COMPARISON OF VERTICAL NO2 PROFILES MEASURED IN-SITU FROM A QUADCOPTER, RETRIEVED FROM MAX-DOAS OBSERVATIONS AND COMPUTED USING THE CHIMERE MODEL

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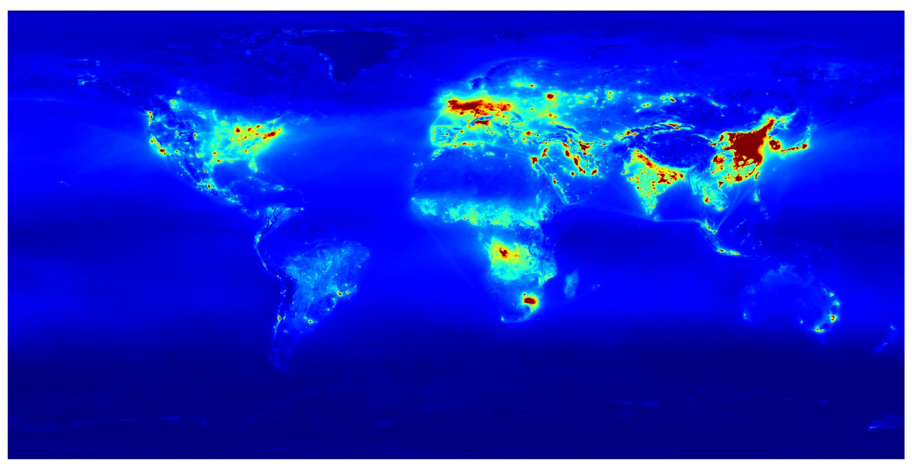
Abstract

During the Research on the Simulation and Mechanism of the impacts of Black Carbon on Climate and Environment atmospheric measurement campaign carried out near Nanjing, China in June 2018, a lightweight, accurate nitrogen dioxide (NO_2) sensor was attached to a quadcopter to measure vertical profiles of NO_2. Between 1 and 14 June 2018, ~50 vertical NO2 profiles were measured inside the planetary boundary layer up to an altitude of 900-1300 meters during 13 subsequent measurement days. Six NO_2 soundings were conducted on a daily basis at approximately 8 AM (morning), 12 & 4 PM (afternoon), 8 PM (evening) and 12 & 4 AM (night). The NO2 measurements were calibrated using a scaling factor derived from a side-by-side inter comparison with a commercial NO2 analyzer operated by NUIST prior to the start of the campaign. These measurements clearly demonstrate the diurnal cycle of NO2, including the emergence of elevated concentrations close to the surface during the night and early morning and the mixing of the boundary layer from sunrise onward resulting in flat NO2 vertical profile shapes with lower concentrations. The in-situ NO2 vertical profile shapes were compared to NO2 profile information retrieved from nearby MAX-DOAS observations as well as computed using the CHIMERE chemistry-transport model. This comparison demonstrates that in-situ quadcopter measurements could play an important role in the validation of future geostationary satellites since the diurnal cycle of NO2 will have an impact on the accuracy of the satellite retrievals and is not always flawlessly captured by commonly used measurement techniques and models.

Introduction

Nitrogen dioxide (NO_2) is a key component of air pollution worldwide.
The KNMI Research & Development Satellite Observations department monitors trace gases such as NO_2 from space using satellite instruments such as the TROPOspheric Monitoring Instrument (TROPOMI).



Global distribution of tropospheric NO_2 as observed in 2019-2021 by TROPOMI. Figure by Henk Eskes and Bas Mijling, KNMI.

The amount of NO_2 in the atmosphere derived from TROPOMI observations is often divided in a tropospheric and a stratospheric amount.

Large uncertainty in satellite derivations:
about 30-40% of the tropospheric amount.

One of the major sources of uncertainty:
the assumed vertical NO_2 profile shape in the troposphere.

