The impact of resolution on meteorological, chemical and aerosol properties in regional simulations with WRF-Chem

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16   **Abstract**

17   Limited area (regional) models applied at high resolution over specific regions of interest are

18   generally expected to more accurately capture the spatio-temporal variability of key

19   meteorological and climate parameters. However, improved performance is not inevitable, and

20   there remains a need to optimize use of numerical resources, and to quantify the impact on

21   simulation fidelity that derives from increased resolution. The application of regional models

22   for climate forcing assessment is currently limited by the lack of studies quantifying the

23   sensitivity to horizontal spatial resolution and the physical-dynamical-chemical schemes

24   driving the simulations. Here we investigate model skill in simulating meteorological, chemical

25   and aerosol properties as a function of spatial resolution, by applying the Weather Research

26   and Forecasting model with coupled Chemistry (WRF-Chem) over eastern North America at

27   different resolutions. Using Brier Skill Scores and other statistical metrics it is shown that

28   enhanced resolution (from 60 to 12 km) improves model performance for all of the

29   meteorological parameters and gas phase concentrations considered, in addition to both mean

30   and extreme Aerosol Optical Depth (AOD) in three wavelengths in the visible relative to

31   satellite observations, principally via increase of potential skill. Some of the enhanced model

32   performance for AOD appears to be attributable to improved simulation of meteorological

33   conditions and the concentration of key aerosol precursor gases (e.g. SO_2 and NH_3). Among

34   other reasons, a dry bias in the specific humidity in the boundary layer and a substantial

35   underestimation of total monthly precipitation in the 60 km simulations are identified as causes

36   for the better performance of WRF-Chem simulations at 12 km.

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1 Motivation and Objectives

Aerosols alter Earth's radiation balance primarily by scattering or absorbing incoming solar radiation (direct effect, dominated by accumulation mode (diameters ~ wavelength (λ), where total extinction is often quantified using AOD), or regulating cloud formation/properties by acting as cloud condensation nuclei (CCN) (indirect effect, dominated by diameters $\geq$ 100 nm, magnitude = f(composition)). Most aerosols (excluding black carbon) have a larger scattering cross-section than absorption cross-section, and act as CCN thus enhancing cloud albedo and lifetimes. Hence increased aerosol concentrations are generally (but not uniformly) associated with surface cooling (offsetting a fraction of greenhouse gas warming) (Boucher, 2013;Myhre et al., 2013b) to a degree that is principally dictated by the aerosol concentration, size and composition, in addition to the underlying surface and height of the aerosol layer (McComiskey et al., 2008). Despite major advances in measurement and modeling, both the current global mean aerosol direct effect (possible range: -0.77 to +0.23 W m^{-2}) and the indirect effect (possible range: -1.33 to -0.06 W m^{-2}) remain uncertain (Stocker, 2013), as does their future role in climate forcing (Rockel et al., 2008) and regional manifestations (Myhre et al., 2013a). Specific to our current study region (eastern N. America), one analysis using the NASA GISS global model found that the "regional radiative forcing from US anthropogenic aerosols elicits a strong regional climate response, cooling the central and eastern US by 0.5–1.0 °C on average during 1970–1990, with the strongest effects on maximum daytime temperatures in summer and autumn. Aerosol cooling reflects comparable contributions from direct and indirect radiative effects" (Leibensperger et al., 2012). A recent comparison of multiple global models conducted under the AEROCOM-project indicated this is also a region that exhibits very large model-to-model variability in simulated AOD (<AOD> ~ 0.5, σ(AOD) ~ 1) (Myhre et al., 2013a).

Major reasons why aerosol radiative forcing on both the global and regional scales remains uncertain include short atmospheric residence times and high spatio-temporal variability of aerosol populations, and the complexity of the processes that dictate aerosol concentrations, composition and size distributions (Seinfeld and Pandis, 2016). Although aerosol processes and properties are increasingly being treated in the global Earth System Models (ESMs) (Long et al., 2015;Tilmes et al., 2015) applied in the Coupled Model Intercomparison Project Phase 6 (CMIP-6) (Meehl et al., 2014), the scales on which such models are applied remain much coarser than those on which aerosol population properties are known to vary (Anderson et al., 2003). Therefore, limited area atmospheric models (regional models) applied at higher

74　　resolution over specific regions of interest are expected to 'add value' (i.e. improve the fidelity)

75　　of the physical-dynamical-chemical processes that induce extreme events and dictate climate

76　　forcing. There is empirical evidence to suggest a strong resolution dependence in simulated

77　　aerosol particle properties. For example, WRF-Chem simulations with spatial resolution

78　　enhanced from 75 km to 3 km exhibited higher correlations and lower bias relative to

79　　observations of aerosol optical properties over Mexico likely due to more accurate description

80　　of emissions, meteorology and of the physicochemical processes that convert trace gases to

81　　particles (Gustafson et al., 2011;Qian et al., 2010). This improvement in the simulation of

82　　aerosol optical properties implies a reduction of the uncertainty in associated aerosol radiative

83　　forcing (Gustafson et al., 2011). Further, WRF-Chem run over the United Kingdom and

84　　Northern France at multiple resolutions in the range of 40-160 km, underestimated AOD by

85　　10-16% and overestimated CCN by 18-36% relative to a high resolution run at 10 km, partly

86　　as a result of scale dependence of the gas-phase chemistry and differences in the aerosol uptake

87　　of water (Weigum et al., 2016).

88　　However, debate remains regarding how to objectively evaluate model performance, quantify

89　　the value added by enhanced resolution (Di Luca et al., 2015;Rockel et al., 2008) and on

90　　possible limits to the improvement of climate representation in light of errors in the driving

91　　"imperfect lateral boundary conditions" (Diaconescu and Laprise, 2013). Nevertheless,

92　　although "it is unrealistic to expect a vast amount of added values since models already

93　　performs rather decently" (Di Luca et al., 2015) and global ESMs are now run at much higher

94　　resolution than in the past, it is generally assumed that high resolution regional models will add

95　　value via more realistic representation of spatio-temporal variability than global coarser-

96　　resolution simulations. Further, "the main added value of a regional climate model is provided

97　　by its small scales and its skill to simulate extreme events, particularly for precipitation"

98　　(Diaconescu and Laprise, 2013).

99　　It is particularly challenging to assess the added-value from enhanced resolution in the context

100　　of climate-relevant aerosol properties since they are a complex product of the fidelity of the

101　　simulation of meteorological parameters, gas-phase precursors, emissions and the treatment of

102　　aerosol dynamics. Here we quantify the value added by enhanced resolution in the description

103　　of physical and chemical atmospheric conditions using year-long simulations from WRF-Chem

104　　over eastern North America, and investigate how they impact AOD. The primary performance

105　　evaluation of aerosol properties focuses on AOD at different wavelengths (λ = 470, 550 and

106　　660 nm, where the AOD at different λ is used as a proxy of the aerosol size distribution (Tomasi

107 et al., 1983), see details in Sect. 2.3) and is measured relative to observations from satellite-
108 borne instrumentation. Thus the term "value-added" is used here in the context of columnar
109 aerosol properties to refer to an improvement of model performance in simulation of
110 wavelength specific AOD as measured by the MODerate resolution Imaging
111 Spectroradiometer (MODIS) instrument aboard the polar-orbiting Terra satellite. To attribute
112 sources of the enhanced fidelity of AOD, our analysis also incorporates evaluation of the value-
113 added by enhanced resolution in terms of key meteorological and gas-phase drivers of aerosol
114 concentrations and composition and is conducted relative to the MERRA-2 reanalysis product
115 for the physical variables and columnar gas concentrations from satellite observations (see
116 details of the precise data sets used given below). The meteorological parameters considered
117 are air temperature at 2 m (T_{2m}), total monthly precipitation (PPT), planetary boundary-layer
118 height ($PBLH$) and specific humidity in the boundary layer (Q_{PBL}). The gas phase
119 concentrations considered are sulfur dioxide (SO_2), ammonia (NH_3), nitrogen dioxide (NO_2)
120 and formaldehyde (HCHO).

121 We begin by quantifying the performance of WRF-Chem when applied over eastern North
122 America at a resolution of 60 km (WRF60) (~ finest resolution likely to be employed in CMIP-
123 6 global simulations) and then compare the results to those from simulations conducted at 12
124 km (WRF12) (simulation details are given in Table 1). Quantification of model skill is
125 undertaken by mapping the WRF12 output to the WRF60 grid (WRF12-remap) and computing
126 Brier Skill Scores (BSS) using MODIS as the target, WRF60 as the reference forecast and
127 WRF12-remap as the forecast to be evaluated. We also evaluate the performance of the WRF-
128 Chem simulations of 2008 relative to climatology as represented by MODIS observations for
129 2000-2014. We additionally assess the impact of simulation resolution on extreme AOD values
130 that are associated with enhanced impacts on climate and human health. This analysis uses both
131 *Accuracy* and *Hit Rate* as the performance metrics and focuses on the co-occurrence of extreme
132 values in space from the model output and MODIS.

