Supporting figures for

Comprehensive evaluations of diurnal NO_2 measurements during DISCOVER-AQ 2011: Effects of resolution dependent representation of NO_x emissions

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Figure S1. The locations of surface and P-3B aircraft observations during the DISCOVER-AQ 2011 campaign. We mark the 36-km REAM grid cells with red lines and the 4-km REAM grid cells with black lines. Gray shading denotes land surface in the nested 4-km WRF domain, while white denotes ocean/water surface. Black squares denote surface O_3 observation sites. Circles denote surface NO_2 observation sites, and their colors denote different measurement instruments: blue for the Thermo Electron 42C-Y NO_y analyzer, dark orchid for the Ecotech Model 9841/9843 T-NO_y analyzers, black for the Thermo Model 42C NO_x analyzer, and chocolate for the Teledyne API model 200eup photolytic NO_x analyzer. Triangles denote Pandora sites, and the cyan triangle denotes a Pandora site (USNA) on a ship. Green dots denote the inland P-3B aircraft spiral locations.

Figure S2. Distributions of NO_x emissions for the (a) 36-km and (b) 4-km REAM simulations. Here NO_x emissions refer to the mean values (molecules km^{-2} s^{-1}) in one week (Monday – Sunday).

Figure S3. Comparisons of (a, b) U wind, (c, d) V wind, and (e, f) temperature (T) between P-3B spirals and coincident WRF simulation results in July 2011. For P-3B observations, we derive U wind and V wind from wind speed and wind direction (Figure S4), which were measured via a Honeywell INS sensor. The accuracies of P-3B wind speed and wind direction are 1 m s^{-1} and ±5°, respectively. Temperature from P-3B was measured by using a Rosemount model 102 sensor with an accuracy of ±0.2 °C. The left panel is for comparisons between P-3B and the 36-km WRF simulation, and the right panel is for comparisons between P-3B and the nested 4-km WRF simulation. WRF wind components have been rotated to earth coordinates.

Figure S4. Wind roses for P-3B observations and coincident WRF simulation results in July 2011, (a) for the 36-km WRF simulation, (b) for the nested 4-km WRF simulation, and (c) for P-3B observations. WRF wind components have been rotated to earth coordinates. "SpdAve" denotes the average of wind speed, "SpdStd" denotes the standard deviation of wind speed, and "DirAve" denotes wind direction derived from averaged U-wind and V-wind.

Figure S5. Distributions of (a, c) precipitation amounts and (b, d) precipitation durations in July 2011 for (a, b) Stage IV and (c, d) the 36-km WRF simulation. (e) and (f) are the differences in precipitation amount and precipitation duration between Stage IV and the 36-km WRF simulation. The original resolution of Stage IV is about 4 km over polar stereographic grids, and we regrid the dataset to the 36-km WRF pixels. Triangles denote the inland Pandora sites in Figure S1.

Figure S6. Distributions of (a, c) precipitation amounts and (b, d) precipitation durations in July 2011 for (a, b) Stage IV and (c, d) the nested 4-km WRF simulation. (e) and (f) stand for the differences in precipitation amount and precipitation duration between Stage IV and the nested 4-km WRF simulation. The original resolution of Stage IV is about 4 km over polar stereographic grids, and we regrid the dataset to the nested 4-km WRF pixels. Due to significantly lower precipitation amounts in the nested 4-km WRF simulation, we use different color bars for WRF and Stage IV data. Triangles denote the inland Pandora sites in Figure S1.

Figure S7. Distributions of (a, b) large-scale and (c, d) convective precipitation (a, c) amounts and (b, d) durations in July 2011 for the 36-km WRF simulation. Due to significantly lower large-scale precipitation amounts compared to convective precipitation, we use different color bars for them. Triangles denote the inland Pandora sites in Figure S1.