Next to satellite validation, measurement of the vertical distribution of NO_2 is essential to:

- Study NO_x (NO_2 + nitrous oxide (NO)) photochemistry and (aerosol) dynamics
- Validate results from chemical models
- Understand effects of regional transport
- Identify emitting sources and sinks[1]

Objective

Investigate whether the currently widely available **MAX-DOAS** observations and chemical transport **models** provide **enough information** about the **vertical NO_2 distribution** to validate satellite observations.

Investigate whether - under certain atmospheric conditions - additional in-situ **drone or balloon measurements** of NO_2 are needed to **improve** the **a priori vertical NO_2 profile** that plays an important role in the processing of satellite observations.

Methods

KNMI NO_2-sonde: Deployment of a self-developed, accurate in-situ NO_2 sensor based on wet luminol chemiluminescence, mounted on a quadcopter. Performing 4-hourly soundings up to ~1 km, resulting in 36 calibrated high resolution vertical NO_2 profiles recorded throughout the day and night.[2]

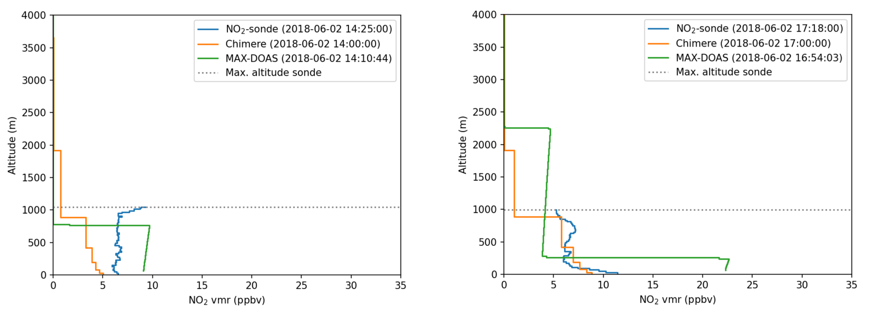
MAX-DOAS: spectral measurements were performed with a mini-MAXDOAS instrument installed on top of the xx building at the NUIST campus. Subsequently, a retrieval method developed at the KNMI was used to produce tropospheric columns and low-resolution vertical profiles of NO_2.[3]