133 Based on the performance evaluation of the WRF-Chem simulations that indicate substantial
134 dry bias in the WRF60 simulations and large seasonality in the skill-scores for AOD as a
135 function of resolution, we conducted two further year-long simulations at 60 km. In the first
136 we held all other simulation conditions constant but selected a different cumulus
137 parameterization. In the second, we held all simulation conditions constant but employed a
138 different set of lateral boundary conditions for the meteorology. In the context of the
139 precipitation biases reported herein it is worthy of note that discrepancies in simulated

140   precipitation regimes are key challenges in regional modelling (both physical and coupled with

141   chemistry). Although the Grell 3D scheme has been successfully applied in a number of prior

142   analysis wherein the model was applied at resolutions in the range of 1-36 km (e.g. (Grell and

143   Dévényi, 2002;Lowrey and Yang, 2008;Nasrollahi et al., 2012;Sun et al., 2014;Zhang et al.,

144   2016)), the North American Regional Climate Change Assessment Program (NARCCAP)

145   simulations with WRF at 50-km were also dry biased in the study domain (Mearns et al., 2012).

146   Although there have been a number of studies that have sought to evaluate different cumulus

147   schemes over different regions at different resolutions, no definitive recommendation has been

148   made regarding the dependence of model skill on resolution and cumulus parameterization

149   (Arakawa, 2004;Jankov et al., 2005;Nasrollahi et al., 2012;Li et al., 2014). Hence, further

150   research is needed to identify the optimal cumulus scheme for use over North America at

151   coarser resolution. Thus, we performed a sensitivity analysis on the cumulus scheme at 60 km

152   by applying the Grell-Freitas parameterization (Grell and Freitas, 2014), which is the next

153   generation of the Grell 3D scheme.

154   **2 Materials and Methods2.1 WRF-Chem simulations**

155   WRF-Chem (version 3.6.1) simulations were performed for the calendar year 2008 over eastern

156   North America, in a domain centered over southern Indiana (86°W, 39°N) at two resolutions,

157   one close to the finest resolution designed for CMIP-6 global model runs (i.e. 60 km, WRF60)

158   and the other one at much higher resolution (12 km, WRF12). Simulation settings are identical

159   for the two runs except for the time-step used for the physics (Table 1). Physical and chemical

160   parameterizations were chosen to match previous work using WRF-Chem at 12 km on the same

161   region which showed good performance relative to observations and the year 2008 was selected

162   because it is representative of average climate and aerosol conditions during 2000 - 2014

163   (Crippa et al., 2016). More specifically the simulations adopted the RADM2 chemical

164   mechanism (Stockwell et al., 1990) and a modal representation of the aerosol size distribution

165   (MADE/SORGAM, (Ackermann et al., 1998;Schell et al., 2001)) with three lognormal modes

166   and fixed geometric standard deviations (i.e. 1.7, 2 and 2.5 for Aitken, accumulation and coarse

167   mode, respectively (Ackermann et al., 1998;Grell et al., 2005)). Aerosol direct feedback was

168   turned on and coupled to the Goddard shortwave scheme (Fast et al., 2006). A telescoping

169   vertical grid with 32 model layers from the surface to 50 hPa and 10 layers up to 800 hPa was

170   selected. Meteorological initial and boundary conditions from the North American Mesoscale

171   Model at 12 km resolution (NAM12) are applied every 6 hours, while initial and chemical

172   boundary conditions are taken from MOZART-4 (Model for Ozone and Related chemical

173   Tracers, version 4) with meteorology from NCEP/NCAR-reanalysis (Emmons et al., 2010).

174   Anthropogenic emissions are specified for both WRF60 and WRF12 from the US National

175   Emission Inventory 2005 (NEI-05) (US-EPA, 2009) which provides hourly point and area

176   emissions at 4 km on 19 vertical levels. The simulation settings and specifically the use of a

177   modal representation of the aerosol size distribution were selected to retain computational

178   tractability. Accordingly, the 60 km simulations for the year 2008 completed in 6.4 hours

179   whereas the 12 km simulations completed in 9.5 days (230 hours) on the Cray XE6/XK7

180   supercomputer (Big Red II) owned by Indiana University, using 256 processors distributed on

181   8 nodes.

182   As described in detail below, in the WRF60 simulations configured as described in Table 1,

183   simulated precipitation during the summer months exhibits substantial dry bias, and the

184   analysis of value added by enhanced simulation resolution exhibited strong seasonality. We

185   performed a sensitivity analysis to the cumulus scheme, by conducting an additional year-long

186   simulation at 60 km using the Grell-Freitas parameterization (Grell and Freitas, 2014), which

187   is an evolution of Grell 3D that is scale-aware and treats some aspects of aerosol-cloud

188   interactions. We also tested the sensitivity of the simulation results to the meteorological

189   boundary conditions, by repeating the WRF60 simulations using output from the Global

190   Forecast System (GFS) at 0.5° resolution every 6 hours to provide the lateral boundary

191   conditions.

192   **2.2 Observations**

193   Model aerosol optical properties are evaluated relative to the MODIS Collection 6 dark-target

194   land aerosol product from aboard the Terra satellite (~1030 overpass local solar time (LST))

195   (Levy et al., 2013). To provide a consistent assessment of model skill, the evaluation of AOD

196   is conducted only on land areas since the MODIS dark-target ocean aerosol product is based

197   on a retrieval algorithm different from the one over land (Levy et al., 2013). Trace gas

198   concentrations are evaluated relative to measurements from the Ozone Monitoring Instrument

199   (OMI; version 3) (Chance, 2002) and the Infrared Atmospheric Sounding Interferometer (IASI;

200   NN version 1) (Whitburn et al., 2016) aboard the Aura (~1345 LST) and MetOp satellites

201   (~0930 LST), respectively. MODIS retrieves AOD at multiple λ including 470, 550, and 660

202   nm, and the MODIS algorithm removes cloud-contaminated pixels prior to spatial averaging

203   over 10×10 km (at nadir). OMI and IASI have nadir resolutions of 13×24 km and 12 km

204   (circular footprint), respectively, and have been filtered to remove retrievals with cloud

205　fractions > 0.3 (Fioletov et al., 2011;McLinden et al., 2014;Vinken et al., 2014) and OMI pixels

206　affected by the row anomalies. MODIS, OMI, and IASI provide near daily global coverage,

207　although the row anomalies render portions of the OMI viewing swath unusable. Uncertainty

208　in AOD from MODIS is spatially and temporally variable. It has been estimated as ± (0.05 +

209　15%) for AOD over land (Levy et al., 2013), and prior research has reported 71% of MODIS

210　Collection 5 retrievals fall within 0.05 ± 20% for AOD relative to AERONET in the study

211　domain (Hyer et al., 2011). The accuracy of OMI ("root sum of the square of all errors,

212　including forward model, inverse model, and instrument errors" (Brinksma et al., 2003)) is 1.1

213　DU or 50% for SO_2, 2×10^{14} cm^{-2}/30% for background/polluted NO_2 conditions, and 35% for

214　HCHO. This uncertainty is typically reduced by spatial and temporal averaging, as employed

215　herein (Fioletov et al., 2011;Krotkov et al., 2008). IASI NH_3 retrievals do not use an a priori

216　assumption of emissions, vertical distribution, or lifetime of NH_3 (i.e. no averaging kernel);

217　therefore, NH_3 accuracy is variable (Whitburn et al., 2016), and thus only retrievals with

218　uncertainty lower than the retrieved concentrations are used herein.

219　For the model evaluation, satellite observations for each day are regridded to the WRF-Chem

220　discretization. This is done by averaging all valid retrievals within: 0.1° and 0.35° of the WRF-

221　Chem grid-cell center for the 12×12 km and 60×60 km resolutions, respectively for MODIS;

222　0.125° × 0.18° (along-track/latitudinal × cross-track/longitudinal) and 0.365° × 0.42° for OMI;

223　0.12° and 0.36° for IASI. To avoid issues from under-sampling, we require at least 10 valid

224　MODIS granules for the 60×60 km daily average to be computed and at least 5 daily averages

225　to compute a monthly average for each grid cell. Model evaluation of gaseous species is

226　performed on a seasonal basis using standard scores (z-scores), which are computed as the

227　difference between the seasonal mean within a grid cell and the seasonal spatial mean, divided

228　by the seasonal spatial standard deviation. Use of z-scores allows comparison of the spatial

229　patterns of satellite observations and model output in terms of standard deviation units from

230　the mean.

231　The simulated meteorological properties are evaluated using Modern-Era Retrospective

232　analysis for Research and Applications (MERRA-2) reanalysis data as the target. MERRA-2

233　is a homogenized and continuous in time description of atmospheric properties on a 3-

234　dimensional global grid (horizontal resolution of 0.5°×0.625°, L72), developed by NASA and

235　was released in Fall 2015 (Molod et al., 2015). MERRA-2 provides hourly values of T_{2m} and

236　*PBLH*, and vertical profile of 3-dimensional variables every 3 hours on a large number of

237 pressure levels. Here we compute the total specific humidity (Q_{PBL}) of the lowest 8 pressure

238 levels (i.e. in the boundary-layer approximated as the layer from 1000 to 825 hPa) in MERRA-

239 2, assuming an average air density in the PBL of 1.1 kg m^{-3}. For the evaluation of simulated

240 precipitation we use accumulated monthly total values.

2.3 Spectral dependence of AOD

242 Three properties dictate the actual aerosol direct radiative forcing: AOD, single scattering

243 albedo and asymmetry factor, all of which are a function of the wavelength (λ) of incident

244 radiation. The first property is related to the total columnar mass loading, typically dominates

245 the variability of direct aerosol effect (Chin et al., 2009) and is the focus of the current research.

