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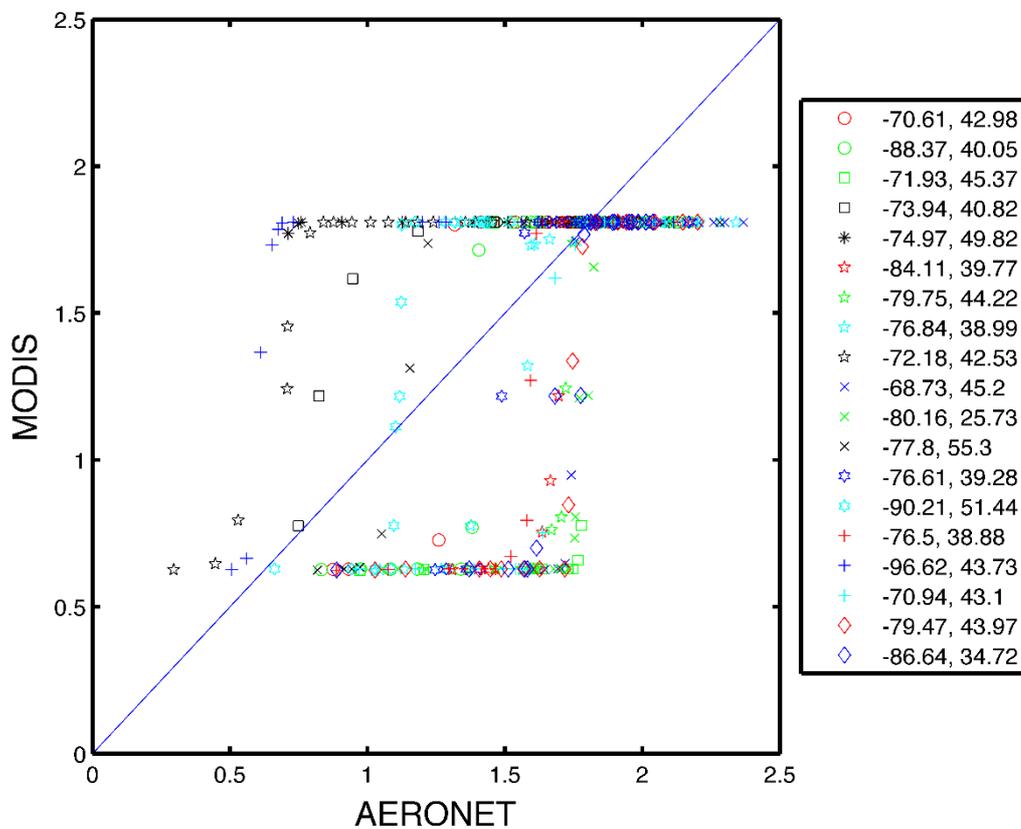
Evaluating the skill of high-resolution WRF-Chem simulations in describing drivers of aerosol direct climate forcing on the regional scale

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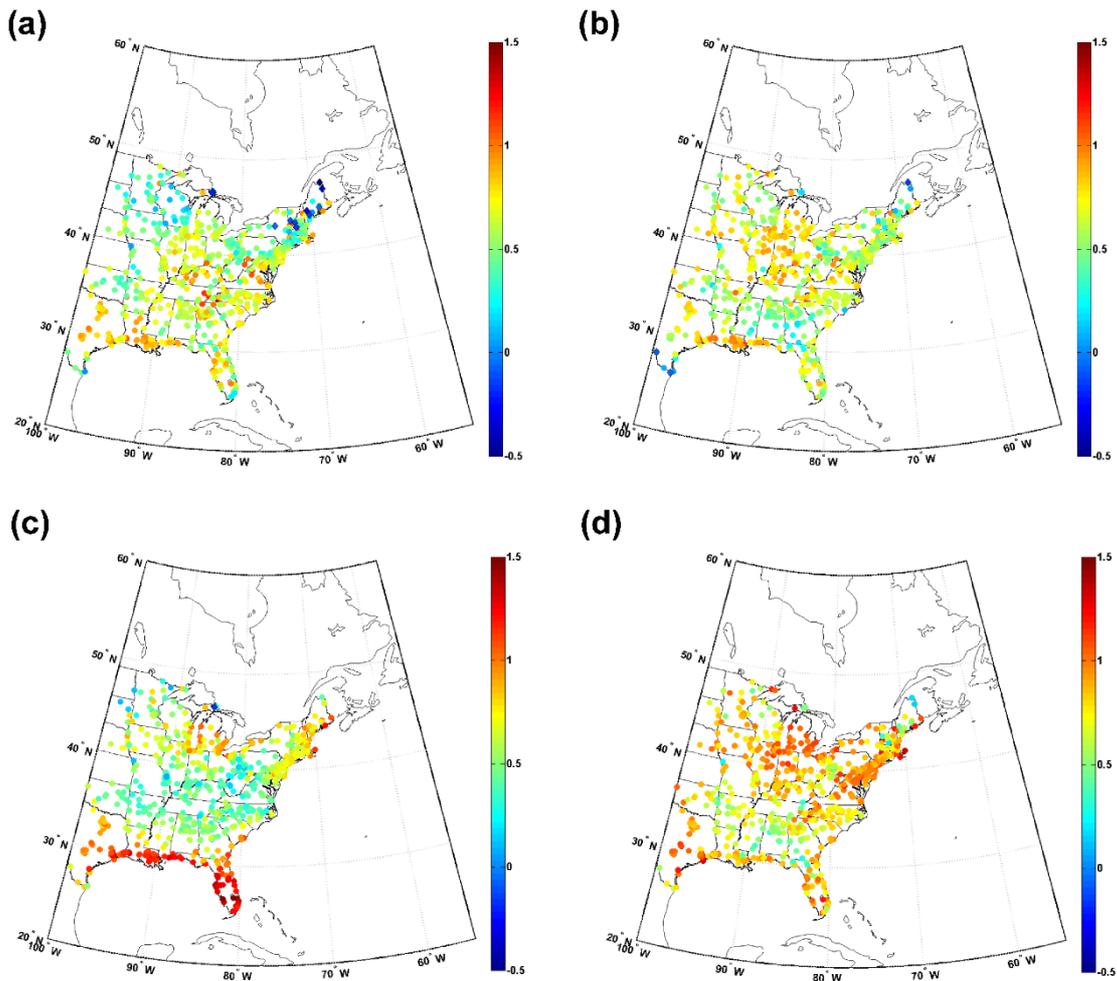
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4 Figure S1. Empirical quantile-quantile plot of simultaneous measurements of AE at 470-660
 5 nm between MODIS (Terra) and AERONET (where the AERONET station longitude (E) and
 6 latitude (N) are given in the legend). As shown, the MODIS data tend to a bimodal distribution,
