

Evaluation of on-road mobile NO_x emissions using modeling and satellite NO₂ retrievals: Application to the Great Lakes Region

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Background

- A reliable NO_x (NO+NO₂) emission inventory is very important
 - NO₂ is a criteria air pollutant
 - NO_x participate in the formation of O₃ and particulate matter
- Possible overestimates of NO_x emissions in the 2011 US National Emission Inventory (NEI), likely associated with mobile sources (~30%) (Travis et al., 2016; Souri et al., 2016; McDonald et al., 2018)
- 50% reduction of NO_x emissions from on-road mobile sources in the 2011 NEI led to better agreement of the CMAQ simulation with ground-based measurements of NO_x and O₃ in the Great Lakes Region (Qin et al., 2018)

Methodology

Model-satellite comparison

- CMAQv5.1**
 - One-way nested: 12 km/4 km (Fig. 1)
 - Period of interest: July 2011
 - 2011 NEI with in-line calculations for BEIS and point sources
 - CB05e51
- Two runs
 - Base case
 - 50% of NO_x emissions from mobile sources
- NO₂ vertical columns** calculated using the 4-km simulation between 13:00 and 16:00 EST



Fig.1 Modeling domains

OMI Level 2 NO₂ data product (OMNO2) version 3.0

- Resolution: 13km×26km to 40km×250km
- Pass-over time: approximately 13:45 Local Time
- Global coverage every other day
- Vertical column densities (VCDs)** derived using tropospheric NO₂ slant column densities (SCDs) from OMNO2 and air mass factors (AMFs):

$$AMF = \frac{SCD}{VCD}$$

where AMFs depends on the position of the Sun (Solar Zenith Angle), the viewing angle of OMI, vertical distribution of NO₂, surface albedo, etc.

- NO₂ vertical profiles in CMAQ (instead of GMI for the operational retrieval) used in the calculation of AMFs (Goldberg et al., 2017)

Source apportionment of NO_x using Decoupled Direct Method (DDM)

- The first-order sensitivity (S⁽¹⁾) of NO_x concentration to NO_x emission from an individual sector reflects contribution of the NO_x emission source to overall NO_x concentration (with Δε of -1):

$$C_{(1+\Delta\epsilon)} = C_{E_0} + \frac{1}{11} \Delta\epsilon E_0 \frac{\partial C}{\partial E} \Big|_{E=E_0} = C_{E_0} + \Delta\epsilon S^{(1)}$$

- Onroad, ptegu, nonroad, ptonipm (point sources not included in EGU or oil/gas), c1c2 rail (C1 and C2 commercial marine emissions plus railroad emissions) and beis (> 80% of NO_x emission)
- Convert to contributions to NO_x columns

Results and discussions

Tropospheric NO₂ Columns in CMAQ vs. OMI

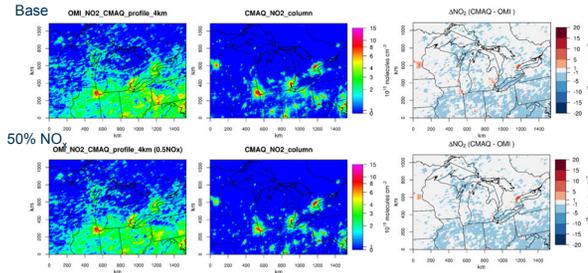


Fig. 2 Tropospheric NO₂ columns derived from OMI retrievals (left) & CMAQ simulations (middle), and differences between OMI and CMAQ (right). (Top: base simulation; Bottom: 50% reduction in NO_x emissions from mobile sources in the US)

Table 1 Overall evaluation of CMAQ simulations compared to OMI retrievals.

Case	N	Mean Bias (10 ¹⁵)	Mean Error (10 ¹⁵)	Fractional Bias (%)	Fractional Error (%)	r ²
Base	104829	-0.69	0.83	-65%	76%	0.41
50% NO _x	104829	-0.68	0.78	-68%	77%	0.39

Model-satellite Gap ← ? → On-road Emissions

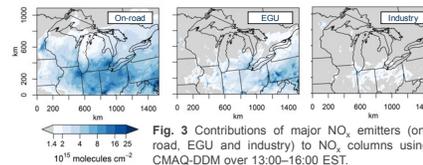


Fig. 3 Contributions of major NO_x emitters (on-road, EGU and industry) to NO₂ columns using CMAQ-DDM over 13:00-16:00 EST.

- CMAQ simulated higher NO₂ columns than OMI retrievals above the 90th (85th) percentile in the base case at grid cells where on-road (other) sources dominate (Fig. 4)
- Reduction in on-road NO_x emissions led to better agreement between CMAQ and OMI at the high end of NO₂ columns (>5×10¹⁵ molecules/cm²) in locations where on-road sources dominate, while no improvement was observed at other grid cells

Compared to Ground-based Measurements

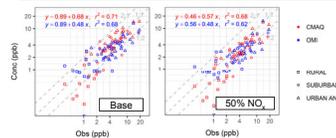


Fig. 5 Comparison of NO₂ surface concentrations derived using CMAQ and OMI with ground-based measurements at the AQS sites. NO₂ columns from OMI are converted to concentrations with the concentration-to-column ratios in CMAQ.

Base simulation

- Rural: CMAQ < OMI (sensitive to upper tropospheric NO_x) (Napelenok et al., 2008; Goldberg et al., 2017)
- Urban: CMAQ > OMI (likely due to overestimation of anthropogenic NO_x emissions)
- Overall: CMAQ < OMI (Table 1)

50% NO_x emission reduction (US mobile)

- Reduced model-satellite differences in urban areas, e.g., Chicago, Detroit
- No significant changes in evaluation statistics (MB, ME, FB, FE and r²) compared to the base case

Results and discussions

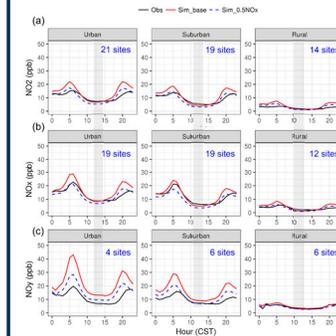


Fig. 6 Diurnal trends of NO₂, NO_x and NO_y in CMAQ simulations compared to ground-based measurements at the AQS sites. Shaded area indicates OMI pass-over time

- OMI mostly underestimates NO₂ concentrations, regardless of location
- Reduction in NO_x emissions decreases high biases of CMAQ NO₂ either in urban/suburban areas and at peaks (Fig. 6)
- Good agreement with measured NO₂/NO_x at the surface during satellite pass-over time

Conclusions

- CMAQ shows low biases in NO₂ columns / surface concentrations against OMI retrievals / ground-based measurements in rural areas, with high biases in urban areas (not in all locations)
- Decreased emissions from on-road mobile sources in CMAQ reduce differences of CMAQ simulations with OMI retrievals & ground-based measurements at the high end of NO₂ columns / concentrations in urban areas
- Overestimation of NO₂ columns in CMAQ relative to OMI occurs in locations where other sources (e.g., EGU) dominate as well, which needs further investigation

Acknowledgements

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References

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