

Model evaluation and ensemble modelling of surface-level ozone in

Europe and North America in the context of AQMEII

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40 **Abstract.** More than ten state-of-the-art regional air quality models have been applied as part of the
Air Quality Model Evaluation International Initiative (AQMEII). These models were run by twenty
42 independent groups in Europe and North America. Standardised modelling outputs, over a full year
(2006), from each group have been shared on the web distributed ENSEMBLE system, which
44 allows for statistical and ensemble analyses to be performed. The simulations of ground-level ozone
concentrations issued from the models are collectively examined in an ensemble fashion, and
46 evaluated with a large set of observations in both continents. The scale of the exercise is
unprecedented and offers a unique opportunity to investigate methodologies for generating skilful
48 ensembles of models. Despite the remarkable progress of ensemble air quality modelling over the
past decade, there still are outstanding questions regarding this technique. Among them, what is the
50 best and most beneficial way to build an ensemble of members? How to determine the optimum
size of the ensemble in order to capture data variability as well as keeping the error low? We try to
52 address these questions by looking at optimal ensemble size and quality of the members. The
analysis carried out is based on systematic minimization of the model error and it is of direct
54 relevance for diagnostic/probabilistic model evaluation. We show that the most commonly used
multi-model approach, namely the average over all available members, can be outperformed by
56 subsets of members optimally selected in terms of bias, error, and correlation. More importantly,
this result does not strictly depend on the skills of the individual members, but requires the
58 inclusion of low ranking-skill members. We apply a methodology to discern among members and to
build a skilful ensemble based on model association and data clustering, which makes no use of
60 priori knowledge of model skill. Results show that while the methodology needs further
refinements, by optimally selecting the cluster distance and association criteria, this approach can
62 be useful for model applications beyond those strictly related to model evaluation, such as air
quality forecasting.

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Keywords: AQMEII, Clustering, Error minimisation, Multi-model ensemble, Ozone

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1. Introduction

76 Regional air quality models have undergone considerable development over the past three decades,
mainly driven by the increased concern regarding the impact of air pollution on human health and
78 ecosystems (Rao et al., 2011). This is particularly true for ozone and particulate matter (e.g.,
Holloway et al., 2003; Rao et al, 2006; Jacob and Winner 2009). Regional air quality models are
80 now widely used for supporting emission control policy, test efficacy of abatement strategies, real-
time forecasts, and integrated monitoring strategies. Moreover, ozone estimates have been used in
82 assimilation schemes to provide further information on meteorological variables such as wind speed
(e.g., Eskes, 2003). The combination of outcomes by several models (regardless of their field of
84 application), in what is typically defined as *ensemble modelling*, has been proven to enhance skills
when compared against individual model realisation (e.g. Galmarini et al. 2004; Delle Monache and
86 Stull, 2003; van Loon et al., 2007). Although ensemble modelling is well established (both from the
applied and the theoretical perspectives) and routinely used in weather forecasting, it is only during
88 the last decade that a growing number of air quality modelling communities have joined their model
outputs in multi-model (MM) combinations (Galmarini et al., 2001; Carmichael et al., 2003; Rao et
90 al., 2011). The advantages of ensemble modelling vs. individual model reside in *i)* the mean (or
median) of the ensemble is, in effect, a new model which is expected to lower the error of
92 individual members due to mutual cancellation of errors, and *ii)* the spread of the ensemble
represents a measure of the variability of the model prediction (Galmarini et al., 2004; Mallet and
94 Sportisse, 2006; Vautard et al., 2006, 2009; van Loon et al., 2007). Potempski and Galmarini (2009)
also point out the scientific consensus around MM ensemble techniques as a way of extracting
96 information from many sources and synthetically assessing their variability. In particular, mean and
median offer enhanced performance, on average, compared with single model (SM) realisations
98 (Delle Monache and Stull, 2003; Galmarini et al., 2004; McKeen et al., 2005 and others).

A MM ensemble can be generated in many ways (see, e.g., Galmarini et al., 2004), for example by
100 varying some SM internal parameters, or by using different input data, or also by applying several
different models to the same scenario. This latter case is of direct relevance in the case of model
102 evaluation, and is one of the main focii of the Air Quality Model Evaluation International Initiative
(AQMEII) (Rao et al., 2011), an international project aiming at joining the knowledge and the
104 experiences of modelling groups in Europe and North America. Within AQMEII, standardised
modelling outputs have been shared on the web distributed ENSEMBLE system, which allows
106 statistical and ensemble analyses to be performed (Bianconi et al., 2004; Galmarini et al., 2012). A

108 common exercise was launched for modelling communities to use their regional air quality models
110 to retrospectively simulate the entire year 2006 for the continents of Europe and North America.
Outputs from numerous regional air quality models have been submitted in the form of hourly
112 average concentrations on a grid of points and at specific locations, allowing direct comparison with
air quality measurements collected from monitoring networks (details are given in Rao et al., 2011).
114 The focus of the AQMEII project is to test the ability of models to retrospectively simulate and not
forecasting air quality. This type of evaluation, with large temporal and spatial scales, is essential to
determine model performance and assessing model deficiencies (Dennis et al., 2010; Rao et al.,
2011).

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118 In this study, we analyse ozone concentration data produced by over ten state-of-the-art regional air
quality models, run with different configuration and versions by twenty independent groups from
both continents. Model's data have been made available along with observational data, on the
120 ENSEMBLE system. We examine the ability of the ensemble mean and median to reduce the error
and bias of SM outputs, and draw some deductions about the *size* of the ensemble and its *quality*.
122 We quantify the level of repetition brought by individual model to the ensemble by applying a
clustering analysis to examine whether the improvement of ensemble of models mean and median
124 in reducing the error is merely due to the increased ensemble size, or if information carried by each
model contributes to the MM superiority.

126
128 We remark that the aim of this study is to study the statistical properties of ensemble of models in
relation to individual model realisation, for a range of cases. Each model has imperfections, and we
try not to speculate as to why the bias of each individual model is generated, which is the topic of a
130 number of detailed papers dealing with sensitivity analyses and evaluation. The ensemble represents
a collective model perspective in which errors and biases result from a collective misrepresentation
132 of the system (McKeen et al 2005).

134 The paper is organised as follows. In Section 2 the air quality models and network data are
presented, and the methodology applied is highlighted, with the strategy for the MM ensemble
136 described in Section 3. Section 4 is devoted to the analysis of combinations of ensemble members
that minimise the error and bias, and a clustering technique for selecting and generating an
138 ensemble of independent members is then outlined in Section 5. Conclusions are drawn in Section
6. Finally, Appendix A briefly summarises the definition of the statistical indicators adopted.

140

2. Models and data

142 2.1 Experiment set up

144 In order to carry out a comprehensive evaluation of regional-scale air quality models, simulations
146 are compared to observations for the year of 2006, and various modelling groups provided hourly
concentration of ozone and other compounds. Surface concentrations were then interpolated to the
monitoring locations to provide model evaluation. The focus of this study is on ozone, whilst a
companion study is devoted to the particulate matter (Solazzo et al., 2012).

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2.2 Participating models

150 Table 1 summarises the meteorological and air quality models participating to the AQMEII
intercomparison exercise , providing ozone concentrations at receptor sites for the European (EU)
152 and North American (NA). In some cases, the same model is used with a different configuration by
different research groups (or in some cases the same group). In total, eleven groups for EU and
154 seven for NA participated to analyse ozone. No a-priori screening on the worst performing model
has been performed on the participating members (considered that the models have gone through, at
156 least, operational model evaluation as defined in Dennis et al. (2010) in the past); hence, all data
providers are participating in this study.

158

AQMEII participants were provided with a reference simulation for the year 2006, generated with
160 the WRF v3.1 (Skamarock et al., 2008) and the MM5 (Dudhia, 1993) models, for NA and EU
respectively, which were applied by the majority of groups. Several other groups conducted
162 separate simulations with other meteorological drivers (Table 1). Skills and shortcoming of the
meteorological models within AQMEII are described by Vautard et al. (2012).

164

The models participating to the exercise, listed below, have been extensively documented in the
166 scientific literature (including sensitivity tests and evaluation studies):

- CMAQ (developed by the U.S. Environmental Protection Agency)
- 168 - CAMx (ENVIRON, 2010)
- CHIMERE (Schmidt et al., 2001; Bessagnet et al., 2004)
- 170 - MUSCAT (Wolke et al., 2004; Renner and Wolke, 2010)
- DEHM (Brandt et al., 2007)
- 172 - POLYPHEMUS (Mallet et al. 2007; Sartelet et al. 2012)
- EUROS (Schaap et al., 2008)
- 174 - SILAM (Sofiev et al., 2006)

- AURAMS (Gong et al., 2006; Smyth et al., 2009)
- EMEP (Simpson et al., 2003; Jeričević et al., 2010).

178 The combination of meteorological and transport models varies for each group (with the only
exception of WRF with WRF-Chem in Europe), thus allowing analysis of diversified modelling
180 data, a diversity which is necessary to sample the spectrum of uncertainty within an ensemble.

182 Emissions and chemical boundary conditions used by the various AQMEII groups are summarised
in Table 1. AQMEII provided a set of emissions (referred to as “standard”) for each continent,
184 focusing on the evaluation of the air quality and meteorological models. The EU standard emissions
were prepared by TNO, which provided a gridded emission database for the year 2005 and 2006.
186 This dataset has been widely used, and partly developed in the framework of the European MACC
project (<http://www.gmes-atmosphere.eu/>) and is the update of an earlier TNO emission database
188 prepared for the GEMS project (<http://gems.ecmwf.int>). This inventory does not include biogenic
emissions, thus models adopted different inventories to supply for this, as summarised in Table 1.
190 The standard emissions for NA are based on the U.S. National Emissions Inventory 2005, Canadian
national inventory 2006, Mexican BRAVO inventory 1999, biogenics from BEISv3.14, fire
192 emissions daily estimates from HMS fire detection and SMARTFIRE system (year 2006), point
sources (EGUs) from Continuous Emissions Monitoring data for the year 2006. Full details are
194 given in Pouliot et al. (2012), where the standard inventories for EU and NA are described and
compared. The standard emission inventories were used by the vast majority of the participating
196 AQMEII groups (Table 1). Data generated with other emission inventories have also been
submitted, and will provide useful comparisons in interpreting the results of model estimated ozone
198 mixing ratios.

200 AQMEII also made available chemical concentrations at boundaries to modelling groups, as
obtained from the GEMS re-analysis data provided by European Centre for Medium-range Weather
202 Forecast (see Schere et al. (2012) for details). Different boundary conditions for ozone used by
other AQMEII models were based on satellite measurements assimilated within the IFS system.
204 LMDZ-INCA, which couples the general circulation model Laboratoire de Meteorologie
Dynamique and the Interaction with Chemistry and Aerosol model (Hauglustaine et al., 2004) was
206 used for CAMx and CHIMERE in one set of simulations (NA simulations), with another
CHIMERE simulation using standard boundary conditions (Table 1).

208

2.3 *Observational data for ozone*

210 The European and North-American continental area have been divided into four subregions (EU1 to
EU4 and NA1 to NA4). Figure 1 displays the subregions for both continents, along with the spatial
212 coverage of ozone receptors. Overlaid are the contours of “standard” anthropogenic NO_x emissions
averaged over the summer months of June-July-August (JJA) of 2006. Only rural receptors below
214 an altitude of 1000 m have been examined (dots in Figs. 1), with, at least, 75% data availability over
the 2006. The choice of analysing only rural receptors is dictated by the need to provide comparison
216 with spatial scales consistent with models resolution (see e.g., Vautard et al., 2009). Moreover,
ozone measured by monitoring stations in urban areas is sensitive to reaction with NO_x, which
218 might reduce ozone production.

220 The selection of the subregions is based on emissions and altitudinal aspects, as well as practical
constrains (data coverage, computational time). Extension of EU subregions are similar to those of
222 two other AQMEII analyses of meteorological forcing (Vautard et al., 2012) and particulate matter
(Solazzo et al., 2012). Subregion EU1, the British Isles, France and North Spain, was selected for its
224 mid-latitude, mixed maritime-continental climate and large conurbations (London, Paris).
Subregion EU2, Central Europe, has a continental climate with marked seasonality, many large
226 cities, and large emissions areas. Subregion EU3, the Po Valley up to the Alps area of Italy and
south-eastern France has a mixed climate, poor air quality, and is influenced by the Alpine barrier.
228 The Southern European domain covers the Mediterranean area (south Italy, east coast of Spain and
Greece), with typical Mediterranean climate and large cities (Barcelona, Rome). The number of
230 rural receptors for EU subregions is of 201, 225, 77, and 140 and (domain 1 to 4) respectively. For
NA, the number of rural receptors in each subregion is between 134 and 150. NA subregions are
232 broadly derived from previous studies (e.g., Eder et al., 1993), also considering the NO_x emission
intensity, with the additional constraint of a uniform number of receptors. The eastern part of US
234 (Domain 1), with marked high emissions along the coast of California, and milder emission towards
the continent, has high solar radiation, low relative humidity, large cities with poor air quality (Los
236 Angeles). NA2 is to the east of the Rocky Mountains and is characterised by a hot, humid climate,
with large cities with poor air quality (Houston, Dallas). Subregion NA3, northeastern NA
238 including parts of Canada, has a marked seasonal cycle, three of the North American Great Lakes,
among the highest emissions areas in NA, and large cities (New York City, Philadelphia, Toronto,
240 Montreal). Finally, the south-east US (NA4), with high emissions and strong solar radiation.

242 Ozone data for EU were derived from hourly data collected by the AirBase and EMEP (European
Monitoring and Evaluation Programme, <http://www.emep.int/>) networks, for a total of 1563
244 stations, of which over 1400 have a percentage of data validity higher than 80%. Ozone data for NA
were prepared from hourly data collected by the AIRS (Aerometric Information Retrieval Systems,
246 <http://www.epa.gov/air/data/aqsdb.html>) and CASTNet (Clean Air Status and Trends Network,
<http://java.epa.gov/castnet/>) networks in USA and the NAPS (National Air Pollution Surveillance,
248 <http://www.ec.gc.ca/rnspa-naps/>) network in Canada. A total of 1445 stations are available, more
than half with a percentage of data validity higher than 80%.

250

Furthermore, AQMEII participants also provided vertical profiles of ozone at specific airport
252 stations in NA and EU (Galmarini et al., 2012). Measurements for vertical profiles of ozone from
the MOZAIC campaign (<http://mozaic.aero.obs-mip.fr/web/>) were made available on ENSEMBLE
254 system to provide model evaluation.

