

Examination of the Community Multiscale Air Quality (CMAQ) model
performance over the North American and European domains

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1 **Abstract**

2

3 The CMAQ modeling system has been used to simulate the air quality for North America and
4 Europe for the entire year of 2006 as part of the Air Quality Model Evaluation International
5 Initiative (AQMEII) and the operational model performance of O₃, fine particulate matter
6 (PM_{2.5}) and PM₁₀ for the two continents assessed. The model underestimates daytime (8am –
7 8pm LST) O₃ mixing ratios by 13% in the winter for North America, primarily due to an
8 underestimation of daytime O₃ mixing ratios in the middle and lower troposphere from the lateral
9 boundary conditions. The model overestimates winter daytime O₃ mixing ratios in Europe by an
10 average of 8.4%. The model underestimates daytime O₃ by 4-5% in the spring for both
11 continents, while in the summer daytime O₃ is overestimated (NMB = 9.8%) for North America
12 but only slightly underestimated (NMB = -1.6%) for Europe. The model overestimates daytime
13 O₃ in the fall for both continents, grossly overestimating daytime O₃ by over 30% for Europe.
14 The performance for PM_{2.5} varies both seasonally and geographically for the two continents. For
15 North American, PM_{2.5} is overestimated in the winter and fall, with an average NMB greater
16 than -30%, while performance in the summer is relatively good, with an average NMB of -4.6%.
17 For Europe, PM_{2.5} is underestimated throughout the entire year, with the NMB ranging from -
18 24% in the fall to -55% in the winter. PM₁₀ is underestimated throughout the year for both North
19 America and Europe, with remarkably similar performance for both continents. The domain
20 average NMB for PM₁₀ ranges between -45% and -65% for the two continents, with the largest
21 underestimation occurring in the summer for North American and the winter for Europe.

22

23 **Keywords:** CMAQ; Ozone; Particulate Matter; Air Quality Modeling; Model Evaluation;
24 AQMEII

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27 **1. Introduction**

28

29 The Air Quality Model Evaluation International Initiative (AQMEII) is a model evaluation effort
30 involving numerous research groups from North American and Europe with the goal of
31 advancing the methods for evaluating regional-scale air quality modeling systems. As part of the
32 AQMEII project, the Community Multiscale Air Quality (CMAQ; Foley et al., 2010) model has
33 been applied to simulate air quality over North America (NA) and Europe (EU) for the year
34 2006.

35 The CMAQ simulation performed for NA for this project is unique compared to the
36 CMAQ simulations performed in the past for several reasons. First, the simulation was
37 performed over a single domain that covers the entire CONUS and a large portion of Canada
38 using 12-km by 12-km horizontal grid spacing. In the past, two separate simulations covering
39 the eastern and western U.S. have been used instead of single, continuous domain. Second, the
40 simulation utilizes meteorology provided by the latest version of the Weather Research and
41 Forecasting (WRF) model, whereas previous CMAQ annual simulations have typically utilized
42 meteorology provided by the 5th Generation Mesoscale Model (MM5; Grell et al., 1994).
43 Finally, the CMAQ simulation utilizes boundary conditions provided by the Global and regional
44 Earth-system Monitoring using Satellite and in-situ data (GEMS) product.

45 The analysis presented here focuses primarily on ozone (O₃) and particulate matter (PM_{2.5}
46 and PM₁₀), as these are pollutants for which both the NA and EU have established criteria for

47 acceptable limits (e.g. National Ambient Air Quality Standards) and instituted numerous control
48 strategies to reduce precursor emissions. The analysis presented here is intended to provide a
49 broad overview of the operational performance of the CMAQ model for these pollutants for NA
50 and EU, and compare and contrast significant similarities or differences in model performance
51 for the two continents.

52

53 **2. Data**

54 *2.1 Model Inputs and Configuration*

55

56 The CMAQ model requires gridded meteorological and emissions data to simulate the
57 formation, transport and fate of numerous atmospheric pollutants, including O₃ and PM.
58 Meteorological data for the NA and EU simulations were provided by the Weather Research and
59 Forecast (WRF) model (Skamarock et al., 2008). For NA, the WRF domain covered the
60 CONUS and portions of Canada and Mexico using 12-km by 12-km horizontal grid spacing and
61 34-vertical layers extending up to 50 hPa. The simulation utilized the Pleim-Xu land surface
62 model (LSM), ACM2 planetary boundary layer (PBL) scheme, Morrison mixed phase (MP)
63 scheme, Kain-Fritsch2 cumulus parameterization (CuP) scheme and the RRTMG long-wave
64 radiation (LWR) scheme. Lateral boundary conditions (BCs) were provided by the North
65 American Model (NAM), available from the National Centers for Environmental Prediction.

66 For the EU CMAQ simulation, the WRF model was also used, but with a slightly
67 different configuration to that of the NA WRF simulation more appropriate for simulating the
68 Europe continent. The EU WRF simulation was performed using 18-km by 18-km horizontal
69 grid spacing with 52 vertical layers, 11 of which were below 1-km. The simulation utilized the

70 NOAH LSM, Morrison (MP) scheme, Grell and Devenyi CuP scheme, and RRTMG LWR
71 scheme. Initial and lateral BCs were provided by the European Center for Medium-Range
72 Weather Forecasts (ECMWF) model. Outputs from the WRF simulations for both continents
73 were preprocessed for input into CMAQ using v3.6 of the Meteorology-Chemistry Interface
74 Processor (MCIP; Otte et al., 2005). More specific details regarding the WRF simulations,
75 including references for the various schemes used and an operational performance evaluation of
76 the simulations can be found in Vautard et al. (this issue).

77 The NA CMAQ model simulation used the AQMEII standard NA emissions dataset,
78 which is based on a 12-km national U.S. domain with speciation for the Carbon-Bond 05 (CB05)
79 chemical mechanism (Yarwood et al., 2005). The emission inventory and ancillary files were
80 based on the 2005 emission modeling platform. The fire emissions were based on 2006 daily
81 fire estimates using the Hazard Mapping System Fire detections and Sonoma Technology
82 SMARTFIRE system. Continuous Emission Monitoring System (CEMS) data from 2006 was
83 used for the electric generating units sector. Plume rise was calculated within the CMAQ model
84 (in-line). Temporal allocation was done monthly for each day of the week with all holidays
85 ignored. Emissions were preprocessed for the CMAQ model using the Sparse Matrix Operator
86 Kernel Emissions (SMOKE; Houyoux et al., 2000).

87 The AQMEII standard EU emissions data were used for the EU CMAQ simulation and
88 are based on the TNO (<http://www.tno.nl/>) inventory for 2005, which consists of anthropogenic
89 emission from ten Selected Nomenclature for Air Pollution (SNAP) sectors and international
90 shipping. The ten SNAP sectors are energy transformation, small combustion sources, industrial
91 combustion, industrial processes, extraction of fossil fuels, solvent and product use, road
92 transport, non road transport, waste handling, and agriculture. Biogenic emissions of isoprene

93 and terpene, calculated using the Model of Emissions of Gases and Aerosols from Nature
94 (MEGAN; Guenther and Wiedinmyer, 2007; Sakulyanontvittaya et al., 2008), are included on
95 the same resolution as the anthropogenic emissions. The fire emissions were based on 2006 daily
96 fire estimates from the MODIS fire radiative power product using the FMI Fire Assimilation
97 System FAS-FRP (Sofiev et al., 2009). Plume rise was calculated offline with SMOKE. A more
98 detailed description of the emission used for the two continents is available in Pouliot et al. (this
99 issue).

100 The CMAQ model configurations were similar for NA and EU, with both simulations
101 utilizing version 4.7.1 (Foley et al., 2010) of the model. The NA simulation used 34-vertical
102 layers (matched to the WRF model vertical layers) and 12-km horizontal grid spacing covering
103 the CONUS, southern Canada and northern Mexico, while the EU simulation used 34 vertical
104 layers (52 WRF vertical layers collapsed to 34 CMAQ vertical layers in MCIP) and 18-km
105 horizontal grid spacing covering most of EU. Other model options employed that were common
106 to both simulations include the CB05 chemical mechanism with chlorine chemistry extensions
107 (Yarwood et al., 2005), the AERO5 aerosol module (Carlton et al., 2010), the Asymmetric Cloud
108 Model 2 (ACM2) PBL scheme (Pleim, 2007a,b).

109 Both the NA and EU simulations utilized the standard AQMEII BCs provided by the
110 Global and regional Earth-system Monitoring using Satellite and in-situ data (GEMS) product
111 (<http://gems.ecmwf.int/about.jsp>), which assimilates modeled data and observations (surface and
112 satellite) to provide data for meteorology and atmospheric gases including greenhouse gases,
113 global reactive gases and global aerosols. A more detailed description of the GEMS data as used
114 as boundary conditions can be found in Schere et al. (this issue).

115

116 *2.2 Air Quality Observations*

117

118 For NA the observed data used to assess the CMAQ model estimates are obtained from
119 several observational networks available across the U.S. that measure a combination of gas,
120 aerosol, wet deposition and meteorological variables. The primary sources of ground level O₃,
121 PM_{2.5} and PM₁₀ mass measurements for the U.S. is the USEPA's Air Quality System (AQS). The
122 AQS network is geographically diverse and spans the entire U.S. and is an excellent source of
123 quality assured air quality measurements. Measurements of O₃ are hourly, while measurements
124 of PM can be either hourly or daily averages (available every 1, 3 or 6 days), depending on the
125 particular site configuration. For observations of PM_{2.5}, measurements from the AQS, the
126 Chemical Speciation Network (CSN) and the Interagency Monitoring of Protected Visual
127 Environments (IMPROVE) network are used. In addition to total PM_{2.5}, the CSN and
128 IMPROVE networks provide measurements of particulate SO₄⁼, NO₃⁻, NH₄⁺, EC and OC, along
129 with a large number of other trace elements. The AQS is used to provide PM₁₀ measurement
130 data. For Canada, the National Air Pollution Surveillance (NAPS) network provides
131 measurements of O₃ and PM_{2.5}.

132 The air quality networks in EU used to provide data for the present analysis are the
133 AirBase network (<http://www.eea.europa.eu/themes/air/airbase>), the Automatic Urban and Rural
134 (AURN; <http://uk-air.defra.gov.uk/interactive-map>) network and the EMEP
135 (http://www.emep.int/index_facts.html) network. Each of these networks provides hourly and
136 daily average data for a number species, including O₃, PM_{2.5} and PM₁₀. Assessment of the model
137 performance was accomplished using the Atmospheric Model Evaluation Tool (AMET; Appel et
138 al., 2010), which can perform a vast number of different analyses and produce many different

139 plots useful for assessing model performance. AMET was originally designed for the U.S. based
140 air quality networks, but has been extended to incorporate observations available from air quality
141 networks in EU.

142

143 **3. Results**

144 *3.1 Ozone*

145

146 Ozone is an important criteria pollutant for both NA and EU. Ozone mixing ratios are
147 the highest in the summer as the production of O₃ is a photo-chemically driven reaction and the
148 reactions are more efficient under higher temperatures. In the U.S., O₃ mixing ratios generally
149 peak in July and August (Fig. 1), when temperatures are the highest and the sun angle is high.
150 The pattern of O₃ mixing ratios in EU is similar to that of NA, with a peak in O₃ mixing ratios in
151 June and July (Fig. 2). The current daily thresholds for O₃ in the U.S. and EU are based on the
152 maximum daily 8-hr average O₃ value and are currently set to 75 ppb for the U.S. and 120 µg⁻³
153 (~60 ppb) for EU. Since the O₃ standards for each continent are based on the daily maximum 8-
154 hr average O₃, the analysis here is limited to just the daytime hours, where daytime is defined as
155 8am to 8pm local standard time (LST), when O₃ mixing ratios are the highest.

