

# 1 How bias-correction can improve air quality forecast over Portugal

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7

## 8 Abstract

9 Currently three air quality modelling systems routinely operate with high resolution over  
10 mainland Portugal for forecasting purposes, namely MM5-CHIMERE, MM5-EURAD and  
11 CALIOPE. Each of one operates daily using different horizontal resolutions (10 km x 10 km; 5  
12 km x 5 km and 4 km x 4 km, respectively), specific physical and chemical parameterizations,  
13 and their own emission pre-processors (with common EMEP emission database source, but  
14 different spatial disaggregation methodologies). The operational BSC-DREAM8b model is  
15 offline coupled within the aforementioned air quality systems to provide Saharan dust  
16 contribution to particulate matter. Bias-correction studies have demonstrated the benefit of  
17 using past observational data to reduce systematic model forecast errors. The present  
18 contribution aims to evaluate the application of two bias-correction techniques - multiplicative  
19 ratio and Kalman filter in order to improve the air quality forecast over Portugal. Both  
20 techniques are applied to the three modelling systems over the full year 2010. Raw and unbiased  
21 model results for the main atmospheric pollutants (O<sub>3</sub>, NO<sub>2</sub>, SO<sub>2</sub>, PM<sub>10</sub> and PM<sub>2.5</sub>) are  
22 analysed and compared against 18 monitoring stations distributed within inland Portugal in an  
23 hourly basis. Statistical analysis shows that both bias-correction techniques improve the raw  
24 forecasts skills (for all the modelling systems and pollutants). In the case of O<sub>3</sub> max-8h,  
25 correlation coefficients improve in 19-45 %, from 0.56-0.81 (raw models) to 0.78-0.86  
26 (corrected models). PM<sub>2.5</sub> also present significant improvements, e.g., correlation coefficients  
27 increase in more than 50% (both techniques) reaching values between 0.50-0.64. The corrected  
28 primary pollutant NO<sub>2</sub> and SO<sub>2</sub> demonstrate significant relative improvements compared to O<sub>3</sub>,  
29 mostly because the original modelling system skills are lower for those species. Despite the  
30 applied techniques have different mathematic formulation and complexity level, there are  
31 comparable answers for all of the forecasting systems. Analysis performed over specific  
32 situations, such as air quality episodes, not-validated or missing data reveals different behaviour  
33 of the bias-correction techniques under study. The results confirm the advantage of the  
34 application of bias-correction techniques for air quality forecast. Both techniques can be applied

35 routinely in an operational forecast system and they will be useful to alerts for the population  
36 about accurate exceedances.

37

38 **KEYWORDS:** air quality forecast, modelling systems, bias correction, multiplicative ratio,  
39 Kalman filter.

40

## 41 **1. INTRODUCTION**

42 Air quality forecasting is both a challenge and a scientific problem, which has recently emerged  
43 as a major priority in many urbanised and industrialised countries due to the increasing  
44 consciousness of the effect, on health and environment, caused by airborne pollutant emissions.  
45 Furthermore, is one of the requirements of the Air Quality Framework Directive (2008/50/EC)  
46 and a key issue of the Clean Air for Europe (CAFE) Programme (Cuvelier et al., 2007). The  
47 goals of reliable air quality forecasts are obvious: population exposure can be more efficiently  
48 reduced and protected by means of information and short-term action plans.

49 For that, European legislation settled ambient air quality standards for acceptable levels of air  
50 pollutants (like O<sub>3</sub>, NO<sub>2</sub>, SO<sub>2</sub>, PM<sub>2.5</sub> and PM<sub>10</sub>) and also recommended the use of modelling  
51 tools to assess and to forecast the air quality, in order to develop emission abatement plans and  
52 alert the population when health-related issues occur. In some European member states, like  
53 Portugal, air pollution limit values, namely for PM<sub>10</sub> and ground-level O<sub>3</sub>, are being exceeded  
54 every year and during long-term periods (Monteiro et al., 2007a; Carvalho et al., 2010; EEA,  
55 2010).

56 Several operational air quality forecasting systems already exist over Europe (see  
57 <http://gems.ecmwf.int> or <http://www.chemicalweather.eu>). Some of them forecast at the  
58 national level as in Portugal. In particular the MM5-CHIMERE (Monteiro et al., 2005), the  
59 MM5-EURAD-IM (Elbern et al., 2007; Strunk et al., 2010) and the CALIOPE (Baldasano et al.,  
60 2008a) forecasting systems are advancing our understanding of atmospheric dynamics in  
61 Portugal as follows. First, they are applied with a higher resolution over Portugal. Meanwhile  
62 most European models use a horizontal resolution of at least 25 x 25 km<sup>2</sup>, the MM5-CHIMERE,  
63 the MM5-EURAD-IM and the CALIOPE systems use horizontal resolution of 10x10 km<sup>2</sup>, 5x5  
64 km<sup>2</sup> and 4x4 km<sup>2</sup>, respectively. Second, they include contribution of Saharan dust emissions on  
65 an hourly basis from the BSC-DREAM8b model. Third, there are several evaluation studies that  
66 support the confidence on the three selected systems (MM5-CHIMERE in Monteiro et al.  
67 2007a,b; MM5-EURAD-IM in Monteiro et al., 2011; and CALIOPE in Baldasano et al., 2008a,  
68 2011 and Pay et al., 2011).

69 Air quality forecast modelling, which rely not only on the meteorological prediction but also on  
70 a chemical-transport modelling and on highly uncertain emission inventories, are likely to have  
71 significant (systematic) model errors (Borrego et al., 2003, 2008; Chang and Hanna, 2004). In  
72 order to improve each model forecast skill, different bias-correction techniques have been  
73 recently applied and examined (McKeen et al., 2005; Wilczak et al., 2006; Pagowski et al.,  
74 2006; van Loon et al., 2007; Djalalova et al., 2010; Sicardi et al., 2011).

75 The objective of the present study is to examine the efficacy of two bias-correction techniques,  
76 multiplicative ratio and Kalman filter methods, to improve the air quality forecasts (ground-  
77 based concentrations of O<sub>3</sub>, NO<sub>2</sub>, SO<sub>2</sub>, PM<sub>10</sub> and PM<sub>2.5</sub>) calculated from the three operational  
78 modelling systems available at high resolution over Portugal mainland domain. The model  
79 evaluation exercise covers the full year 2010 and observation from 18 air quality monitoring  
80 stations.

81 The present work is organized as follows. Section 2 describes the different forecast modelling  
82 systems. Section 3 presents the observational dataset selected and used within this study. The  
83 applied bias techniques are described in Section 4 and the analysis and discussion of the results  
84 are presented in Section 5. In Section 6, classical/categorical statistics are addressed to  
85 investigate the forecast skills after bias correction. Finally, the conclusions are drawn in Section  
86 7.

87

## 88 **2. THE AIR QUALITY FORECASTING SYSTEMS**

89 There are three air quality forecasting systems operating over Portugal with high resolution.  
90 Both MM5-CHIMERE (Monteiro et al., 2005) and MM5-EURAD-IM (Elbern et al., 2007;  
91 Strunk et al., 2010) modelling systems are being applied by the University of Aveiro's research  
92 group using an European/Iberian Peninsula coarse domain as boundary and initial conditions for  
93 the nested domain over Portugal with a 10x10 km<sup>2</sup> and a 5x5 km<sup>2</sup> horizontal resolution,  
94 respectively. The MM5-CHIMERE modelling system is operational with daily forecasts  
95 available since 2007: [http://adamastor.dao.ua.pt/previsao\\_qar/](http://adamastor.dao.ua.pt/previsao_qar/). The MM5-EURAD-IM is only  
96 operational for Portugal since 2010, with also daily forecasts in an hourly basis, as a result of a  
97 scientific collaboration between the University of Aveiro and the Rhenish Institute for  
98 Environmental Research at the University of Cologne. The CALIOPE system (Baldasano et al.,  
99 2008a) provides high-resolution air quality forecast for Spain. CALIOPE system encompasses a  
100 set of models: WRF-ARW meteorological model, the High-Effective Resolution Modelling  
101 Emission System (HERMES, Baldasano et al., 2008b) and the chemical transport model  
102 CMAQ. CALIOPE is applied over Iberian Peninsula with a 4 x 4 km<sup>2</sup> horizontal resolution and

103 also with an hourly basis (Baldasano et al., 2011). Current forecasts and near real-time  
104 evaluation are available through the CALIOPE system website (<http://www.bsc.es/caliope>).  
105 CMAQ, CHIMERE and EURAD-IM are all regional-scale three-dimensional chemical  
106 transport models (CTM) designed for short-term and long-term simulations of oxidants and  
107 aerosol formation. Both CHIMERE and EURAD-IM CTM are forced by the MM5  
108 meteorological fields (Dudhia, 1993), meanwhile CMAQ uses the outputs of the WRF-ARW  
109 model (Michalakes et al., 2004). Both MM5 and WRF-ARW are non-hydrostatic models.  
110 The three modelling system have different degrees of complexity and spatial resolution. A  
111 summary of their key features, including emissions and boundary conditions, is listed in Table  
112 1. Additional descriptions can be consulted on the online Model Documentation System  
113 (<http://pandora.meng.auth.gr/mds/mds.php>). CALIOPE configurations in both European and  
114 Iberian Peninsula domains are described in detail in Pay et al. (2010) and Baldasano et al.  
115 (2011), respectively.