256 **3. Single model and multi-model ensemble: operational evaluation and general statistics**

3.1. Operational and ensemble statistics for the continental-wide domains

258 van Loon et al. (2007) showed that the ensemble mean ozone daily cycle over Europe, obtained by
averaging over all monitoring stations for the entire year of 2001, agrees almost perfectly with the
260 observations, and better than any member of the ensemble. This result provides substantial evidence
of the superiority of the MM approach versus the SM approach. Such a result, while encouraging,
262 poses some additional questions, as for example on the role of repeated averaging (in time and
space) in smoothing out peaks and hide data variability, and also on the possibility to prove the
264 superior MM skills for any ensemble combinations. Galmarini and Potempski (2004) show that, for
the ETEX-1 case study, the MM did not show significant superior skill (and anyway poorer than an
266 air quality case due to the instantaneous character of the prediction), and thus concluded that, in
absence of a method for discriminating between members, the MM improved performance might be
268 just coincidental and depends on the ‘lucky shot’ of having the right collection of models around the
measured data. No methodology exists to pre-select nor to discriminate ensemble members, and
270 therefore the result is purely dependant on opportunity.

272 Let us consider the plots in Fig. 2, where ozone concentrations predicted by AQMEII models for
EU (Fig 2a) and NA (Fig 2b) are shown, continent-wide averaged for the full year of 2006. Whisker
274 representation has been adopted to describe the frequency distribution, where the rectangle
identifies the interquartile range (25th to 75th percentile), the square is the mean, the continuous

276 horizontal line the median, crosses are the 1st and 99th percentiles, and the whiskers extend between
minimum and maximum value. Measurements, mean and median are also shown. The top row
278 displays the distribution of hourly values (i.e., each bar is the distribution over 8760 hourly values),
the middle row is the daily average distribution (over 365 values), and the bottom row is the mean
280 diurnal range (each bar reflects the distribution over 24 values), in which same hours of each day of
the year are averaged. Depending on the averaging period, ozone concentrations are reduced by a
282 factor of two, for both continents. This translates into a dramatic reduction of the spread (min and
max values are within the inter-quantile ranges for the diurnal cycle) and a clustering of the diurnal
284 time series which results in enhanced statistical agreement. (To maintain anonymity, each
participating model has been assigned a random model number, Mod 1 to 11 for EU, and Mod 12 to
286 18 for NA, which do not correspond to the order of models in Table 1). Thus, averaging over
extended areas (continent) and periods (year) has a dramatic effect in reducing the spread of the
288 data.

290 The ability of the ensemble to sample measurements uncertainty in both continents is analysed by
means of the rank histograms in Figs. 3, which are a measure of the ensemble reliability (Talagrand
292 et al., 1998; Joliffe and Stephenson, 2003). The rank histogram is a widely adopted diagnostic tool
to evaluate the spread an ensemble of members. In a rank histogram, the population of the k -th rank
294 is the fraction of time observations falls between the sorted member $k-1$ and k . Ideally, the
frequency for each bin should be the same, meaning that the ozone estimate from each ensemble
296 member is as probable as any other member, and observations have an equal probability to belong
to any bin (Hamill, 2000). In such case, in fact, the observation and the ensemble members are
298 derived from the same probability distribution, and the probability of the observation falling into a
particular bin is the same for all bins. In Fig. 3, unbiased hourly ozone data from the full year,
300 continents-wide averaged are used. For EU (Fig 3a), bins population is rather uniform for the first
ten bins (between 6 and 11%), with bins 11 and 12 having a frequency of ~18% each, indicating a
302 difficulty of the ensemble mean to simulate high hourly concentration, which is a bias of the
ensemble mean (underestimation). The ranked histogram for NA (Fig 3b) shows the intermediate
304 bins more populated than the side bins, indicating the possible presence of outlying members.

306 It is not straightforward to assess whether the deviation from flatness in both cases (EU and NA) is
due to chance (ensemble members and observations derived from the same distribution) or if there
308 is a compensation effect over such large domains and long time scale. These aspects will be further
detailed in Section 3.4.

310

3.2 Subregional SM and MM ensemble analyses

312 Regional air quality models are often used on limited spatial and temporal scales (months or season
over a few hundreds of kilometres wide, Boynard et al., 2011; Hogrefe et al., 2011; Bloomer et al.,
314 2009; Camalier et al., 2007), for which mutual cancelation of models' errors might not be as
effective as in the case of continental and yearly scales, as discussed for the results of Fig. 2. The
316 analyses presented in this study focus on the ozone spatial variability of ozone concentration
statistics in four distinct subregions of the continental domains of Fig. 1, examining the temporal
318 variability for the summer months JJA, i.e., when the ozone mixing ratios are at maximum and are
of major concern for the public health. Analysis and evaluation of single models performance over
320 the whole year are presented elsewhere.

322 Subregional ozone diurnal cycles are shown in Fig. 4a (EU) and Fig. 4b (NA), including ensemble
mean and median (hourly data have been used for the analysis). Examining the observational data
324 trends, there is an ample intra-continental variability of the ozone maxima, with the North Italian
and Mediterranean regions (EU3 and EU4) reaching 60 ppb and over, and peaks of ~45-50 ppb
326 occur in the other EU subregions. For EU1,2,3 the maximum occurs at LT 17, while it is detected
two hours earlier in EU4, due to the higher insulation. Minimum values, between LT 7 and 8, range
328 between 20 and 30 ppb, with the Mediterranean area having the highest minimum due to the
relative abundance of biogenic emissions (see e.g., Sartelet et al., 2012). Maximum values for NA
330 are of the same general magnitude as EU, between 45 and 55 ppb for subregions 1, 2 and 4 and only
reaching ~35 ppb for NA3 in the North-East region, and occur at LT 16. Minimum values, at LT 07,
332 range between 20 to 25 ppb for NA1,2,3 and less than 20 ppb for NA4. This latter (South-East
region) exhibits a steep rise of ozone mixing ratios that is indicative of strong daytime
334 photochemical activity in this region.

336 The majority of models (thin lines in Figs. 4) exhibit highly regional-dependant behaviours,
although some common patterns can be detected. The Muscat model is severely biased high at all
338 regions (probably due to ...), whilst the majority of the other models have a tendency to
underestimate (even significantly) the peak concentration (and the time of the peak), as well as to
340 under predict the night hour mixing level (especially for EU2, continental EU), probably due to
high daily temperature gradient in this region. The CMAQ model, as emerge from previous works
342 (Herwehe et al., 2011; Smyth et al., 2009), has the tendency of overestimating ozone concentration
at night, due to difficulties in dealing with stable conditions. Another source of performance

344 variability for EU is identified in the different biogenic emissions adopted by each model. This
hypothesis is confirmed by the performance of the Chimere model with MEGAN biogenic emission
346 (and standard EMEP anthropogenic), which is the best scoring model for all EU subregions. The
DEHM model has the highest bias (low) for EU4 and especially during night time. Brandt et al.
348 (2012) suggest DEHM underestimation of summer months ozone in Europe to be due to isoprene
emission being too low. Sartelet et al. (2012), in evaluating the impact of biogenic emission in the
350 context of AQMEII also point out the impact of biogenic emission for estimating ozone. In
particular they found that the best model performance for ozone is obtained when using the
352 MEGAN biogenic inventory.

354 Model's results for NA regions exhibit a lower spread throughout the cycle (Fig. 4b), with the
exception of a clear outlying model for NA1,2,3, which is consistently biased low, especially at
356 night. However, the night over prediction is, to various degrees, a common feature to the majority
of models, indicating the difficulties in dealing with stable conditions, despite the variety of vertical
358 mixing schemes implemented by the models. The case of the south-east area (NA4), with consistent
model overestimation, indicates that...

360
Ensemble mean and median generally underestimate the amplitude of the diurnal ozone cycle in
362 Europe, despite one largely biased high outlying model. By contrast, mean and median accurately
mimic the ozone amplitude for NA1,2,3 (whilst largely overestimate for NA4) thanks to the mutual
364 compensation of the biased low outlier and the general tendency of the other member to over
predict. It should be observed that the mean and the median curve overlapping is a consequence of
366 the repeated data averaging (spatially and temporally) that has smoothed out the peaks of the
distribution, as previously shown in Fig 2.

368
Figures 5 quantify the error statistics for EU (Fig 5a) and NA (Fig 5b), in form of a "soccer plot"
370 (Appel et al., 2011). The NMSE vs NMB (see Appendix A for definition) is reported for each
individual model, together with the ensemble mean and median, for the four subregions (numbered
372 1 to 4). Models data falling within the dotted lines indicate compliance with the performance
criteria set by Russel and Dennis (2000) for ozone (bias within $\pm 15\%$ and error within $\pm 30\%$).
374 The majority of points lie in the left region of the soccer area, indicating underprediction for EU,
with the exception of Mod1, substantially overestimating the mixing ratio for all subregions. Model
376 results for the NA are well within the 15% box (mainly overestimation), with the exception of NA4,
where three models show overestimation between 15 and 20%. Points are mainly grouped by model

378 for EU and by region for NA, suggesting a stronger dependence on local conditions for the latter
continent. The ensemble mean and median (approximately identical for NA, as already noted for the
380 diurnal cycle) are within the 15% area, indicating compliance of performance criteria for both
continents.

382

3.3 Sources of model bias

384 Vautard et al. (2012) have shown that there is a clear model overestimation of the 10 m wind speed
across the European continent (up to a factor of two), especially in the EU2 (continental Europe).
386 Solazzo et al. (2012) have also shown that, in the case of particulate matter, there is a significant
correlation between the bias of wind speed and that of particulate concentration (also
388 underestimated), meaning that the poor model prediction for wind speed are partially responsible
for the bias of primary pollutants. Although ozone is not emitted at ground, we can infer that the
390 positive bias for wind has a diluting effect on the primary species (NO_x, VOC, etc), help explaining
– partially – the underestimation of ozone across Europe.

392

From investigating Fig. 5, there seems to exist, especially for NA, a direct correlation between bias
394 and error, suggesting that most of the uncertainty is due to biases. Schere et al. (2012); Appel et al.,
(2012); Nopmongcol et al. (2012) have shown that such bias is likely introduced by the standard
396 GEMS BC for ozone, which are overestimated, especially in winter. NA-Mod13 uses the same
chemistry transport model as NA-Mod17, but uses boundary conditions from a global model, which
398 provided ozone mixing ratios along the boundary that are significantly lower than the standard
boundary data, a difference which propagates across the entire NA domain (Schere et al., 2012).
400 During summer months, with O₃ largely produced by atmospheric chemistry, the influence of the
boundary conditions is reduced, but not disappeared. To quantify the bias due to boundary
402 conditions for ozone over the NA continent, in Fig. 6 models predictions of vertical ozone
concentration are compared against measurements collected along flight paths during MOZAIC.
404 The focus is put on the west coast of NA, where the oceanic air masses moved by the westerly
winds first encounter the land. Concentration of pollutants aloft in this coastal region should also be
406 less influenced by the ground emissions and they should then be comparable with the boundary
conditions for ozone. Each MOZAIC flight gives a set of measurements along a trajectory, from
408 which the closest values to the model predictions are used for comparison. Both, MOZAIC
measurements and model predictions are then averaged in time to obtain an average value at each
410 model vertical level. Sixteen flights made over Portland airport for the month of August 2006 have
been used (lon/lat: -122.75W/45.5N). For comparison, we use outcomes from three models,

412 adopting GEMS boundary conditions. We are not interested in the inter-model comparison here, but
rather in the distance between the MOZAIC measurements and the BCs, and to what extent this
414 translates into bias. The black thick line is the observation (with the standard deviation), the grey
thick line is the GEMS BCs at the node 120.833W/47.00E, and the thin grey lines are the models.
416 For completeness, we have reported the ground level concentration of ozone over six ground base
regional receptors of the AIRS network within the MOZAIC domain defined for Portland (the
418 projection to the ground of all flight trajectories up to 8500 m, see Galmarini et al. (2012) for
details). We notice that the independent ground level and the MOZAIC measurements are in perfect
420 agreement, supporting the validity of the observations. In the boundary layer, below 1000 m, there
is a difference between GEMS and MOZAIC of ~ 13 ppb, leading to a positive model bias (models
422 to receptors). The bias turns into negative above that height, and thus we might expect a model
underestimation above the boundary layer, but this is not investigated here.

424

3.4. Reliability of the Multi-model Ensemble

426 Consistently biased rank histograms, displayed for all subregions of the two continents, exhibit a
sloped shape, as in Figs 7a (EU) and 7b (NA). Analysis is based on hourly data for the period JJA.
428 EU subregions 1,2, have histograms significantly deviating from flatness, with most populated bins
towards the end of the ranks (model underprediction). EU4 shows empty initial and final bins,
430 indicating an excess of variability. The histogram for the entire EU domain, the biased trends for
EU1,2 are compensated by those of EU3 and 4, resulting in a flat rank histogram, as observed for
432 the entire year (Fig. 3a). As discussed at the beginning of section 3.2, by looking at long time and
large spatial scales, the seasonal and intra-continental variability is hidden by the averaging and
434 mutual compensation. Strong biases are also observed for the NA subregions (Fig 7b), with over-
prediction (left bins most populated) for all sub regions, as seen in Fig. 5b. The spread also suffers
436 from deficiencies of the ensemble in all cases, with excess of spread for NA1 (middle ranks more
frequent) or insufficient spread, such as in NA2,3,4 (side-bins more populated). This latter case is
438 typically due to not having captured all sources of error properly (Vautard et al., 2009). This might
be due to many members of the ensemble using the same meteorological drivers and/or emissions.
440 Comparing the histograms of Fig. 7b for the entire NA domain for JJA and that of Fig. 3b for the
entire NA domain for the full year highlights that for the full year the bins were more uniform, with
442 a tendency to a “bell” shape, whereas for JJA the distribution is drastically biased and bin
populations uneven. This is due to large underestimation in the winter months by models adopting
444 the GEMS boundary conditions for ozone (Appel et al., 2012) which compensates for the
overestimation in the summer.

446

448 **4. Multimodel analysis: selected vs. unselected model ensembles**

4.1 Ensemble size

450 In this section we evaluate whether an ensemble built with a subset of models can outperform the
ensemble mean of all available members, as anticipated by the theoretical analysis of Potempski
452 and Galmarini (2009). The analysis is done for the subregions of EU and NA separately, using
hourly ozone data for the period JJA.

454

Let us consider the distribution of some statistical skills (RMSE, PCC, MB, MGE, defined in the
456 Appendix A) of the mean of all possible combinations of available members, n (n is 11 for EU and
7 for NA). The number of combinations of any k members is $\binom{n}{k}_{k=2,\dots,n-1}$. For example there are as
458 many as 462 combinations of 5 models for EU, and 35 combinations of 3 models for NA. Results
are presented in Figs 8a (EU) and 8b (NA). The first column is the RMSE, the middle column is the
460 MB, and the last is PCC, as function of the number of members of the MM ensemble.

