

Supplement of Atmos. Chem. Phys., 16, 397–416, 2016  
<http://www.atmos-chem-phys.net/16/397/2016/>  
doi:10.5194/acp-16-397-2016-supplement  
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*Supplement of*

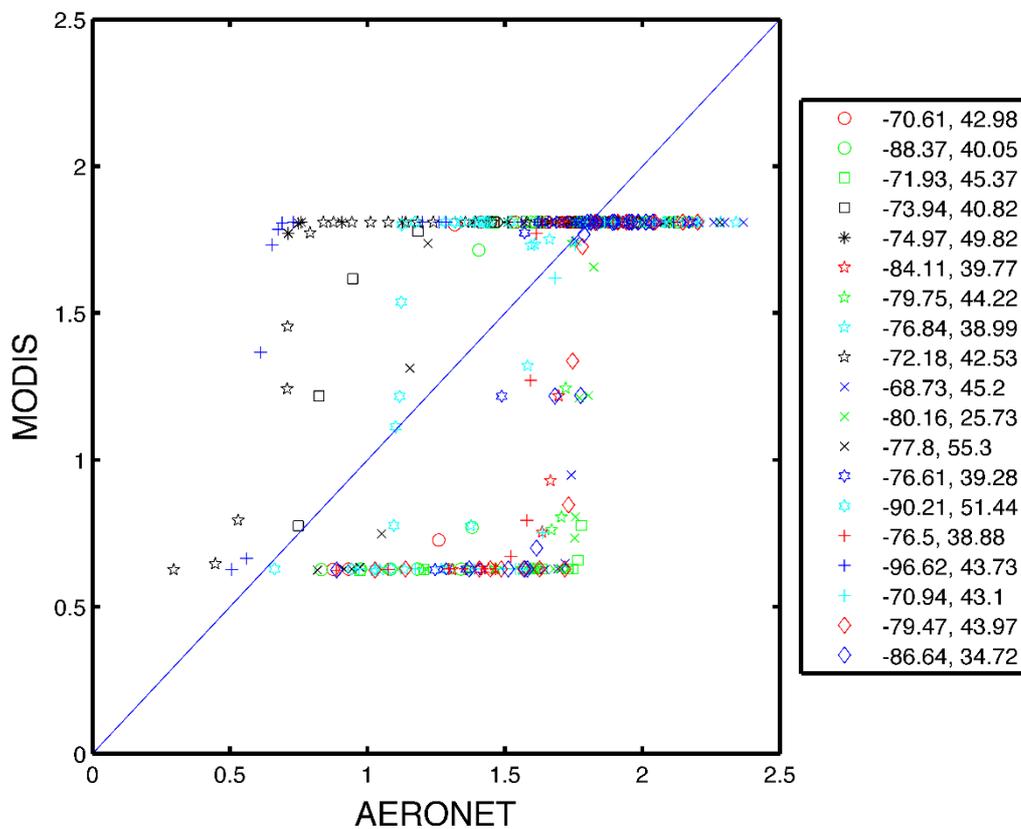
## **Evaluating the skill of high-resolution WRF-Chem simulations in describing drivers of aerosol direct climate forcing on the regional scale**