116

117 Table 1

118

119 Since episodic natural of dust outbreaks are frequently observed over all Iberian Peninsula  
120 (Rodríguez et al., 2001; Silva et al., 2003; Basart et al., 2009), and because the representation of  
121 these events cannot be well simulated with solely the information of aerosol boundary  
122 conditions (Vautard et al., 2005, Jiménez-Guerrero et al., 2008; Menut and Bessagnet, 2010),  
123 the long-range transport of mineral dust from Sahara desert is modelled on an hourly basis by  
124 the BSC-DREAM8b model (Nickovic et al., 2001; Pérez et al., 2006a,b). The BSC-DREAM8b  
125 is fully embedded within the NCEP/Eta meteorological driver (Janjic, 1994). Dust aerosols are  
126 represented by 8 bins size distribution within the 0.1-10  $\mu\text{m}$  radius range. Dust-radiation  
127 interactions are calculated online. The modelled domain in this study comprises Northern  
128 Africa, the Mediterranean basin, Europe and Middle East. It is applied with a  $0.3^\circ \times 0.3^\circ$   
129 horizontal resolution using 24 vertical layers extending up to 15 km. In the present study the  
130 BSC-DREAM8b model is offline coupled within the hourly forecast PM<sub>10</sub> and PM<sub>2.5</sub>  
131 concentrations from CALIOPE, MM5-CHIMERE and MM5-EURAD-IM.

132

### 133 3. MONITORING DATA

134 The air quality monitoring network of mainland Portugal (<http://www.qualar.org/>) includes 68  
135 stations of which 42 are background, 19 traffic and 7 industrial, following the classification in  
136 Garber et al. (2002). The spatial coverage, together with the background influence and a

137 minimum data collection efficiency of 75% are three of the criteria used for the stations  
138 selection. A fourth criterion is related with the measured pollutants. Stations that measure O<sub>3</sub>  
139 also measure NO<sub>2</sub> and the stations that measure PM10 also do it for PM2.5. As a result, a total  
140 of 18 stations (8 rural, 5 urban and 5 suburban) are selected for the present study, 13 stations for  
141 O<sub>3</sub>/NO<sub>2</sub>, 9 for SO<sub>2</sub> and 6 for PM10/PM2.5. Figure 1 shows the location and main characteristics  
142 of the selected stations over the study domain. Note that the measured data are in an hourly  
143 basis and the data are not validated since they refer to year 2010.

144

145 Figure 1

146

147 Despite the spatial coverage criteria, there is an evident concentration of monitoring stations  
148 over the coastal area and the two metropolitan areas of Porto and Lisbon (see Figure 1).  
149 Nevertheless, all the regions of Portugal are covered by at least one rural background station. In  
150 terms of topography, the mountainous regions are not so well represented by monitoring sites.  
151 The majority of the stations, which are located near/over the coast, have altitudes lower than  
152 300 meters.

153

#### 154 **4. BIAS-CORRECTION TECHNIQUES**

155 As discussed in previous works, the applied forecast systems are found to have significant  
156 biases (Monteiro et al., 2007a; Baldasano et al., 2010) that could be removed through bias-  
157 correction techniques. There are several techniques by which bias correction can be applied as  
158 mean subtraction (McKeen et al., 2005; Wilczak et al., 2006), multiplicative ratio adjustment  
159 (McKeen et al., 2005), hybrid forecast (Kang et al., 2008) and Kalman filter (Delle Monache et  
160 al., 2006; Kang et al., 2008; Djalalova et al., 2010), model ensembles (van Loon et al., 2007;  
161 Wilczak et al., 2006; Djalalova et al., 2010) among others. The bias correction does not try to  
162 gain additional insight into model deficiencies or performance neither to correct them  
163 artificially, but intends to remove potential errors intrinsic to each model formulation or input  
164 data. In the present study two post-processing methods are used to correct the bias of the three  
165 forecasting system for all the considered pollutants: a multiplicative ratio correction (McKeen et  
166 al., 2005) and the Kalman filter method (Delle Monache et al., 2006; Kang et al., 2008, 2010).  
167 Both techniques are site-specific approaches since they use past ground-based measurements  
168 and simulated data at each monitoring site to revise and improve the current hourly forecasts for  
169 the entire year of 2010.

170

#### 171 **4.1 The multiplicative ratio correction**

172 The multiplicative ratio correction (RAT, McKeen et al., 2005) is a simple approach that can be  
173 mathematically expressed by equation 1.

$$174 \quad C_{model}^{corrected}(h, day) = \frac{\sum_{day=1}^n C^{obs}(h, day)}{\sum_{day=1}^n C_{model}^{raw}(h, day)} \times C_{model}^{raw}(h, day) \quad (1)$$

175  
176 The corrected concentration with RAT ( $C_{model}^{corrected}$ ) is estimated based on the application of a  
177 correction factor to the raw modelled concentration ( $C_{model}^{raw}$ ). The correction factor is calculated  
178 as the quotient between the additions of observed ( $C^{obs}$ ) and modelled  $C_{model}^{raw}$  concentrations at a  
179 particular hour (h) of the n previous days. To estimate the number of previous days (n),  
180 Monteiro et al. (2011) tested different training periods and chosen a 4 day training period as a  
181 compromise between having a sufficiently long period to gather adequate statistics, but not too  
182 long to mask seasonal variations (for O<sub>3</sub>, for e.g.). According to Stull (1988) and also Tchepel  
183 and Borrego (2010), synoptic conditions are characterized by a 3-4 day period, which supports  
184 the chosen training period. Thus, the current multiplicative ratio correction approached was  
185 applied with a 4 day period (RAT04).

186

#### 187 **4.2. Kalman filter**

188 The Kalman filter (KF) is a recursive, linear, and adaptive method that has been used recently to  
189 improve air quality forecast of ground-based O<sub>3</sub> (Delle Monache et al., 2006, 2008; Kang et al.,  
190 2008; Djalalova, et al., 2010; Sicardi et al., 2011) and PM<sub>2.5</sub> (Dajalalova, et al., 2010; Kang et  
191 al., 2010). KF performance is sensitive to the error ratio ( $\sigma_{\eta}^2/\sigma_{\epsilon}^2$ ) which indicates the way in  
192 which the KF responds to the variations in biases at prior steps. There exists an optimal error  
193 ratio to generate the best forecast given the forecast modelling system and the dynamic of the  
194 study area. We follow the methodology of Kang et al. (2008) for estimating the optimal error  
195 ratio which consists in minimizing the root mean square error and maximizing the correlation  
196 coefficient for all the stations. Therefore, optimal errors ratios are selected for each modelling  
197 system and for all the selected stations over the year 2010. Only in the case of O<sub>3</sub>, optimal errors  
198 ratios are selected seasonality because it was found that corrected O<sub>3</sub> simulation improved when  
199 using seasonally varying values. Table 2 presents the optimal error ratios selected for each  
200 pollutant.

201

202 Table 2

203

## 204 **5. BIAS-CORRECTION ASSESSMENT**

205 The evaluation of the different bias-correction approaches applied to the three modelling system  
206 is carried out using classical statistical indicators (Tilmes et al., 2002; Borrego et al., 2008;  
207 Denby et al., 2010; Dennis et al., 2010). The global skills of the bias-correction approaches are  
208 represented by means Taylor diagrams. Additionally, this evaluation is complemented with  
209 analysis of the most important critical points of each bias-correction technique find on the air  
210 quality forecast of the three modelling systems under study.

211 The Taylor diagram (Taylor, 2001) is a powerful tool frequently used in model evaluation  
212 studies (Cuvelier et al., 2007; Denby et al., 2010; Dennis et al., 2010) for the simultaneous  
213 visualization of three statistical indicators, in the present study we present the observed and  
214 modelled standard deviation (SD), the centred root mean square error (CRMSE) and the  
215 correlation coefficient (R) in a single point. Together these statistical parameters provide a quick  
216 outline of the degree of pattern correspondence among the raw and the unbiased simulated  
217 values of each forecasting system and the observed data.

218 Figure 2 shows the Taylor diagrams for each pollutant. O<sub>3</sub> is expressed in maximum daily  
219 concentration (O<sub>3</sub> max-1h) (Figure 2a) and in maximum daily eight-hour running average (O<sub>3</sub>  
220 max-8h) (Figure 2b) following the current 2008/50/EC European directive (European  
221 Commission, 2008). NO<sub>2</sub>, SO<sub>2</sub>, PM10 and PM2.5 are expressed in daily mean concentrations  
222 (Figure 2c-f, respectively). Each Taylor diagram shows the annual performance of the two bias-  
223 correction techniques, KF and RAT04, applied to the three forecasting systems and the  
224 corresponding raw modelling systems over all the studied stations.