 7 while in the AERONET observations AE is a continuous variable (or nearly so). Thus, while
 8 in comparison with WRF-Chem simulations AE from AERONET is treated as a continuous
 9 variable, in the majority of comparisons with MODIS a threshold of 1 is applied to identify the
 10 dominance of coarse mode ($AE < 1$) versus fine mode ($AE > 1$).



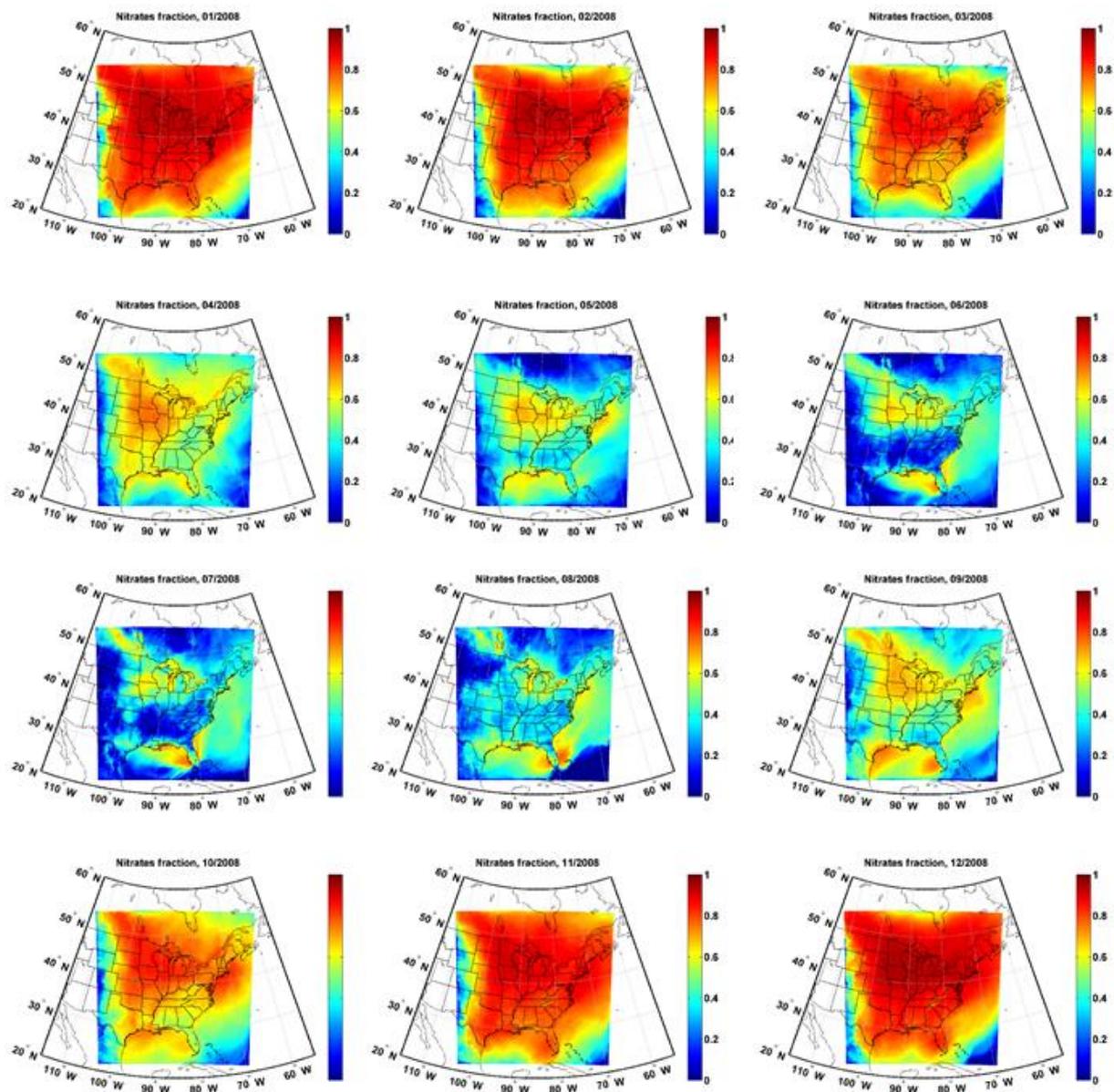
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12 Figure S2. Mean fraction bias (MFB) of near-surface daily mean $PM_{2.5}$ concentrations as
13 simulated by WRF-Chem relative to observations at EPA sites during (a) winter, (b) spring, (c)