246 The relationship between the aerosol size distribution and spectral dependence of AOD is

247 described by a power law function:

$$\beta\left(\lambda_1\right)=\beta\left(\lambda_2\right)\times\left(\frac{\lambda_1}{\lambda_2}\right)^{-\alpha} \quad (1)$$

249 where β is the particle extinction coefficient at a specific wavelength λ, and α is the Ångström

250 exponent (Ångström, 1964) which describes the wavelength dependence of AOD (and is

251 inversely proportional to the average aerosol diameter):

$$\alpha=\frac{ln\dfrac{AOD\left(\lambda_1\right)}{AOD\left(\lambda_2\right)}}{ln\dfrac{\lambda_2}{\lambda_1}} \quad (2)$$

253 The aerosol volume distribution usually conforms to a multi-lognormal function with n modes:

$$\frac{dV(r)}{d\ln r}=\sum_{i=1}^{n}\frac{C_i}{\sqrt{2\pi}\sigma_i}\exp\left[\frac{-\left(\ln r-\ln R_i\right)^2}{2\sigma_i^{\,2}}\right] \quad (3)$$

255 where r is the particle radius and C_i, R_i and σ_i are the particle volume concentration, the

256 geometric mean radius and the standard deviation in the mode i respectively.

257 We can thus compute AOD for a polydisperse distribution of aerosols with refractive index m

258 in an atmospheric column of height Z as:

$$AOD(\lambda)=\int\frac{3\beta(m,r,\lambda)}{4r}\frac{dV(r)}{d\ln r}d\ln rdZ \quad (4)$$

260 As indicated in (Schuster et al., 2006), "the spectral variability of extinction diminishes for

261 particles larger than the incident wavelength", thus fine mode particles contribute more to AOD

262 in the visible ($\lambda \sim 0.5$ µm) than at longer wavelengths, whereas coarse mode particles provide a

263 similar AOD both at short and long wavelengths. This is reflected in the Ångström parameter

264 which can be thus used as a proxy for the fine mode fraction or fine mode radius (Schuster et

265 al., 2006).

2.4 Quantification of model performance and added-value

267 Taylor diagrams summarize three aspects of model performance relative to a reference: the

268 spatial correlation coefficient (i.e. Pearson correlation of the fields, r), the ratio of spatial

269 standard deviations of the two spatial fields ($\sigma_{wrf}/\sigma_{sat}$) and the root mean squared difference

270 (RMSD) (Taylor, 2001). Here Taylor diagrams are presented for monthly mean AOD from

271 WRF60, WRF12 and WRF12-remap relative to MODIS at different wavelengths (Fig. 1 d-f).

272 Because AOD is not normally distributed, Spearman's rank correlation coefficients (ρ) of the

273 mean monthly AOD spatial fields are also computed to reduce the impact of a few outliers and

274 the small sample size during cold months (Table 2). To assess the significance of ρ while

275 accounting for multiple testing, we apply a Bonferroni correction (Simes, 1986) in which for

276 m hypothesis tests, the null hypothesis is rejected if $p \le \dfrac{\alpha}{m}$, where p is the p-value and α is the

277 confidence level (0.05 is used here).

278 We further quantify the value added (or lack of thereof) of the high-resolution simulations

279 using the following metrics:

(i) Brier Skill Score

281 Value added is quantified using Brier Skill Scores (BSS) and is evaluated in two ways: first by

282 evaluating the model performance as a function of simulation resolution and then using

283 climatology as the reference 'forecast'. In these analyses the hourly output from the 12 km

284 resolution simulation is degraded (averaged) to 60 km (hereafter WRF12-remap) as follows:

285 the 12 km domain is resized excluding 2 grid cells at the border to exactly match the 60 km

286 resolution domain. For example, in the analysis of AOD each coarse grid cell thus includes 5×5

287 12 km resolution cells and its value is the mean of all valid 12 km grid cells inside it if at least

288 half of those cells contain valid AOD (i.e. no cloud cover), otherwise the whole coarse cell is

289 treated as missing. In all comparisons of AOD only cells with simultaneous (i.e. model and

290 MODIS) clear sky conditions are considered. A daily value from WRF-Chem is computed as

291   an instantaneous value for the hour nearest to the satellite overpass time. When the comparison

292   is done on a monthly basis, a monthly mean value is computed from the daily values obtained

293   under clear sky conditions, only if there are at least five valid observations in the month.

294   The primary metric used to quantify the added value of WRF12-remap versus WRF60 is the

295   Brier Skill Score (BSS) (Murphy and Epstein, 1989):

296
$$BSS = \frac{r_{F'P'}^{2} - \left(r_{F'P'} - \frac{\sigma_{F'}}{\sigma_{P'}}\right)^{2} - \left(\frac{\langle P'\rangle - \langle F'\rangle}{\sigma_{P'}}\right)^{2} + \left(\frac{\langle P'\rangle}{\sigma_{P'}}\right)^{2}}{1 + \left(\frac{\langle P'\rangle}{\sigma_{P'}}\right)^{2}} \quad (5)$$

297   where F is the "forecast" (i.e. the 12 km simulations mapped to 60 km, WRF12-remap); P is

298   the "target" (i.e. for AOD this is MODIS at 60 km) and output from WRF60 are used as the

299   reference forecast; F' the difference between 12 km estimates regridded to 60 km and MODIS;

300   P' the difference between the 60 km simulation and the 'target' (i.e. for the AOD MODIS

301   observations regridded to 60 km). In the analysis of BSS relative to the long-term (15-year)

302   climatology of AOD from MODIS, the monthly mean climatological value of AOD is used as

303   the reference forecast, while WRF60 and WRF12-remap are used as the forecasts, and monthly

304   mean AOD from MODIS at 60 km is the target.

305   BSS measures by how much a test simulation (WRF12-remap) more closely (or poorly)

306   reproduces observations (from MODIS, MERRA-2 or other satellite products) relative to a

307   control (WRF60) run. For example, a BSS>0 indicates WRF12, even when regridded to 60 km,

308   does add value. The first term in (5) ranges from 0 to 1, is described as the potential skill, and

309   is the square of the spatial correlation coefficient between forecast and reference anomalies to

310   MODIS. It is the skill score achievable if both the conditional bias (second term) and overall

311   bias (third term) were zero, and for most of the variables considered herein (particularly AOD)

312   it contributes to a positive BSS in most calendar months (and seasons). The second term (the

313   conditional bias, > 0), is the square of the difference between the anomaly correlation

314   coefficient and the ratio of standard deviation of the anomalies and is small if for all points F'

315   is linear to P'. The third term is referred to as the forecast anomaly bias, and is the ratio of the

316   difference between the mean anomalies of WRF12-remap and the observations relative to

317   WRF60 and the standard deviation of WRF60 anomaly relative to observed values. The fourth

318   term is the degree of agreement and appears in both the numerator and denominator. It is

319 computed as the square of the ratio of the mean anomaly between WRF60 and observations
320 and the standard deviation of the anomalies.

(ii) Pooled paired t-test

322 To identify which areas in space contribute most to the AOD added-value, we compare daily
323 mean AOD fields from WRF-Chem at different resolutions and MODIS. We perform a pooled
324 paired t-test to evaluate the null hypothesis that those differences come from normal
325 distributions with equal means and equal but unknown variances (the test statistic has a
326 Student's t distribution with df = n + m − 2, and the sample standard deviation is the pooled
327 standard deviation, where n and m are the two sample sizes). The test is conducted by
328 climatological season (e.g. winter = DJF) since there are fewer than 20 valid AOD observations
329 in most 60 km grid cells for each calendar month (Fig. 2). Given the large number of hypothesis
330 tests performed (i.e. one for each 60 km grid cell), we adjust the p-values using the False
331 Discovery Rate (FDR) approach (Benjamini and Hochberg, 1995). In this approach, p-values
332 from the t-tests are ranked from low to high ($p_1,p_2,\ldots,p_m$), then the test with the highest rank, j,
333 satisfying:

334 $$p_j \leq \frac{j}{m}\alpha \quad \textbf{(6)}$$

335 is identified. Here all p-values satisfying Eq. 6 with $\alpha=0.1$ are considered significant.

(iii) Accuracy and Hit Rate in identification of AOD extremes

337 For each month we identify grid cells in which the wavelength specific AOD exceeds the 75th
338 percentile value computed from all grid cells and define that as an extreme. Thus grid cells
339 with extreme AOD are independently determined for MODIS and WRF-Chem at different
340 resolutions. The spatial coherence in identification of extremes in the fields is quantified using
341 two metrics: the *Accuracy* and the *Hit Rate* (*HR*). The *Accuracy* indicates the overall spatial
342 coherence and is computed as the number of grid cells co-identified as extreme and non-
343 extreme between WRF-Chem and MODIS relative to the total number of cells with valid data.
344 The *HR* weights only correct identification of extremes in MODIS by WRF-Chem.

3   Results

3.1 Model performance as a function of spatial resolution

347 When WRF-Chem is applied at 60 km resolution the degree of association of the resulting
348 spatial fields of mean monthly AOD at the three wavelengths with MODIS varies seasonally.

12

349   Smallest RMSD and highest Spearman spatial correlations (ρ) with MODIS observations

350   generally occur during months with highest mean AOD (i.e. during summer, Fig. 1 d-f and Fig.