225

226 Figure 2

227

228 Visualization of every single polar plots shows that the application of both KF and RAT04  
229 techniques improve the raw forecasts for all the modelling systems and pollutants, bringing  
230 unbiased SD closer to the observed SD than raw modelled SD, reducing errors and increasing  
231 correlation coefficients close to the unit. For O<sub>3</sub> max-1h the improvements in annual  
232 performance is significant after applying bias-correction techniques. The maximum variability  
233 increases with KF (SD = 25.2-28.0 μg.m<sup>-3</sup>) and RAT04 (SD = 26.4-29.5 μg.m<sup>-3</sup>) falling closer to  
234 the observed SD (29.3 μg.m<sup>-3</sup>) than raw modelled SD (22.1-26.9 μg.m<sup>-3</sup>), which means that  
235 techniques adjust high O<sub>3</sub> peaks although they are still slightly underestimated. Annually,

236 unbiased error decreased in 21-22% (KF) and 16-26%, from 20.4-25.2  $\mu\text{g.m}^{-3}$  (raw model) to  
237 16.2-19.6  $\mu\text{g.m}^{-3}$  (KF) and 17.1-19.2  $\mu\text{g.m}^{-3}$  (RAT04); and correlation coefficient increase in  
238 16-34% (KF) and 13-37% reaching 0.75-0.84 (KF) and 0.77-0.83 (RAT04). The same tendency,  
239 but with slight better skills, is found in the case of  $\text{O}_3$  max-8h. Although the variability is  
240 improved, the unbiased standard deviations are usually smaller than their observed field.  
241 CRMSE is reduced in 25-26% (KF) and 25-33% (RAT04) and correlation coefficient range  
242 between 0.78-0.85 and 0.81-0.86, respectively.

243 In the case of  $\text{NO}_2$  daily mean, after applying bias-correction techniques unbiased concentration  
244 increase the daily variability getting closer to the observed SD (14.0  $\mu\text{g.m}^{-3}$ ) from 7.7-11.2  
245  $\mu\text{g.m}^{-3}$  (raw model) to 12.4-12.9  $\mu\text{g.m}^{-3}$  (KF) and 14.1-14.6  $\mu\text{g.m}^{-3}$  (RAT04) showing slightly  
246 more daily variability with RAT04. CRMSE decreases from 10.6-12.2  $\mu\text{g.m}^{-3}$  (raw model) to  
247 7.2-7.3  $\mu\text{g.m}^{-3}$  (KF) and 5.9-6.7  $\mu\text{g.m}^{-3}$  (RAT04); and temporal correlations increase from 0.55-  
248 0.66 (raw model) to 0.85-0.86 (KF) and 0.89-0.91 (RAT04).

249 As for  $\text{NO}_2$  primary pollutant, annual unbiased modelled  $\text{SO}_2$  daily means present higher skills  
250 than raw modelled concentrations. Raw modelled  $\text{SO}_2$  concentrations present higher daily  
251 variability (SD = 4.1-12.7  $\mu\text{g.m}^{-3}$ ) than observed field (2.4  $\mu\text{g.m}^{-3}$ ). In this sense, both bias-  
252 correction techniques get to deduced raw modelled SD till 3.3-7.3  $\mu\text{g.m}^{-3}$  (KF) and 3.1-7.3  
253  $\mu\text{g.m}^{-3}$  (RAT04) which means that high  $\text{SO}_2$  peaks have been reduced and decreased the daily  
254 concentration. Annual CRMSE are reduced in 34-75% after applying bias-correction techniques  
255 in the range of 1.6-5.8  $\mu\text{g.m}^{-3}$  (KF) and 2.1-5.7  $\mu\text{g.m}^{-3}$  (RAT04). Unbiased models also improve  
256 temporal annual correlation in more than 100%, reaching 0.17-0.50 and 0.14-0.59 with KF and  
257 RAT04, respectively.

258 Raw modelled PM present higher daily variability than observations which is reduced after  
259 applying bias-correction techniques. For  $\text{PM}_{10}$ , raw modelled SD are reduced from 13.1-22.3  
260  $\mu\text{g.m}^{-3}$  to 1.3-18.3  $\mu\text{g.m}^{-3}$  (KF) and 14.1-16  $\mu\text{g.m}^{-3}$  (RAT04) closer to 13.6  $\mu\text{g.m}^{-3}$  (observed  
261  $\text{PM}_{10}$  SD).  $\text{PM}_{2.5}$  daily mean presents the same tendency, raw modelled concentrations are  
262 reduced from 8.0-13.7  $\mu\text{g.m}^{-3}$  to 7.1-10.2  $\mu\text{g.m}^{-3}$  (KF) and 6.8-8.9  $\mu\text{g.m}^{-3}$  (RAT04) closer to 7.5  
263  $\mu\text{g.m}^{-3}$  (observed  $\text{PM}_{2.5}$  SD). The higher variability observed with  $\text{PM}_{10}$  and  $\text{PM}_{2.5}$ , even after  
264 applying bias-correction techniques, is deviated by the high overestimation urban stations such  
265 as CAM. Temporal variability improves for  $\text{PM}_{10}$ , in the range of 7-20 % (KF) and 12-33%  
266 (RAT), reaching correlation of 0.49-0.58 (KF) and 0.58-0.61 (RAT04). Improvements are  
267 higher with  $\text{PM}_{2.5}$  for temporal variability, (>50% with both KF and RAT04) reaching R in the  
268 range of 0.50-0.64 (KF) and 0.57-0.62 (RAT04).

269 Based on annual statistics indicator, the biggest percentage of improvement after applying bias-  
270 correction techniques are found for  $\text{SO}_2$  daily concentration, R increase in more than 100% and



271 error (CRMSE) decrease in the range of 34-51 % (for both KF and RAT04), following by NO<sub>2</sub>  
272 daily concentration where R increase in 30-65% and error decrease in 32-51% (for both KF and  
273 RAT04). The percentage of improvement is smaller for O<sub>3</sub> max-1h and max-8h, although with  
274 significant impact in correlation that reach 0.78-0.86 (both KF and RAT04) in the case of O<sub>3</sub>  
275 max-8h, since the raw modelled present high skills. Note that to get high skills after applying  
276 bias-correction techniques modelling systems has to demonstrate their relative accuracy.  
277 Overall, Taylor diagrams (Figure 2) point out that despite the applied techniques have different  
278 mathematic formulation and complexity level, there is comparable answers for all of the  
279 forecasting systems (see e.g. Figure 2c). There is a slightly superiority of RAT04 technique over  
280 Kalman filter in terms of statistical indicator and graphical representation of Taylor diagrams.  
281 However the aforementioned evaluation has the limitation that it is done over all the stations in  
282 annual basin and it gives no information whether the unbiased concentrations are correct for the  
283 right or wrong reason. Therefore, in order to go more in detail on the skills of bias-correction  
284 techniques specific examples of the successes/failures of both techniques are illustrated  
285 following, since is important to know how RAT04 and KF behave in specific situations, such as  
286 air quality episodes, not-validated or missing data, in order to choose the most convenient bias-  
287 correction technique to apply on air quality forecast over Portugal.

288 In Figure 3 (top) where the hourly observed O<sub>3</sub> concentrations (red points) at the CAL station is  
289 presented along with the raw CALIOPE outputs (blue) and the post-processed KF and RAT04  
290 values (orange and green, respectively) during a summer period (June month). This example  
291 demonstrates how both KF and RAT04 techniques improve the forecasted O<sub>3</sub> daily cycles, since  
292 they agree with the observed hourly variability in both diurnal maximum and night minimum,  
293 reducing the persistent overestimation with respect to measurements (Figure 3, bottom). Hourly  
294 statistical analyses (not shown here) quantify that maximum and minimum annual bias are in  
295 the range of  $\pm 5 \mu\text{g.m}^{-3}$  after post-processing with both KF and RAT04. That means a bias  
296 improvement of more than 80% in the maximum overestimation (from 40-20  $\mu\text{g.m}^{-3}$  to less than  
297 5  $\mu\text{g.m}^{-3}$ ) for all the system.

298

299 Figure 3

300

301 Figure 4 shows PM10 time series at FUN station during an air quality episode in August 2010.  
302 In the first part of time series, from August 7<sup>th</sup> to 10<sup>th</sup>, a desert dust outbreaks arrives to Portugal  
303 due to a North Africa advection (Figure 4 c). The raw CALIOPE system reproduces the event  
304 thanks to the contribution of the BSC-DREAM8b model (Figure 4b) although the  
305 concentrations are slightly underestimated. After applying bias-correction techniques, unbiased

306 outputs are closer to the hourly observed concentrations. In the second part, from August 10<sup>th</sup> to  
307 13<sup>th</sup>, the wind changes the trajectory to northwest (see Figure 4c) and the observed  
308 concentrations reach ~170  $\mu\text{g}\cdot\text{m}^{-3}$ . According to the Portuguese Forest Authority (Autoridade  
309 Florestal Nacional, 2010) nine forest fires occurred during this period in a radius of 100 km  
310 from FUN station where more than 10,000 ha were burned. In the described fire episode both  
311 bias-correction techniques do not reproduce the event since the raw CALIOPE modelling  
312 system, as MM5-CHIMERE and MM5-EURAD-IM systems, does not include forest fire  
313 emissions. The high bias estimated for this episode generates that both techniques overestimate  
314 observed concentration four days later after the fire is finished. KF gets closer to the  
315 observations faster than RAT04 since KF gradually spreads the error and RAT04 present high  
316 sensitivity to the magnitude of the modelled values.