156 For NA, operational model performance for O₃ was generally consistent with previous
157 CMAQ simulations (Eder and Yu, 2006; Tesche et al., 2006; Appel et al., 2007), with several
158 notable exceptions. Performance of maximum 8-hr average O₃ in the winter (January - March)
159 underperformed previous CMAQ simulations (Appel et al., 2007), with the model demonstrating
160 a large underestimation of daytime O₃ (-13.4% domain-wide average) for that period (Table 1).
161 Fig. 1 illustrates the large underestimation of O₃ for NA in the winter, while Fig. 3a presents a

162 spatial plot of Normalized Mean Bias (NMB) for AQS sites for winter. The underestimation of
163 O₃ in the winter is largest in the Northeast and Great Lakes regions of the U.S. and for most of
164 the Canadian sites, with smaller underestimations in the southern U.S. For EU, the CMAQ
165 system overestimates daytime O₃ in southwestern half of the domain and underestimates daytime
166 O₃ in the northeastern half of the domain, including the United Kingdom, in the winter (Fig. 4a).
167 The largest overestimations occur in northern Italy, primarily in Po River Valley, where a large
168 number of sites have NMBs greater than 100%. The largest underestimations occur in the Czech
169 Republic and Poland, where some sites have NMBs exceeding -70%.

170 Investigation of the poor wintertime performance for O₃ in the NA CMAQ simulation
171 suggests that the lateral BCs used in the AQMEII CMAQ simulation are largely responsible for
172 the poor performance (Schere et al. this issue). In order to determine the impact of the later BCs
173 on the winter O₃ model estimates, the CMAQ simulation was repeated using BCs provided by
174 the global model GEOS-Chem (Bey et al., 2001) instead of the AQMEII default BCs which used
175 GEMS. The O₃ time-series for the NA and EU CMAQ simulations using lateral BCs provided
176 by the GEOS-Chem model are presented in Figs. 1 and 2 along with the base AQMEII CMAQ
177 simulation. The large wintertime underestimation of daytime O₃ that is clearly evident in
178 CMAQ simulation for NA using the GEMS derived BCs is not present in the CMAQ simulation
179 that utilized GEOS-Chem BCs. Similarly, the CMAQ estimated O₃ in the simulation for EU
180 using GEOS-Chem BCs is much higher in the winter and spring than the simulation using
181 GEMS BCs.

182 Further comparison of the vertical profiles of observed and CMAQ estimated O₃ (not
183 shown) indicated that the mid to lower tropospheric O₃ mixing ratios in the GEMS BCs were
184 significantly underestimated, while the same comparison to the CMAQ estimated O₃ from the

185 simulation using GEOS-Chem BCs showed no significant underestimation of lower tropospheric
186 O₃ (see also Schere et al., 2011 for additional discussion of the GEMS data). The lower O₃
187 mixing ratios in the troposphere in the GEMS BCs result in lower ground-level O₃ mixing ratios,
188 particularly in the winter when O₃ provided from the lateral boundaries contributes a significant
189 portion of the CMAQ estimated ground-level O₃. In the summer, O₃ mixing ratios in the lower
190 troposphere in the GEMS BCs are much more similar in magnitude to the mixing ratios in the
191 GEOS-Chem BCs, which results in better agreement with observations. Schere et al. (this issue)
192 describe similar results for a comparison between the CMAQ simulations for EU using GEMS
193 and GEOS-Chem BCs, and note that the performance degrades in the lower troposphere when
194 using the GEMS BCs.

195 For the spring, the site specific NMBs typically range between $\pm 10\%$ for much of North
196 America, with slightly larger NMBs in the Northeast, Canada and California, where daytime O₃
197 is underestimated at some sites by 20% or more (Fig. 3b). For EU, there continues to be a strong
198 differentiation in performance in the spring between the southwest and northeast portions of the
199 domain that was seen in the winter, with O₃ being relatively unbiased (NMB within $\pm 10\%$) in the
200 southwest (the exception being northern Italy where O₃ is overestimated/underestimated by 50%
201 at several sites). The daytime O₃ for sites in Germany, Poland and the Czech Republic is
202 frequently underestimated by 10-30% in the spring (Fig. 4b). Similar to the simulation for NA, a
203 contributing factor to the underestimation of O₃ in the spring is the underestimation of O₃ in the
204 GEMS lateral BCs (see Schere et al. in this issue).

205 For the summer, daytime O₃ is overestimated over the majority of NA (domain average
206 NMB = 9.8%), with the largest overestimations in California, Florida and along the Gulf of
207 Mexico (Fig. 3c). The NMB for the Canadian NAPS sites in summer tends to be lower than that

208 of the AQS sites. For EU, the daytime O₃ performance is generally better than that of NA, with a
209 large number of sites having NMBs within $\pm 10\%$ and the majority of sites having NMBs with
210 $\pm 20\%$ (Fig. 4c). The largest biases occur in France and northern Italy (Po River Valley), where
211 O₃ tends to be underestimated by 10-20% for the majority of the sites, and along the coast of
212 Spain, where the model typically overestimates daytime O₃ by 20% or more (slightly smaller
213 overestimations occur along the coast of Italy as well). The overestimation of O₃ in the summer
214 along coastal areas is seen in the CMAQ simulation for NA as well (Fig. 3c), suggesting that the
215 source of the large biases may be due to errors in the meteorological inputs to the CMAQ
216 system, particularly in regards to the meteorological model's ability to accurately represent the
217 sea-breeze and land-breeze effects along the coast. The CMAQ model performance for the
218 summer is consistent with a previous study by Eder et al. (2009) that reported CMAQ
219 overestimated O₃ during the summer by about 9% and also noted very large overestimations
220 along the Gulf of Mexico.

221 Daytime O₃ is overestimated in the fall for both NA and EU (Figs. 3d and 4d). The
222 largest overestimations in NA occur in the eastern U.S. (including the eastern NAPS sites),
223 where the NMB frequently exceed 20% at a large number of sites, and in the Northwest, where
224 the NMB exceeds 80% at several of the NAPS sites. The fall has the worst overall performance
225 for daytime O₃ for EU, with the model grossly overestimating O₃ across most of the domain
226 (domain average NMB = 32.3%). The majority of sites have NMBs greater than 20%, with a
227 large number of sites in northern Italy having NMBs exceeding 100%.

228

229 *3.2 PM_{2.5}*

230

231 Particulate matter, including both $PM_{2.5}$ with a diameter less than $2.5 \mu m$ and coarse
232 PM_{10} with a diameter less than $10 \mu m$, is an important air pollutant for which standards exist for
233 both the U.S. and EU. The U.S. limits on PM are based on $PM_{2.5}$, with the current annual limit
234 set at $15 \mu g m^{-3}$, while for EU the primary PM standard is based on PM_{10} , with the current annual
235 limit set at $40 \mu g m^{-3}$. Since the two continents use different standards for regulating PM, the
236 monitoring networks are also different, with North American (U.S. and Canada) networks
237 focused primarily on measuring $PM_{2.5}$ and European networks focused on measuring PM_{10} . As
238 such, PM_{10} measurements for NA are not as widely available as $PM_{2.5}$ measurements, and
239 likewise there are limited $PM_{2.5}$ measurements available for EU. On average, there are
240 approximately 870 AQS sites in the U.S. and 160 AirBase sites in EU with $PM_{2.5}$ measurements,
241 and 580 AQS sites and over 1000 AirBase sites with PM_{10} measurements.

242 Unlike O_3 , which has a large seasonal dependency, $PM_{2.5}$ concentrations in NA do not
243 vary as much throughout the year (Fig. 5), while for EU high concentrations of $PM_{2.5}$ are
244 observed from January through March, after which the concentrations are considerably lower
245 and relatively constant throughout the remainder of the year (Fig. 6). The CMAQ model
246 generally does well representing the small seasonal trends in $PM_{2.5}$ for both continents, and
247 captures the synoptic forcing features. Note that there are a limited number of $PM_{2.5}$
248 observations available for EU, with the majority of the observations sites in Portugal, Spain,
249 France, Italy, Belgium, Germany, and the Czech Republic.

250 For the winter, there is a large overestimation of $PM_{2.5}$ in NA (Table 2), with a domain-
251 wide average NMB of 30.4% and Mean Bias (MB) of $3.4 \mu g m^{-3}$, but underestimates $PM_{2.5}$ to an
252 even greater extent in EU, with a NMB of -55% (MB = $-12.9 \mu g m^{-3}$). The largest
253 underestimations in the NA occur in the west, where a large number of sites report NMBs greater

254 than 100% (Fig. 7a). The northeastern U.S. also has a number of sites with NMBs exceeding
255 30%. For EU, the underestimation in $PM_{2.5}$ is systematic across the domain, with only a handful
256 of sites reporting an overestimation (Fig. 8a). The largest underestimations occur in the Czech
257 Republic, Germany and Italy, with the majority of sites reporting NMBs greater than -60%. The
258 performance for France, the United Kingdom, Spain and Portugal is better, with a number of
259 sites reporting NMBs smaller than -30%.

260 The overestimation in $PM_{2.5}$ in NA is primarily due to an overestimation of the
261 unspiciated $PM_{2.5}$ mass, along with a smaller overestimation of elemental and organic carbon
262 (Appel et al, 2008). The unspiciated $PM_{2.5}$ mass, sometimes referred to as PM_{other} , is comprised
263 primarily of the non-carbon atoms associated with OC, along with trace elements (e.g. Fe, Mg,
264 Mn, etc.), primary ammonium and other unidentified mass in the speciation profiles. Since this
265 unspiciated mass makes up a significant portion of the total $PM_{2.5}$ mass and is often largely
266 under or overestimated in the CMAQ model, efforts were made to include speciation of the
267 unidentified mass, in particular the trace elements, in the model. The next version of the CMAQ
268 model, due to be released in the fall of 2011, will include the speciation of the trace metals,
269 allowing for a comparison of the model estimates to observations, which will hopefully lead to
270 an improvement in the model estimates for those elements and reduction in the bias for PM_{other} .

271 The model estimates for $PM_{2.5}$ improve significantly in the spring, with a domain-wide
272 average NMB of 18.9% ($MB = 2.0 \mu g m^{-3}$) for NA and -36.9% ($MB = -5.8 \mu g m^{-3}$) for EU (Table
273 2). For NA, $PM_{2.5}$ tends to be underestimated in the southern portion of the domain, with most
274 sites having a NMB less than -20%, while $PM_{2.5}$ continues to be overestimated by the model in
275 the Northeast and in the west, where most sites have a NMB of 20% or greater (Fig. 7b). For
276 EU, $PM_{2.5}$ continues to be significantly underestimated in the east (Czech Republic and Italy),

277 with the underestimation in Germany, France and the United Kingdom improved from the winter
278 (Fig. 8b). The performance in Spain and Portugal is relative good, with most sites having a
279 NMB within $\pm 20\%$.