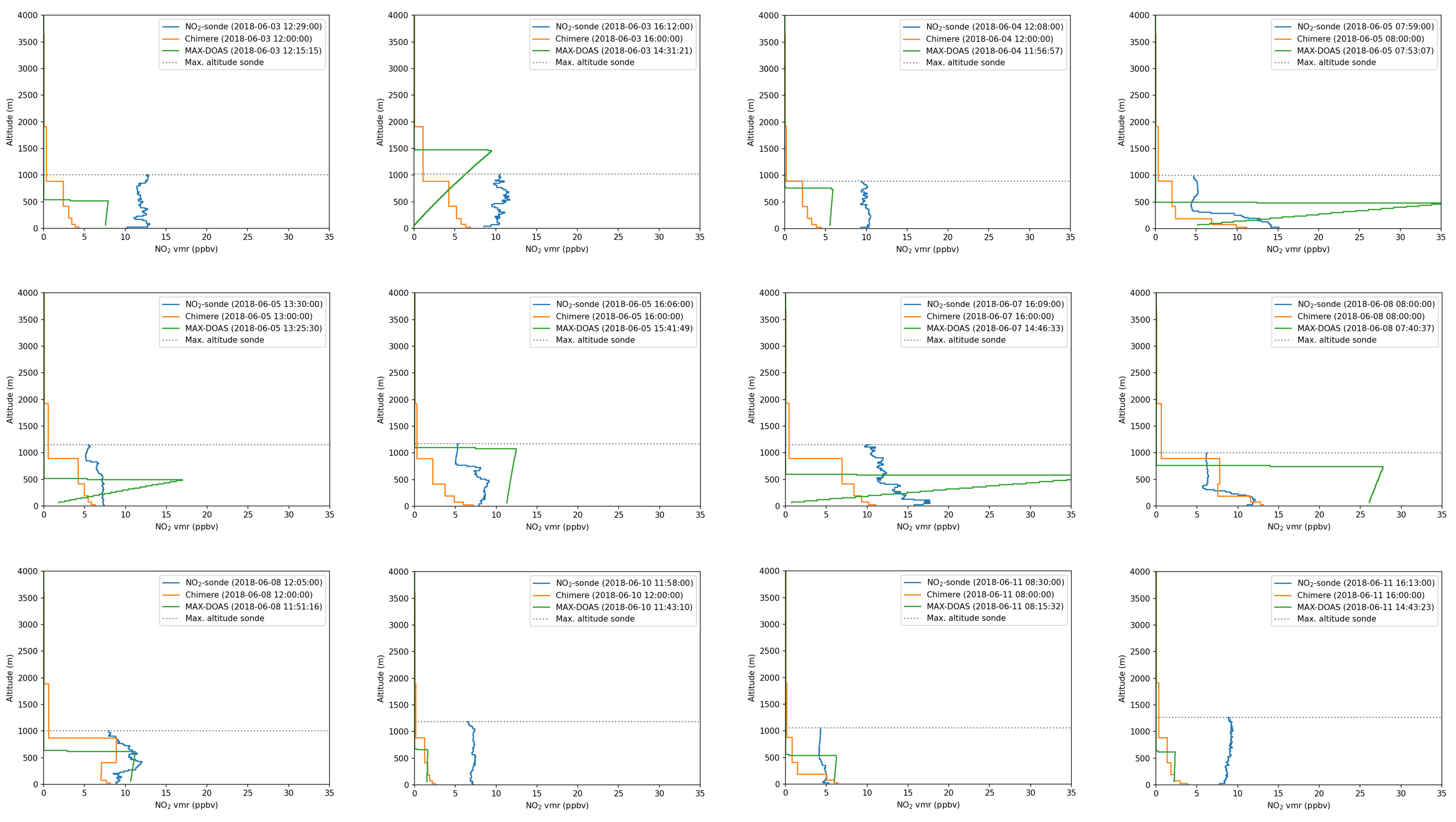
CHIMERE: The open-source multi-scale chemistry-transport model CHIMERE was run for the research region with a spatial resolution of 0.1° to produce hourly simulations of the vertical distribution of NO_2 over 8 atmospheric layers between 0 and ~6 km. Meteorological data from ECMWF was used.

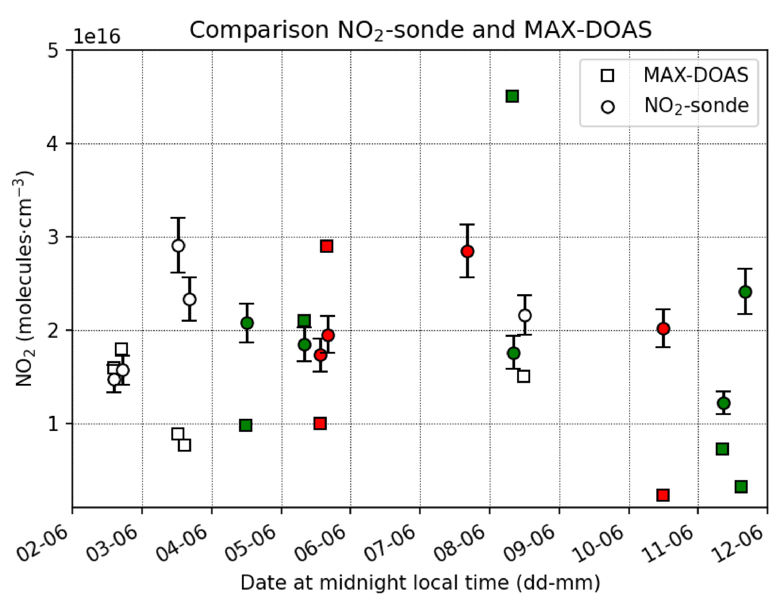
Analysis: Effects of wind direction, wind speed, time and cloud cover on the relative error between NO_2-sonde and MAX-DOAS NO_2 column concentrations were tested using logistic regression and Python 3.6.