317

318 Figure 4

319

320 Frequent problems in the forecast of  $\text{SO}_2$  are associated to high underestimations of  $\text{SO}_2$  peaks.  
321 The main activity sources of  $\text{SO}_2$  emissions are related to power plants and  
322 transformation/manufacturing industry (source: <http://www.emep.int/>). Besides a high level of  
323 control of the  $\text{SO}_2$  emissions, these point sources can episodically generate large plumes of  
324 high- $\text{SO}_2$  content affecting the air quality in urban and regional scales downwind the sources.  
325 Accurate  $\text{SO}_2$  forecasts depend on the accuracy in the meteorological patterns, the variability on  
326 the sub-grid scale with respect to measured data (Stern et al., 2008; Baldasano et al., 2011), and  
327 the accurate representation of emissions sources.

328 The Figure 5 illustrates an episode of high  $\text{SO}_2$  concentrations at the CHA station, on March  
329 27<sup>th</sup> from 6:00 to 12:00 where any of the forecast systems were able to predict the observed  
330 event (only MM5-EURAD-IM is shown in Figure 5). This example demonstrates that both KF  
331 and RAT04 produce an error due to high concentrations observed on March 27<sup>th</sup> which is  
332 propagated to the same hour during the days after. The propagated error is higher for RAT04  
333 than KF since RAT04 is a simple technique by which simulated and observed data have the  
334 same weight. RAT04 applies a correction on the same hour of the next days and if there is no  
335 other high concentration during 4 days, the hourly correction factor error will not be reproduced  
336 on the 5<sup>th</sup> day after. On the other hand, the optimal ratio of KF to MM5-EURAD-IM is low  
337 (0.04, see Table 2) which means that KF has more confidence on model simulations than  
338 observations data. In this sense, the propagated error by KF is less than RAT04 error. In  
339 addition, if no other high concentration is recorded, KF error will decrease over the next days,  
340 meaning that corresponding bias will be getting closer to 0. The propagation of an error

341 produced by model simulations or observations data (both by a high recorded concentration and  
342 by not validated data) is a common characteristic of both techniques. This example illustrates  
343 that despite RAT04 has a better performance in general terms, KF can generate a correction  
344 with less error in these specific situations.

345

346 Figure 5

347

348 The Figure 6 shows an episode registered in October 25<sup>th</sup> - 30<sup>th</sup> at the MVE station where the  
349 raw CALIOPE system forecasted high SO<sub>2</sub> concentrations that actually did not occur. The same  
350 behaviour was obtained with the raw MM5-CHIMERE and MM5-EURAD-IM forecasting  
351 systems (not shown here). The figure demonstrates the limitations of the KF technique against  
352 high overestimation of the models. Meanwhile, the RAT04 technique (green) corrects the raw  
353 forecast following the hourly observation with a bias reduction of 80%. This poor performance  
354 of KF is related with two facts. First, SO<sub>2</sub> optimal error ratio ( $\sigma_{\eta}^2/\sigma_{\varepsilon}^2$ ) for the three models result  
355 between 0.13 and 0.20, higher compared to the other pollutants ratios (see Table 2). When ratio  
356 is high, the forecast-error white-noise variance ( $\sigma_{\varepsilon}^2$ ) will be relatively small compared to the true  
357 forecast-bias white-noise variance ( $\sigma_{\eta}^2$ ). Therefore, the filter will put excessive confidence on  
358 the previous forecast and the predicted bias will respond very quickly to previous forecast  
359 errors. Second, KF bias-adjustment is a linear and recursive algorithm. KF predicts the future  
360 bias with a linear relationship given by the previous bias estimate plus a quantity proportional to  
361 the difference between the present forecast error and the previous bias estimates. Therefore KF  
362 is unable to correct large bias due to model overestimations when all the biases for the past few  
363 days have been small.

364

365 Figure 6

366

367 The absence of monitoring data is frequently a problem for data assimilation or bias-correction  
368 procedures. In case of the RAT04 approach, if there are no measurements, the unbiased outputs  
369 will be equal to the raw modelled data. On the other hand, KF has a capacity to learn the  
370 behaviour of simulations data relatively to monitoring data, which means that KF is designed to  
371 apply the same correction as that estimated for the previous days. Figure 7 illustrates this  
372 problem with an example of two different periods of absence of measurement data registered at  
373 the CAL station, from April 10<sup>th</sup> to May 1<sup>st</sup> 2010. Once all of the forecast systems presented the  
374 same behaviour, just the MM5-EURAD-IM simulation is shown here. In the first half period

375 (from April 10<sup>th</sup> to the half of April 14<sup>th</sup>) KF and RAT04 produce a reasonable corrections with  
376 bias values closer to 0 (Figure 6, bottom). During the periods of April 14<sup>th</sup> - 18<sup>th</sup> and April 23<sup>rd</sup> -  
377 25<sup>th</sup>, there are no monitoring data, In this case, KF applies the same correction from previous  
378 days and RAT04 does not correct the simulated data, taking the same raw modelled outputs.  
379 When data start to be available, KF continues to apply the bias correction base on previous days  
380 and after four days the recent measurement have an effective effect on bias correction (observed  
381 and simulated data). With the RAT04 technique the simulated data is only possible to be  
382 corrected after 4 days of monitoring data availability. In future work RAT04 can be  
383 improved/designed in order to minimize this problem, applying the previous correction to the  
384 hour without observed data, as KF does.

385

386 Figure 7

387

388 Both techniques are sensitive to not validated data which is a frequent problem for time  
389 forecasting mode working. Figure 8 shows an example of not validated data, specifically, when  
390 the station presents a calibration problem. Time series of hourly SO<sub>2</sub> concentrations (red) at the  
391 MVE station present two clear tendencies in Figure 8. In the first part, SO<sub>2</sub> measured  
392 concentrations present a background level  $\sim 8 \mu\text{g.m}^{-3}$ , and on June 22<sup>sd</sup> at 12:00 observed data  
393 decrease sharply  $7 \mu\text{g.m}^{-3}$  to be oscillating around  $\sim 1 \mu\text{g.m}^{-3}$  the rest of the year. This suggests  
394 that MVE station registered/exhibited a calibration problem in the first part of the time series  
395 that is corrected in the second part. In this situation both KF and RAT04 correct the raw forecast  
396 to agree with observations in the both aforementioned situations. On one hand KF presents a  
397 robust response against a systematic bias. KF gives more confidence to the observations based  
398 on persistent systematic bias, and adjusts the background levels to  $\sim 8 \mu\text{g.m}^{-3}$  in the first part,  
399 and to  $\sim 1 \mu\text{g.m}^{-3}$  in the second part, with a transition period of 4 days till the bias are reduce to  
400 0 (orange line, Figure 8 bottom). On the other hand RAT04 tries to adjust background levels in  
401 both situations, but produces overestimations during these periods. These instabilities show its  
402 sensitivity to high gradient of concentrations, and it is a limitation of multiplicative techniques  
403 (Wilczak et al., 2006).

404

405 Figure 8

406

## 407 **6. FORECAST MODELS PERFORMANCE**

408 The categorical statistical skills (Kang et al, 2005; Eder et al., 2006) are computed in order to  
409 evaluate how the two bias correction techniques improve the three air quality forecasts daily  
410 produced over Portugal in terms of exceedances and non-exceedances events. Exceedances  
411 analysis is based on a comparison with a fixed threshold concentration (T). The present work  
412 uses as thresholds those established by the European directive 2008/50/EC on air quality  
413 (European Commission, 2008). Only O<sub>3</sub> and PM10 are evaluated in terms of categorical  
414 statistics because neither PM2.5, NO<sub>2</sub> nor SO<sub>2</sub> exceeded the European limit values at the  
415 selected stations in 2010. The 2008/50/EC directive sets an information threshold of 180 µg.m<sup>-3</sup>  
416 for maximum daily concentrations (max-1h) and a target value of 120 µg.m<sup>-3</sup> for maximum  
417 daily eight-hour running average (max-8h) not to be exceeded on more than 25 days per year. In  
418 the case of PM10, it establishes a limit value of 50 µg.m<sup>-3</sup> for daily average (Mean-24h) not to  
419 be exceeded more than 35 times per year. Table 3 shows the annual categorical parameters for  
420 all the selected Portuguese stations. The calculated statistics are the accuracy (A), the bias (B),  
421 the probability of detection (POD), the false alarm ratio (FAR) and the critical success index  
422 (CSI). Kang et al. (2005) shows the formulas of the aforementioned categorical statistics.