14 summer and (d) fall. As shown, near-surface $PM_{2.5}$ concentrations from WRF-Chem exhibit a
15 positive bias (MFB > 0) for most sites and in all seasons, but the bias is largest over the southern
16 states during summer. Note also that the MFB in $PM_{2.5}$ concentrations greatly exceeds that for
17 either AOD or AE (see Fig. 2).



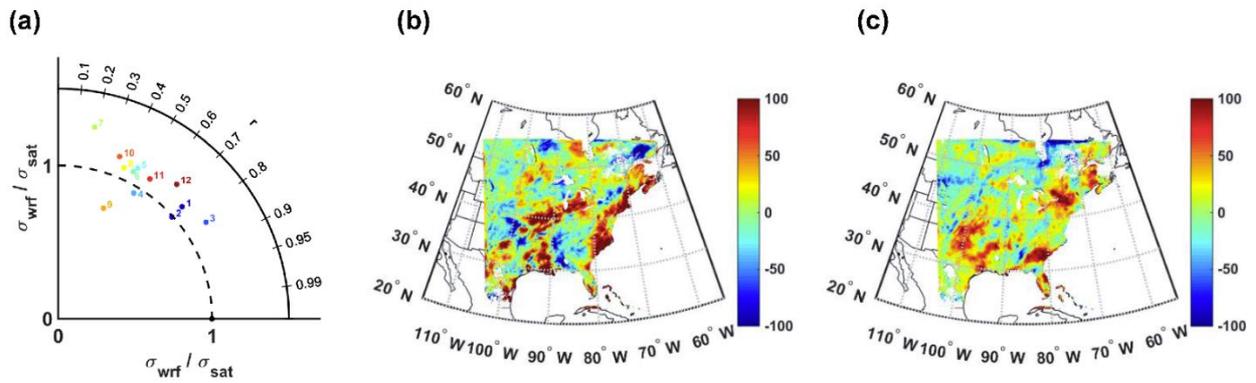
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20 Figure S3. Fraction of near-surface monthly averaged mass concentration of nitrate versus the
21 sum of aerosol nitrate and sulfate as simulated in the accumulation mode by WRF-Chem for
22 each calendar month during 2008.