462 The continuous lines on each plot are the mean and median of the distribution of any k -model
combinations. Mean and median have similar behaviour decaying as $O(1/k)$, (Potempski and
464 Galmarini, 2009). These curves move toward more skilful model combinations as the number of
members k increases, which confirms the common practice to average over all available members to
466 obtain enhanced performance with respect to the mean of the available members. For MB, the mean
trend is flat, because of the quasi-symmetric error fluctuations about the mean value. Mean RMSE
468 curves decrease steeply from two to four models (all subregions, except NA4). A further striking
feature is that the best SM has similar (EU1,2, NA1,3) or lower (EU3,4, NA2,4) RMSE than the
470 ensemble mean with all members. This is most probably due to having included members with large
variances in the ensemble (Potempski and Galmarini, 2009).

472

Analysis of mean RMSE for EU subregions (Fig 8a), for which a large set of members is available,
474 shows a plateau for $k > 5$. This would indicate that there is no advantage, on average, to combine
more than six members, as the benefit in minimizing the RMSE is negligible. Investigating the
476 maximum of RMSE, it results $\max_k RMSE(k) > \max_k RMSE(k+1)$. The mean of ensembles with a
large number of members have the properties of reducing the maximum error. For example, RMSE
478 of EU3 has a large error span, between 2.5 and 15 ppb for $k = 2$, which reduces between 4 and 7

ppb for $k = 10$ (Fig 8a). A similar trend is replicated for PCC (all subregions), with a monotonic
480 improvement of minimum PCC values when increasing k .

482 Values of minimum RMSE (lower bound) exhibit a more complex behaviour. A minimum, among
all combinations, systematically emerges for ensembles with number of members $k < n$. Similarly, a
484 maximum of PCC is achieved by combinations of a subset of members. This result tells that
ensembles of a few members outperform –systematically– the whole ensemble; further to that,
486 adding new members to such an optimal ensemble (thus moving towards higher value of k in Figs.
8) deteriorates the quality of the ensemble, as the error increases.

488

4.2 Ensemble combinations of minimum RMSE and MB

490 In Table 2, the MM combination of minimum RMSE is reported for any k , where models are
identified by the RMSE-ranking (for example, 2-5 is the ensemble mean of the second and fifth best
492 SM in terms of RMSE). The SM RMSE ranking is defined by domain. Models may not have the
same SM RMSE ranking over the different subregions. In bold face are the global minimum, i.e. the
494 minimum among all possible combinations (a five and a six-model combination gave the same
minimum RMSE for EU4).

496

The main aspect worth noticing is that the RMSE-ranking shows that the optimum is, in some
498 cases, achieved by MM ensemble containing low ranking members. This indicates that all members
are needed to build a skilful ensemble. This means that an ensemble of top ranking model results
500 can be worse than an ensemble of top ranking and low ranking ones. Also outliers need to be
included in the ensemble to obtain the best performance.

502

It can be argued that large ensembles are needed to capture extreme events with high
504 concentrations. Figure 9 displays a scatter plot of the 1-hour daily maximum ozone concentration
for the EU subregions (analysis for NA with fewer members produced similar results and is not
506 reported). On the x -axes is the 1-hour maximum of the ensemble of all available members, and the
 y -axes the 1-hour maximum of the ensemble of the selected members with minimum RMSE (bold-
508 face combinations of Table 2). Data distribution along the diagonal line for each region shows that
ensembles of selected models and full ensembles have the same probability to capture the extreme
510 concentrations. In particular for EU1 and EU3, maximum predicted are higher with the small
ensemble. This is because poor performing SM added to an ensemble can improve RMSE and even
512 lead to optimal RMSE. This is due to biases compensating one another.

514 As an example, let us consider the case of Fig. 10, in which ozone concentrations of: observations
(Fig 10a), the ensemble of ranked models 1 and 5, (Fig 10b), ranked model 2 (Fig 10c), and ranked
516 model 11 (Fig 10d), are displayed at receptor positions. With reference to Table 2, the ranked
combination 1-5-11 is that of minimum RMSE for EU1. One could wonder as to why the least
518 ranked model (11) gives a better contribution to the ensemble than a high ranking one, for example
the second best. Looking at the British Isles and France receptors (Domain 1 of Fig 1a), the MM
520 mean of Fig 10b clearly underestimates the observations in the south of France. When the 11th
ranked model (Fig. 10d) is added to the ensemble of Fig 10b, it allows error compensation and
522 RMSE to be lower than the combination with the 2nd best ranked model of Fig. 10c. This is because,
the 2nd best model has a performance very similar to the best one (the 1st already included in the
524 ensemble), and thus brings no new information to the existing ensemble, while the 11th model,
whilst scoring bad at all domain, matched the high concentration of southern France, i.e. the only
526 place where the best models are failing. RMSE penalises large errors, and model ranked 11 is the
model that, on a spatially averaged sense, allows the ensemble to match the concentration at a larger
528 number of stations and larger number of hour with minimum error.

530 Statistical results and whisked plots for the full and for the selected members ensemble are reported
in Table 3 and Fig. 11, for each subregion. The first row of Table 3 is the full members ensemble
532 mean, the second row is the MM of minimum RMSE (bold face combinations of Table 2). RMSE
is, of course, lower for the latter at all subregions. PCC varies only slightly, indicating that the
534 association between observation and ensemble is not strictly related to model's error. The minimum
RMSE combinations also improve the estimation of the spread (the standard deviation of the MM
536 ensemble, σ), in almost all regions (Table 3) and especially for NA regions. Thus, having reduced
the number of the members does not degrade the ensemble variability, which actually becomes
538 closer to the spread of the observations. This is most probably due to the reduced variability
induced by members sharing similar emissions and boundary conditions. Graphical representation
540 in Fig. 11 also shows the how the selected member compares against the full member ensemble in
terms of spread, maximum and minimum, and percentile distribution. The improvement of the
542 selected ensemble to model the spread and the high concentrations are most visible, particularly for
EU regions.

544

5. Reduce data complexity: a clustering approach

546 Results of previous section 4 have shown that a skilful ensemble is built with an optimal number of
members between two (NA) and five (EU), also including low ranking skill score members. Is there

548 any methodology that allows discerning among members? To this scope, we adopted here a
methodology for clustering highly associated models and discard redundant information, using the
550 PCC as metric (we note that PCC is independent to model bias, therefore the analysis would be the
same for unbiased models). The most representative models of each cluster, chosen based on a
552 distance metric, are then used to generate a reduced ensemble. In this way, the information each
member brings to the ensemble is “unique” to the maximum possible degree.

554
The metric used to calculate the distance between the PCC of any two models, and between
556 clusters, is the squared Euclidean distance. First the points which are furthest apart are identified,
and used as the initial cluster centres. Then, the other cases are allocated to the closest centre by
558 Euclidean distance from each centre. Results for this procedure are displayed in Figure 12 (EU) and
Fig 13 (NA). The height of each inverted U-shaped line represents the distance between the two
560 clusters being connected. Independent clusters are identified by different colours. Sensitivity
analysis to other distance metrics (not shown), have shown that the clustering of models is
562 independent on the metric adopted for the distance (leaving the group associations unaltered). The
distance itself, however, changes, but this does not affect the results of this study. On the vertical
564 axis of Figs 12 and 13, models are identified by their number and by their RMSE-ranking
(discussed in Section 4.2). The ranking information allows to track the models positioning and
566 analysing whether aggregation results from difference in the models themselves (transport model,
meteorological drivers, emission, etc.), or if the models’ performance (RMSE) have an influence.

568
For EU (Fig 12), the maximum PCC distance (degree of model’s disassociation) varies between
570 0.12 (EU4) to 0.28 (EU2). By contrast, analysis of NA regions (Fig. 13) shows the maximum
distance is of 0.08 for all regions, except for NA2 (~0.03). Association between models is thus
572 stronger for NA, indicating a lower degree of independence. This can be explained by considering
that four out of seven models used the same meteorological driver, and six models shared the same
574 emissions.

576 Moreover, for EU it is possible to isolate two repeating groups of models whose PCC distance is
very small: Mod6 and Mod7, Mod11 and Mod3. Models of the former group are essentially the
578 same, as they share the transport and meteorological model (WRF-Chem), as well as emission and
boundary conditions. They also have similar RMSE-ranking. Mod11 and Mod3 differ in the air
580 quality model but share the MM5 input and the standard anthropogenic emissions. The NA
analysis, with fewer members, shows repeated association of groups of two models: Mod15 and16

582 (same meteorological driver WRF, anthropogenic emission and boundary conditions), Mod13 and
Mod17 (share the meteorological and air quality models), and Mod14 and Mod18 (same
584 meteorological driver). Mod12 is associated with Mod14 and Mod18, with the exception of NA3.

586 In order to find an optimal set of clusters, we can define a threshold at which models are said to be
independent (imagine cutting vertically the dendrograms). The selection of the cutting height is
588 partially arbitrary. The common practice suggests cutting the dendrogram at the height where
the distance from next clustered groups is relatively large, and the retained number of
590 clusters is small compared to the original number of models (Riccio et al., 2012). Members of
ensemble generated with higher threshold are more distant, thus more independent. Cluster
592 representative and ensembles are summarised in Table 4, for both continents and for different PCC
distance. For clusters composed by two members only, and for cluster with a symmetric structure
594 (same mutual distance among all members such as the third cluster of EU2, Fig 12b), it was no
possible to identify a model having minimum distance or whose distance from the centre of the
596 cluster was minimum in terms of RMSE. In these cases more than one model, in turn, is selected to
represent the cluster.

598
The number of independent members varies between 3 and 6 for EU, and between 2 and 4 for NA
600 (this difference is probably due to the smaller number of models for NA). It is remarkable that the
number of independent clusters matches the number of models to generate MM ensemble of
602 minimum RMSE of Figs. 8, for both continents. The two investigations are in fact independent, as
the clustering analysis makes no use of observational data. Looking at the bold-face combinations
604 of Table 2 (minimum RMSE combination), it can be deduced that the ensembles of minimum
RMSE have two or more members belonging to the same cluster, and for NA4 all members from
606 the same cluster. This result is driven by not having many independent members, due to models
sharing of boundary conditions, meteorology, and emissions.

608
We have also compared the RMSE of MM ensembles of Table 4 with the curves of RMSE
610 discussed in Fig. 8, and reported the results in Fig 14a (EU) and Fig 14b (NA). Imagine connecting,
for any number of models, the minimum and the maximum RMSE of Figs. 8. The curves obtained
612 are those of Figs. 14 (thick lines: minimum; dotted lines: maximum). RMSE of combinations of
Table 4 (obtained with the clustering technique) are the short lines in Figs. 14, reported along with
614 the ranked combination. In the cases of clusters with two members only (symmetric clusters), it was
not possible to identify the cluster representative, and thus two members have been retained for the

616 analysis. What is interesting to compare in Figs. 14 is the position of the cluster's combination
against the RMSE of the full member ensemble, to infer whether the new methodology is able to
618 produce reduced ensembles more skilful than the full ensemble mean. In fact, we can notice that
independent model combinations have, in most cases, lower RMSE than the full ensemble, and that
620 for all subregions there are ensembles that clearly outperform it. For example, the combinations 1-
2-3-8-11, 2-6-7-8, 1-3-6-9-11, 1-2-4-8-9-11 for EU1,2,3, and 4, respectively, have lower RMSE
622 than the mean of all ensemble members and close to the minimum curve. There are, on the other
hand, situations in which the ambiguous definition of cluster representatives leads to high-RMSE
624 MM combinations, as for the four-member combination of EU4 (1-2-4/5-11) and NA1 (2-4-5).
Further work needs to be devoted to remove such ambiguity.

626

6. Conclusions

628 This study collectively evaluates and analyses the performance of over ten regional air quality
models and their ensembles in the context of the AQMEII intercomparison exercise. The scale of
630 the exercise is unprecedented, with two continent-wide domains being modelled, for a full year.
Focus of this paper is on the *collective* analysis of surface ozone concentration, rather than on inter-
632 comparing metrics for each individual model. We start with analysing time series for ozone in
subregions of the continental Europe and North America, and proceed with the interpreting
634 uncertainties for individual model and ensemble. Analysis of model bias and error in each
subregion demonstrate that most of the model's error is introduced by bias, from emissions,
636 boundary conditions and meteorological drivers.

638 We then show that there are ensemble combinations with a reduced number of members whose
mean produces an error smaller than the full member ensemble mean. Thus, we have shown that a
640 skilful ensemble is not necessarily generated by all available members, but rather by selecting
members that can contribute to minimise the error.

642

We find that an ensemble of top ranking model results can be worse than an ensemble of top
644 ranking and low ranking ones. Until now it was assumed that, as long as a large set of results were
treated statistically in one ensemble, then the ensemble would have been better than any individual
646 member. Furthermore, it was supposed that the better the results the better the ensemble. What we
demonstrate here is that such hypothesis is not necessarily true, as also outliers need to be included
648 in the ensemble to enhance performance. Further to that, we have shown that the score does not
improve with just increasing the number of models in the ensemble. By contrast, the level of

650 dependence of model results may lead to a deterioration of the results and to an overall worsening
of performance. Despite the remarkable progress of ensemble air quality modelling over the past
652 decade and the effort spent to build a theoretical foundation, there still are many outstanding
questions regarding this technique. Among them, what is the best and most beneficial way to build
654 an ensemble of members? How to determine the optimum size of the ensemble in order to capture
data variability as well as keeping the error low?

656
To try addressing these questions, we adopt a methodology for reducing data complexity, known as
658 *clustering technique*, which has the purpose of simplifying information provided by the large
amounts of data (such as air quality model outputs) by classifying, or clustering, the data into
660 groups based on a select metric, where there is no prior knowledge of grouping. Results show that,
whilst this methodology needs further refinements, by opportunely selecting the cluster distance and
662 association criteria, we can generate ensemble of selected members with errors significantly lower
than the full members ensemble mean. The results of the clustering analysis have a general
664 character and impact, and are of direct relevance for applications not only strictly related to
ensemble model evaluation, but also to other ensemble communities, for example air quality
666 forecasting, climate, oceanography, and others.

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676

Appendix A: Statistical Measures

678 Defining y the vector of model output and obs the vector of observations (n -component both), having mean
value \bar{y} and \overline{obs} , respectively.