280 For the summer, CMAQ estimated $PM_{2.5}$ concentrations are slightly underestimated on
281 average, with a domain-wide average NMB of -4.6% and MB of $-0.6 \mu\text{gm}^{-3}$ (Table 2). Spatially,
282 $PM_{2.5}$ is underestimated by 20-30% for majority of sites in the eastern U.S., the exceptions being
283 Florida, where $PM_{2.5}$ is overestimated, and the Great Lakes region, where most sites have NMBs
284 within $\pm 10\%$ (Fig. 7c). The underestimations in the southeastern U.S. may be due in part to an
285 underestimation of secondary organic aerosol, which can make up a large portion of the total
286 $PM_{2.5}$ in the southeast (Carlton et al., 2010). Large underestimations of $PM_{2.5}$ in the desert
287 southwest (New Mexico, Arizona, Colorado and Utah) of -50% or more may be due to a lack of
288 wind-blown dust in the model. The next version of the CMAQ model will include a method for
289 representing wind-blown dust, which may improve the underestimations of $PM_{2.5}$ in the
290 southwestern U.S. in the summer. For EU, the performance for the summer is similar to the
291 spring, with a domain-wide average NMB of -37.2% (MB = $-4.9 \mu\text{gm}^{-3}$), and a similar spatial
292 distribution of bias as the spring (Fig. 8c).

293 For the fall, $PM_{2.5}$ is again overestimated for NA, with a domain-wide average NMB of
294 36.3% (MB = $4.0 \mu\text{gm}^{-3}$). The spatial pattern of bias is similar to that of the winter, with the
295 largest overestimations in the northeast and northwest U.S. (Fig. 7d). As with the winter, the
296 overestimation of the unspciated $PM_{2.5}$ mass is largely responsible for the overestimation of
297 $PM_{2.5}$ in the fall, along with smaller overestimations of particle nitrate and ammonium. For EU,
298 $PM_{2.5}$ continues to be underestimated, however the bias is smaller than any of the other seasons,
299 with an average NMB of -24.2% (MB = $-3.8 \mu\text{gm}^{-3}$). The largest underestimations continue to

300 be in the Czech Republic and Italy, with most sites having NMBs of – 20% to -50% (Fig. 8d).
301 Performance for sites in Germany, France the United Kingdom improves again, with most sites
302 having NMBs within $\pm 20\%$, while in Spain and Portugal several of the sites now show an
303 overestimation of $PM_{2.5}$, generally within 30-50%.

304

305 *3.3 PM_{10}*

306

307 The PM_{10} mass is composed of all the PM less than 10 μm in diameter, and therefore
308 includes all the $PM_{2.5}$ mass and coarse PM ($PM_{10} - PM_{2.5}$). Fig. 9 presents the domain-average
309 time series for observed and CMAQ estimated PM_{10} for NA, while Fig. 10 presents a similar
310 time-series plot for EU. The model systematically underestimates PM_{10} for both continents
311 throughout the year, with the largest underestimation occurring in the winter for EU when
312 observed PM_{10} is very high. For EU in the winter, the domain average NMB is -64.8% ($MB = -$
313 $21.5 \mu g m^{-3}$), compared to only -47.6% ($MB = -11.5 \mu g m^{-3}$) for NA. For the other seasons, the
314 underestimation for both continents is nearly identical and relatively consistent through the year,
315 with the model underestimating PM_{10} by between 45-60% ($11-16 \mu g m^{-3}$) for each continent
316 (Table 3).

317 Spatially, the model tends to demonstrate a similar bias pattern throughout the year for
318 both continents. In the winter, when the PM_{10} underestimation is the smallest for NA, the model
319 generally overestimates PM_{10} by 20-50% along the east coast of the U.S. (Fig. 11a). For the rest
320 of country, PM_{10} is largely underestimated, particularly in the western U.S. (with the exception
321 of areas right along the coast). For EU, almost every site shows an underestimation of PM_{10} ,
322 with most sites having NMBs exceeding -50% (Fig. 12a). The smallest biases are in northern

323 France, where most sites have NMBs less than 30%. In the spring, the bias pattern is similar to
324 the winter, with the smallest biases for NA occurring along the east and west coasts, while in EU
325 the bias spatial pattern is nearly identical to that of winter (Figs. 11b and 12b).

326 For the summer, the majority of sites in NA now show some level of underestimation of
327 PM_{10} , with almost all the sites in the western U.S. having NMBs greater than -20% (Fig. 11c).

328 For EU, the bias pattern is again similar to the winter and spring, with only northern France and
329 Portugal having any significant number of sites showing NMBs smaller than 40% (Fig. 12c).

330 The bias tends to improve in the fall for both continents compared to the summer, with a large
331 number of sites in the eastern U.S. having NMBs between $\pm 30\%$, while in the western U.S. most
332 sites continue to show large underestimations of PM_{10} of 50% or more (Fig. 11d). For EU, the
333 majority of sites continue to show significant underestimations of PM_{10} in the fall (Fig. 12d),
334 however a large number of sites in France and Germany now have NMBs between -20 to -30%,
335 an improvement of the -40% or more NMBs seen in the other seasons. Additional analysis is
336 needed to diagnose the cause for the large biases in CMAQ PM_{10} estimates, which are likely due
337 to a combination of errors in the emissions inventory and chemical transport model.

338

339 **4. Summary**

340

341 The CMAQ modeling system has been used to simulate NA and EU for the entire year of
342 2006. The model performance for O_3 varies seasonally, with the model underestimating daytime
343 O_3 mixing ratios in the winter by about 13% for NA and overestimating daytime O_3 for EU by
344 roughly 8%. Analysis suggests that lower O_3 mixing ratios in the middle and lower troposphere
345 from the chemical boundary conditions are primarily responsible for the lower ground-level O_3

346 mixing ratios in the winter in NA and EU. For the spring, daytime O₃ is slightly underestimated
347 for both NA and EU (4-5%), likely due in part to an underestimation of O₃ from the boundaries.
348 For the summer, when O₃ mixing ratios are the highest, CMAQ overestimates daytime O₃ for
349 NA by about 10% on average, while for EU the model underestimates daytime O₃ by less than
350 2% on average. Daytime O₃ continues to be overestimated in the fall for NA by 8% on average,
351 while for the EU the model grossly overestimates O₃ by more than 30% on average. Overall, the
352 model demonstrates relatively similar performance for daytime O₃ in both modeling domains,
353 with the exception of the fall.

354 The model performance for PM_{2.5} varies between the two continents, with the model
355 overestimating PM_{2.5} in the winter, spring and fall, and being relatively unbiased in the summer
356 for NA, while for EU the model underestimates PM_{2.5} throughout the entire year. While it is not
357 clear what is driving the bias in PM_{2.5} for the two continents, likely sources of error for both
358 continents is the lateral boundary conditions and emissions. It would be helpful to examine any
359 speciated PM_{2.5} data available in EU to determine what components of PM_{2.5} are primarily
360 responsible for the underestimation. The model performance for PM₁₀ was also examined for
361 both continents, with the model systematically underestimating PM₁₀ for both continents.
362 Outside of the winter months, when PM₁₀ was grossly underestimated for EU, the model
363 performance for PM₁₀ for both continents is very similar, with model generally underestimating
364 PM₁₀ between 45-60% on average. More investigation is needed to determine what is driving
365 the poor PM₁₀ estimates from the modeling system (e.g. emissions or meteorology). Segregating
366 the data by different synoptic regimes (e.g. Appel et al., 2007) may highlight the role
367 meteorology plays in the PM₁₀ estimates, while the addition of trace metals and a method for

368 tracking wind-blown dust available in the next release of the CMAQ model may help illuminate
369 errors in the emission inventory.

370 The analysis presented here represents only a broad overview of the operational model
371 performance of three pollutants for NA and EU. The analysis describes some the similarities and
372 differences in model performance between the two continents and highlights aspects of the
373 modeling system that need improvement (e.g. PM₁₀). Further analysis is needed to determine the
374 factors driving these differences in model performance. Future work will include comparing the
375 model performance for other species, such as NO₂ and SO₂, between the two continents, as well
376 as examining the performance of the model wet deposition estimates, which are important
377 outputs used in ecological studies.

378

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380

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414

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419

420 **References**

- 421
- 422 Appel, K.W., Bhave, P.V., Gilliland, A.B., Sarwar, G., Roselle, S.J., 2008. Evaluation of the
423 Community Multiscale Air Quality (CMAQ) model version 4.5: Sensitivities impacting
424 model performance; Part II - particulate matter, *Atmospheric Environment* 42, 6057-6066.
425
- 426 Appel, K.W., Gilliland, A.B., Sarwar, G., Gilliam, R.C., 2007. Evaluation of the Community
427 Multiscale Air Quality (CMAQ) model version 4.5: Sensitivities impacting model
428 performance; Part I – ozone, *Atmospheric Environment* 41, 9603-9615.
429
- 430 Appel, K. W., Gilliam, R. C., Davis, N., Zubrow, A., and Howard, S. C., 2010. Overview of the
431 Atmospheric Model Evaluation Tool (AMET) v1.1 for evaluating meteorological and air
432 quality models, *Environmental Modeling and Software*, [doi:10.1016/j.envsoft.2010.09.007](https://doi.org/10.1016/j.envsoft.2010.09.007).
433
- 434 Bey, I., Jacob, D.J., Yantosca, R.M., Logan, J.A., Field, B.D., Fiore, A.M., Li, Q., Liu, H.Y.,
435 Mickley, L.J., and Schultz, M.G., 2001. Global modeling of tropospheric chemistry with
436 assimilated meteorology: Model description and evaluation, *Journal of Geophysical Research*,
437 106, 23073-23096.
438
- 439 Carlton, A. G., Bhave, P. V., Napelenok, S. L., Edney, E. O., Sarwar, G., Pinder, R. W., Pouliot,
440 and G. A., Houyoux, M., 2010. Model representation of secondary organic aerosol in
441 CMAQv4.7, *Environmental Science and Technology*, 44, 8553-8560.
442

443 Eder, B., Kang, D., Mathur, R., Pleim, J., Yu, S., Otte, T., and Pouliot, G., 2009. A performance
444 evaluation of the National Air Quality Forecast Capability for the summer 2007, Atmospheric
445 Environment, 43 (14), 2312-2320.

446

447 Eder, B., Yu, S., 2006. A performance evaluation of the 2004 release of Models-3 CMAQ.
448 Atmospheric Environment 40, 4811–4824.

449

450 Foley, K. M., Roselle, S. J., Appel, K. W., Bhave, P. V., Pleim, J. E., Otte, T. L., Mathur, R.,
451 Sarwar, G., Young, J. O., Gilliam, R. C., Nolte, C. G., Kelly, J. T., Gilliland, A. B., and Bash,
452 J. O., 2010. Incremental testing of the Community Multiscale Air Quality (CMAQ) modeling
453 system version 4.7, Geoscientific Model Development 3, 205-226.

454

455 Grell, G. A., Dudhia, A. J., and Stauffer, D. R., 1994. A description of the Fifth-Generation
456 PennState/NCAR Mesoscale Model (MM5). NCAR Technical Note NCAR/TN-398+STR.
457 Available at <http://www.mmm.ucar.edu/mm5/doc1.html>.

458

459 Guenther, A. and Wiedinmyer, C., 2007. User's guide to the Model of Emissions of Gases and
460 Aerosols from Nature (MEGAN), Version 2.01.

461

462 Houyoux, M. R., Vukovich, J. M., Coats Jr., C. J., Wheeler, N. J. M., Kasibhatla, P., 2000.
463 Emission inventory development and processing for the seasonal model for regional air
464 quality, Journal of Geophysical Research, 105 (D7), 9079 – 9090.

