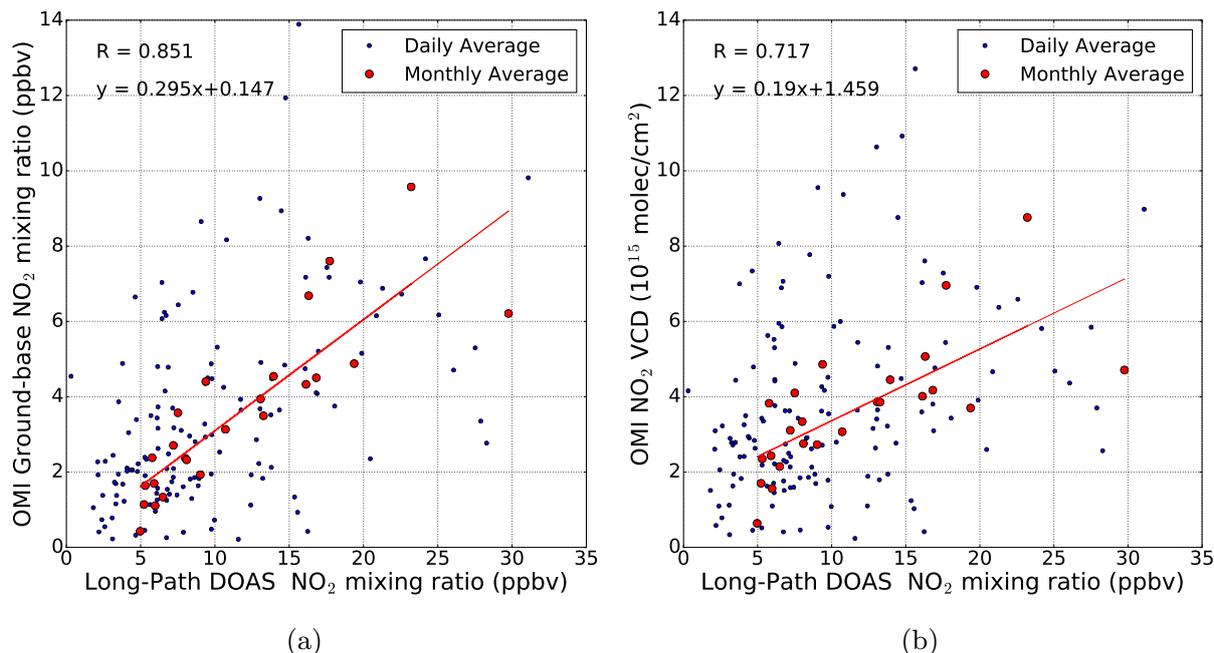


Figure S2. Correlation of LP DOAS measurements with retrieved NO₂ mixing ratio (ppbv) in (a) and VCD in (b) of OMI observation.

conversion of VCDs to ground level mixing ratios. In order to show the influence of temporal averaging and assess the temporal representativeness of the governmental monitoring station data, monthly averages for all LP DOAS data and the data of three governmental monitoring stations are shown in Figure 1 as well. In addition, the data of the governmental monitoring stations were 15.4 ppb on average higher than the LP DOAS measurements. The Pearson correlation between the two data sets was 0.32. The low correlation may be due to the different measurement areas, measurement heights and measurement resolution.

Average variability of the NO₂ mixing ratios in winter (November to February) and summer (June to August) were 16.5 ppb and 5.5 ppb. The errors bars of OMI data do not overlap with the LP DOAS data for most months, neither within 10 km nor within 50 km, which demonstrated that there might be systematic errors in the conversion of VCDs to ground level mixing ratios or in the OMI retrieval process itself (Wenig et al., 2008). As the previous study also suggests, lower OMI values over cities compared to ground measurements could be due to the OMI a-priori profile used for both, the VCD retrieval and converting the tropospheric NO₂ VCDs to ground level mixing ratios, was taking an average over a larger area, not only urban areas, but also rural areas with a lower ground level mixing ratio to total column ratios. A total underestimation for the ground level NO₂ of about 69 % can be observed. A similar result was found by Kuhlmann et al. (2015). However, because of the good correlation it is safe to assume that relative temporal changes captured by the LP DOAS can be regarded as representative for area covered by OMI that spans the entire city of Munich.

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