Figure S8. Diurnal variations of the solar zenith angles of Pandora measurements in July 2011. Here we use monthly averages of the 11 inland Pandora sites (Table S1 and Figure S1). Error bars denote standard deviations.

Figure S9. Daily variations of Pandora NO_2 TVCDs at the 11 inland sites for three randomly selected days in July 2011. Blue dots denote Pandora measurements on July 16, 2011 (Saturday), black dots denote July 18, 2011 (Monday), and red dots denote July 27, 2011 (Wednesday). Here Pandora NO_2 TVCDs are calculated by subtracting stratospheric NO_2 VCDs from Pandora total NO_2 VCDs. See the main text for details.

Figure S10. Stratospheric NO_2 VCD diurnal variations at the Greenbelt station in Maryland (39° N, 76.89° W) from the GMI MERRA-2 1° × 1.25° simulation (https://gmi.gsfc.nasa.gov/merra2hindcast/) for July 2011, and the corresponding satellite stratospheric NO_2 VCDs in the DISCOVER-AQ region (about 39.2° N, 76.3° W) (Figure S1). "NASA-OMI" denotes the OMI NO_2 VCDs from NASA, "KNMI-OMI" denotes the OMI NO_2 VCDs from KNMI, and "KNMI-GOME2" denotes the GOME-2A NO_2 VCDs from KNMI. Gray shading and vertical bars denote the standard deviations of the GMI results and satellite stratospheric VCD products, respectively.

Figure S11. Diurnal variations of boundary-layer vertical velocities for the (a) 36-km and (b) nested 4-km WRF simulations in the DISCOVER-AQ region (Figure S1) for July 2011.

Figure S12. Contributions of emission, chemistry, transport, and dry deposition to NO_x VCD diurnal variations in the surface layer of the 36-km REAM simulation in the DISCOVER-AQ region on weekdays in July 2011. "Chem" refers to net NO_x chemistry production in the surface layer; "Emis" refers to NO_x emissions in the surface layer with the impact of vertical turbulent mixing; "Drydep" denotes NO_x dry depositions in the surface layer; "Transport" includes advection, turbulent mixing, lightning NO_x production, and wet deposition in the surface layer. "Total (NO_x)" is the hourly change of surface-layer NO_x VCDs ($\Delta(VCD) = VCD_{t+1} - VCD_t$). "Total ($NO_2$)" is the hourly change of surface-layer NO_2 VCDs, and "Total (NO)" is the hourly change of surface-layer NO VCDs.

Figure S13. Contributions of emission, chemistry, transport, and dry deposition to NO_x TVCD diurnal variations over the 11 inland Pandora sites (Table S1 and Figure S1) on weekdays in July 2011 for the (a) 36-km and (b) 4-km REAM simulations. "Chem" refers to net NO_x chemistry production; "Emis" refers to NO_x emissions; "Drydep" denotes NO_x dry depositions; "Transport" includes advection, turbulent mixing, lightning NO_x production, and wet deposition. "Total (NO_x)" is the hourly change of NO_x TVCDs ($\triangle(TVCD) = TVCD_{t+1} - TVCD_t$). "Total ($NO_2$)" is the hourly change of NO_2 TVCDs, and "Total (NO)" is the hourly change of NO TVCDs.

Figure S14. (a) Percentages of NO_2 VCDs below the heights of Pandora instruments in July 2011 based on 36-km REAM results; (b) the comparison between original Pandora TVCDs ("unscaled") and updated Pandora TVCDs ("scaled") with the inclusion of VCDs below the Pandora instruments. Here we use monthly averages in July 2011. Error bars in (b) denote standard deviations.

Figure S15. Temporal evolutions of derived-NO_y vertical profiles from the (a, b) P-3B aircraft and (c, d) REAM simulations at (a, c) 36-km and (b, d) 4-km resolutions during the DISCOVER-AQ campaign. Error bars denote the corresponding standard deviations. Due to the limited number of P-3B derived-NO_y observations and slightly different heights between 36- and 4-km grid cells from different WRF simulations (Table S2), small differences exist between the 36-km and 4-km observations when we bin them vertically to REAM grid cells (see also Table 1).