465

466 Otte, T. L., Pouliot, G., Pleim, J. E., Young, J. O., Schere, K. L., Wong, D. C., Lee, P. C. S.,
467 Tsidulko, M., McQueen, J. T., Davidson, P., Mathur, R., Chuang, H. Y., DiMego, G., and
468 Seaman, N. L., 2005. Linking the Eta model with the Community Multiscale Air Quality
469 (CMAQ) modeling system to build a national air quality forecasting system, *Weather and*
470 *Forecasting*, 20, 367–384.

471

472 Pleim, J. E., 2007a. A combined local and nonlocal closure model for the atmospheric boundary
473 layer. Part I: model description and testing, *Journal of Applied Meteorology and Climate*, 46,
474 1383-1395.

475

476 Pleim, J. E., 2007b. A combined local and nonlocal closure model for the atmospheric boundary
477 layer. Part II: application and evaluation in a mesoscale meteorological model, *Journal of*
478 *Applied Meteorology and Climate*, 46, 1396–1409.

479

480 Pouliot, G., Pierce, T., van der Gon, H. D., Schapp, M., Moran, M., and Nopmongcol, U., 2011.
481 Comparing emission inventories and model-ready emission datasets between Europe and
482 North America for the AQMEII project, *Atmospheric Environment*.

483

484 Sakulyanontvittaya, T., Duhl, T., Wiedinmyer, C., Helmig, D., Matsunaga, S., Potosnak, M.,
485 Milford, J., and Guenther, A., 2008. Monoterpene and sesquiterpene emission estimates for the
486 United States, *Environmental Science and Technology*, 42, 1623-1629.

487

488 Schere, K., Flemming, J., Vautard, R., Chemel, C., Colette, A., Hogrefe, C., Bessagnet, B.,
489 Meleux, F., Mathur, R., Roselle, S., Hu, R., Sokhi, R. S., Rao, S.T., Galmarini, S., this issue.
490 Trace gas/aerosol boundary concentrations and their impacts on continental-scale AQMEII
491 modeling domains, Atmospheric Environment (this issue).
492

493 Skamarock, W. C., Klemp, J. B., Dudhia, J., Gill, D. O., Barker, D. M., Duda, M. G., Huang, X-
494 Y, Wang, W., and Powers, J. G., 2008. A description of the advanced research WRF version
495 3. NCAR Tech Note NCAR/TN 475 STR, 125 pp, [Available from UCAR Communications,
496 P.O. Box 3000, Boulder, CO 80307.].
497

498 Sofiev, M., Vankevich, R., Lotjonen, M., Prank, M., Petukhov, V., Ermakova, T., Koskinen, J.,
499 and Kokkonen, J., 2009. An operational system for the assimilation of satellite information on
500 wild-land fires for the needs of air quality modeling and forecasting. Atmospheric Chemistry
501 and Physics, 9, 6833-6847.
502

503 Tesche, T. W., Morris, R., Tonnesen, G., McNally, D., Boylan, J., and Brewer, P. 2006.
504 "CMAQ/CAMx annual 2002 performance evaluation over the eastern US." Atmospheric
505 Environment 40 (26), 4906-4919.
506

507 Vautard, R., Moran, M. D., Solazzo, E., Gilliam, R. C., Matthias, V., Bianconi, R., Chemel, C.,
508 Ferreira, J., Geyer, B., Hansen, A. B., Jericevic, A., Prank, M., Segers, A., Silver, J. D.,
509 Werhahn, J., Wolke, R., Rao, S. T., and Galmarini, S., (this issue). Evaluation of the

510 meteorological forcing used for the Air Quality Model Evaluation International Initiative
511 (AQMEII) air quality simulations, Atmospheric Environment (this issue).

512

513 Yarwood, G., Roa, S., Yocke, M., and Whitten, G., 2005. Updates to the carbon bond chemical
514 mechanism: CB05. Final report to the US EPA, RT-0400675, available at

515 <http://www.camx.com>.

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517

518 Table 1. Seasonal, domain-wide MB, ME, NMB and NME for daytime (8am – 8pm LST)

519 average O₃ for the North America (NA) AQS network and Europe (EU) AirBase network.

520

Season	MB (ppb)	NMB (%)	ME (ppb)	NME (%)
Winter (NA)	-3.5	-13.4	9.0	34.7
Winter (EU)	1.5	8.4	10.4	58.1
Spring (NA)	-1.8	-4.1	9.3	29.4
Spring (EU)	-1.8	-4.8	10.5	27.7
Summer (NA)	4.4	9.8	11.0	24.2
Summer (EU)	-0.7	-1.6	10.8	24.4
Fall (NA)	2.6	8.4	8.8	28.0
Fall (EU)	7.8	32.3	11.0	45.8

521

522

523 Table 2. Seasonal, domain-wide MB, ME, NMB and NME for daily average PM_{2.5} for the North
 524 America (NA) AQS network and Europe (EU) AirBase network.

525

Season	MB (μgm^{-3})	NMB (%)	ME (μgm^{-3})	NME (%)
Winter (NA)	3.4	30.4	6.0	52.9
Winter (EU)	-12.9	-55.0	15.8	67.3
Spring (NA)	2.0	18.9	4.5	42.2
Spring (EU)	-5.8	-36.9	8.2	52.3
Summer (NA)	-0.6	-4.6	4.4	30.5
Summer (EU)	-4.9	-37.2	6.9	52.2
Fall (NA)	4.0	36.3	5.6	51.6
Fall (EU)	-3.8	-24.2	7.7	49.1

526

527

528 Table 3. Seasonal, domain-wide MB, ME, NMB and NME for daily average PM₁₀ for the North
 529 America (NA) AQS and Europe (EU) AirBase network.

530

Season	MB (μgm^{-3})	NMB (%)	ME (μgm^{-3})	NME (%)
Winter (NA)	-11.5	-47.9	16.0	66.8
Winter (EU)	-21.5	-64.8	23.2	69.8
Spring (NA)	-14.5	-56.5	17.1	66.4
Spring (EU)	-14.0	-56.2	15.6	59.5
Summer (NA)	-16.1	-57.4	17.8	63.4
Summer (EU)	-15.1	-61.2	16.3	66.1
Fall (NA)	-11.4	-46.5	15.3	62.3
Fall (EU)	-12.2	-46.8	15.1	57.8

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532

533 **Figure Captions**

534 Fig. 1. Time series of NA daytime (8am – 8pm LST) average ozone (ppb) for AQS observed
535 (black), CMAQ using GEMS (CMAQ-GEMS) data for boundary conditions (dashed; dark grey)
536 and CMAQ using GEOS-Chem (CMAQ-GC) data for boundary conditions (dot-dashed; light
537 grey). The bottom plot shows the corresponding bias (ppb) for the CMAQ-GEMS simulation
538 (solid) and CMAQ-GC simulation (dashed).

539

540 Fig. 2. Time series of EU daytime (8am – 8pm LST) average ozone (ppb) for AirBase observed
541 (black), CMAQ using GEMS (CMAQ-GEMS) data for boundary conditions (dashed; dark grey)
542 and CMAQ using GEOS-Chem (CMAQ-GC) data for boundary conditions (dot-dashed; light
543 grey). The bottom plot shows the corresponding bias (ppb) for the CMAQ-GEMS simulation
544 (solid) and CMAQ-GC simulation (dashed).

545

546 Fig. 3. Normalized mean bias (%) for daytime (8am – 8pm LST) average ozone for the North
547 America AQS (triangles) and NAPS (circles) networks for a) winter b) spring c) summer and d)
548 fall. Warm colors indicate positive NMBs; cool colors indicate negative NMBs; grey shading
549 indicates NMBs less than $\pm 10\%$.

550

551 Fig. 4. Normalized mean bias (%) for daytime (8am – 8pm LST) average O₃ for the Europe
552 AirBase (circles), AURN (triangles) and EMEP (squares) networks for a) winter b) spring c)
553 summer and d) fall. Warm colors indicate positive NMBs; cool colors indicate negative NMBs;
554 grey shading indicates NMBs less than $\pm 10\%$.

555

556 Fig. 5. Time series of daily average $PM_{2.5}$ (μgm^{-3}) for AQS observed (solid) and CMAQ
557 estimated (dashed) for the entire U.S. The bottom time series plot shows the corresponding bias
558 (μgm^{-3}).

559

560 Fig. 6. Time series of daily average $PM_{2.5}$ (μgm^{-3}) for AirBase observed (solid) and CMAQ
561 estimated (dashed) for Europe. The bottom time series plot shows the corresponding bias ($\mu\text{g m}^{-3}$).
562 ³).

563

564 Fig. 7. Normalized mean bias (%) for $PM_{2.5}$ for the North America IMPROVE (circles), CSN
565 (triangles), NAPS (squares) and AQS (diamonds) networks for a) winter b) spring c) summer
566 and d) fall. Warm colors indicate positive NMBs; cool colors indicate negative NMBs; grey
567 shading indicates NMBs less than $\pm 10\%$.

568

569 Fig. 8. Normalized mean bias (%) for $PM_{2.5}$ for the Europe AirBase (circles), AURN (triangles),
570 and EMEP (squares) networks for a) winter b) spring c) summer and d) fall. Warm colors
571 indicate positive NMBs; cool colors indicate negative NMBs; grey shading indicates NMBs less
572 than $\pm 10\%$.

573

574 Fig. 9. Time series of daily average PM_{10} (μgm^{-3}) for AQS observed (solid) and CMAQ
575 estimated (dashed) for North America. The bottom plot shows the corresponding bias (μgm^{-3}).

576

577 Fig. 10. Time series of daily average PM_{10} (μgm^{-3}) for AirBase observed (solid) and CMAQ
578 estimated (dashed) for Europe. The bottom plot shows the corresponding bias (μgm^{-3}).

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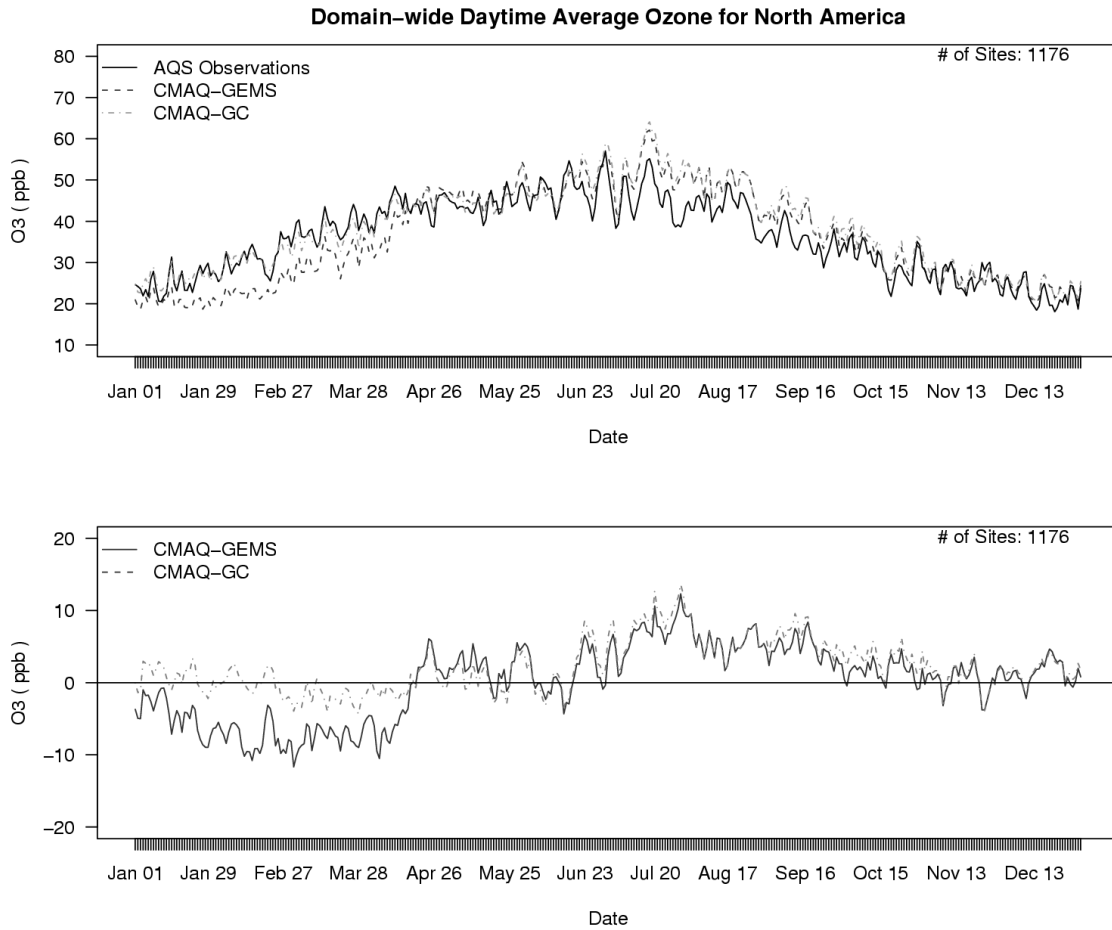
580 Fig. 11. Normalized mean bias (%) for daily average PM_{10} for the North America AQS network
581 for a) winter b) spring c) summer and d) fall. Warm colors indicate positive NMBs; cool colors
582 indicate negative NMBs; grey shading indicates NMBs less than $\pm 10\%$.