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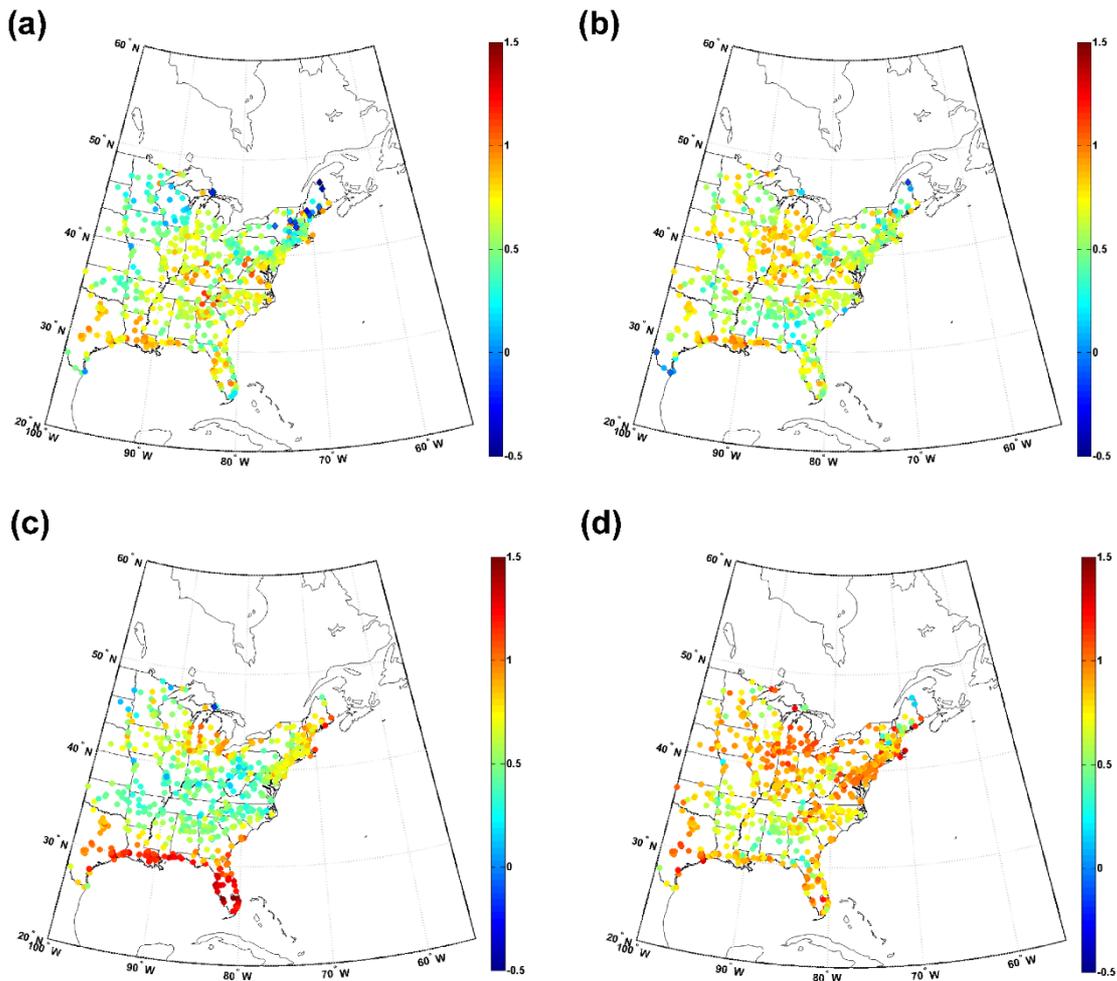
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4 Figure S1. Empirical quantile-quantile plot of simultaneous measurements of AE at 470-660  
 5 nm between MODIS (Terra) and AERONET (where the AERONET station longitude (E) and  
 6 latitude (N) are given in the legend). As shown, the MODIS data 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 ( $AE > 1$ ).