680 Mean bias:

$$MB = \frac{\sum_i (y_i - obs_i)}{n} \quad (A1)$$

682 Root mean square error:

$$RMSE = \sqrt{\frac{\sum_i (y_i - obs_i)^2}{n}} \quad (A2)$$

684 Mean Gross Error:

$$MGE = \frac{\sum_i |y_i - obs_i|}{n} \quad (A3)$$

686 Normalised mean square error

$$NMSE = \frac{\sum_i (y_i - obs_i)^2}{n \overline{y \text{ obs}}} \quad (A4)$$

688 Fractional Bias

$$FB = 2 \frac{\overline{obs} - \overline{y}}{\overline{obs} + \overline{y}} \quad (A5)$$

690 Normalised Mean Bias:

$$NMB = \frac{\sum_i (y_i - obs_i)}{n \overline{y \text{ obs}}} \quad (A6)$$

692

Pearson correlation coefficient:

$$PCC = \frac{\sum_i (y_i - \overline{y})(obs_i - \overline{obs})}{\sqrt{\sum_i (y_i - \overline{y})^2 \sum_i (obs_i - \overline{obs})^2}} \quad (A7)$$

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Captions

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Table 1. Participating models and features

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Table 2. RMSE-ranked combinations of models that give minimum RMSE. In bold the minimum of all combinations

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Table 3. Statistical skills for all members ensemble (first row of each domain), ensemble of minimum RMSE (second row). σ is the standard deviation in $\mu\text{g m}^{-3}$ for EU and ppb for NA.

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Table 4. Ranking of cluster representatives for EU and NA subregions for varying PCC distance.

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Figure 1 – Continental maps of *a)* Europe and *b)* North America and subregions by colours. The dots are the positions of ozone receptors used in the analysis. The contours are the anthropogenic NO_x emissions (in kg km⁻²) using the standard inventories.

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Figure 2. Ozone ranges at receptors, averaged in space over *a)* EU domain and *b)* NA domain and in time for the whole 2006 year.

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Figure 3. Ranked histogram for the whole subregions of *a)* EU and *b)* NA, full model ensemble, hourly data for the whole 2006 year.

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Figure 4. Time series (JJA) of diurnal ozone cycle for *a)* EU and *b)* NA subregions

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Figure 5. Mean Bias vs Normalised Mean Square Error for *a)* EU and *b)* NA. Subregions 1 to 4 represented by number, coloured by model or ensemble. Mean and median for each subregion are highlighted

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Figure 6. Average vertical profiles of ozone in the vicinity of Portland airport. Measurements are the monthly mean MOZAIC for August with the standard deviation; the thick grey line is the GEMS BC at a node close to Portland; thin grey lines are the ozone predictions by three models employing GEMS BCs; the brown square is the ozone concentration at surface receptors in the vicinity of Portland.

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Figure 7. Ranked histogram for *a)* EU and *b)* NA by subregions, full model ensemble, hourly data for the period JJA.

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Figure 8. RMSE, MGE, MB, PCC of the ensemble mean of any possible combination of members for *a)* EU, and *b)* NA. Continuous lines are the mean and the median of the distributions

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Figure 9. Daily maximum concentrations for EU subregions, for the period JJA. Horizontal axis: ensemble maximum of all available members. Vertical axis: ensemble maximum of model combinations with minimum RMSE.

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Figure 10. Ozone concentrations ($\mu\text{g m}^{-3}$) for the period JJA at receptors position. *a)* observations, *b)* ensemble of ranked models 1 and 5; models ranked *c)* 5th and *d)* 11th.

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Figure 11. Box-plot of observed ozone concentration, full model ensemble and selected (combinations with minimum RMSE) model ensemble. Top row: EU subregions; bottom row: NA subregions.

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Figure 12 Dendrograms of models clustering as function of mutual PCC distance for EU subregions

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Figure 13 Dendrograms of models clustering as function of mutual PCC distance for NA subregions

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Figure 14 Curves of minimum (thick lines) and maximum (dotted lines) RMSE obtained by connecting min and max of Fig. 8. The short lines are the RMSE of MM ensembles from clustering analysis (combinations of Table 4). The labels are the individual RMSE-ranking of MM members. Different colours correspond to different subregions for *a)* EU and *b)* NA.

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924 **Tables**
Table 1

| | Model | | Res (km) | n.Vertical layers | Emission | Chemical BC |
|-------------------------------|-----------|------------|----------|-------------------|---|--------------------------|
| | Met | AQ | | | | |
| European Domain | MM5 | DEHM | 50 | 29 | Global emission databases, EMEP | Satellite measurements |
| | MM5 | Polyphemus | 24 | 9 | Standard [§] | Standard |
| | PARLAM-PS | EMEP | 50 | 20 | EMEP model [§] | From ECMWF and forecasts |
| | WRF | CMAQ | 18 | 34 | Standard [§] | Standard |
| | WRF | WRF-Chem | 22.5 | 36 | Standard [§] | Standard |
| | WRF | WRF-Chem | 22.5 | 36 | Standard [§] | Standard |
| | ECMWF | SILAM | 24 | 9 | Standard anthropogenic In-house biogenic | Standard |
| | MM5 | Chimere | 25 | 9 | MEGAN, Standard | Standard |
| | LOTOS | EUROS | 25 | 4 | Standard [§] | Standard |
| | COSMO | Muscat | 24 | 40 | Standard [§] | Standard |
| | MM5 | CAMx | 15 | 20 | MEGAN, Standard | Standard |
| North American Domain* | GEM | AURAMS | 45 | 28 | Standard ⁺ | Climatology |
| | WRF | Chimere | 36 | 9 | Standard | LMDZ-INCA |
| | MM5 | CAMx | 24 | 15 | Standard | LMDZ-INCA |
| | WRF | CMAQ | 12 | 34 | Standard | Standard |
| | WRF | CAMx | 12 | 26 | Standard | Standard |
| | WRF | Chimere | 36 | 9 | Standard | standard |
| | MM5 | DEHM | 50 | 29 | global emission databases, EMEP | Satellite measurements |

926 [§] Standard anthropogenic emission and biogenic emission derived from meteorology (temperature and solar radiation) and land use distribution implemented in the meteorological driver (Guenther et al., 1994; Simpson et al., 1995).
928 *Standard inventory for NA includes biogenic emissions (see text).
930 ⁺Standard anthropogenic inventory but independent emission processing, excluding wildfire and different version of BEIS (v3.09) used.

932 **Table 2**

| | | Number of Models | | | | | |
|----|------|------------------|---------------|----------------|--------------------|----------------------|-----------------|
| | | 2 | 3 | 4 | 5 | 6 | 7 |
| EU | dom1 | 1-2 | 1-5-11 | 1-2-7-11 | 1-2-5-7-11 | 1-2-4-5-6-11 | 1-2-3-4-5-6-11 |
| | dom2 | 3-8 | 2-3-8 | 2-3-5-8 | 1-2-3-5-8 | 1-2-3-4-5-8 | 1-2-3-4-5-6-8 |
| | dom3 | 2-3 | 2-3-5 | 1-2-3-5 | 1-2-3-9-11 | 1-2-3-5-8-11 | 1-2-3-5-8-9-11 |
| | dom4 | 5-9 | 1-6-9 | 2-6-7-9 | 1-6-9-10-11 | 1-2-6-9-10-11 | 1-2-3-6-9-10-11 |
| NA | dom1 | 1-2 | 1-2-3 | 1-2-4-6 | 1-2-3-4-6 | 1-2-3-4-6-7 | |
| | dom2 | 1-2 | 1-3-4 | 1-2-3-4 | 1-2-3-4-5 | 1-2-3-4-5-6 | |
| | dom3 | 2-3 | 1-2-3 | 1-2-3-4 | 1-2-3-4-5 | 1-2-3-4-5-6 | |
| | dom4 | 1-2 | 1-2-3 | 1-2-3-4 | 1-2-3-4-5 | 1-2-3-4-5-6 | |

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Table 3.

| | | Bias | FBias | RMSE | PCC | σ |
|----|----------------------|-------------|--------------|-------------|------------|----------------------------|
| EU | Dom 1 | -5.11 | -0.08 | 12.01 | 0.97 | 18.29 |
| | $\sigma_{obs}=27.4$ | -0.82 | -0.01 | 8.49 | 0.96 | 22.24 |
| | Dom 2 | -8.77 | -0.11 | 13.50 | 0.93 | 17.59 |
| | $\sigma_{obs}=24.5$ | 1.35 | 0.02 | 7.78 | 0.95 | 22.29 |
| | Dom 3 | -4.87 | -0.06 | 17.38 | 0.89 | 20.25 |
| | $\sigma_{obs}=31.7$ | -2.34 | -0.03 | 14.90 | 0.90 | 24.17 |
| NA | Dom 4 | -1.11 | -0.013 | 8.27 | 0.92 | 17.25 |
| | $\sigma_{obs}=20.7$ | -1.25 | -0.014 | 7.27 | 0.94 | 18.34 |
| | Dom 1 | 0.66 | 0.02 | 3.63 | 0.94 | 12.3 |
| | $\sigma_{obs}=10.13$ | -0.11 | -0.003 | 3.45 | 0.94 | 12.1 |
| | Dom 2 | 3.90 | 0.10 | 6.40 | 0.92 | 11.80 |
| | $\sigma_{obs}=12.83$ | 2.05 | 0.05 | 4.82 | 0.92 | 12.6 |
| NA | Dom 3 | 4.51 | 0.13 | 7.34 | 0.85 | 12.5 |
| | $\sigma_{obs}=10.36$ | 2.55 | 0.07 | 5.8 | 0.87 | 10.5 |
| | Dom 4 | 10.55 | 0.26 | 12.35 | 0.90 | 12.3 |
| | $\sigma_{obs}=14.50$ | 5.10 | 0.13 | 7.98 | 0.91 | 14.2 |

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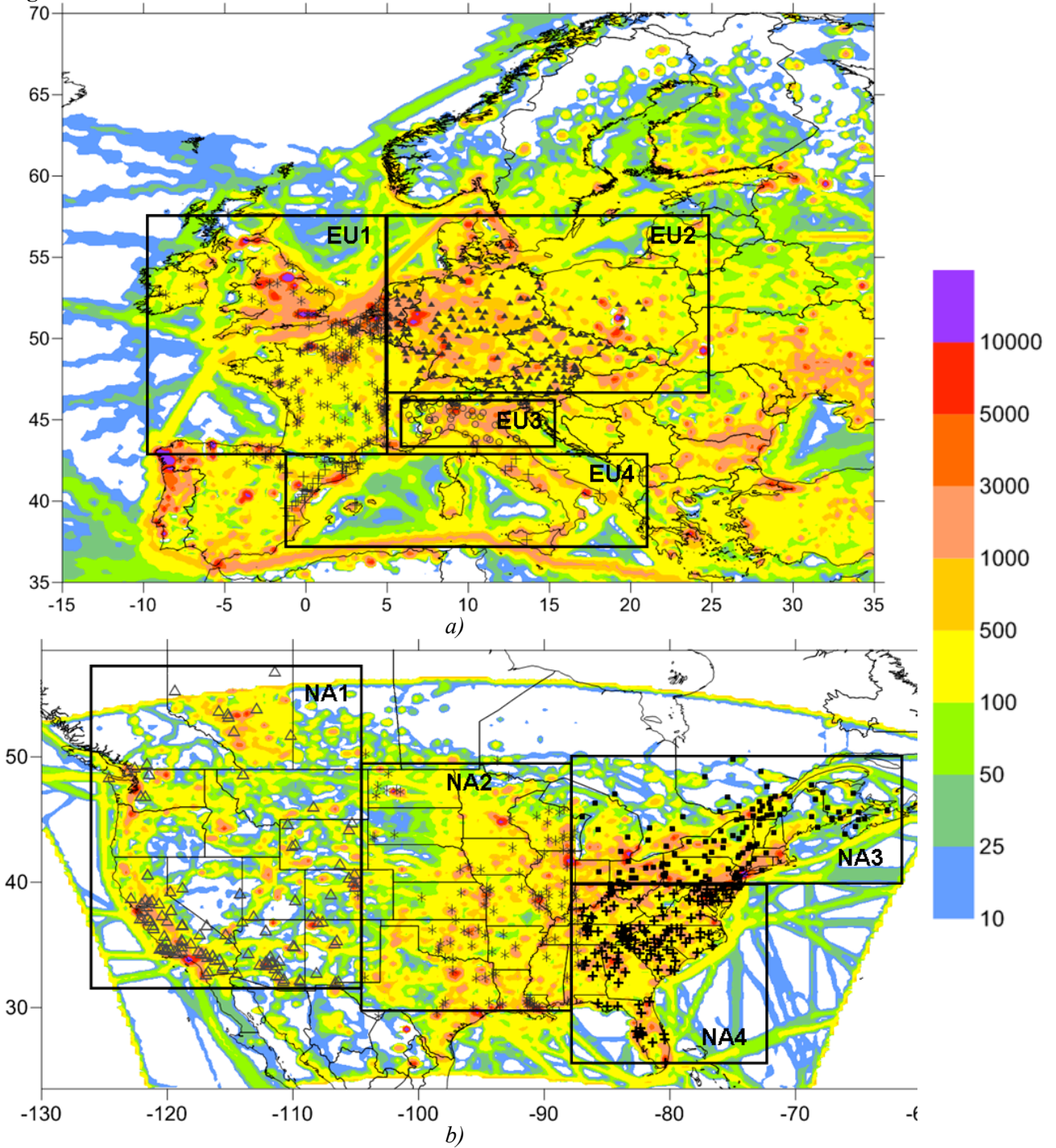
Table 4

| | distance | Number of members | Ranking of cluster representatives |
|------------|-----------------|--------------------------|---|
| EU1 | PCC > 0.06 | 3 | 6-2-8/9 |
| | PCC = 0.05 | 4 | 3-2-8/9-11 |
| | PCC = 0.03 | 5 | 3-2-8/9- 11-1/10 |
| EU2 | PCC>0.045 | 4 | 6-1/8-2-7/9 |
| EU3 | PCC>0.08 | 3 | 3-6/7- 1 |
| | PCC=0.06 | 5 | 3-11- 6/7- 1-9 |
| EU4 | PCC > 0.04 | 4 | 1-4/5-2-11 |
| | PCC =0.02 | 6 | 1-4/5-2/10-9-11/7-8 |
| NA1 | PCC>0.04 | 3 | 3/4-1/5- 2 |
| NA2 | PCC>0.012 | 3 | 3/5-6/7-1 |
| NA3 | PCC>0.035 | 2 | 2/4-6 |
| | PCC=0.03 | 4 | 4-2-3-6/7 |
| NA4 | PCC>0.025 | 3 | 4/7- 5/6-3 |