583

584 Fig. 12. Normalized mean bias (%) for daily average PM_{10} for the Europe AirBase (circles),
585 AURN (triangles) and EMEP (squares) networks for a) winter b) spring c) summer and d) fall.
586 Warm colors indicate positive NMBs; cool colors indicate negative NMBs; grey shading
587 indicates NMBs less than $\pm 10\%$.

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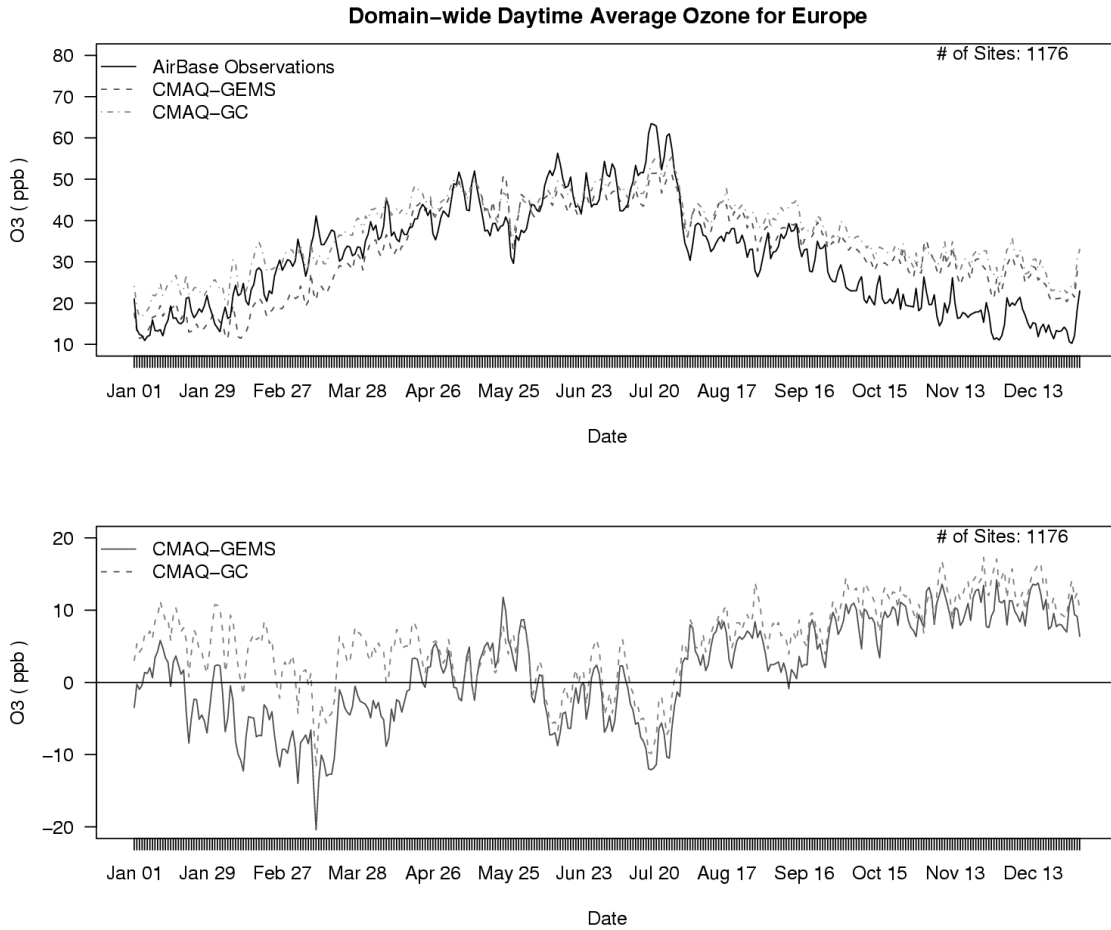
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591 Fig. 1. Time series of NA daytime (8am – 8pm LST) average ozone (ppb) for AQS observed
592 (black), CMAQ using GEMS (CMAQ-GEMS) data for boundary conditions (dashed; dark grey)
593 and CMAQ using GEOS-Chem (CMAQ-GC) data for boundary conditions (dot-dashed; light
594 grey). The bottom plot shows the corresponding bias (ppb) for the CMAQ-GEMS simulation
595 (solid) and CMAQ-GC simulation (dashed).

596



597
598 Fig. 2. Time series of EU daytime (8am – 8pm LST) average ozone (ppb) for AirBase observed
599 (black), CMAQ using GEMS (CMAQ-GEMS) data for boundary conditions (dashed; dark grey)
600 and CMAQ using GEOS-Chem (CMAQ-GC) data for boundary conditions (dot-dashed; light
601 grey). The bottom plot shows the corresponding bias (ppb) for the CMAQ-GEMS simulation
602 (solid) and CMAQ-GC simulation (dashed).
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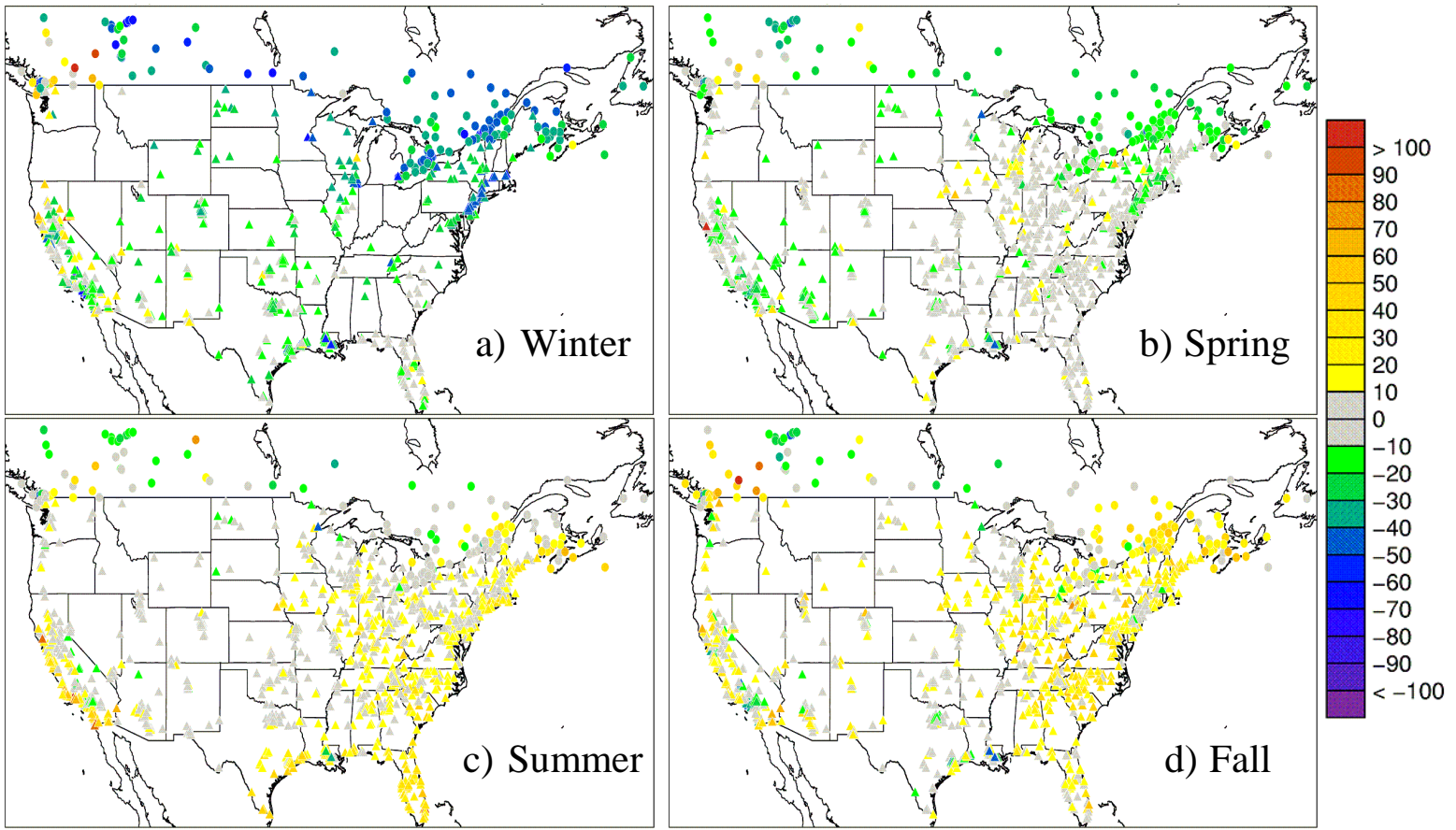


Fig. 3. Normalized mean bias (%) for daytime (8am – 8pm LST) average ozone for the North America AQS (triangles) and NAPS (circles) networks for a) winter b) spring c) summer and d) fall. Warm colors indicate positive NMBs; cool colors indicate negative NMBs; grey shading indicates NMBs less than $\pm 10\%$.