351   3), and reach a maximum in August (ρ = 0.60, Table 2). However, while the patterns of relative

352   AOD variability are well captured, the absolute magnitudes and spatial gradients of AOD

353   during the summer are underestimated by WRF60 (Fig. 1 d-f and Fig. 3, Table S1). High spatial

354   correlations (ρ > 0.40) are also observed in March, April and November (Table 2), when the

355   ratio of spatial standard deviations is closer to 1 (Fig. 1 d-f, Table S1). Only a weak wavelength

356   dependence is observed in the performance metrics as described on Taylor diagrams. The

357   spatial variability is generally more negatively biased for AOD at 660 nm (Table S1), indicating

358   that WRF60 simulations tend to produce larger diameter aerosols homogeneously distributed

359   over the domain, whereas MODIS observations indicate more spatial variability.

360   The performance of WRF60 simulations relative to MODIS contrasts with analyses of WRF12

361   and WRF12-remap. WRF12 and WRF12-remap indicate highest spatial correlations with

362   MODIS observations throughout the summer months (ρ = 0.5-0.7, Table 2), although the bias

363   towards simulation of more coarse aerosols than are observed is consistent across the two

364   simulations and with prior research (see details provided in (Crippa et al., 2016)). However,

365   simulations at 12 km (WRF12) show positive ρ with MODIS for all λ in all calendar months,

366   while mean monthly spatial fields of AOD from WRF60 show low and/or negative correlations

367   with MODIS during May, June, September, October and December, indicating substantial

368   differences in the degree of correspondence with MODIS AOD in the two simulations, and

369   higher fidelity of the enhanced resolution runs (Tables 2 and S1).

370   Monthly mean spatial fields of AOD(λ) as simulated by WRF12 or WRF12-remap exhibit

371   positive Spearman correlation coefficients (ρ) with MODIS observations for all calendar

372   months and range from ~ 0.25 for WRF12-remap (0.20 for WRF12) during winter to ~ 0.70

373   and 0.64, respectively during summer (Table 2). Spearman's ρ are uniformly higher in WRF12-

374   remap than WRF12 indicating a mismatch in space in the high-resolution simulation (i.e. that

375   grid cells with high AOD are slightly displaced in the 12 km simulations possibly due to the

376   presence of sub-grid scale aerosol plumes (Rissman et al., 2013)). Mean monthly fields of AOD

377   (all λ) from both WRF12 and WRF12-remap exhibit lower ρ with MODIS in February-April

378   and November than the 60 km runs (Table 2). These discrepancies appear to be driven by

379   conditions in the south of the domain. For example, differences between WRF60/WRF12-

380   remap vs. MODIS during all seasons are significant according to the paired t-test over Florida

381 and along most of the southern coastlines (Fig. 2). This region of significant differences extends

382 up to ~ 40°N during summer and fall, reflecting the stronger north-south gradient in AOD from

383 MODIS and WRF12-remap that is not captured by WRF60 (see example for $\lambda = 550$ nm, Fig.

384 3). These enhancements in the latitudinal gradients from WRF12-remap are also manifest in

385 the physical variables (particularly specific humidity as discussed further below).

386 The differences in the absolute values of mean monthly AOD deriving from differences in the

387 resolution at which WRF-Chem was applied are of sufficient magnitude (a difference of up to

388 0.2 in regions with a mean AOD value of 0.4), particularly in the summer months (Fig. 4), to

389 raise concerns. However, detailed investigation of the simulations settings and repetition of the

390 60 km simulation resulted in virtually identical results indicating no fault can be found in the

391 analysis. Further, we note that the eastern-half of North America was also identified as a region

392 of high discrepancy in global ESM (Myhre et al., 2013a).

393 To further investigate differences in the simulation output due to spatial discretization we

394 computed Brier Skill Scores (BSS). In this analysis AOD for each λ from WRF12-remap are

395 used as the 'forecast', output from WRF60 are used as the reference forecast and MODIS

396 observations at 60 km are used as the target. BSS exceed 0 during all months except for

397 September and October, and largest BSS (> 0.5) for AOD (all λ) is found during most months

398 between December and July (Fig. 5a-c). This indicates that running WRF-Chem at 12 km

399 resolution yields higher skill in simulated AOD relative to WRF60, even when the WRF12

400 output is remapped to 60 km. BSS do not strongly depend on λ, indicating the added value

401 from enhanced resolution similarly affects aerosol particles of different sizes. Inspecting the

402 terms defining the BSS provides information about the origin of the added value (Fig. 5a-c).

403 The positive BSS derives principally from the potential skill (first term in Eq. 5), which

404 demonstrates a reduction in bias and/or more accurate representation of the spatial gradients in

405 WRF12-remap. This term exhibits weak seasonality with values below 0.5 only during August

406 and fall months. The second and third terms are close to zero during most months, although

407 bigger biases are found during August-October. The substantial conditional bias during late

408 summer and early fall is the result of the large ratio of standard deviations (> 1, i.e. the spatial

409 variability of the anomaly relative to MODIS is larger for WRF12-remap than WRF60, Table

410 S1). It thus contributes to the negative BSS found in September and October, which are also

411 identified as outlier months in WRF12-remap from the Taylor diagram analysis (Fig. 1). Output

412 for these months show modest spatial correlations with AOD from MODIS and higher ratio of

413 standard deviations than in WRF60-MODIS comparisons (Fig. 1, Table S1). Previous work

414   showed that the lower model skill (in WRF12) during September and October may be partially
415   attributable to a dry bias in precipitation from WRF-Chem relative to observations. As a result,
416   simulated AOD and near-surface aerosol nitrate and sulfate concentrations are positively biased
417   over large parts of the domain (Crippa et al., 2016). Although the effects of the boundary
418   conditions appear in some variables (e.g. in Fig. 4 and Figs. S1-S3), the BSS results do not
419   significantly change even when those cells are removed from the analysis.

420   When the BSS is used to assess the skill of each model relative to MODIS AOD climatological
421   mean over the years 2000-2014, WRF12-remap is found to add value relative to the
422   climatology (i.e. BSS >0) during summer months and Nov-Jan whereas BSS for WRF60 is
423   positive from late Fall to early Spring (Fig. 5d). The fact that WRF-Chem does not always
424   outperform the climatology is expected since the model is based on time invariant emissions
425   and skill is assessed relative to a year selected to be representative of the AOD climatology.
426   Mean seasonal AOD from MODIS retrievals over the study region during 2008 lie within ±0.2
427   standard deviations of the climatology (Crippa et al., 2016). Interestingly, BSS for most months
428   (excluding September) are higher for the WRF60 simulations conducted using lateral boundary
429   conditions from NAM12 than GFS.

430   Model resolution also affects the *Accuracy* and *Hit Rate* (*HR*) for identification of areas of
431   extreme AOD (AOD>75th percentile). Highest coherence in the identification of extreme AOD
432   in space identified in WRF12-remap (and WRF12) relative to MODIS is found during May-
433   August (*HR* = 53-77%) vs. WRF60 (*HR* = 17-54%, Table 3). Conversely highest *HR* are found
434   for WRF60 and MODIS during winter and early spring, and indeed exceed those for WRF12
435   and WRF12-remap (Table 3, e.g. Feb: *HR* = 0.78 for WRF60, and 0.67 and 0.68 for WRF12
436   and WRF12-remap, respectively). These differences are consistent with the observation that
437   WRF12-remap overestimates the scales of AOD coherence and AOD magnitude during the
438   cold season along coastlines and over much of the domain in April (Fig. 3).

439   The synthesis of these analyses is thus that the higher resolution simulation increases the
440   overall spatial correlation, decreases overall bias in AOD close to the peak of the solar spectrum
441   relative to MODIS observations and therefore the higher-resolution simulations better
442   represent aerosol direct climate forcing. However, WRF12-remap exhibits little improvement
443   over WRF60 in terms of reproducing the spatial variability of AOD in the visible wavelengths
444   and further that WRF12-remap tends to be more strongly positively biased in terms of mean
445   monthly AOD outside of the summer months (Fig. 2 and Fig. 3). Also the improvement in
446   detection of areas of extreme AOD in the higher resolution simulations (WRF12-remap) is

447　　manifest only during the warm season.

3.2 Investigating sources of error in simulated AOD

449　　As documented above, WRF-Chem applied at either 60 or 12 km resolution over eastern North

450　　America exhibits some skill in reproducing observed spatial fields of AOD and the occurrence

451　　of extreme AOD values. However, marked discrepancies both in space and time are found, and

452　　at least some of them show a significant dependence on model resolution. Thus, we

453　　investigated a range of physical conditions and gas phase concentrations known to be strongly

454　　determinant of aerosol dynamics in terms of the BSS as a function of model resolution and also

455　　in terms of the mean monthly spatial patterns.