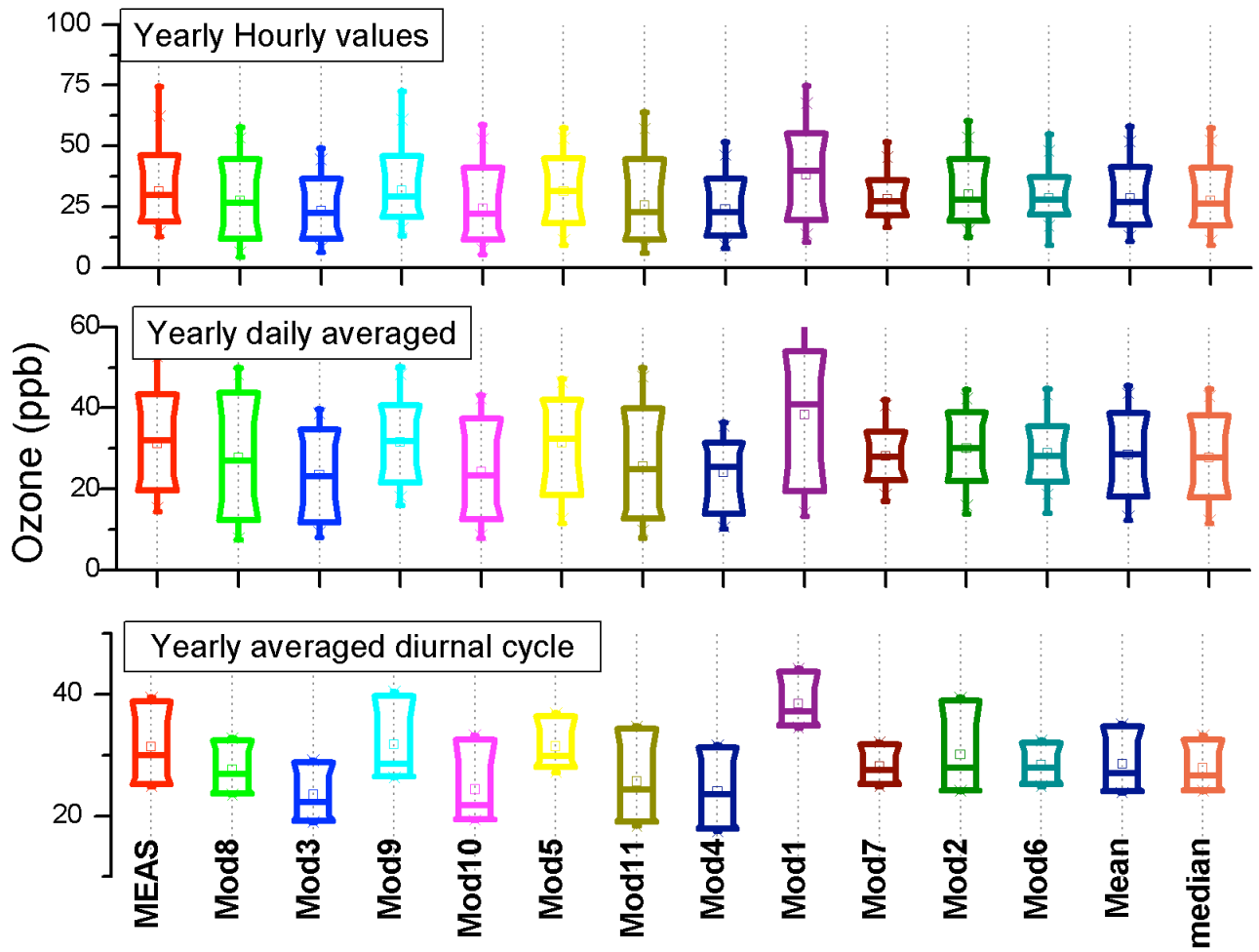
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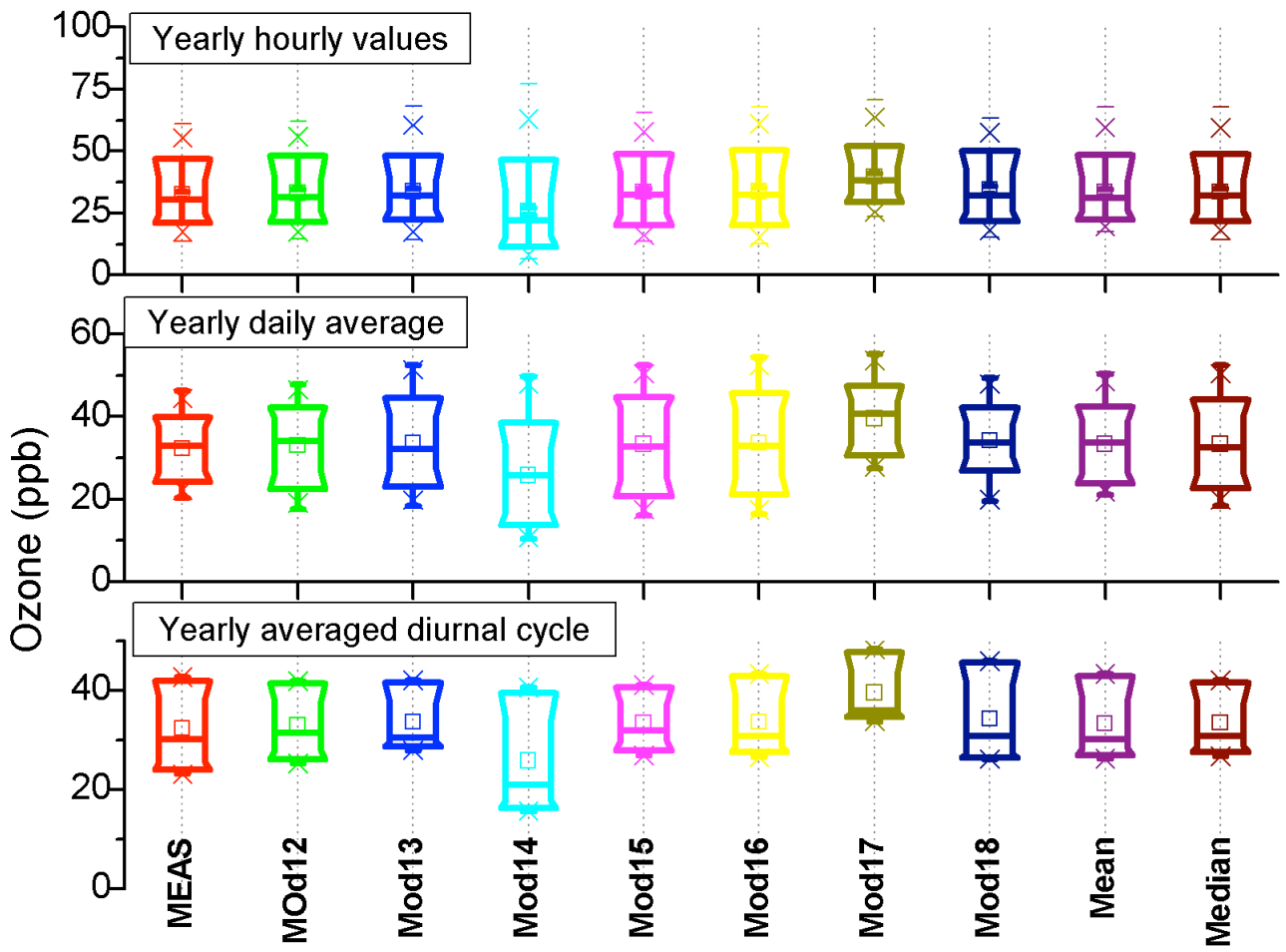
Figures



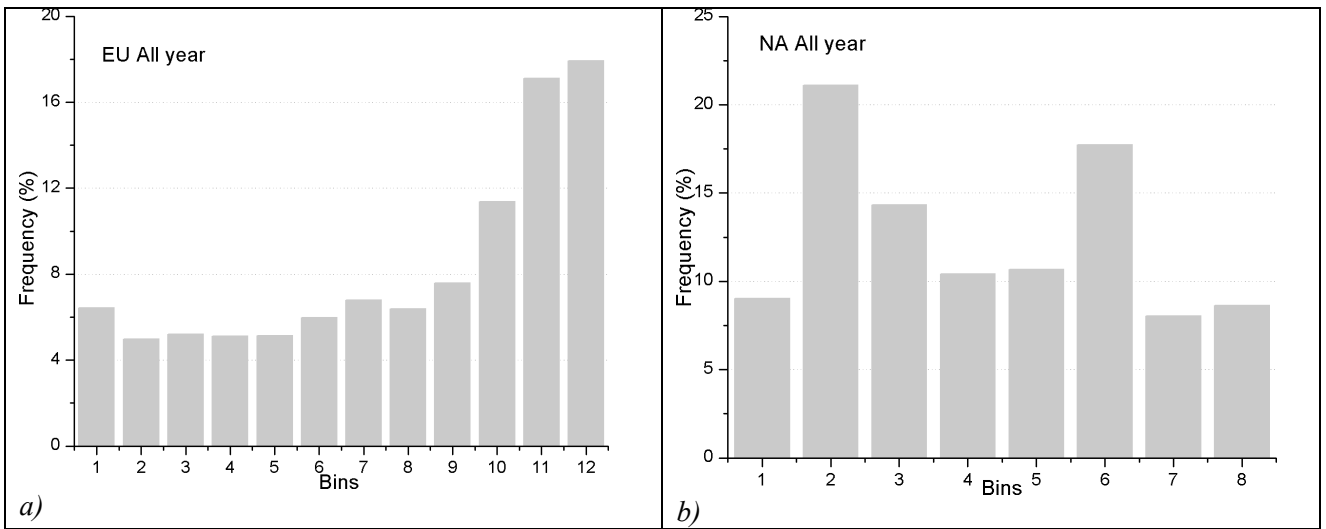
956 Figure 1



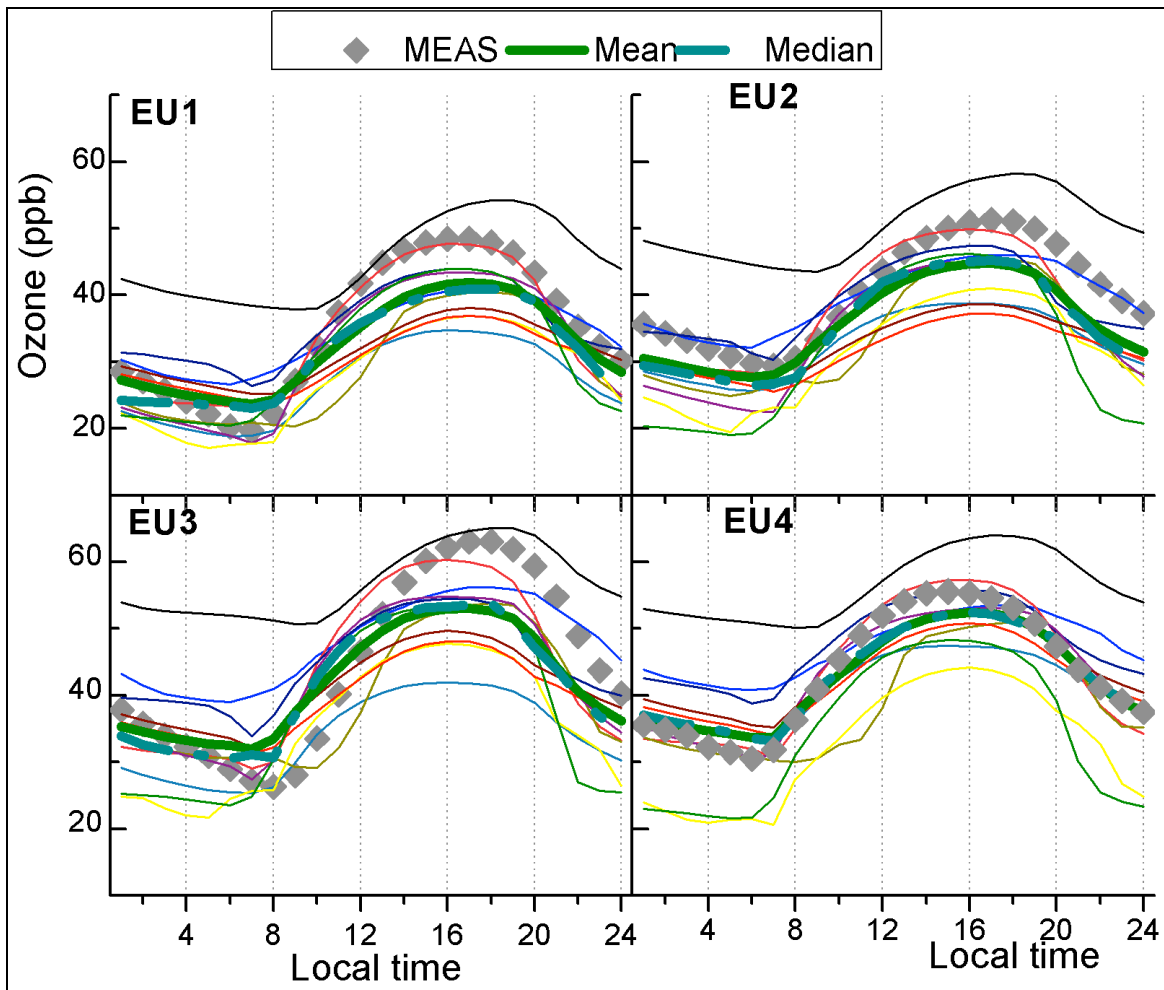
a)