423

424 Table 3

425

426 The percentages of the 2010 exceedances that are actually forecasted are estimated with the  
427 value of POD. For O<sub>3</sub> max-1h, a total of 51 exceedances of the information threshold (T = 180  
428 µg.m<sup>-3</sup>) are observed over the 13 Portuguese stations in 2010 (1/3\*(b+d), in Table 3). The bias-  
429 correction techniques increase the POD from 3% (raw models) to 10 % in KF and 31% in  
430 RAT04. In the case of O<sub>3</sub> max-8h, a total of 297 exceedances of the target value (T = 120 µg.m<sup>-3</sup>)  
431 were observed over all the stations. The POD also improves when bias-correction techniques  
432 are applied from 27% (raw) to 48% (KF) - 54% (RAT04). Overall, POD improves strongly after  
433 the post-processing techniques for both O<sub>3</sub> max-1h and max-8h, reaching an improvement of  
434 more than 100% and 50% for max-1h and max-8h, respectively. This means that by means the  
435 application of bias-correction techniques the forecast alerts for the population about  
436 exceedances would be significant accurate.

437 For PM10 daily mean a total of 68 exceedances of the daily limit value (T = 50 µg.m<sup>-3</sup>) were  
438 measured. The bias-correction techniques increase the POD from 32% (raw) to 34% (KF) - 45%  
439 (RAT04). However the improvement percentage of POD is less than 50%, lower than for O<sub>3</sub>,

440 due to the no significant increase of the number of hits (b) (from 65 (raw) to 70 (KF) and to 92  
441 (RAT04)).

442 The accuracy (A) measures the percentage of simulations that correctly reproduce an  
443 exceedance or no-exceedance (ideally 100%). Actually, A is already high for the raw models for  
444 three variables ( $A > 90\%$ ), and there are no significant improvements after post-processing. In  
445 the present study, careful must be done in the interpretation of the A since the number of the  
446 observed exceedances (b+d) is little respect to the total pair of data (a+b+c+d). The categorical  
447 bias (B) indicates if the forecasts fail by overestimating (false positive) or underestimating  
448 (correct negative) exceedances (ideally 1). For O<sub>3</sub> max-1h and max-8h, B remains below 1  
449 before and after post-processing, which indicates that errors by missing of observed  
450 exceedances are not totally resolved ( $d > a$ ). The better performance is found for O<sub>3</sub> max-8h,  
451 where B improves from 0.7 (raw) to 0.8 (KF and RAT04). Low B performance in O<sub>3</sub> max-1h is  
452 due to the poor capability to reproduce maximum hourly O<sub>3</sub> concentrations. On the other hand,  
453 for PM10 daily mean categorical bias originally presents problem with false alarms ( $B > 1$ ). B is  
454 significant reduced after post-processing, from 2.2 (raw) to 1.7 (KF) – 1.5 (RAT04).  
455 Nevertheless corrected models still present problems with false alarms.

456 FAR is useful to quantify the fails by simulating exceedances that actually did not occur (ideally  
457 0%). Application of the post-processing techniques reduces of almost the half the value of the  
458 FAR for the max-8h. This indicates the ability of the KF and RAT04 techniques to reduces the  
459 number of projected false alarms from 371(raw) to 254 (KF) – 266 (RAT04). For the O<sub>3</sub> max-1h  
460 the false alarms (b) does not improve significantly after the post-processing. However FAR  
461 improves owing to the improving of hits detections (b) from 5 (raw) to 16-48 (KF and RAT04,  
462 respectively). For PM10 daily mean, FAR improves less than 20% with both post-processing  
463 techniques, since the bias-correction techniques do not reduce significantly modelled false  
464 alarms (a) for PM10 daily mean. The CSI indicates how well both forecast exceedances and  
465 actual exceedances are predicted (ideally 100%). For the three analysed variables CSI improves  
466 when both KF and RAT04 techniques are applied. Unlike the POD and the FAR, the CSI takes  
467 into account both false alarms and missed events, and it is thus a more balanced score.

468 Results demonstrate that both techniques improve modelling skills to reproduce exceedances  
469 established by the European directive 2008/50/EC for PM10 daily mean and O<sub>3</sub> max-1h and  
470 max-8h. Better skills are found with RAT04 than for KF in most cases. Nevertheless, it must be  
471 taken into account that the categorical statistics only evaluate the model in terms of  
472 exceedances; therefore caution is needed when interpreted.

473

## 474 **7. SUMMARY AND CONCLUSIONS**

475 The current work performs an exhaustive examination of two different bias-correction  
476 techniques, the Kalman filter method (KF) and a multiplicative ratio with a 4 days training  
477 period (RAT04), within their application inland Portuguese domain. Both approaches have been  
478 applied to the three advanced forecasting systems operated routinely over Portugal in 2010 –  
479 CALIOPE, MM5-CHIMERE and MM5-EURAD-IM. The evaluation is carried on in terms of  
480 ground-based concentrations of gas-phase ( $O_3$ ,  $NO_2$ , and  $SO_2$ ) and particulate matter (PM10 and  
481 PM2.5) pollutants. Statistical parameters were used (classical and categorical) and graphical  
482 techniques (Taylor diagram and temporal series) in order to quantify the abilities of the two  
483 post-processing techniques to improve the air quality forecast over Portugal.

484 Comparative statistical analysis, based on Taylor diagram, show that both KF and RAT04  
485 techniques improve the raw forecasts skills (for all the modelling systems and pollutants),  
486 bringing unbiased SD closer to the observed SD than raw modelled SD, reducing errors and  
487 increasing correlation coefficients close to the unit. In the case of  $O_3$  max-8h, temporal  
488 variability improves in 19-45 % from 0.56-0.81 (raw models) to 0.78-86 (KF and RAT04,  
489 respectively). Similar tendency is found for  $O_3$  max-1h. The primary pollutant  $NO_2$  and  $SO_2$   
490 daily concentrations, demonstrate significant relative improvements compared to  $O_3$ , mostly  
491 because the original modelling system skills are lower for those species.  $NO_2$  correlation  
492 coefficients improve between 30-65% and more than 100% for  $SO_2$  (for both KF and RAT04);  
493 and errors decrease also in both cases in ~30-40% (for both KF and RAT04). For PM,  
494 improvement after applying both KF and RAT04 are higher with PM2.5 where correlation  
495 coefficients increase in more than 50% (both techniques) reaching values between 0.50 – 0.64.  
496 Note that to get high skills after applying bias-correction techniques modelling systems has to  
497 demonstrate their relative accuracy.

498 Despite the applied techniques have different mathematic formulation and complexity level,  
499 there are comparable answers for all of the forecasting systems. There is a slightly superiority of  
500 RAT04 technique over KF in terms of statistical indicator and graphical representation of  
501 Taylor diagrams. However the analysis performed over specific situations, such as air quality  
502 episodes, not-validated or missing data reveals different behaviour for KF and RAT04. In the  
503 case of hourly  $O_3$  concentrations, both bias-correction techniques are efficient tools to improve  
504 simulated  $O_3$  daily cycle remaining bias in the range of  $\pm 5 \mu g.m^{-3}$ . Under desert dust advection  
505 from North Africa, KF and RAT04 are able to correct PM10 bias within slightly overestimation  
506 of RAT04. Nevertheless, under missed pollution events of short-life (< 2 days), as shown with  
507 forest fire or high  $SO_2$  peaks, KF and RAT04 have no efficient corrections of that large bias.

508 RAT04 applies a correction on the same hour of the next days and if there is no other high  
509 concentration during 4 days, the hourly correction factor error will not be reproduced on 5<sup>th</sup> day  
510 after. In the other hand, the propagation of error in KF is less sharp than for RAT04, since give  
511 more confidence to previous persistent bias. This is an advantage of KF under not validated data  
512 or missing data since the capability of response is higher than RAT04. One evident  
513 disadvantage of KF against RAT04 is when the modelling system presents high overestimations  
514 (as shown with hourly SO<sub>2</sub> peaks). KF is unable to correct large bias due to model  
515 overestimations since the filter puts excessive confidence on modelled forecast. Note that both  
516 techniques are sensitive to not validated data.

517 The improvements of the discussed critical points will conduct to a better unbiased model  
518 performance which will be reflected on a higher accuracy of episodes forecasted. Beyond the  
519 discussed weaknesses of the both bias-correction approaches, there is a critical point that is  
520 common to KF and RAT04: both are site-specific dependents. We are currently working to  
521 solve this problem, developing a spatial approach for the bias correction on the overall domain.

522 Categorical analysis has been performed over air quality pollutant that exceed threshold and  
523 limit values establish by the European legislation on air quality which are O<sub>3</sub> max-1h (threshold  
524 = 180 µg.m<sup>-3</sup>), O<sub>3</sub> max-8h (threshold =120 µg.m<sup>-3</sup>) and PM10 daily mean (limit value=50 µg.m<sup>-3</sup>).  
525 Results indicate that the probability of detection (POD) of both techniques improve in more  
526 than 100% for O<sub>3</sub> max-8h and 50% for O<sub>3</sub> max-1h with a total increase from 27% to 48% (KF)  
527 and 54% (RAT04) in the case of O<sub>3</sub> max-8h. However, the improvement percentage of POD is  
528 less than 50%, lower than for O<sub>3</sub>, due to the no significant increase of the number of hits (b)  
529 (from 65 (raw) to 70 (KF) and to 92 (RAT04)), may be related with the fact that some missing  
530 sources (such as forest fires) are not includes in the raw modelling systems.