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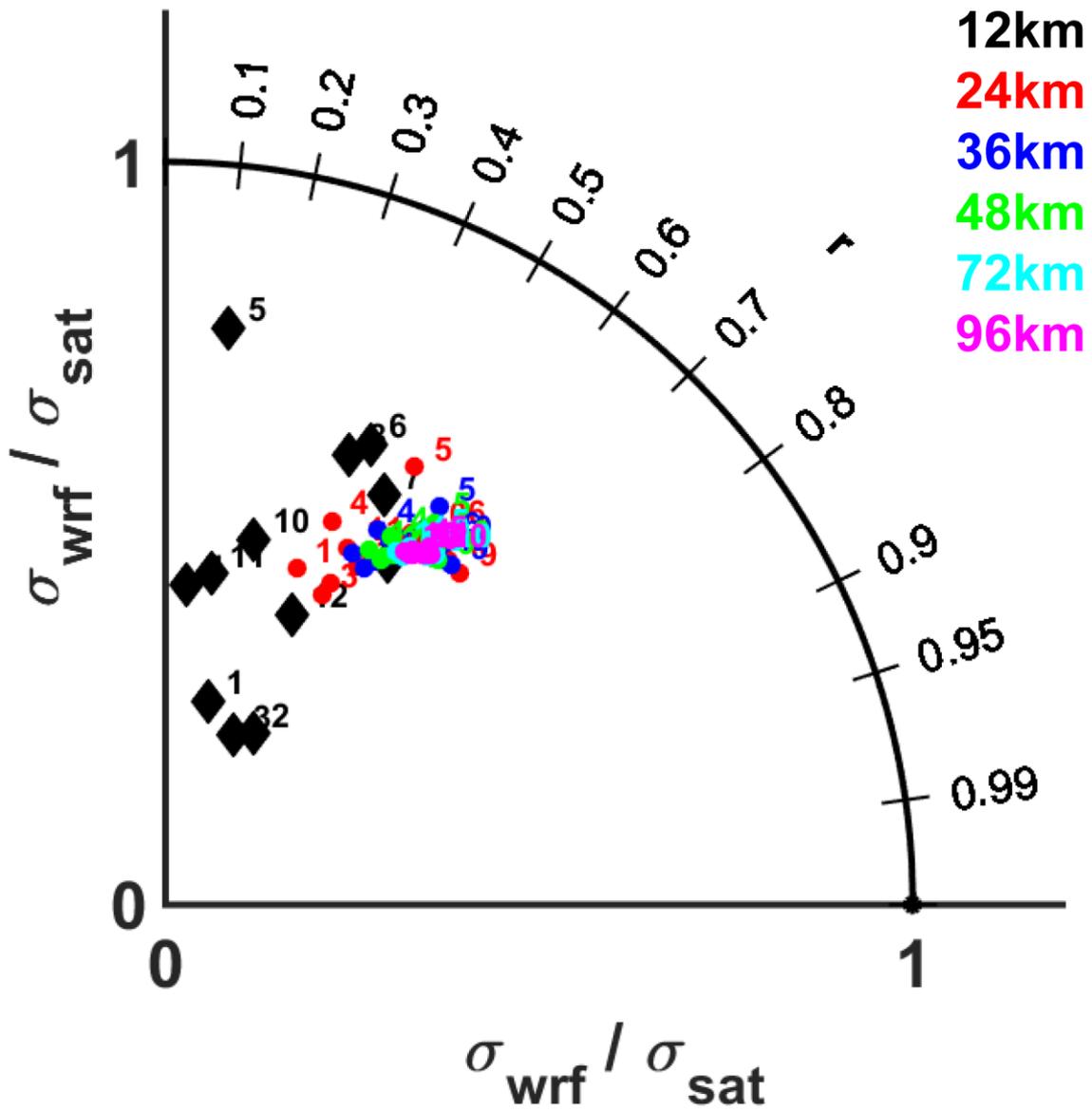
25 Figure S4. (a) Taylor diagram for monthly accumulated precipitation during 2008 as simulated
26 by WRF-Chem and in the gridded observations (Matsuura and Willmott, 2009), after applying
27 a linear interpolation to match the WRF-Chem grid. Results for the individual calendar months
28 are shown by the numbers (1 = January). Panels (b) and (c) show the difference [mm] between
29 observed and simulated accumulated precipitation during the month of (b) September and (c)
30 October 2008. Values larger than zero indicate the observed precipitation is higher than the
31 simulated one.



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34 Figure S5. Taylor diagram for AE when MODIS observations and WRF-Chem simulations at
35 12 km are spatially aggregated to 24, 36, 48, 72 and 96 km. Numbers next to the colored
36 dots/diamonds indicate different months (e.g. 1 = Jan).



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39 Figure S6. Extreme AOD (i.e. occurrence of AOD > 75th percentile) from WRF-Chem and
 40 MODIS Terra (upper panels) and Aqua (lower panels) by month. Green areas denote grid cells
 41 defined as experiencing extreme AOD in WRF-Chem, blue pixels indicate extreme values as
 42 diagnosed using MODIS, while red pixels indicate areas where the occurrence of extreme
 43 values is indicated by both.