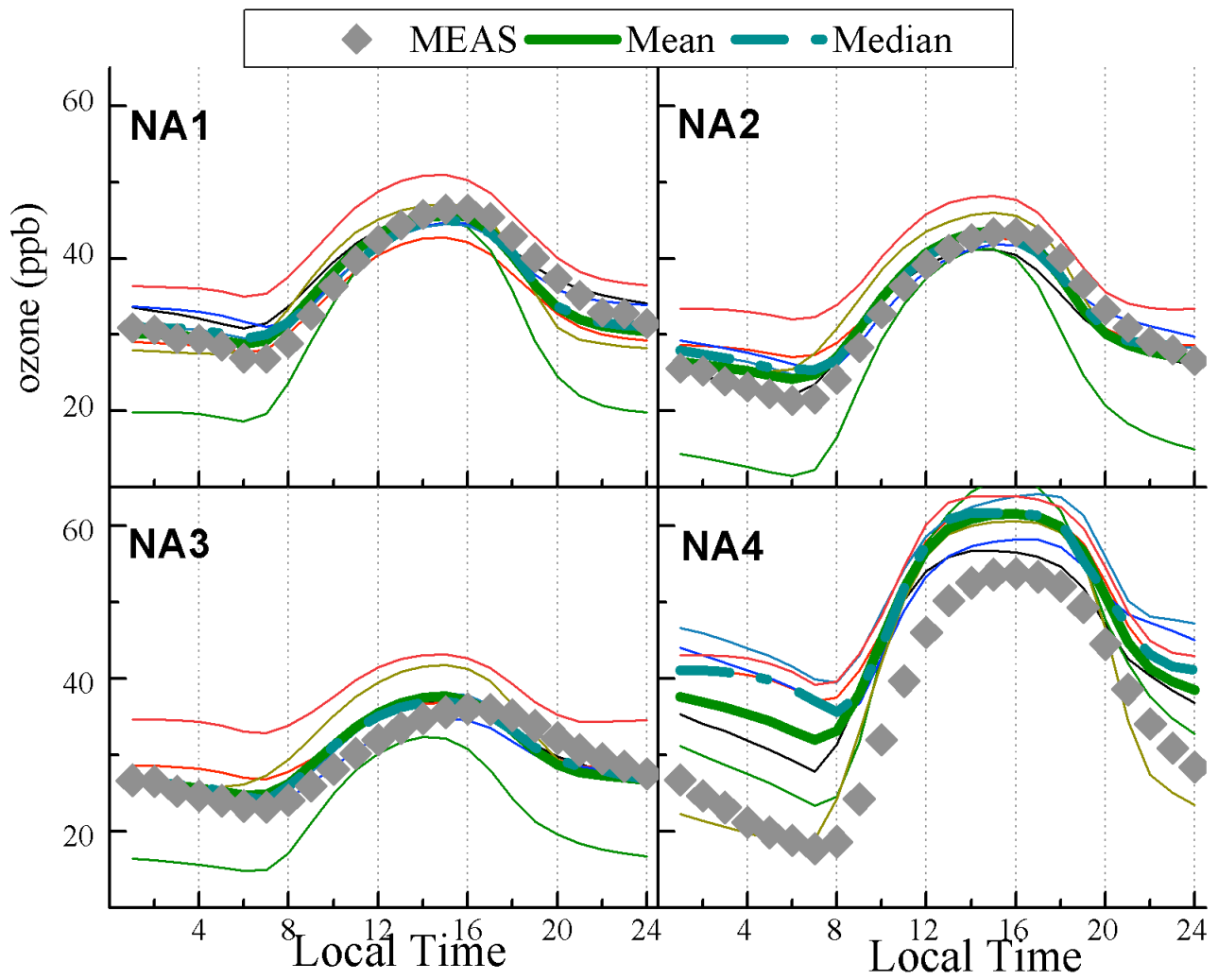
b) **Figure 2**



a) **Figure 3**



a)

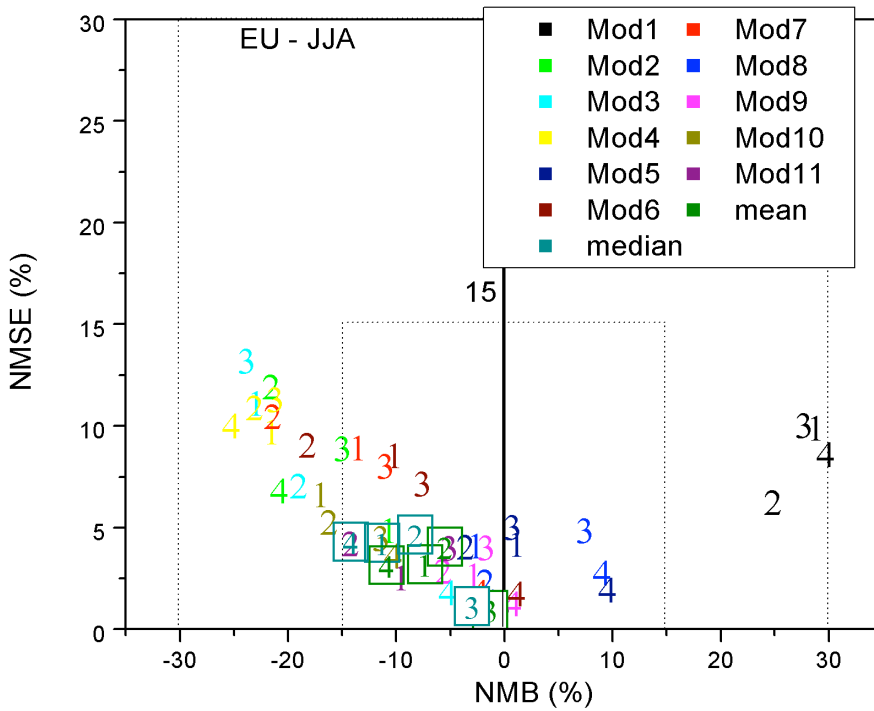


b)

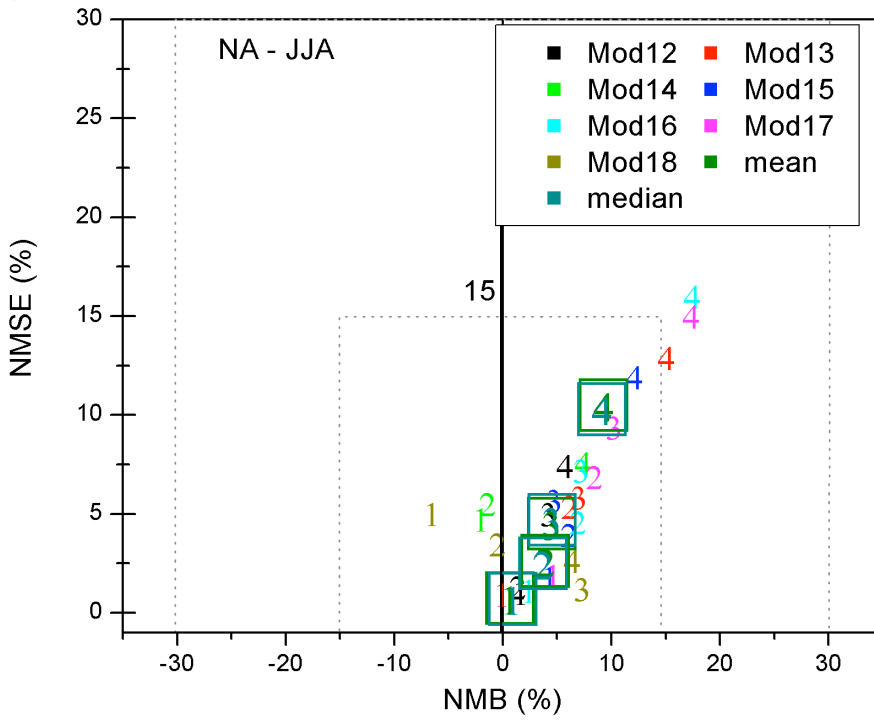
962 **Figure 4.**

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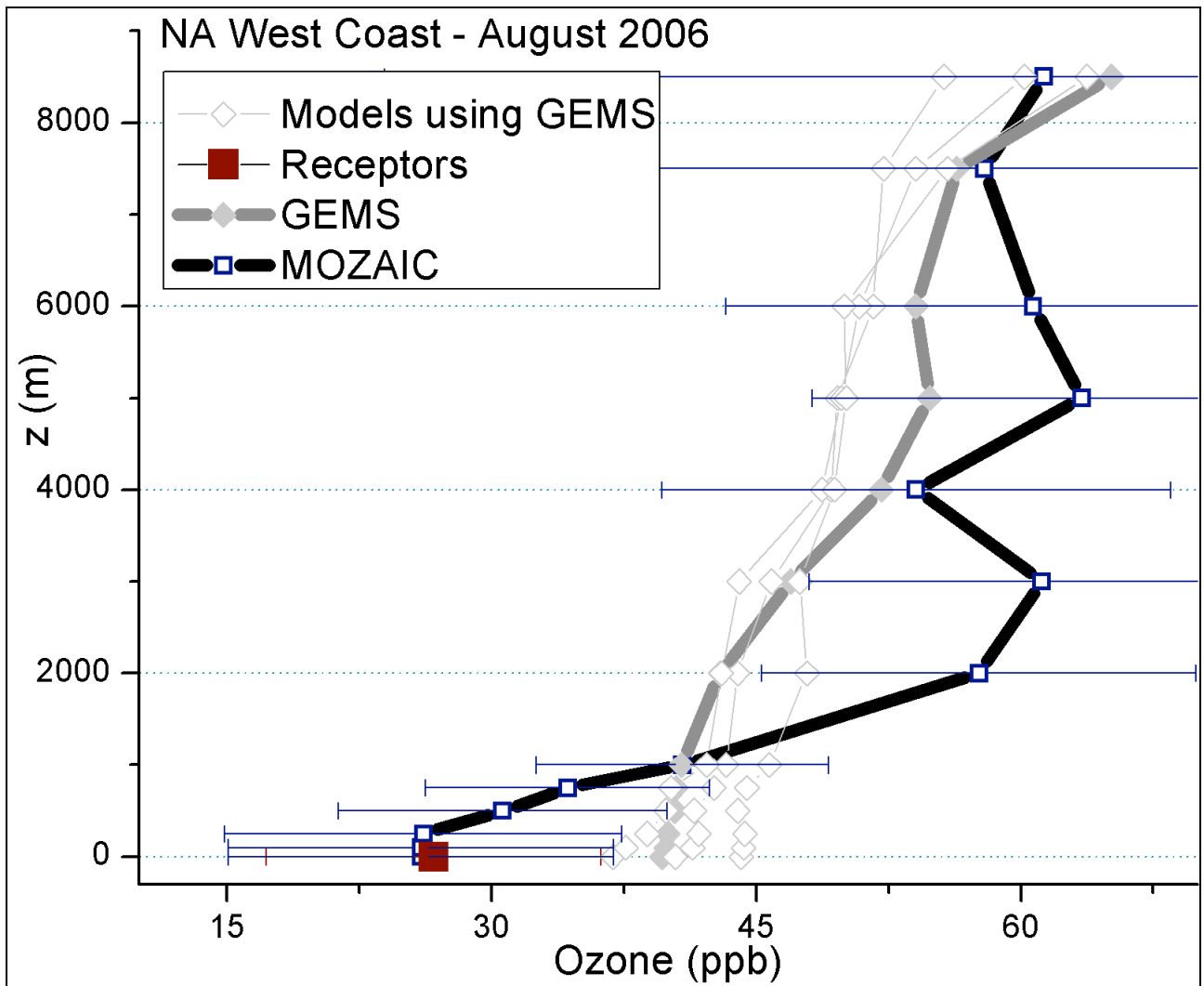


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Figure 5.