531 These above results confirm the advantage of the application of RAT04 and KF bias-correction  
532 techniques for air quality forecast. Both techniques can be applied routinely in an operational  
533 forecast system and they will be useful to alerts for the population about accurate exceedances.

534

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547

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## Figures captions

Figure 1: Location and main characteristics of the selected stations from the mainland Portuguese air quality monitoring network for 2010. (a) Station environments according to Garber et al. (2002) and the terrain elevation (in m). (b) Measured pollutants in each station.

Figure 2: Taylor diagram for each air quality system (CALIOPE, MM5-CHIMERE and MM5-EURAD-IM) and for each bias correction technique (KF and RAT04) over all selected monitoring stations. (a) O<sub>3</sub> max-1h; (b) O<sub>3</sub> max-8h (c) NO<sub>2</sub> daily mean; (d) SO<sub>2</sub> daily mean, and (e) PM10 daily mean; (f) PM2.5 daily mean. Black dots represent the reference point (observed data). The radial distances from the origin (0, 0) to the points are proportional to the standard deviations (in  $\mu\text{g}\cdot\text{m}^{-3}$ ). Azimuthal positions give the correlation coefficients. The distances between single points and reference point give the centred root mean square error (in  $\mu\text{g}\cdot\text{m}^{-3}$ ).

Figure 3: (Top) hourly O<sub>3</sub> time series ( $\mu\text{g}\cdot\text{m}^{-3}$ ) at the CAL station, estimated by the CALIOPE forecasting system (blue) and after the two bias correction techniques KF (orange) and RAT04 (green) from June 9<sup>th</sup> to 30<sup>th</sup>, 2010. (Bottom) hourly bias evolution ( $\mu\text{g}\cdot\text{m}^{-3}$ ) corresponding to CALIOPE forecasting system, KF and RAT04.

Figure 4: (a) hourly PM10 time series ( $\mu\text{g}\cdot\text{m}^{-3}$ ) at the FUN station for the CALIOPE forecasting system (blue line) and the two bias correction techniques KF (orange) and RAT04 (green) from August 5<sup>th</sup> to 16<sup>th</sup>, 2010. Area plot shows the modelled desert dust contribution (DD, light blue area) and anthropogenic contribution (CALIOPE-DD, dark blue area). (b) Desert dust concentration ( $\mu\text{g}\cdot\text{m}^{-3}$ ) forecast with the BSC-DREAM8b at 12h August 9<sup>th</sup> (available at [http://www.bsc.es/plantillaH.php?cat\\_id=521](http://www.bsc.es/plantillaH.php?cat_id=521)). (c) 5 day HYSPLIT back-trajectories ending at FUN station at different levels (500, 1000, 1500 m a.g.l.) for August 10<sup>th</sup>. (d) 3 day HYSPLIT back-trajectories ending at FUN station at different levels (500, 1000, 1500 m a.g.l.) for August 12<sup>th</sup>.

Figure 5: (Top) hourly SO<sub>2</sub> time series at the CHA station, measured and estimated with the MM5-EURAD-IM forecasting system (blue) values and applying the two bias correction techniques KF (orange) and RAT04 (green), from March 26<sup>th</sup> to April 1<sup>st</sup> 2010. (Bottom) bias evolution ( $\mu\text{g}\cdot\text{m}^{-3}$ ) corresponding to MM5-EURAD-IM forecasting system, KF and RAT04.

Figure 6: (Top) hourly SO<sub>2</sub> time series (μg.m<sup>-3</sup>) at the MVE station for the CALIOPE forecasting system (blue) and the two bias correction techniques KF (orange) and RAT04 (green) from October 25<sup>th</sup> to 30<sup>th</sup>. (Bottom) hourly bias evolution (μg.m<sup>-3</sup>) corresponding to CALIOPE forecasting system, KF and RAT04.

Figure 7: (Top) hourly O<sub>3</sub> time series at the CAL station for MM5-EURAD-IM forecasting system (blue) and the two bias correction techniques KF (orange) and RAT04 (green), from April 10<sup>th</sup> to May 1<sup>st</sup>, 2010. (Bottom) hourly bias evolution (μg.m<sup>-3</sup>) corresponding to MM5-EURAD-IM forecasting system, KF and RAT04.

Figure 8: (Top) hourly SO<sub>2</sub> time series (μg.m<sup>-3</sup>) at the MVE station for the MM5-CHIMERE system (blue) and the two bias correction techniques KF (orange) and RAT04 (green) from June 3<sup>th</sup> to July 3<sup>th</sup>. (Bottom) hourly bias evolution (μg.m<sup>-3</sup>) corresponding to MM5-CHIMERE forecasting system, KF and RAT04.



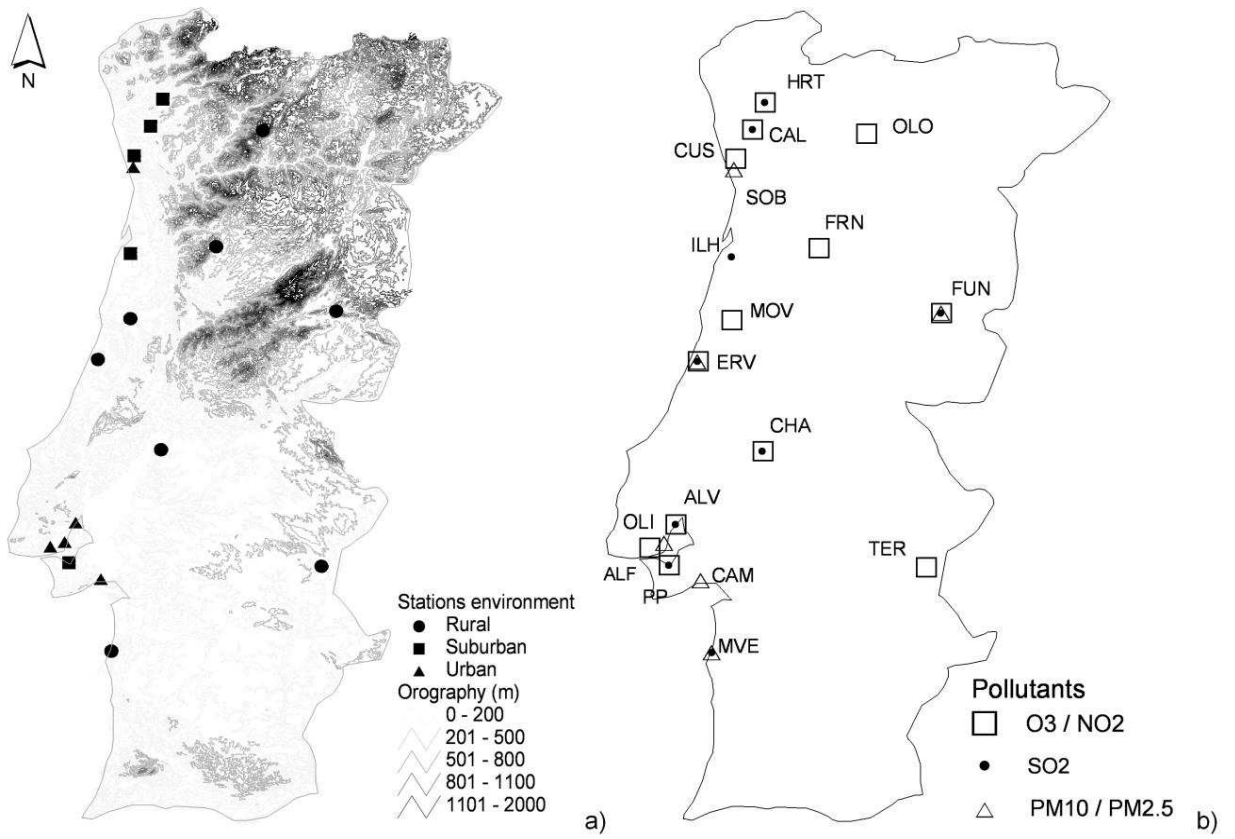


Figure 1.