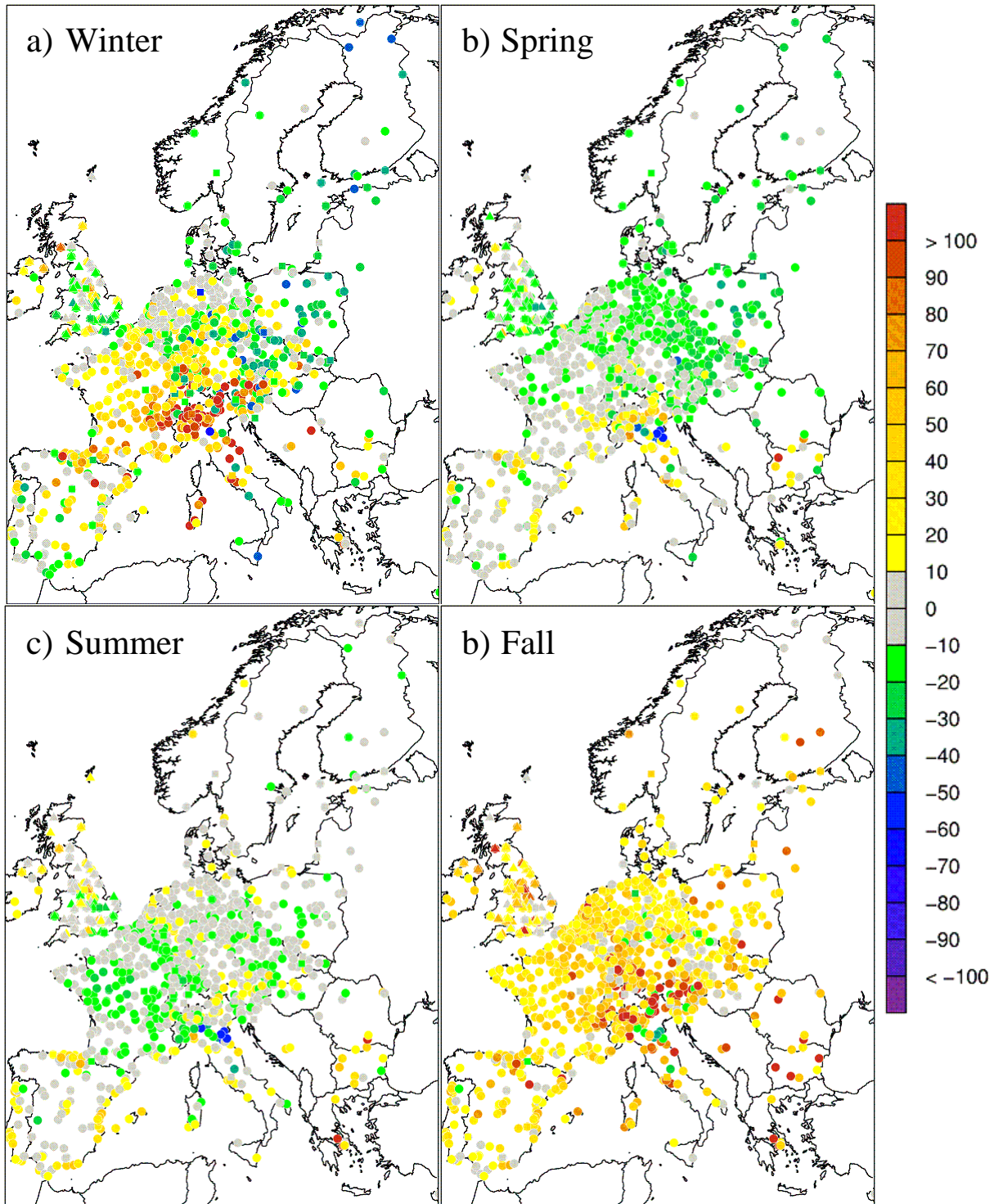
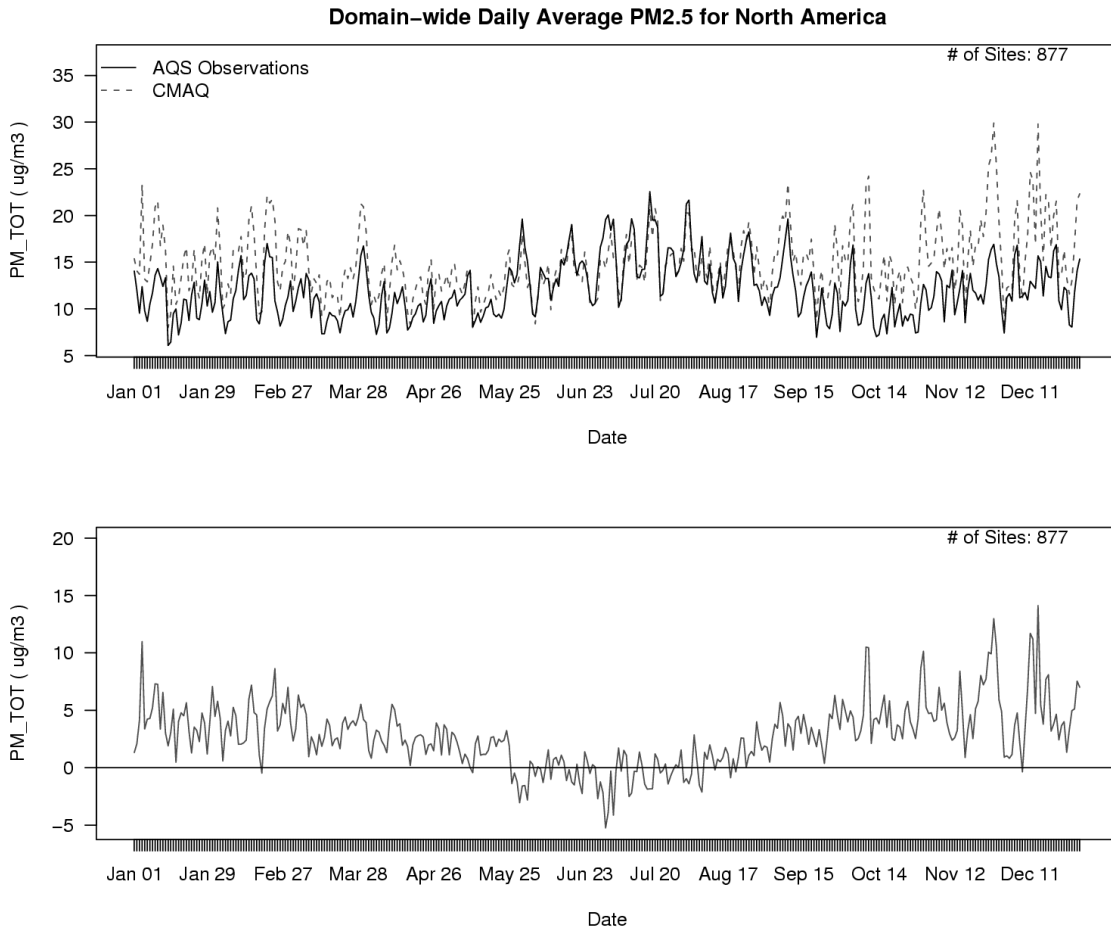


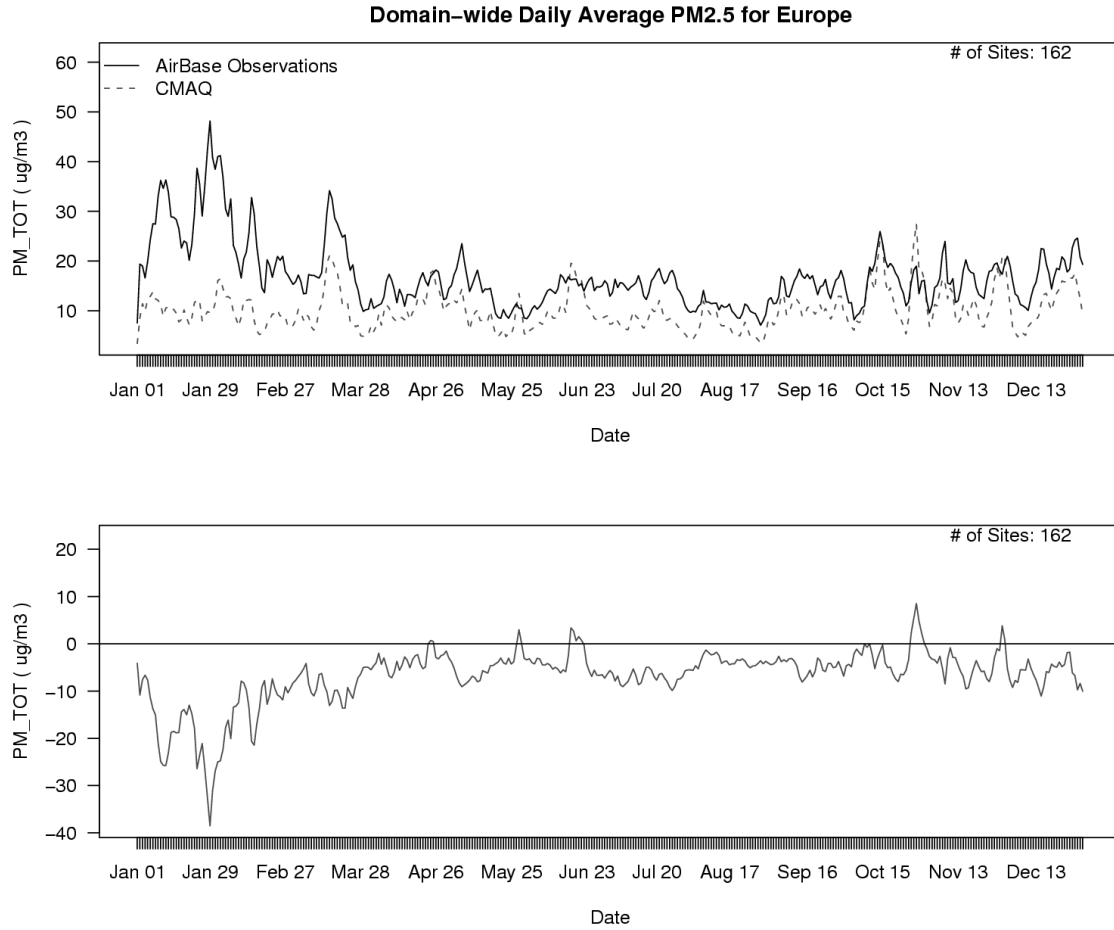
Fig. 4. Normalized mean bias (%) for daytime (8am – 8pm LST) average O₃ for the Europe AirBase (circles), AURN (triangles) and EMEP (squares) networks for a) winter b) spring c) summer and d) fall. Warm colors indicate positive NMBs; cool colors indicate negative NMBs; grey shading indicates NMBs less than $\pm 10\%$.

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Fig. 5. Time series of daily average PM_{2.5} ($\mu\text{g m}^{-3}$) for AQS observed (solid) and CMAQ estimated (dashed) for the entire U.S. The bottom time series plot shows the corresponding bias ($\mu\text{g m}^{-3}$).