456　　WRF12 even when remapped to 60 km provides more accurate description of key

457　　meteorological variables such as specific humidity (Q) within the boundary layer, $PBLH$,

458　　surface temperature and precipitation (see Fig. 6, S1, S2 and S3) when compared to MERRA-

459　　2, as indicated by the positive BSS during almost all months (Fig. 7a). Good qualitative

460　　agreement is observed for the spatial patterns and absolute magnitude of T_{2m} in both WRF60

461　　and WRF12-remap relative to MERRA-2 for all seasons (Fig. S1) leading to only modest

462　　magnitude of BSS (i.e. value added by the higher resolution simulations (Fig. 7a)). The aerosol

463　　size distribution and therefore wavelength specific AOD exhibits a strong sensitivity to Q

464　　(Santarpia et al., 2005) due to the presence of hygroscopic components in atmospheric aerosols

465　　and thus the role of water uptake in determining aerosol diameter, refractivity and extinction

466　　coefficient (Zieger et al., 2013). For example, the hygroscopic growth factor, which indicates

467　　the change of aerosol diameter due to water uptake, is ~ 1.4 for pure ammonium sulfate with

468　　dry diameter of 532 nm at relative humidity of 80%, thus biases in representation atmospheric

469　　humidity may lead to big errors in simulated aerosol size and AOD (Flores et al., 2012). Our

470　　previous analyses of the 12 km resolution simulations indicated overestimation of sulfate

471　　aerosols (a highly hygroscopic aerosol component, and one which in many chemical forms

472　　exhibits strong hysteresis (Martin et al., 2004)) relative to observed near-surface $PM_{2.5}$

473　　concentrations during all seasons except for winter (Crippa et al., 2016), leading to the

474　　hypothesis that simulated AOD and discrepancies therein may exhibit a strong dependence on

475　　Q. Consistent with that postulate, Q_{PBL} from WRF12-remap exhibits a moist bias in cloud-free

476　　grid cells mostly during warm months, whereas WRF60 is characterized by a dry bias during

477　　all seasons (Fig. 6). Despite the positive bias, WRF12-remap better captures the seasonal

478　　spatial patterns of Q_{PBL} in MERRA-2, leading to positive BSS for this variable in all calendar

479　　months. Thus, there is added value by higher-resolution simulations in representation of one of

480   the key parameters dictating aerosol particle growth and optical properties. Spatial patterns of

481   differences in Q_{PBL} from WRF60 and WRF12-remap relative to MERRA-2 (Fig. 6) exhibit

482   similarities to differences in AOD (Fig. 4). WRF60 is dry-biased relative to WRF12

483   particularly during the summer (and fall) and underestimates Q_{PBL} relative to MERRA-2 during

484   all seasons over the southern states and over most of continental US during summer and fall.

485   Conversely, WRF12-remap overestimates Q_{PBL} over most of continental US during summer

486   and fall relative to MERRA-2.

487   *PBLH* is a key variable for dictating near-surface aerosol concentrations but is highly sensitive

488   to the physical schemes applied, and biases appear to be domain and resolution dependent.

489   However, this parameter is comparatively difficult to assess because differences in *PBLH* from

490   WRF-Chem and MERRA-2 may also originate from the way they are computed (i.e. from heat

491   diffusivity in MERRA-2 (Jordan et al., 2010) and from turbulent kinetic energy in WRF-Chem

492   (Janjić, 2002;von Engeln and Teixeira, 2013)). Nevertheless, the Mellor-Yamada-Janjich *PBL*

493   scheme combined with the Noah Land Surface Model applied in this work was found to

494   produce lower *PBL* heights (Zhang et al., 2009) than other parameterizations. Thus, the positive

495   bias in simulated AOD and surface $PM_{2.5}$ concentrations (reported previously in (Crippa et al.,

496   2016)) may be linked to the systematic underestimation of *PBLH* simulated by WRF12-remap

497   over continental US relative to MERRA-2 during all seasons (except winter) with greatest bias

498   over regions of complex topography (Fig. S2). A positive bias (of several hundred meters) in

499   terms of *PBLH* for WRF simulations using the MYJ parameterization was previously reported

500   for high-resolution simulations over complex terrain (Rissman et al., 2013), and a positive bias

501   in *PBLH* is also observed in the 60 km simulations presented herein (Fig. S2). This may provide

502   a partial explanation for the large negative bias in AOD in WRF60 during summer (Fig. 3). In

503   general, the BSS indicate improvement in the simulation of *PBLH* in WRF12-remap than in

504   WRF60 (Fig. 7a).

505   Consistent with the dry bias in Q_{PBL} in WRF60, total accumulated precipitation is also

506   underestimated in WRF60, while WRF12-remap captures the absolute magnitudes and the

507   spatial patterns therein (Fig. S3). Analyses of hourly precipitation rates also show higher skill

508   for WRF12-remap than WRF60 in simulating precipitation occurrence (*HR*) relative to

509   MERRA-2 (Table S2). More specifically WRF12-remap correctly predicts between 40% and

510   70 % of precipitation events in MERRA-2 with highest skill during winter months, whereas

511   WRF60 output exhibits lower HR (~6% during summer and 30% during winter). This result

512   thus confirms our expectation of a strong sensitivity of model performance to resolution due to

513   the inherent scale dependence in the cumulus scheme. Use of the Grell-Freitas parameterization
514   in the WRF60 simulations did not lead to substantially different magnitude and/or spatial
515   patterns of precipitation compared to WRF60 applied with the Grell 3D scheme, and no
516   improvement in agreement with output from MERRA2. The findings of a negative bias in
517   precipitation amounts in WRF60 simulations without a corresponding overestimation of AOD
518   may appear counter-intuitive since aerosol concentrations (and thus AOD) are dependent on
519   aerosol residence times and analyses of sixteen global models from the AeroCom project
520   indicate wet scavenging is the dominant removal process for most aerosol species in the study
521   area (Hand et al., 2012;Textor et al., 2006). However, the negative precipitation bias in WRF60
522   simulations appears to also be linked to poor representation of surface moisture availability,
523   boundary layer humidity (Fig. 6), and ultimately aerosol water content (and hence AOD).

524   Gas phase concentrations (transformed into z-scores) from WRF12-remap show higher
525   agreement with satellite observations during almost all months, as indicated by the positive
526   BSS (Fig. 7b). However given the limited availability of valid satellite observations (especially
527   during months with low radiation intensity), the BSS are likely only robust for the summer
528   months for all species. Nevertheless, with the exception of NH_3 during June, BSS for all months
529   are above or close to zero indicating that on average, the enhanced resolution simulations do
530   exhibit higher skill in the simulation of the gas phase species even when remapped to 60 km
531   resolution. Further, the seasonal average spatial patterns of the total columnar concentrations,
532   expressed in terms of z-scores, also exhibit qualitative agreement with the satellite observations
533   (Fig. S4-S7).

4 Concluding remarks

535   This analysis is one of the first to quantify the impact of model spatial resolution on the spatio-
536   temporal variability and magnitude of meteorological and chemical parameters and how
537   representation of these variables impact AOD, and does so using simulations for a full calendar
538   year. Application of WRF-Chem at two different resolutions (60 km and 12 km) over eastern
539   North America for a representative year (2008) leads to the following conclusions:

540   -   Higher-resolution simulations improve the representation of key meteorological
541   variables such as temperature, near-surface specific humidity, boundary layer height
542   and the occurrence and amount of precipitation. Both spatial patterns and precipitation
543   occurrence are better captured by WRF12-remap, and particularly during the summer
544   months the specific humidity within the boundary-layer exhibits closer agreement with

545 a reanalysis product when WRF is applied at higher resolution. The dry bias in the low-
546 resolution WRF-Chem simulations (60 km) is consistent with previous research over
547 eastern North America, and is manifest in simulations with two different cumulus
548 parameterizations and two different data sets for the LBC (GFS and NAM12).

549 - More accurate representation of spatial patterns and concentration of gaseous species
550 that either play a key role in particle formation and growth or are indicators of primary
551 aerosol emissions is also achieved by running WRF-Chem at high resolution.

552 - Partly/largely due to the improved fidelity of key meteorological parameters and gas-
553 phase aerosol precursor species, higher resolution simulations enhance the fidelity of
554 AOD representation at and near to the peak in the solar spectrum relative to a coarser
555 run. At least some of the improvement in the accuracy with which AOD is reproduced
556 in the higher resolution simulations may be due to improved fidelity of specific
557 humidity and thus more accurate representation of hygroscopic growth of some aerosol
558 components. Spatial correlations of AOD from WRF12 and WRF12-remap with
559 observations from MODIS are higher than AOD from a simulation conducted at 60 km
560 during most months. WRF12 show positive spatial correlations with MODIS for all λ
561 in all calendar months, and particularly during summer (ρ = 0.5-0.7). However, the
562 improvement in model performance is not uniform in space and time.

563 - Output from WRF12 and WRF12-remap exhibit highest accord with MODIS
564 observations in capturing the frequency, magnitude and location of extreme AOD
565 values during summer when AOD is typically highest. During May-August WRF12-
566 remap has *Hit Rates* for identification of extreme AOD of 53-78%.

567 It is worthy of note that even the 12 km resolution WRF-Chem simulations exhibit substantial
568 differences in AOD relative to MODIS over eastern North America, and the agreement varies
569 only slightly with wavelength. This may be partially attributable to use of the modal approach
570 to represent the aerosol size distribution in order to enhance computational tractability. In this
571 application each mode has a fixed geometric standard deviation (σ_g), which can lead to biases
572 in simulated AOD in the visible wavelengths by up to 25% (Brock et al., 2016) (with the model
573 overestimating observations if the prescribed σ_g is larger than the observed one). Setting $\sigma_g =$
574 2 for the accumulation mode (the default in WRF-Chem) may lead to an overestimation of the
575 number of particles at the end of the accumulation mode tail, and there is evidence that a value
576 of $\sigma_{g,acc}$=1.40 leads to higher agreement with observations (Mann et al., 2012). Further possible
577 sources of the AOD biases reported herein derive from selection of the physical schemes (e.g.