970 **Figure 6**

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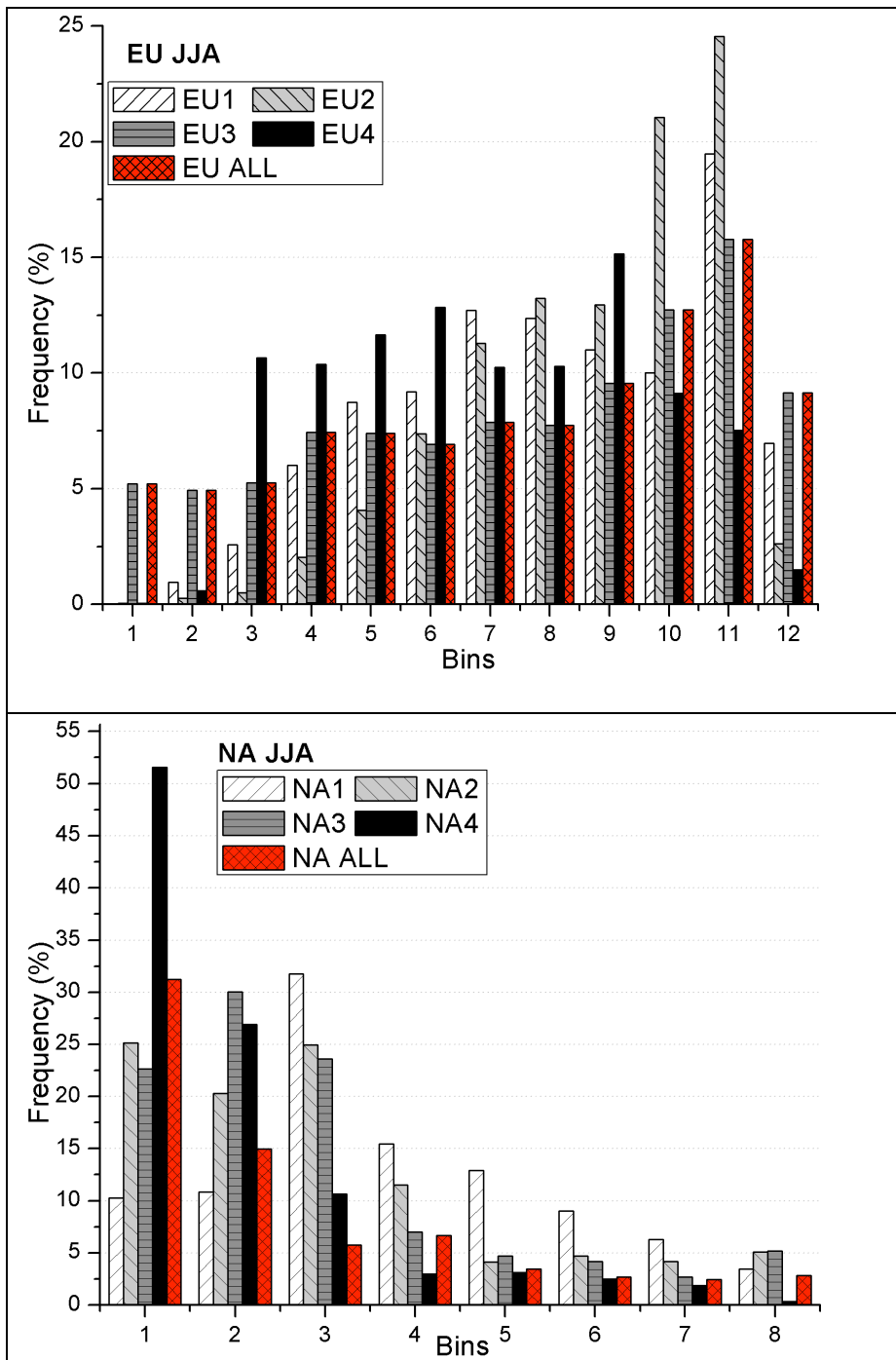
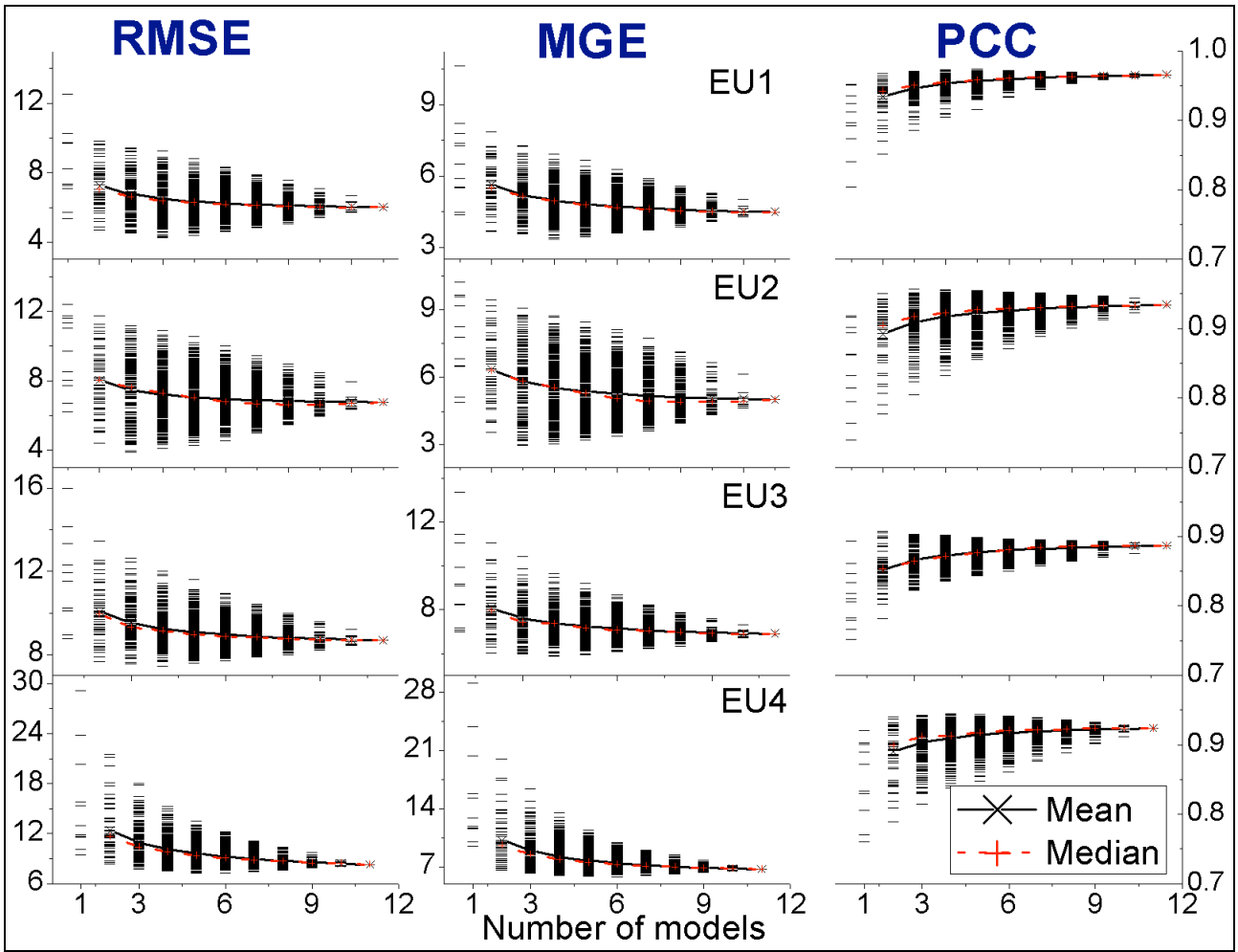
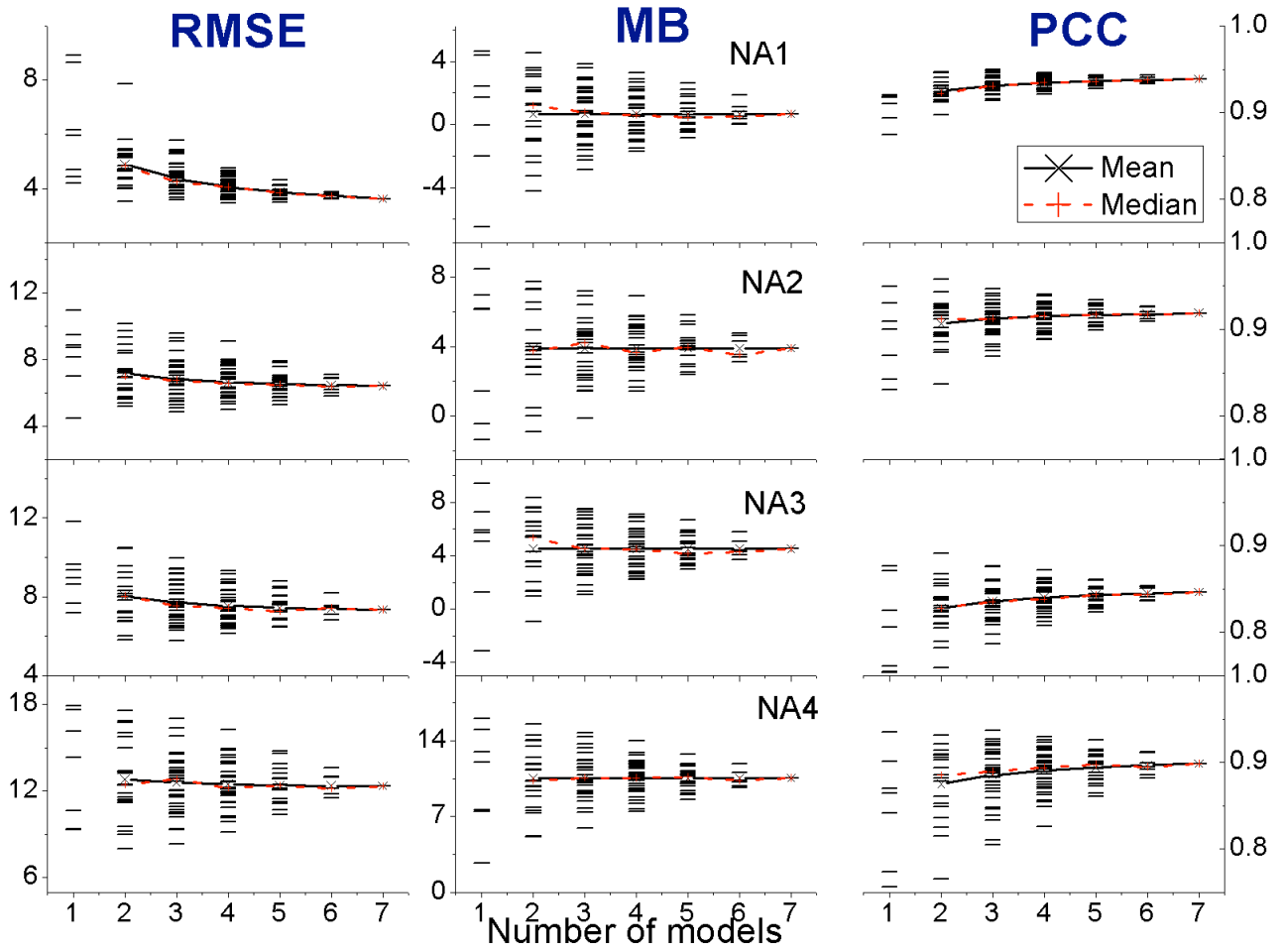


Figure 7



988 **Figure 8a**



990 **Figure 8b**

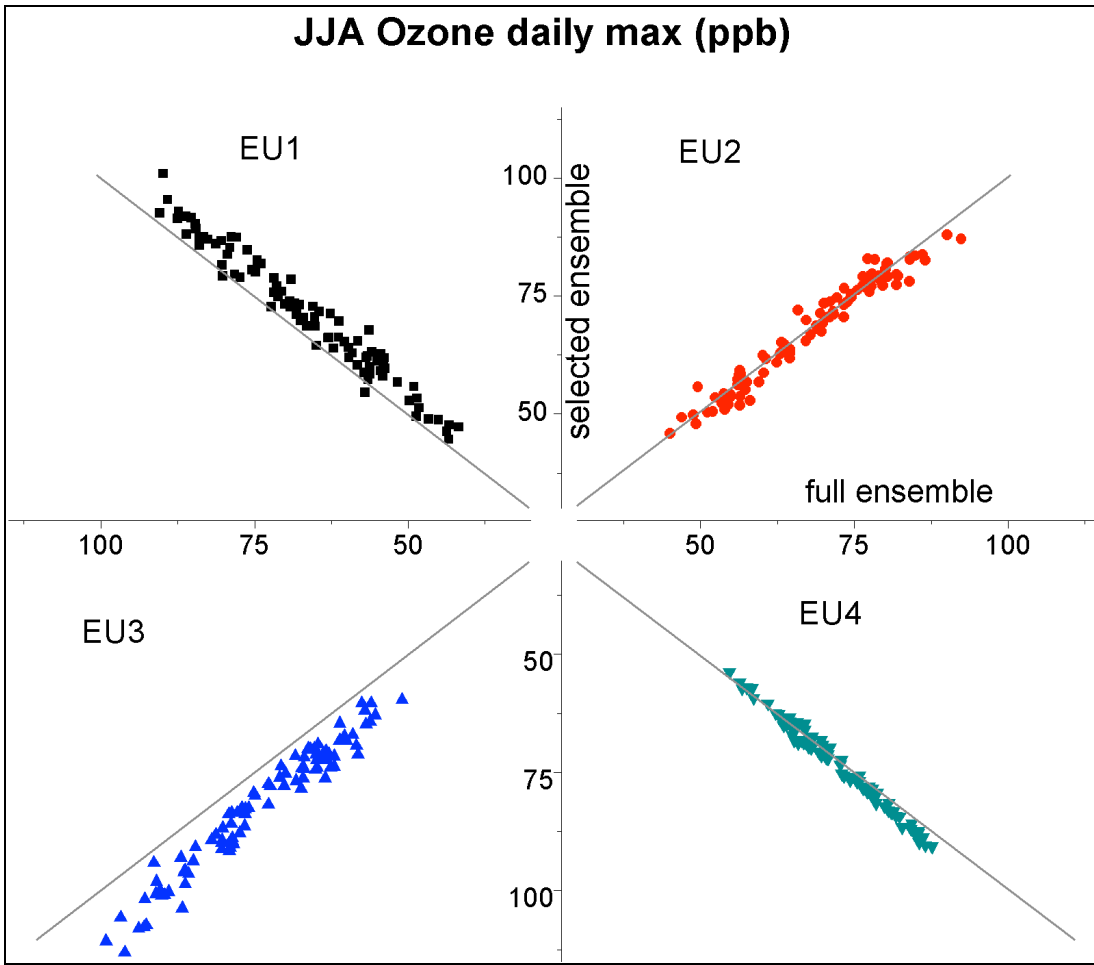


Figure 9

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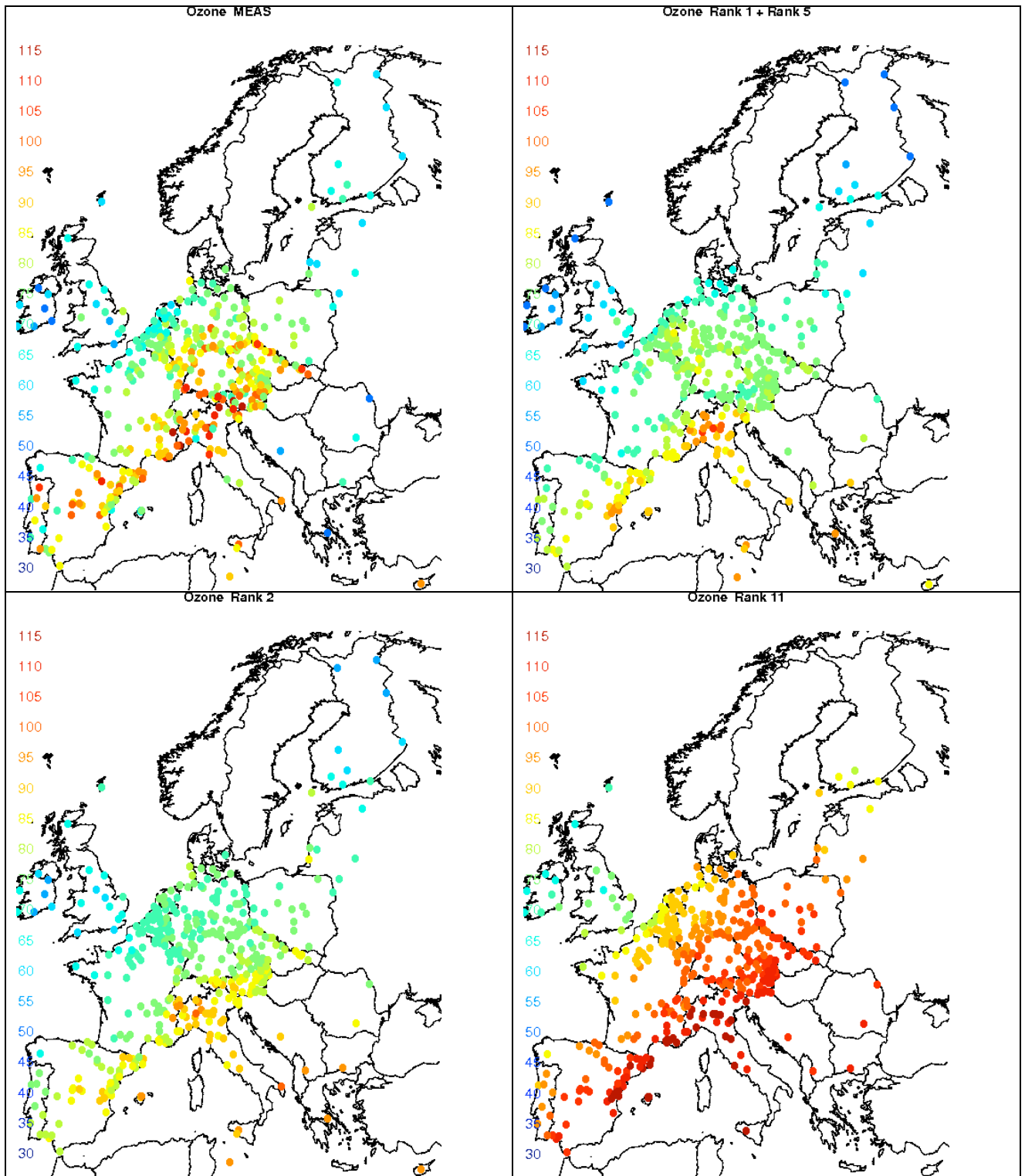