613
614 Fig. 6. Time series of daily average PM_{2.5} ($\mu\text{g m}^{-3}$) for AirBase observed (solid) and CMAQ
615 estimated (dashed) for Europe. The bottom time series plot shows the corresponding bias ($\mu\text{g m}^{-3}$).
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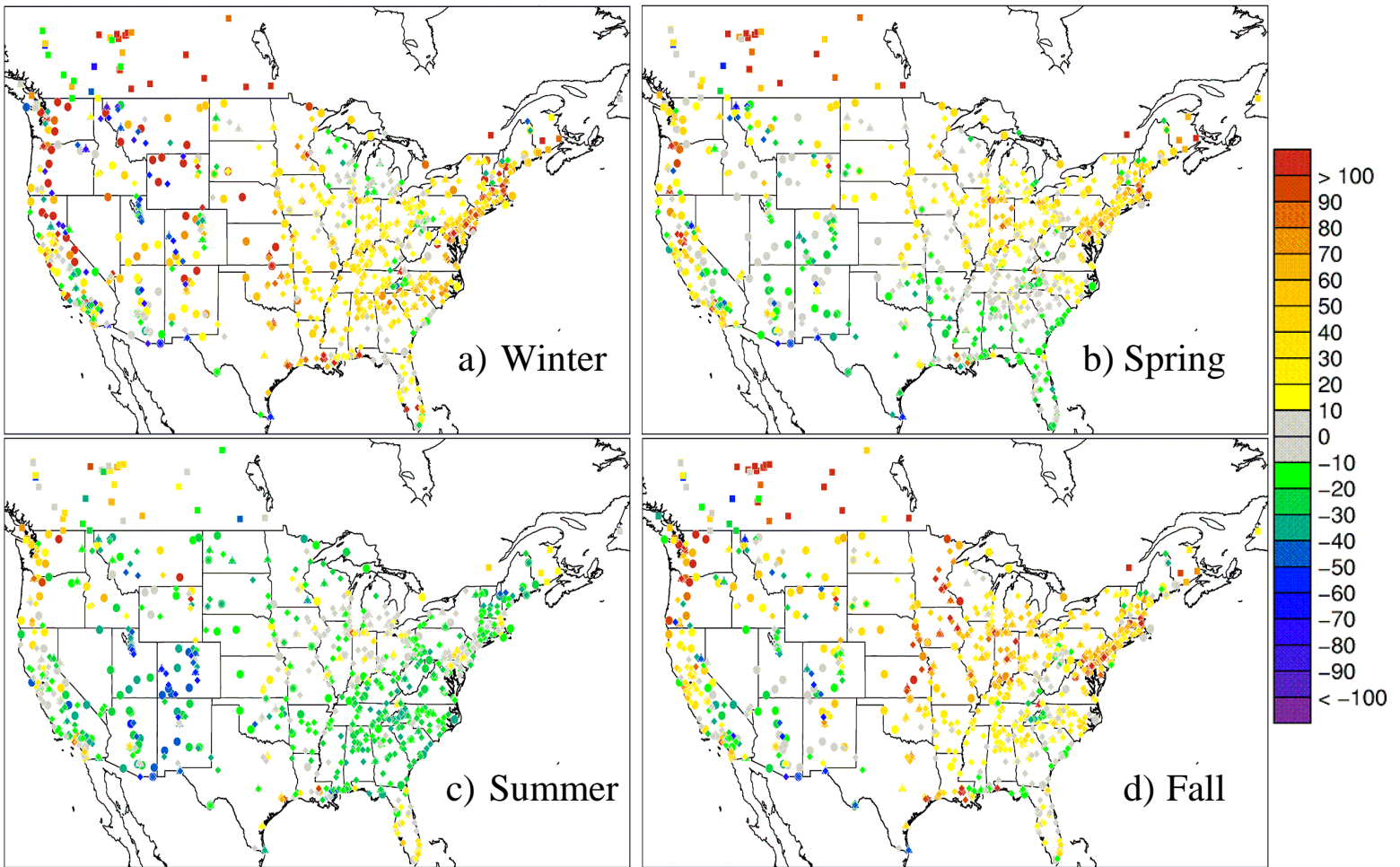


Fig. 7. Normalized mean bias (%) for PM_{2.5} for the North America IMPROVE (circles), CSN (triangles), NAPS (squares) and AQS (diamonds) networks for a) winter b) spring c) summer and d) fall. Warm colors indicate positive NMBs; cool colors indicate negative NMBs; grey shading indicates NMBs less than ±10%.

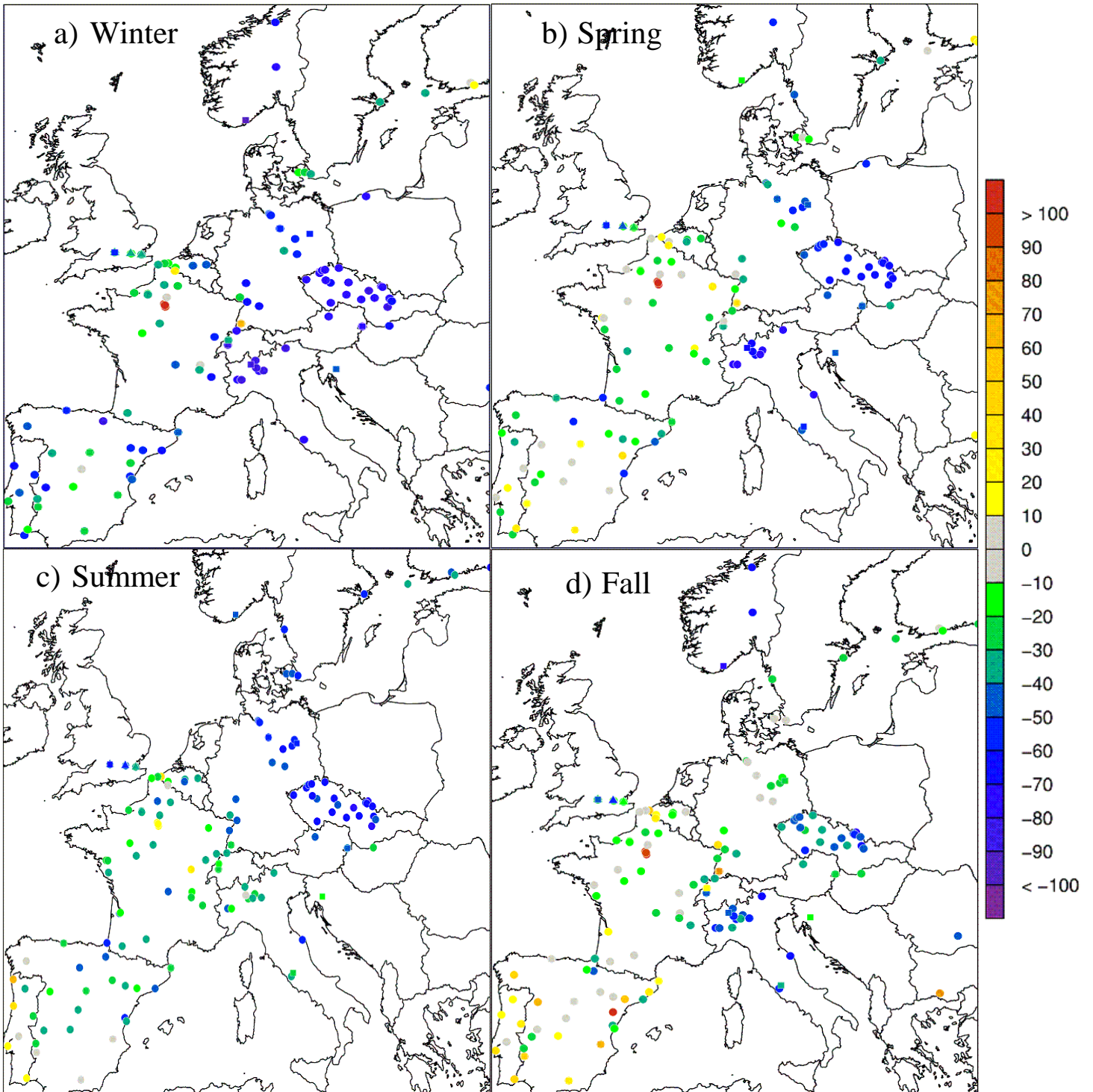
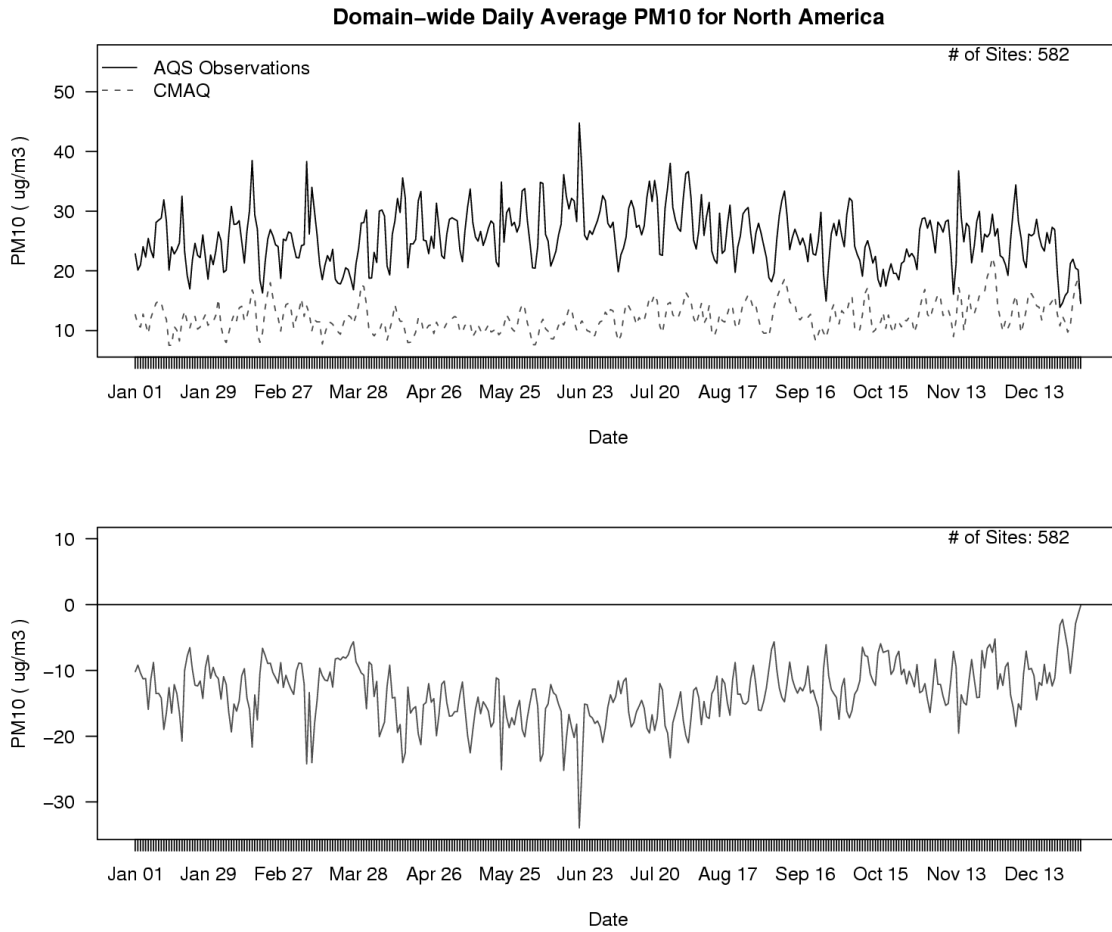
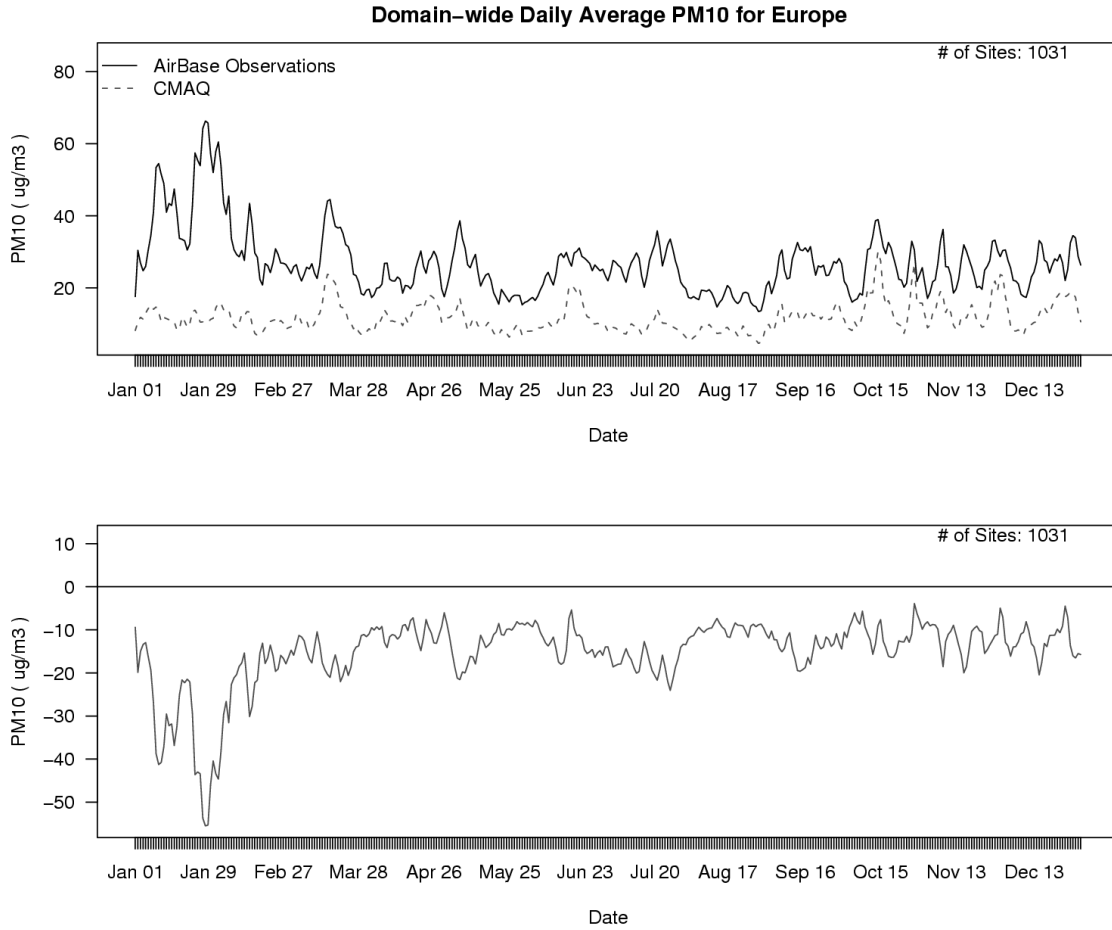