578 planetary boundary layer (*PBL*) schemes and land-surface model (Misenis and Zhang,
579 2010;Zhang et al., 2009)). Further, it is worth mentioning that NEI emissions are specified
580 based on an average summertime weekday, so enhanced model performance might be achieved
581 if seasonally varying emissions were available.

582 Naturally, there is a need for more research regarding the sensitivity of WRF-Chem simulations
583 of climate relevant aerosol properties to the parameterizations used, the lateral boundary
584 conditions employed and the resolution at which the simulations are conducted. Further,
585 attribution of added-value in the simulation of AOD by enhanced spatial resolution is necessary
586 and will be facilitated by identifying simulation settings that minimize bias in the variables
587 affecting AOD. This research will be part of future investigations.

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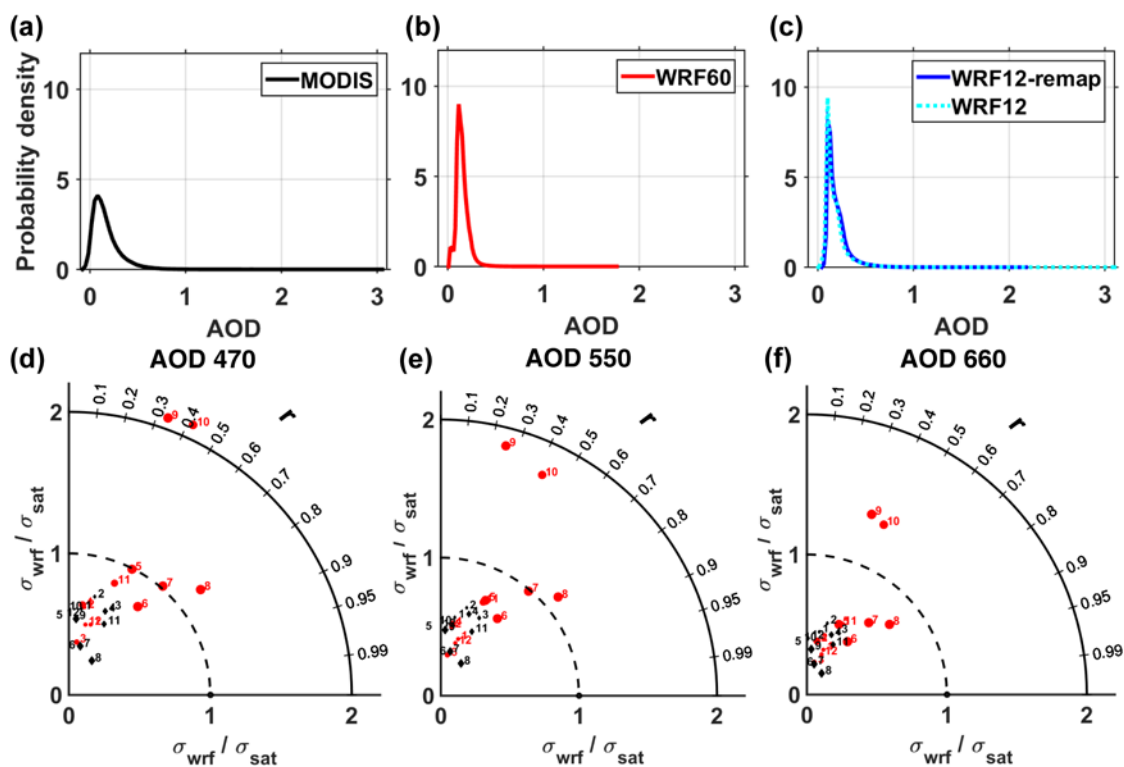
Figure 1. Probability density function of once daily AOD at a wavelength (λ) of 550 nm for (a) MODIS, (b) WRF60 and (c) WRF12 and WRF12-remap during the year 2008. (d-f) Taylor diagrams of mean monthly AOD at wavelengths (λ) of (d) 470, (e) 550 and (f) 660 nm as simulated by WRF-Chem at different resolutions (black diamonds=WRF60 and red dots=WRF12-remap) relative to MODIS observations. The numbers by each symbol denote the calendar month (e.g. 1=January).

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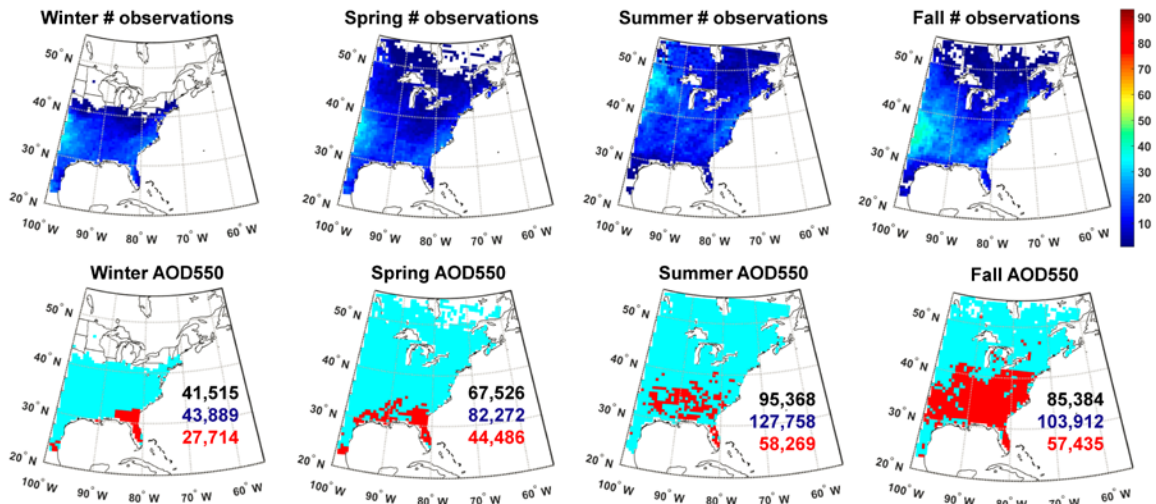
Figure 2. First line: Number of paired AOD observations at a wavelength (λ) of 550 nm (i.e. simultaneous values as output from WRF-Chem and observed by MODIS) used to perform a t-test designed to evaluate whether the difference computed for each grid cell as WRF60-MODIS differs from that computed as WRF12-remap-MODIS on a seasonal basis (columns show Winter (DJF), Spring (MAM), Summer (JJA) and Fall (SON)). Second line: Results of the t-test. Pixels that have p-values that are significantly different at α=0.10 are indicated in red and have been corrected for multiple testing using a False Discovery Rate approach. The number of observations of cloud-free conditions summed across all days in each season and all grid cells is also reported (black=MODIS, blue=WRF60, red=WRF12-remap).