Figure S16. Same as Figure S15 but for coincident HNO_3 concentrations.

Figure S17. Comparisons of NO_2 (a, c) TVCDs and (b, d) surface concentrations over the 11 inland Pandora sites between the 4-km and 36-km REAM simulations on (a, b) weekdays and (c, d) weekends for July 2011. "REAM-36km" (black lines) denotes the 36-km REAM simulation results; "REAM-4km" (red lines) denotes the 4-km REAM simulation results; "4km-regrid" refers to the 36-km values by re-gridding the 4-km REAM simulation results into 36-km REAM grid cells. Error bars denote standard deviations.

Figure S18. Comparisons of wind fields between the (a, c, e) 36-km and (b, d, f) nested 4-km WRF simulations for different height (AGL) bins in July 2011. (a) and (b) are for mean wind fields below about 200 m, (c) and (d) are for about 200 – 1000 m, and (e) and (f) are for about 1000 – 3000 m. Triangles denote the inland Pandora sites in Figure S1. The unit of wind speed is m s^{-1}.

Figure S19. Contributions of emission, chemistry, transport, and dry deposition to NO_x TVCD diurnal variations over the six P-3B spiral sites (Figure S1 and Table S1) on weekdays in July 2011 for the (a) 36-km and (b) 4-km REAM simulations. "Chem" refers to net NO_x chemistry production; "Emis" refers to NO_x emissions; "Drydep" denotes NO_x dry depositions; "Transport" includes advection, turbulent mixing, lightning NO_x production, and wet deposition. "Total (NO_x)" is the hourly change of NO_x TVCDs ($\triangle(TVCD) = TVCD_{t+1} - TVCD_t$). "Total ($NO_2$)" is the hourly change of NO_2 TVCDs, and "Total (NO)" is the hourly change of NO TVCDs. (c), the 36-km and 4-km REAM simulated diurnal cycles of NO_2 TVCDs over the P-3B spiral sites on weekdays in July 2011. The black line in (c) denotes the 36-km REAM simulation results, and the red line denotes the 4-km REAM simulation results.

Figure S20. Comparisons of mean surface NO_y concentrations at Padonia, Edgewood, Beltsville, and Aldino between the 4-km and 36-km REAM simulations on (a) weekdays and (b) weekends for July 2011. "REAM-36km" (black lines) denotes the 36-km REAM simulation results; "REAM-4km" (red lines) denotes the 4-km REAM simulation results; "4km-regrid" refers to the 36-km values by re-gridding 4-km REAM simulation results into 36-km REAM grid cells. Error bars denote standard deviations.

Figure S21. Same as Figure 10 but for weekends in July 2011. The domain averages of ACAM and coincident 4-km REAM NO_2 VCDs are 3.0 ± 1.7 and 3.3 ± 2.8 × 10^{15} molecules cm^{-2}, respectively.

Figure S22. Same as Figure 10 but for weekday ACAM datasets obtained from https://www-air.larc.nasa.gov/cgi-bin/ArcView/discover-aq.dc-2011?UC12=1#LIU.XIONG/. The domain averages of ACAM and coincident 4-km REAM NO_2 VCDs are 5.9 ± 1.8 and 4.5 ± 3.1 × 10^{15} molecules cm^{-2}, respectively.

Figure S23. Same as Figure S22 but for weekend ACAM datasets obtained from https://www-air.larc.nasa.gov/cgi-bin/ArcView/discover-aq.dc-2011?UC12=1#LIU.XIONG/. The domain averages of ACAM and coincident 4-km REAM NO_2 VCDs are 4.7 ± 1.4 and 3.4 ± 2.7 × 10^{15} molecules cm^{-2}, respectively.