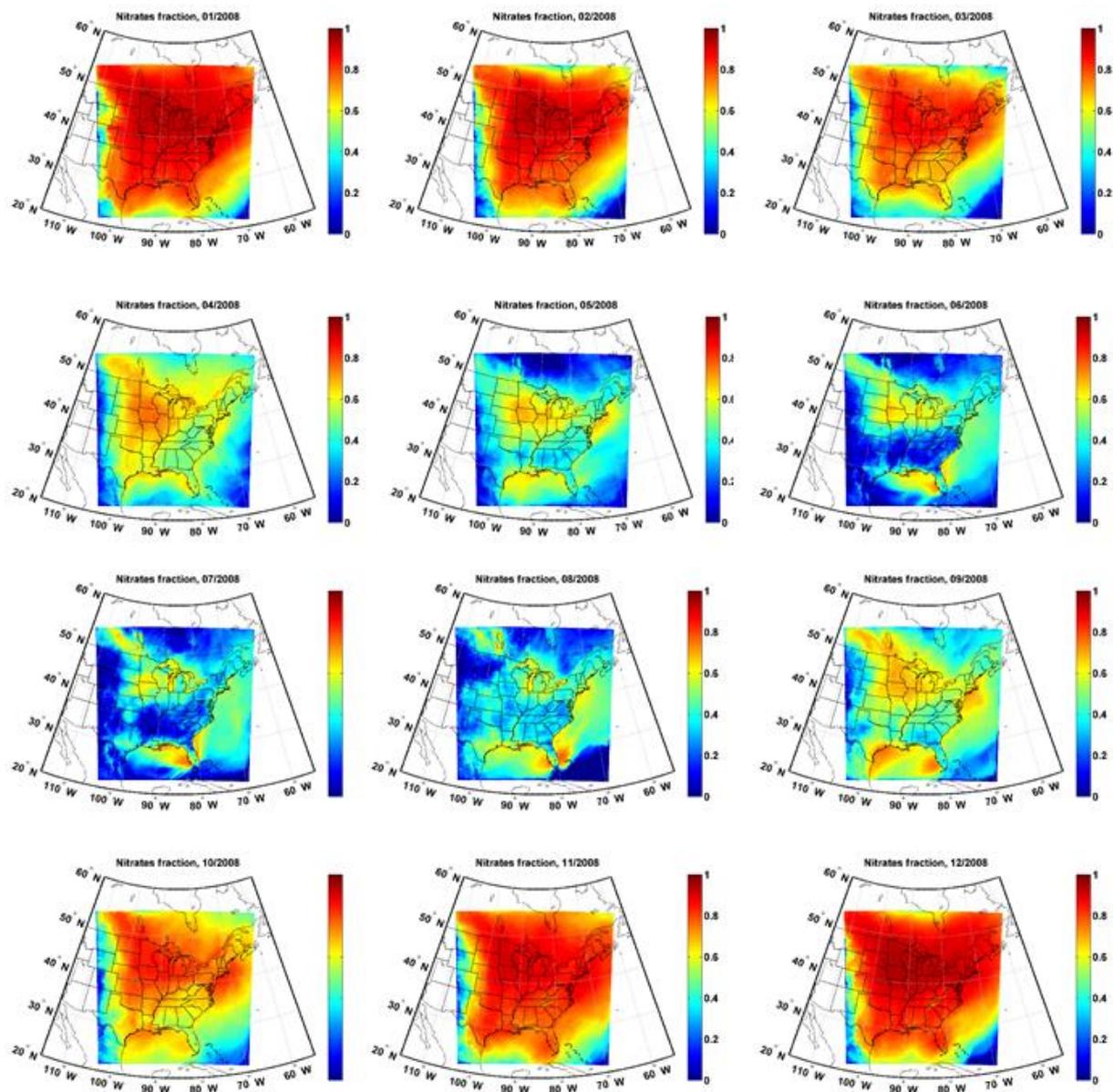
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12 Figure S2. Mean fraction bias (MFB) of near-surface daily mean  $PM_{2.5}$  concentrations as  
13 simulated by WRF-Chem relative to observations at EPA sites during (a) winter, (b) spring, (c)  
14 summer and (d) fall. As shown, near-surface  $PM_{2.5}$  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).



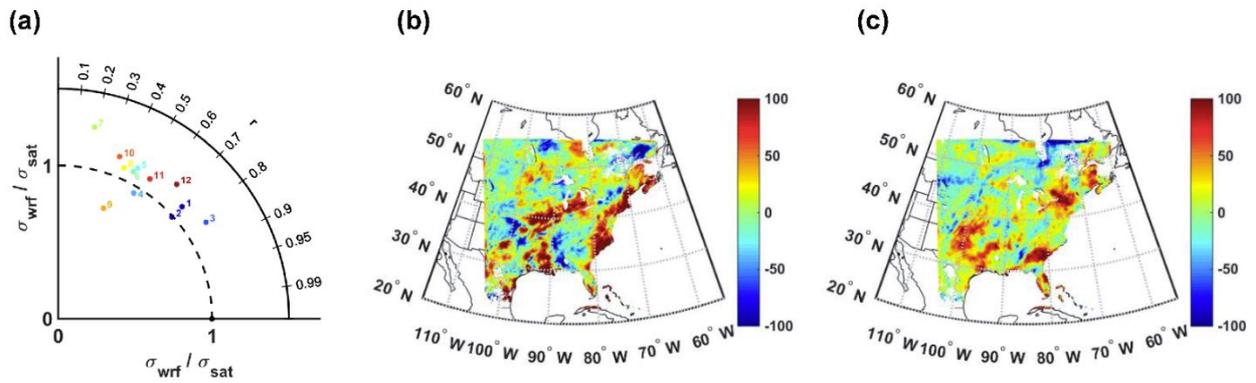
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20 Figure S3. Fraction of near-surface monthly averaged mass concentration of nitrate versus the  
21 sum of aerosol nitrate and sulfate as simulated in the accumulation mode by WRF-Chem for  
22 each calendar month during 2008.