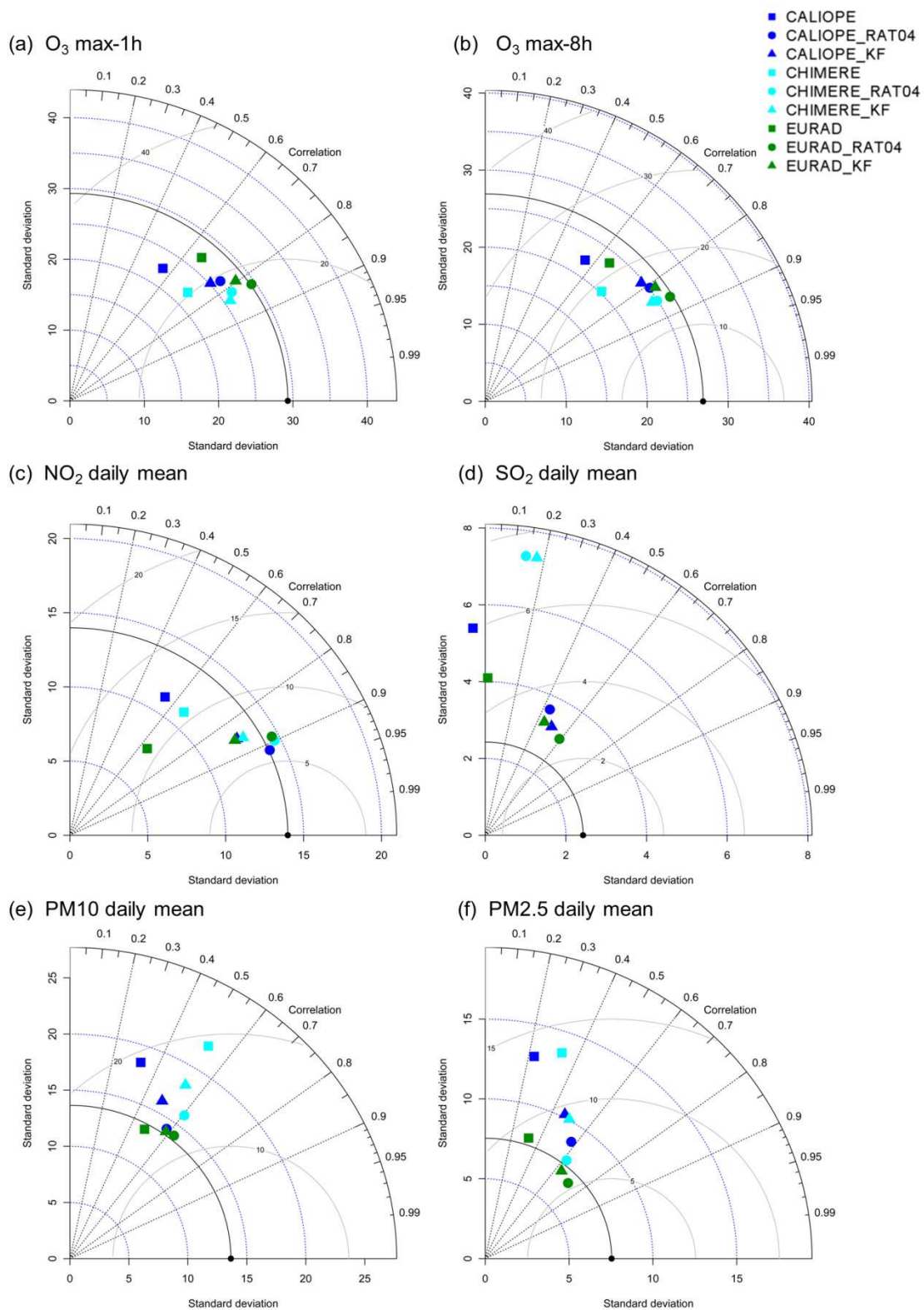


Figure 2.

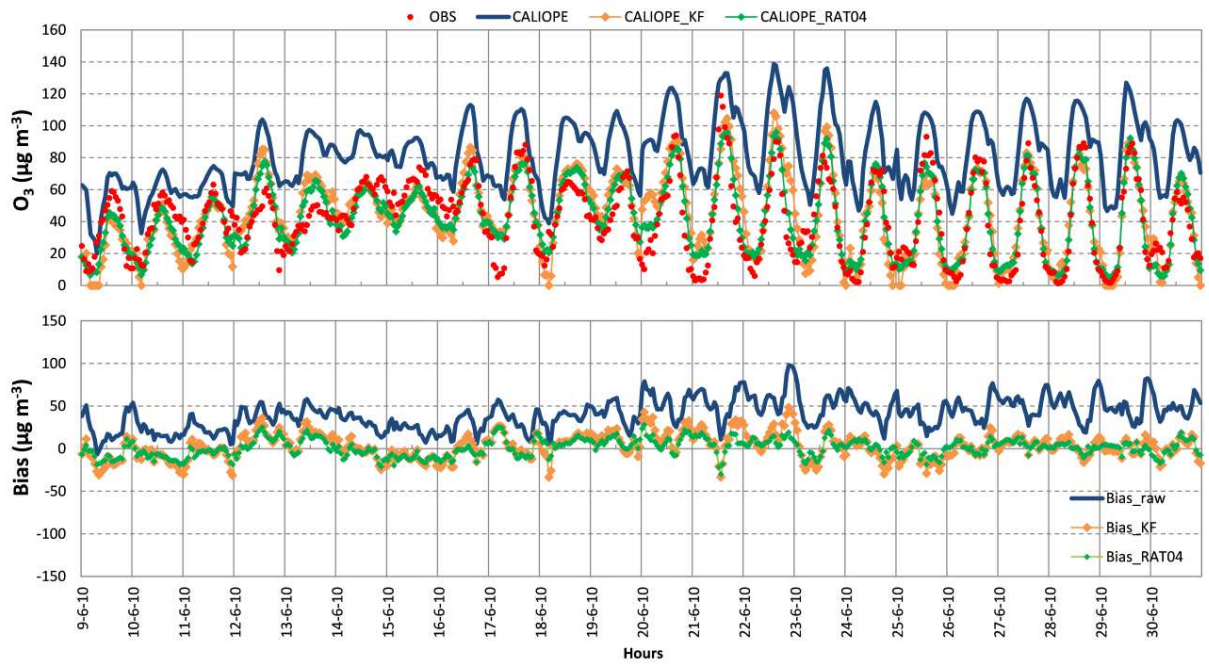


Figure 3.

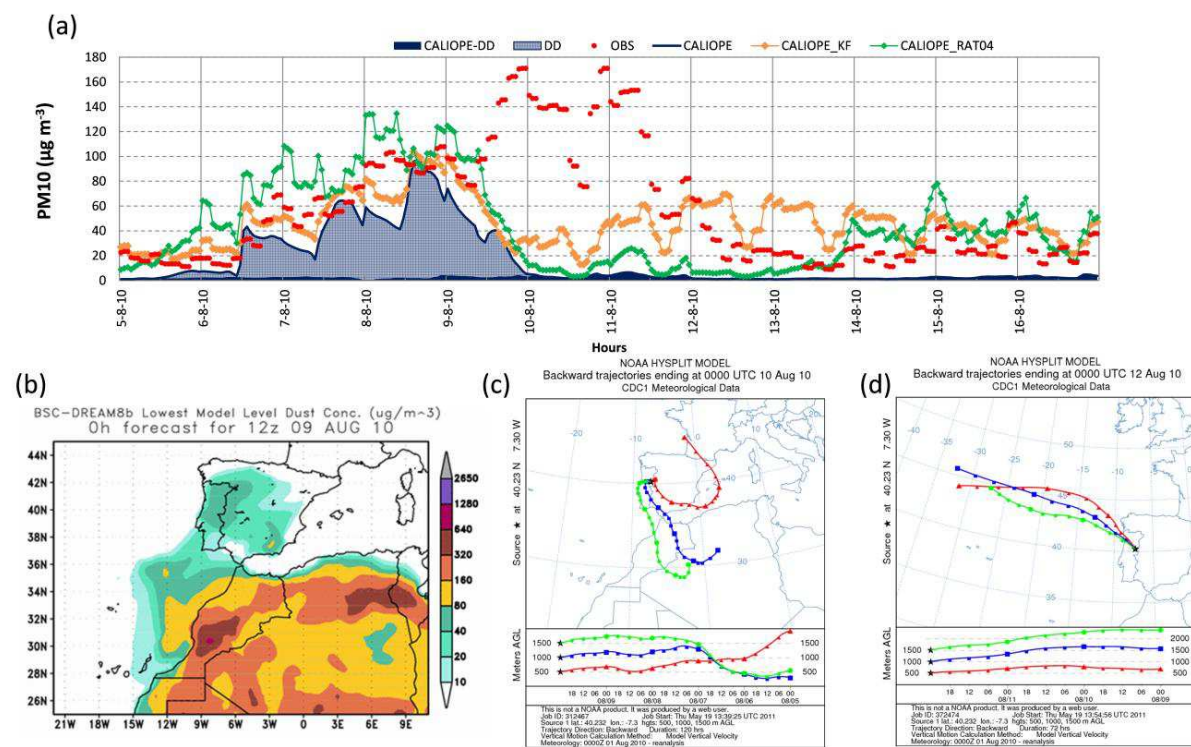


Figure 4.



Figure 5.