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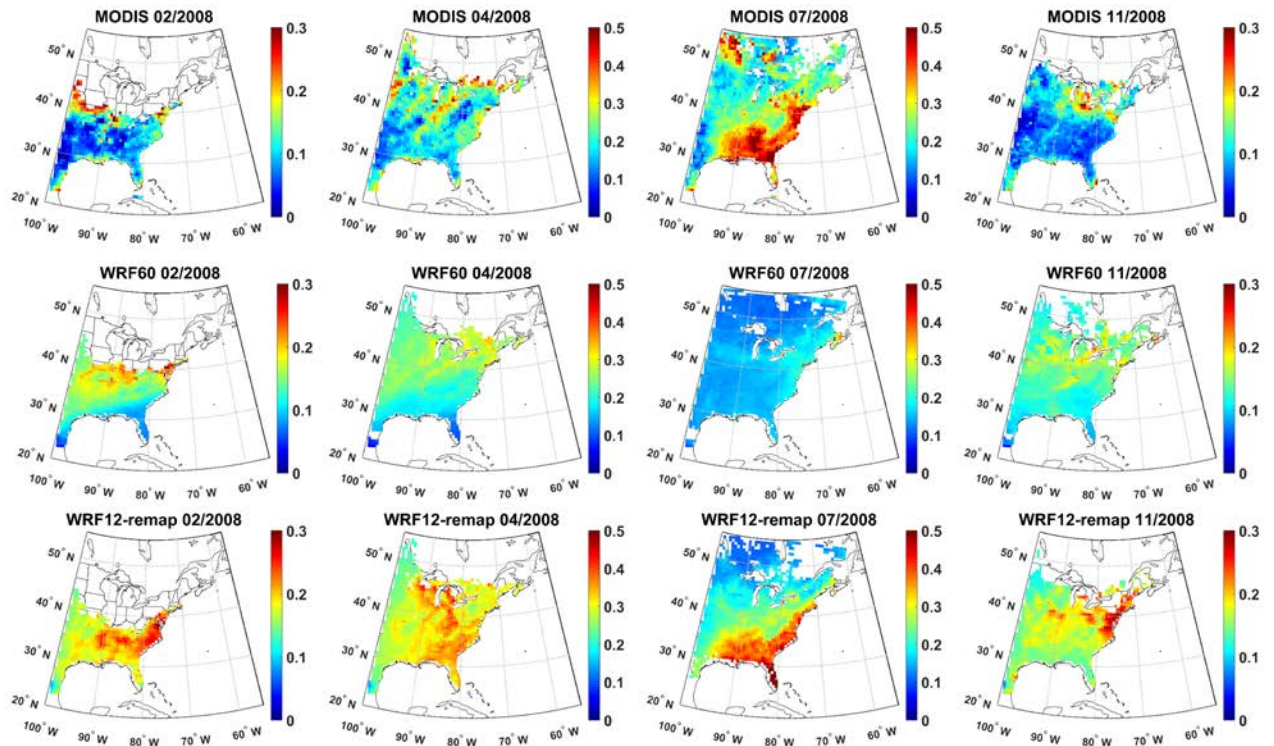
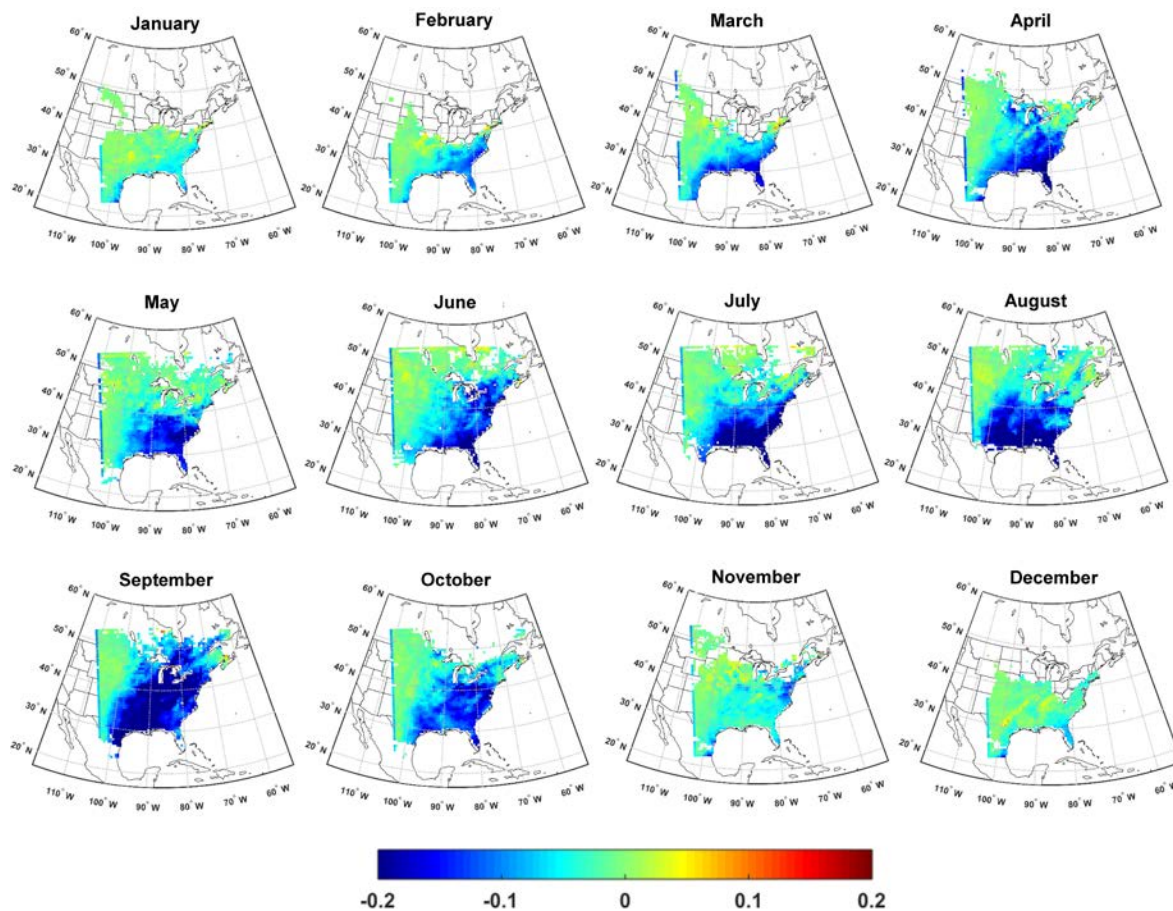**Figure 3. Monthly mean AOD at a wavelength (λ) of 550 nm from MODIS (first line) and**
WRF-Chem at different resolutions (WRF60 and WRF12-remap, second and third line)
during a representative month in each climatological season (columns). Note that a
different color scale is applied for different months. For a monthly mean value for a grid
cell to be shown, there must be at least 5-simultaneous daily values (for the time of the
satellite overpass) available.

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Figure 4. Difference in monthly mean AOD at a wavelength (λ) of 550 nm between WRF-Chem simulations conducted at 60 km resolution (WRF60) and output from WRF-Chem simulations conducted with a resolution of 12 km but remapped to 60 km (WRF12-remap). Differences are computed as WRF60 minus WRF12-remap. Similar spatial patterns and magnitudes of differences are found for λ of 470 and 660 nm. The calendar months of 2008 are shown in the titles of each panel.

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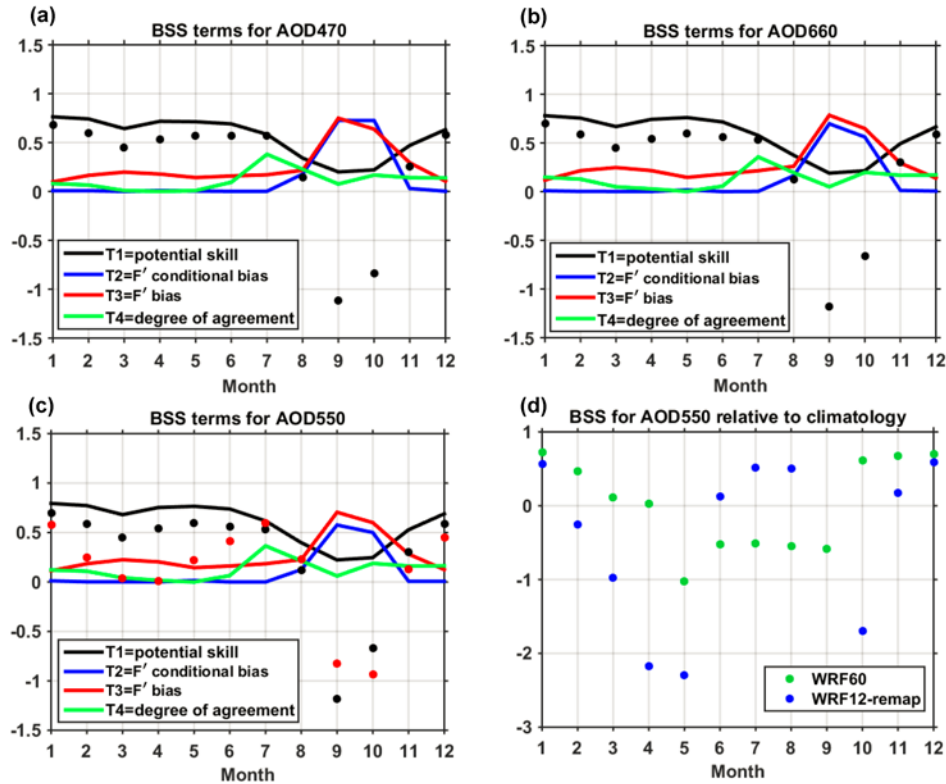
Figure 5. (a-c) Brier Skill Scores (BSS, black dots) for monthly mean AOD by calendar month (1=January) for AOD at 470, 550 and 660 nm. In this analysis of model skill WRF12 output is mapped to the WRF60 grid (WRF12-remap) and BSS are computed using MODIS as the target, WRF60 (driven by NAM12 meteorological boundary conditions) as the reference forecast and WRF12-remap as the forecast. Also shown by the color lines are the contributions of different terms to BSS. In panel c the red dots indicate BSS when the reference forecast is WRF60 driven by GFS meteorological boundary conditions. (d) BSS of monthly mean AOD from WRF60 (green dots) and WRF12-remap (blue dots) relative to MODIS monthly mean climatology during 2000-2014 (reference forecast). Monthly mean AOD from MODIS are used as the target. BSS for WRF12-remap in September is -6.1.
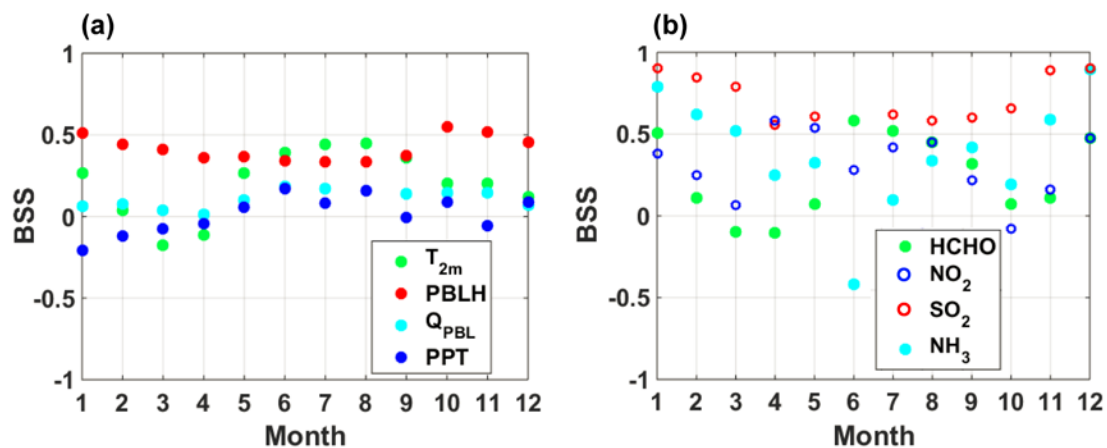
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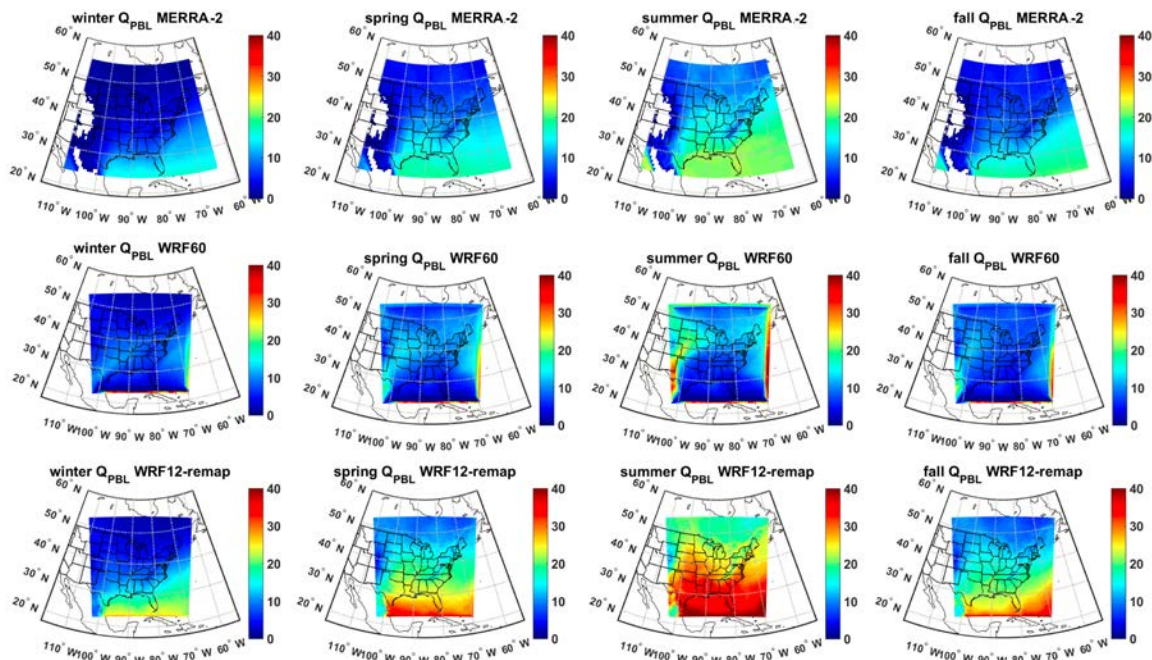
Figure 6. Seasonal mean specific humidity [kg m⁻²] integrated from the surface to 825 hPa
(Q_{PBL}) from MERRA-2 (first row) assuming an average air density in the *PBL* of 1.1 kg
m⁻³, WRF60 (second row), and WRF12-remap (third row). The data are 3-hourly and
show only cloud-free hours in all three data sets.