Fig. 8. Normalized mean bias (%) for PM_{2.5} for the Europe AirBase (circles), AURN (triangles), and EMEP (squares) networks for a) winter b) spring c) summer and d) fall. Warm colors indicate positive NMBs; cool colors indicate negative NMBs; grey shading indicates NMBs less than $\pm 10\%$.



623
624 Fig. 9. Time series of daily average PM₁₀ ($\mu\text{g m}^{-3}$) for AQS observed (solid) and CMAQ
625 estimated (dashed) for North America. The bottom plot shows the corresponding bias ($\mu\text{g m}^{-3}$).
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628 Fig. 10. Time series of daily average PM_{10} ($\mu g m^{-3}$) for AirBase observed (solid) and CMAQ
629 estimated (dashed) for Europe. The bottom plot shows the corresponding bias ($\mu g m^{-3}$).
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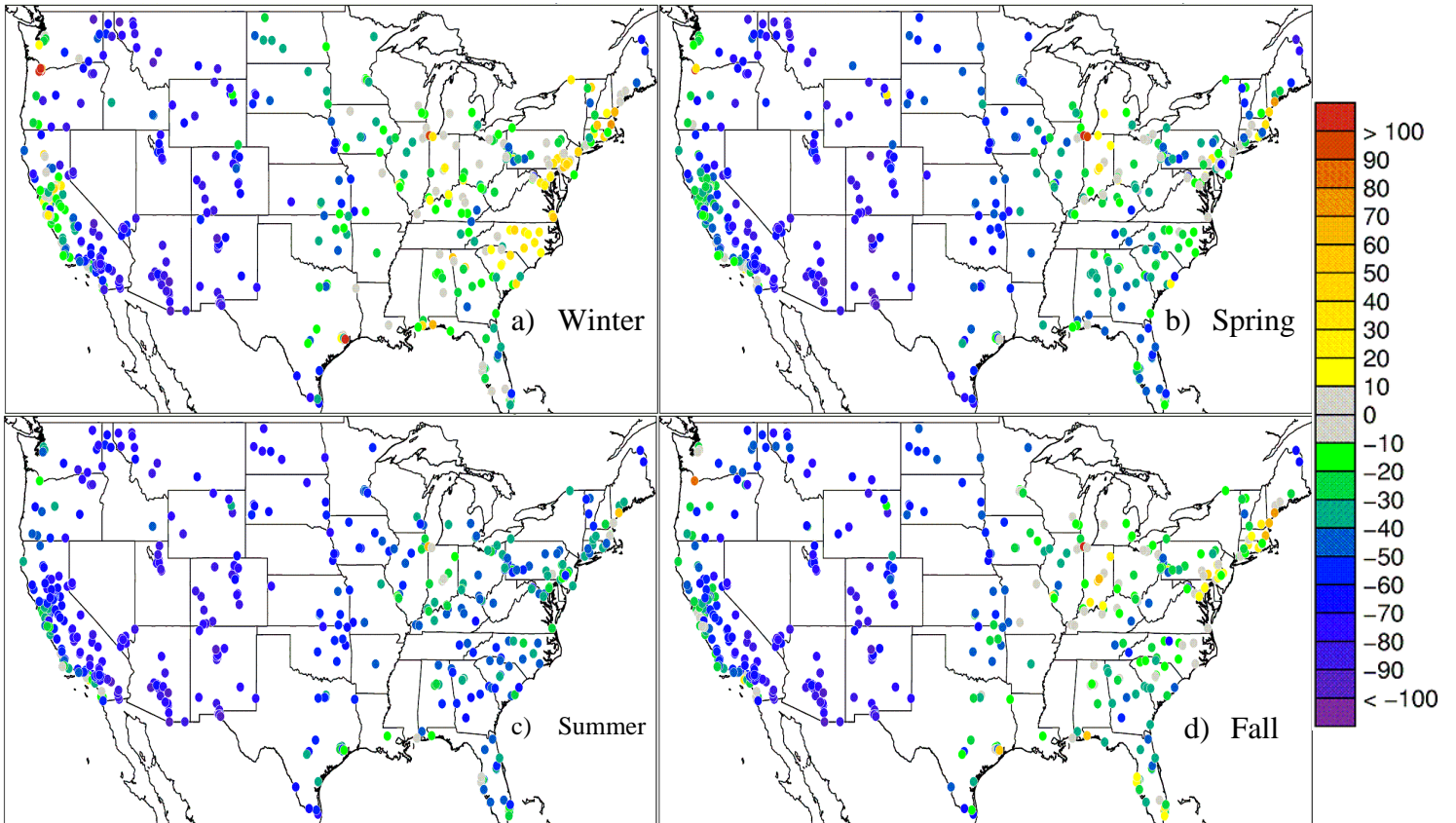


Fig. 11. Normalized mean bias (%) for daily average PM₁₀ for the North America AQS network for a) winter b) spring c) summer and d) fall. Warm colors indicate positive NMBs; cool colors indicate negative NMBs; grey shading indicates NMBs less than $\pm 10\%$.

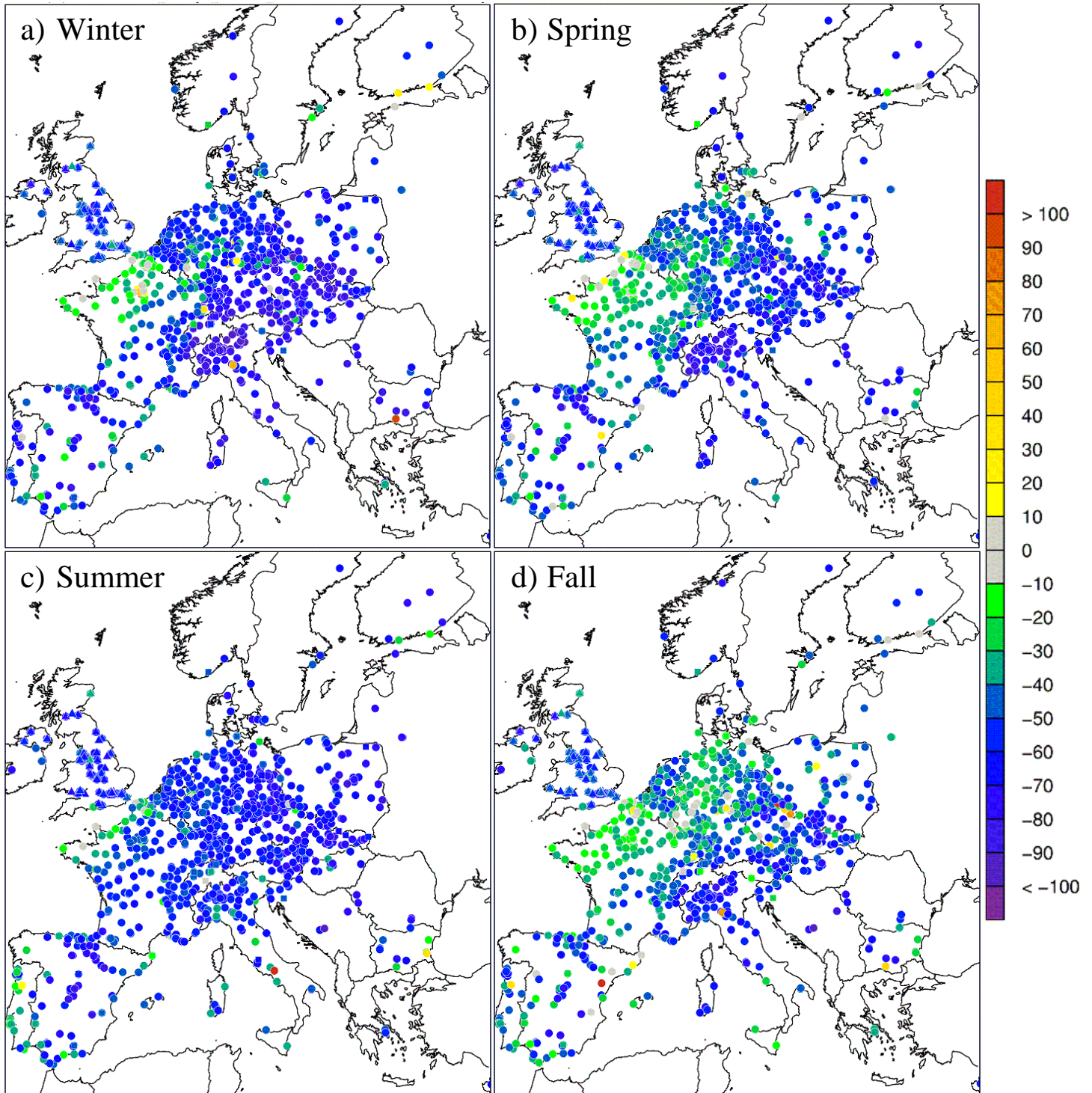


Fig. 12. Normalized mean bias (%) for daily average PM₁₀ for the Europe AirBase (circles), AURN (triangles) and EMEP (squares) networks for a) winter b) spring c) summer and d) fall. Warm colors indicate positive NMBs; cool colors indicate negative NMBs; grey shading indicates NMBs less than $\pm 10\%$.