Figure 10

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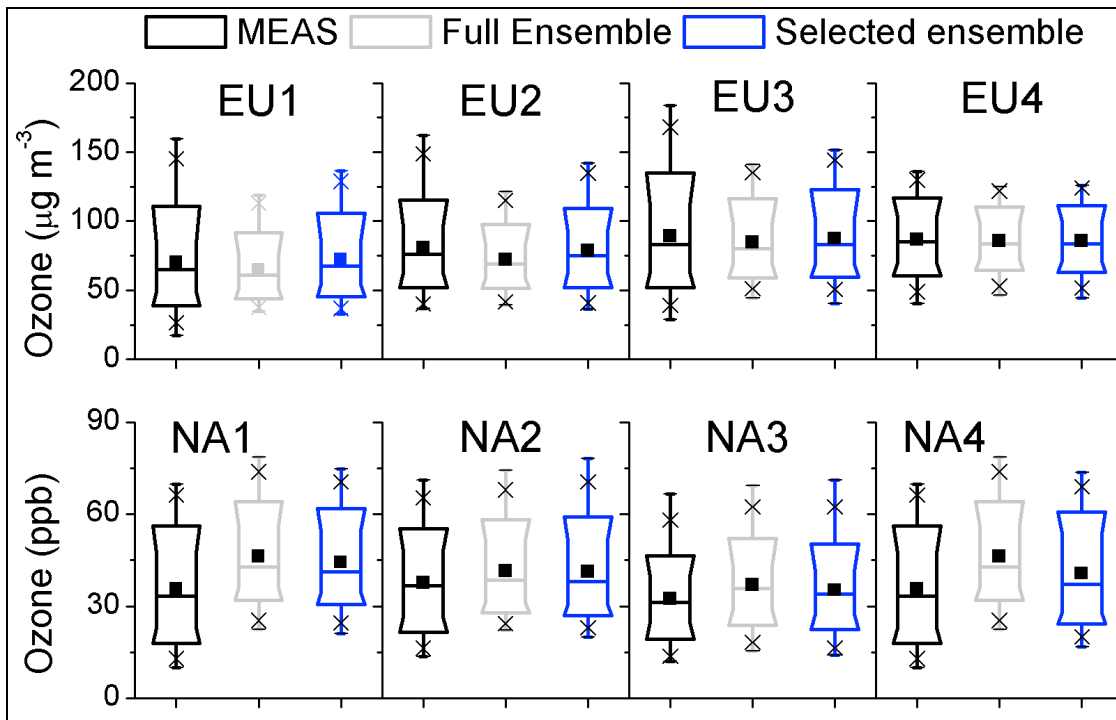
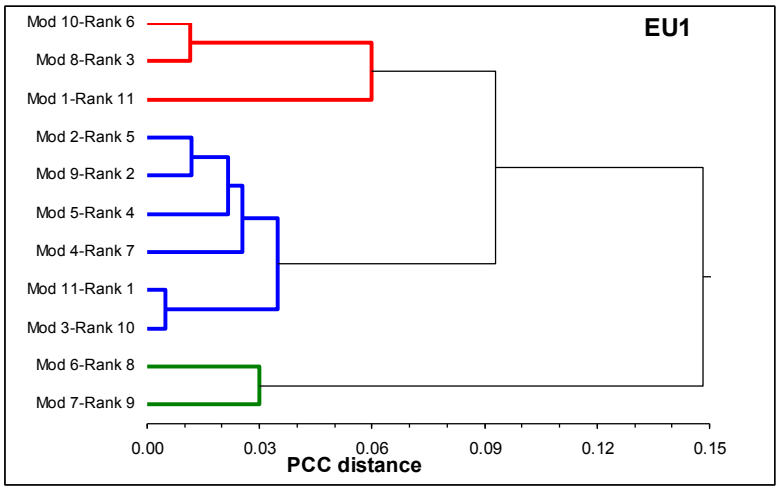
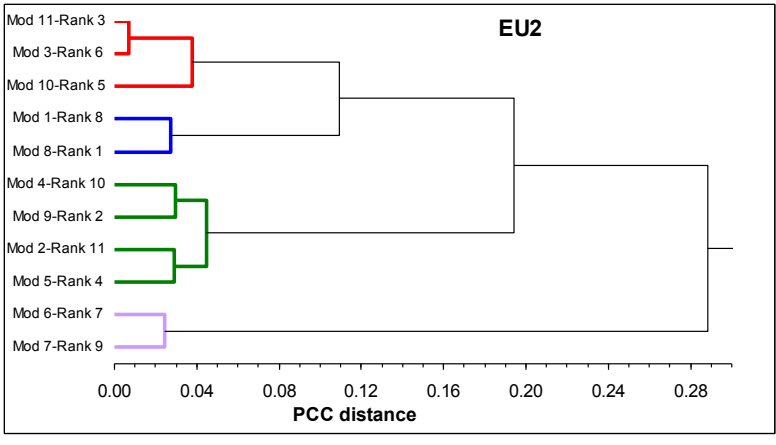


Figure 11

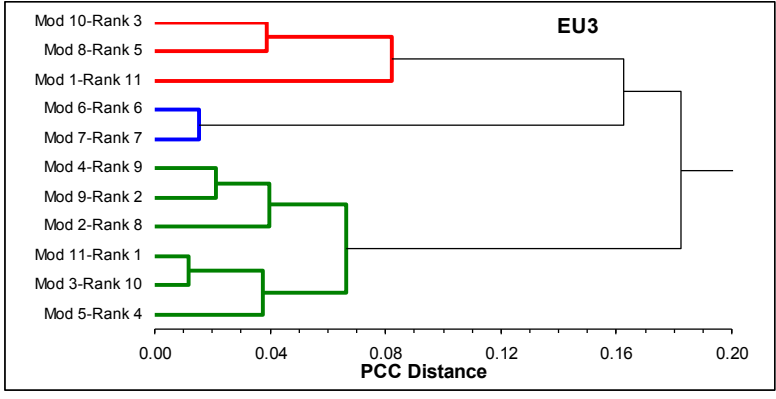
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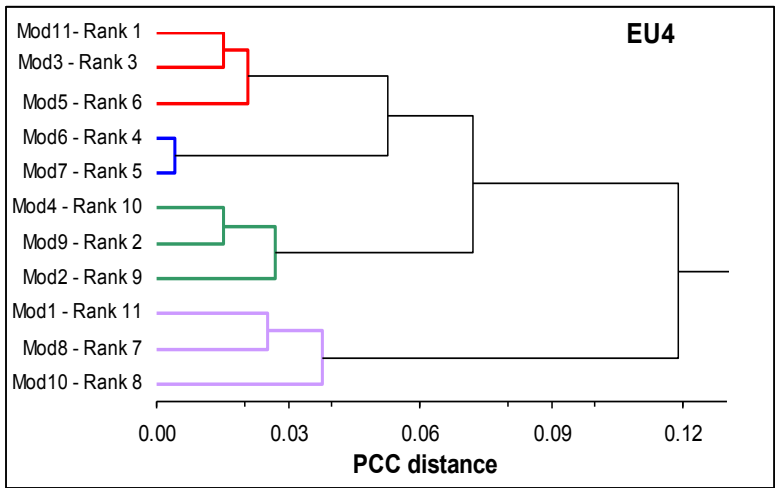
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Figure 12

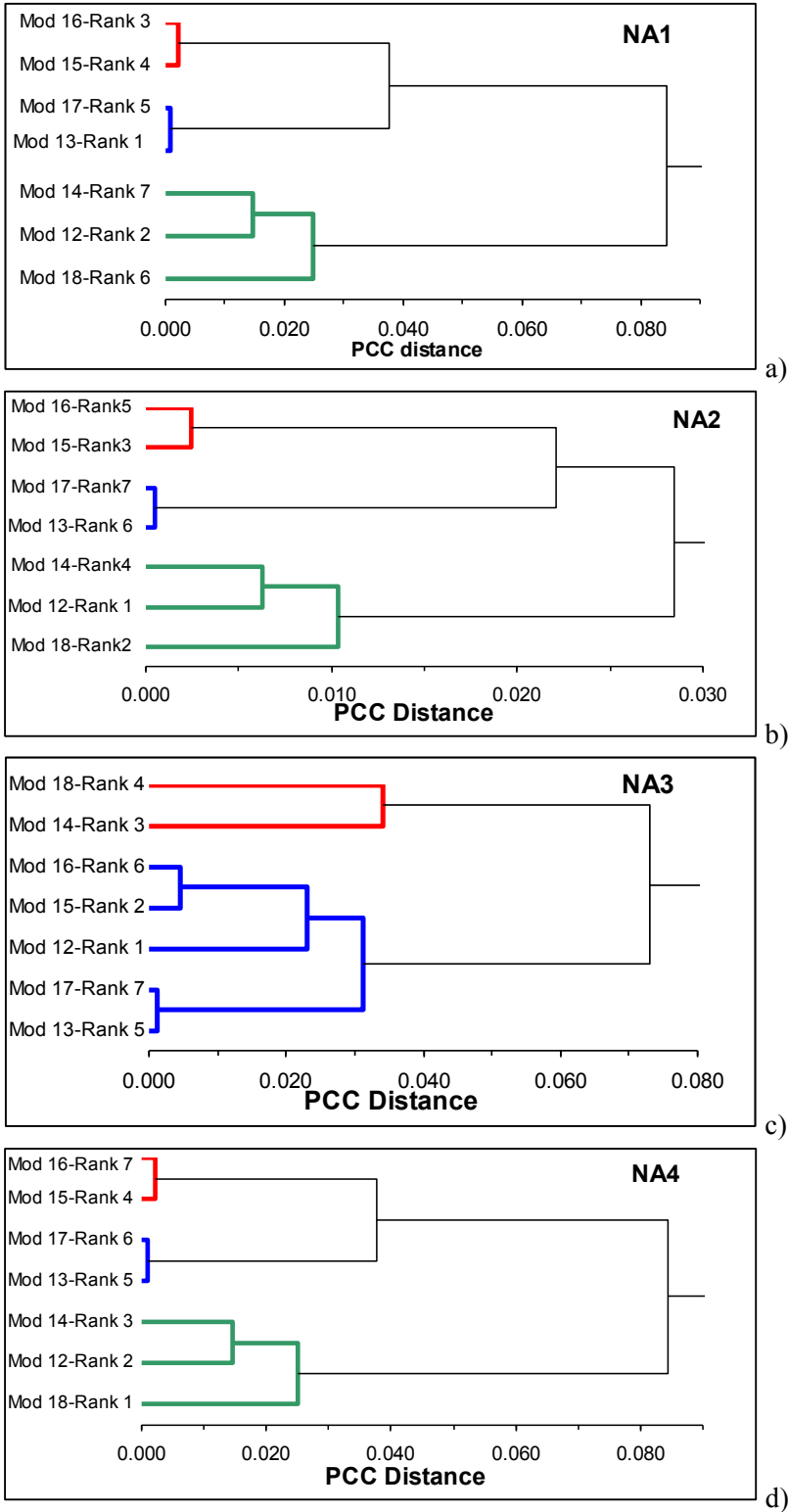
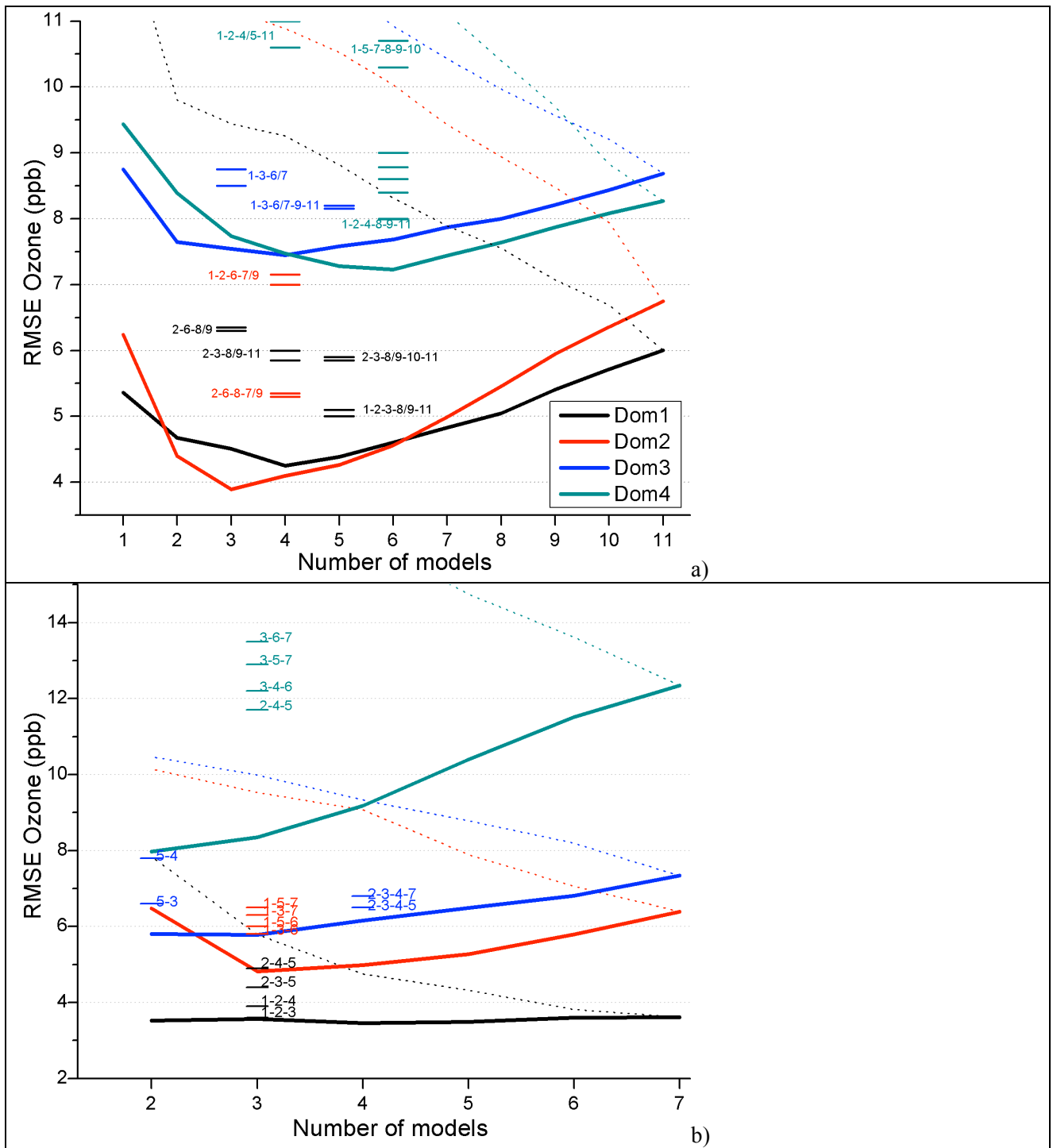


Figure 13



1066 **Figure 14**