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Figure 7. Brier Skill Scores (BSS) for key (a) meteorological and (b) chemical variables.
BSS are computed using hourly data of T at 2m (T_{2m}) and *PBLH*, 3-hourly estimates of
specific humidity in the boundary layer (Q_{PBL}), and z-scores of monthly total precipitation
(*PPT*), and of monthly mean columnar gas phase concentrations.

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933 **Tables**

934 **Table 1. Physical and chemical schemes adopted in the WRF-Chem simulations presented**
935 **herein.**

Simulation settings	Values
Domain size	300×300 (60×60) grid points
Horizontal resolution	12 km (60 km)
Vertical resolution	32 levels up to 50 hPa
Timestep for physics	72 s (300 s)
Timestep for chemistry	5 s
Physics option	**Adopted scheme**
Microphysics	WRF Single-Moment 5-class (Hong et al., 2004)
Longwave Radiation	Rapid Radiative Transfer Model (RRTM) (Mlawer et al., 1997)
Shortwave Radiation	Goddard (Fast et al., 2006)
Surface layer	Monin Obhukov similarity (Janjić, 2002;Janjić, 1994)
Land Surface	Noah Land Surface Model (Chen and Dudhia, 2001)
Planetary boundary layer	Mellor-Yamada-Janjich (Janjić, 1994)
Cumulus parameterizations	Grell 3D (Grell and Dévényi, 2002)
Chemistry option	**Adopted scheme**
Photolysis	Fast J (Wild et al., 2000)
Gas-phase chemistry	RADM2 (Stockwell et al., 1990)
Aerosols	MADE/SORGAM (Ackermann et al., 1998;Schell et al., 2001)
Anthropogenic emissions	NEI (2005) (US-EPA, 2009)
Biogenic emissions	Guenther, from USGS land use classification (Guenther et al., 1994;Guenther et al., 1993;Simpson et al., 1995)

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Table 2. Spearman correlation coefficients (ρ) between AOD at wavelengths (λ) of 470, 550 and 660 nm from MODIS observations averaged over 12 or 60 km and WRF-Chem simulations conducted at 60 km (WRF60, shown in the table as -60), at 12 km (WRF12, shown in the table as -12), and from WRF-Chem simulations at 12 km but remapped to 60 km (WRF12-remap, shown in the table as -remap). Given WRF12-remap is obtained by averaging WRF12 when at least half of the 5×5 12 km resolution cells contain valid data, ρ from WRF60 and WRF12-remap may be computed on slightly different observations and sample size. The bold text denotes correlation coefficients that are significant at α=0.05 after a Bonferroni correction is applied (i.e. $p \leq \dfrac{0.05}{9 \times 12} = 4.63 \times 10^{-4}$ **is significant). The yellow shading is a visual guide that shows for each month and λ the model output that has highest ρ with MODIS.**

Month→/ Variable↓	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
470-12	**0.238**	**0.150**	**0.137**	**0.147**	**0.377**	**0.581**	**0.610**	**0.723**	**0.352**	**0.306**	**0.259**	**0.212**
470-60	0.156	**0.226**	**0.438**	**0.412**	-0.219	-0.146	**0.379**	**0.601**	0.087	-0.051	**0.500**	-0.059
470-remap	**0.295**	**0.197**	**0.250**	**0.182**	**0.516**	**0.637**	**0.675**	**0.777**	**0.368**	**0.441**	**0.315**	**0.274**
550-12	**0.223**	**0.124**	**0.142**	**0.146**	**0.349**	**0.541**	**0.580**	**0.689**	**0.275**	**0.301**	**0.280**	**0.215**
550-60	**0.179**	**0.244**	**0.429**	**0.332**	-0.288	-0.188	**0.324**	0.567	0.073	-0.077	**0.491**	0.002
550-remap	**0.297**	0.164	**0.261**	**0.199**	**0.493**	**0.605**	**0.651**	**0.747**	**0.286**	**0.437**	**0.352**	**0.309**
660-12	**0.217**	**0.136**	**0.165**	**0.152**	**0.324**	**0.476**	**0.540**	**0.644**	**0.183**	**0.290**	**0.292**	**0.221**
660-60	**0.191**	**0.230**	**0.437**	**0.402**	-0.305	-0.189	**0.389**	**0.616**	0.099	-0.137	**0.536**	0.049
660-remap	**0.356**	**0.211**	**0.289**	**0.208**	**0.480**	**0.624**	**0.669**	**0.772**	**0.371**	**0.432**	**0.393**	**0.368**

Table 3. Spatial coherence in the identification of extreme AOD values (i.e. areas with AOD>75th percentile over space for each month) between WRF-Chem at different resolutions relative to MODIS. No significant wavelength dependence is found for model skill in identifying extreme AOD so results are only shown for λ = 550 nm. The different model output is denoted by -60 for simulations at 60 km, -12 for simulations at 12 km resolution, and as –remap for simulations at 12 km but with the output remapped to 60 km. The *Accuracy* (Acc) indicates the fraction of grid cells co-identified as extremes and non-extremes between WRF-Chem and MODIS relative to the total number of cells with valid data. The *Hit Rate* (*HR*) is the probability of correct forecast and is the proportion of cells correctly identified as extremes by both WRF-Chem and MODIS. The yellow shading indicates the model resolution with highest skill in each month for AOD at 550 nm.

Month→/ Metric↓	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
Acc-12	0.673	0.665	0.659	0.638	0.710	0.800	0.855	0.839	0.666	0.679	0.723	0.661
Acc-60	0.707	0.778	0.735	0.730	0.600	0.587	0.658	0.769	0.661	0.637	0.729	0.681
Acc-remap	0.674	0.680	0.694	0.640	0.766	0.824	0.887	0.837	0.667	0.699	0.767	0.641
HR-12	0.346	0.331	0.319	0.275	0.421	0.599	0.711	0.678	0.333	0.358	0.447	0.323
HR-60	0.417	0.558	0.471	0.460	0.200	0.173	0.315	0.538	0.321	0.274	0.458	0.364
HR-remap	0.350	0.361	0.387	0.281	0.532	0.649	0.775	0.674	0.333	0.399	0.535	0.284