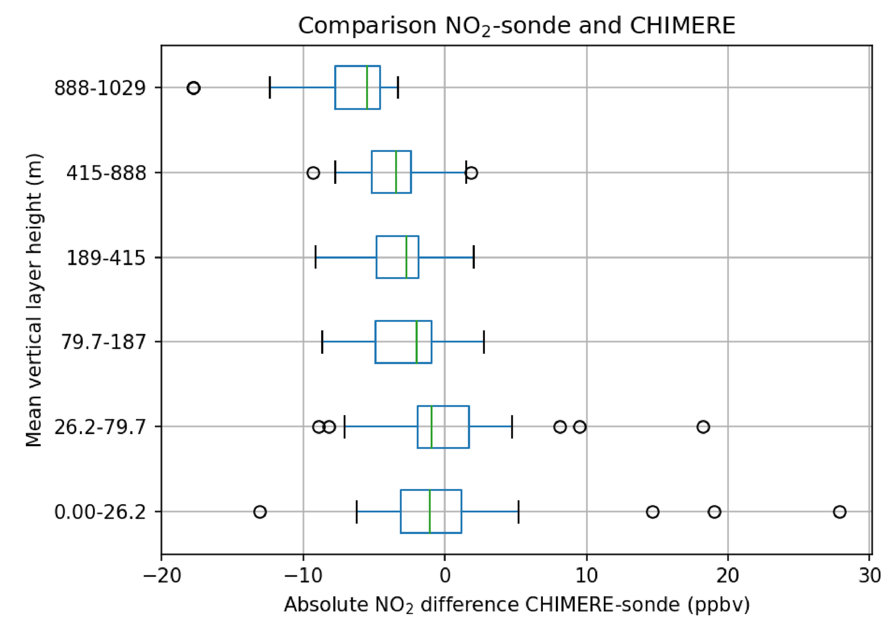
Results

Colocated vertical NO_2 profiles (ppbv). An in-situ NO_2 profile measured with the NO_2-sonde is depicted here (blue) if a MAX-DOAS NO_2 profile could be retrieved within 2 hours (green). The computed CHIMERE NO_2 profile closest in time to the NO_2-sonde measurement is shown in orange.









White: cloud cover/height unknown

- Large differences in **relative error** between NO_2-sonde and MAX-DOAS NO_2 column concentrations ranging from **-89 to 155 %.**
- **3 cases with low relative error (8-15%)**. No common contributing factors could be identified. **2/3 conducted during weekend** (lower local traffic/industrial etc. emissions).
- Effects of **wind direction, wind speed, time, cloud cover not statistically significant.** 13 collocated profiles: insufficient sample sizes.
- **Median absolute NO_2 error is negative:** CHIMERE systematically underestimates the NO_2 vmr for every vertical layer on the drone measurement site. This can be explained by the fact that the modeled vmrs apply to a larger region (0.1°) while NO_2 surface concentrations have a higher spatial variability.
- **More and larger outliers in the lower layers and decreasing IQRs with height** for the NO_2 absolute error: modeled NO_2 vmrs are less accurate closer to the surface where NO_2 is emitted.

Discussion & Conclusion

- **Difficult to draw conclusions about the comparison of NO_2-sonde and MAX-DOAS column concentrations. Too little data** for a representative statistical analysis. Future research will focus on:
  - 1) **improving cloud identification and cloud property characterization** using the color index derived from MAX-DOAS observations (radiance ratio at two selected wavelengths) and
  - 2) **optimizing the MAX-DOAS retrieval for the prevailing climatology in Nanjing**. Applied algorithm rejects NO_2 profiles based on statistics, some of which may still be of sufficient quality.
- **Median absolute NO_2 error (CHIMERE-sonde) ranged from -1 ppbv for the first 2 atmospheric layers (~0-80 m) up to -5 ppbv for the highest comparable layer (~888-1029 m)**. The model had a good overall performance given the fact that the profiles included in the analysis were captured during the entire NO_2 diurnal cycle. Possible differences between PBL phases will be investigated.

Major References

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