Application of OMI NO_2 retrievals to the evaluation of NO_x emissions from on-road mobile sources in the Great Lakes Region

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Background

- A reliable NO_x ($NO+NO_2$) emission inventory is very important
  - NO_2 is a criteria air pollutant
  - NO_x participate in the formation of O_3 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 O_3 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
- NO_2 vertical columns calculated using the 4-km simulation between 13:00 and 16:00 EST

Fig.1 Modeling domains

OMI Level 2 NO_2 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 NO_2 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 NO_2, surface albedo, etc.

- NO_2 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):

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

- Onroad, ptegu, nonroad, ptnonipm (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 NO_2 Columns in CMAQ vs. OMI

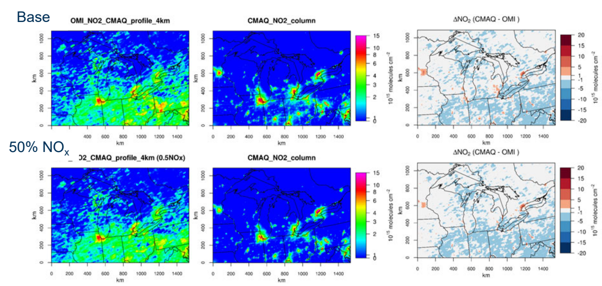


Fig. 2 Tropospheric NO_2 columns derived from OMI retrievals (left) and CMAQ simulations (middle), and differences between OMI and CMAQ (right). (Top: base simulation; Bottom: 50% reduction in NO_x emissions from mobile sources in the US)

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% NOx emission reduction (US mobile)

- Reduced model-satellite differences in urban areas i.e., Chicago, Detroit
- No significant changes in evaluation statistics (MB, ME, FB, FE and r^2) compared to the base case

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

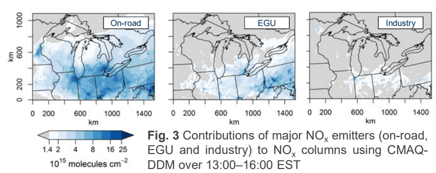
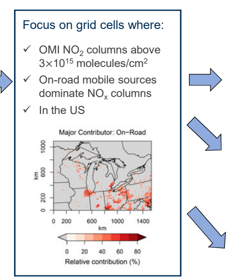


Fig. 3 Contributions of major NO_x emitters (on-road, EGU and industry) to NO_x columns using CMAQ-DDM over 13:00–16:00 EST



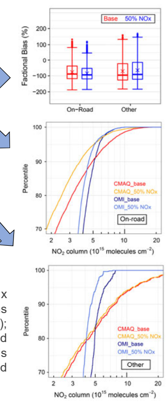


Fig. 4 Fractional biases (boxplot with x indicating mean) of CMAQ simulations compared to OMI retrievals (Top); cumulative distributions of CMAQ- and OMI-derived NO_2 columns at grid cells where on-road sources (middle) and other sources (bottom) dominate

- CMAQ simulates higher NO_2 columns above the 90[th] (85[th]) percentile in the base case than OMI retrievals at grid cells where on-road (other) sources dominate (Fig. 4)
- Reduction in on-road NO_x emissions leads to better agreement between CMAQ and OMI at the high end of NO_2 columns (>5×10^{15} molecules/cm^2) in locations where on-road sources dominate, while the improvement is not seen at other grid cells

Compared to Ground-based Measurements

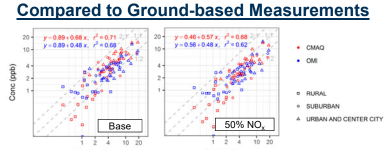


Fig. 5 Comparison of NO_2 surface concentrations derived using CMAQ and OMI with ground-based measurements at the AQS sites. NO_2 columns from OMI are converted to concentrations with the concentration-to-column ratios in CMAQ.

- Significant underestimation of NO_2 surface concentrations at the low end in the CMAQ base case; mostly occurs in rural areas, with overestimation at urban & suburban sites at times (Fig. 5)

Results and discussions

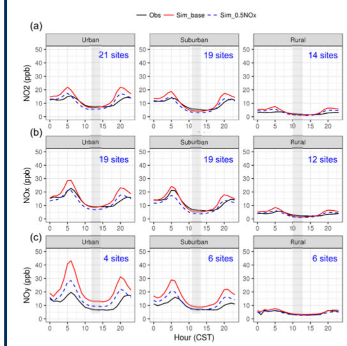


Fig. 6 Diurnal trends of NO_2, NO_x and NO_y in CMAQ simulations compared to ground-based measurements at the AQS sites. Shaded area indicates OMI pass-over time

- OMI mostly underestimates NO_2 concentrations, regardless of location
- Reduction in NO_x emissions decreases high biases of CMAQ NO_2 both in urban/suburban areas and at peaks (Fig. 6)
- Good agreement with measured NO_2 / NO_x at the surface during satellite pass-over time

Conclusions

- CMAQ shows low biases in NO_2 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 NO_2 columns / concentrations in urban areas
- Overestimation of NO_2 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

Goldberg et al., (2017). Doi: https://doi.org/10.5194/acp-17-11403-2017
McDonald et al., (2018). Doi: https://doi.org/10.1021/acs.est.8b00778
Napelenok et al., (2008). Doi: https://doi.org/10.5194/acp-8-5603-2008
Souri et al., (2016). Doi: https://doi.org/10.1016/j.atmosenv.2016.02.020
Travis et al., (2016). Doi: https://doi.org/10.5194/acp-16-13561-2016