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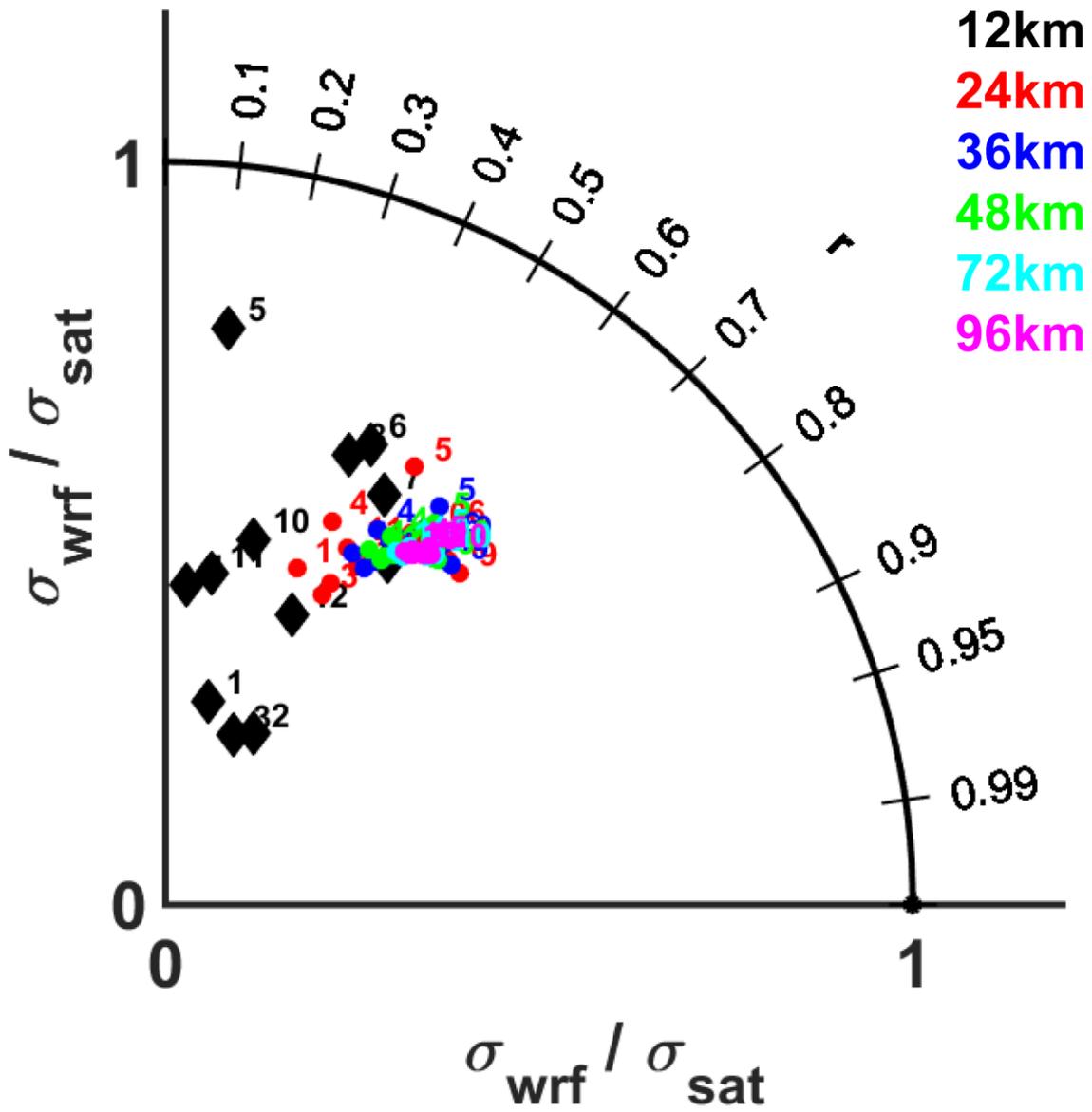
25 Figure S4. (a) Taylor diagram for monthly accumulated precipitation during 2008 as simulated  
26 by WRF-Chem and in the gridded observations (Matsuura and Willmott, 2009), after applying  
27 a linear interpolation to match the WRF-Chem grid. Results for the individual calendar months  
28 are shown 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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33

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 > 75<sup>th</sup> 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.