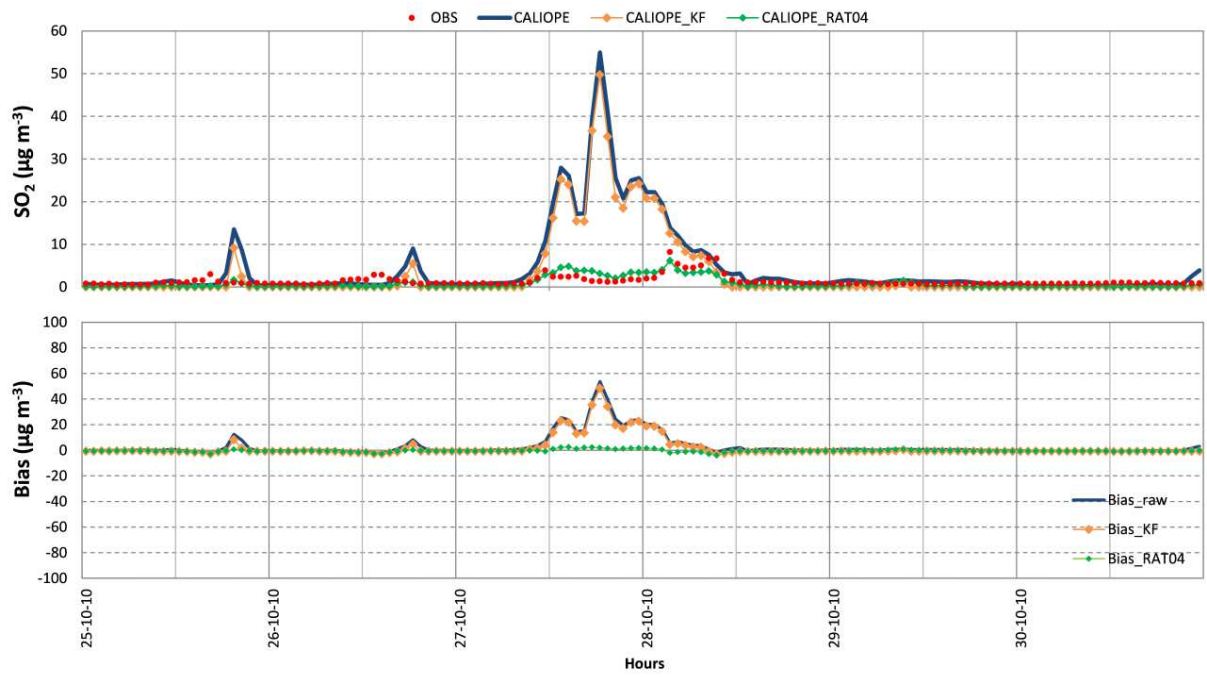


Figure 6.

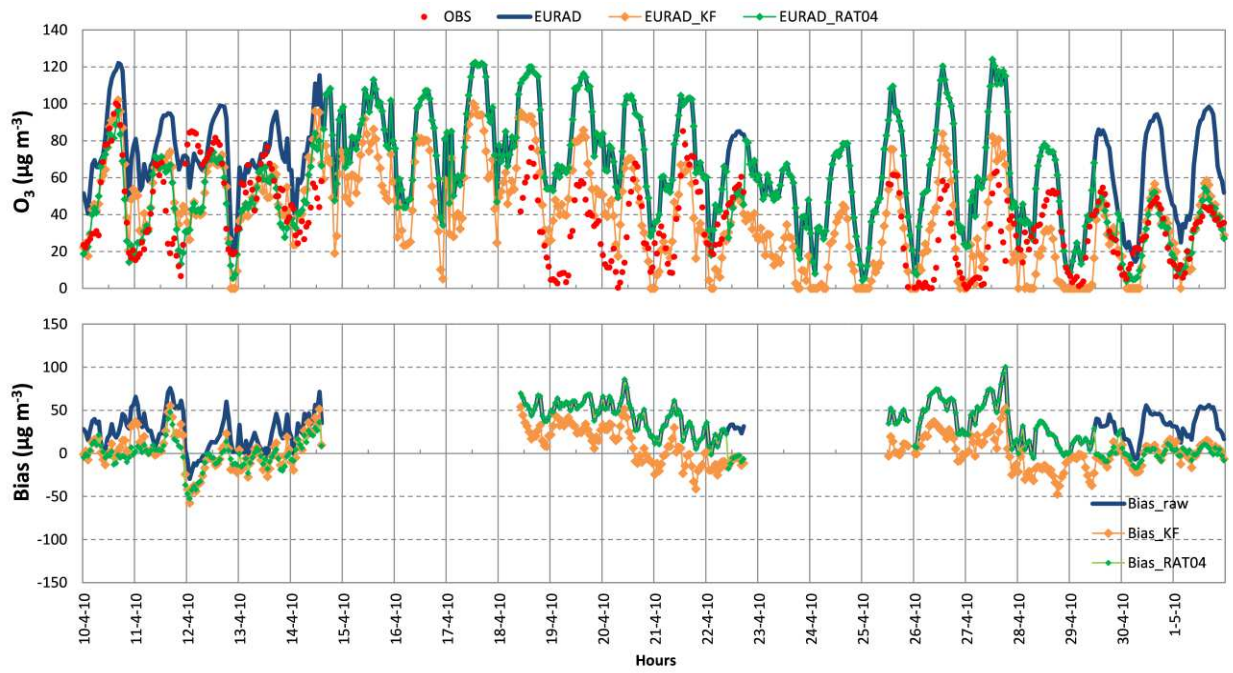


Figure 7.

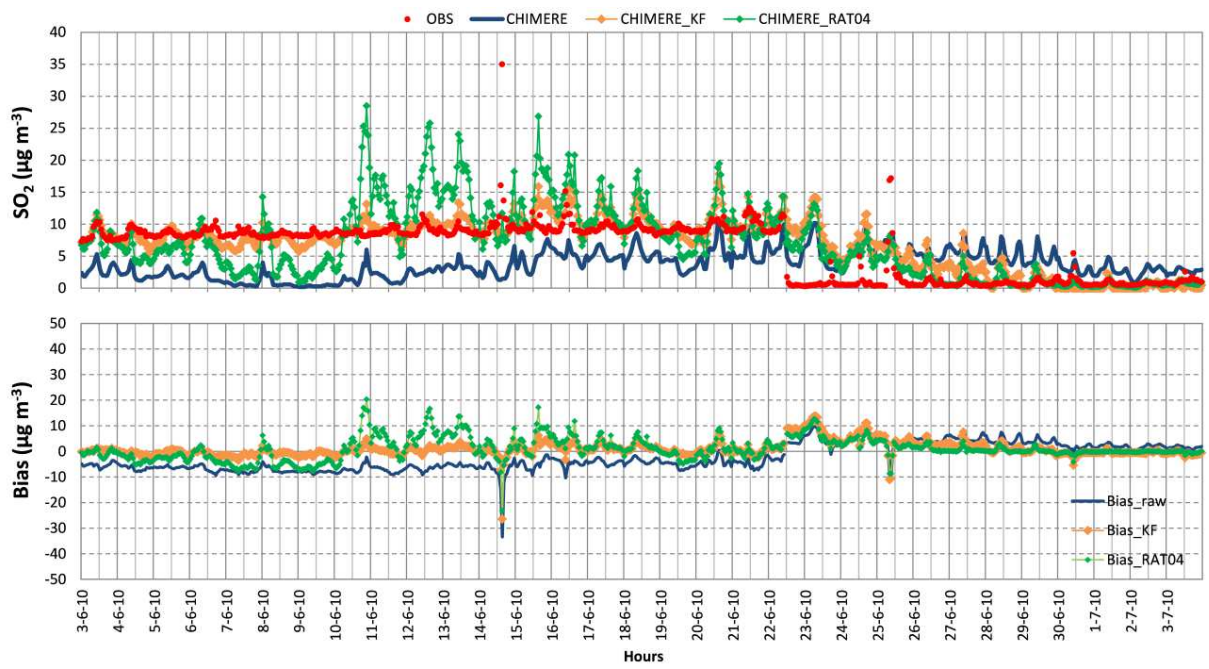


Figure 8.



Table 1: Configurations of the high-resolution air quality forecasting systems which routinely operate over mainland Portugal.

		<b>CALIOPE</b>	<b>MM5-CHIMERE</b>	<b>MM5-EURAD-IM</b>
<b>Domains</b>		<b>Iberian Peninsula</b>	<b>Portugal</b>	<b>Portugal</b>
<b>Meteorology</b>	Model, version	WRF-ARW v3.0.1.1	MM5v3.7	MM5 v3.7
	Horizontal resolution	4 km x 4 km	10 km x 10 km	5 km x 5 km
	Nx, Ny ,Nz	400, 400, 38	35, 70, 32	64, 121, 32
	Mycrophysics	WSM-3 class	Relsner graupel	Relsner graupel
	Radiation	RRTM Dudhia scheme	RRTM Dudhia scheme	RRTM Dudhia scheme
	PBL	YSU	MRF PBL	MRF PBL
	LSM	Noah LSM	Five-layer LSM	Five-layer LSM
	Cumulus	Kain-Fritsch	Grell	Grell
	Initialization and boundary conditions	Nested from European forecast (NCEP-GFS)	Nested from European forecast	Nested from Iberian Peninsula forecast
<b>Emissions</b>	Database source (year)	HERMES+EMEP (2004)*	EMEP (2005)**	EMEP (2005)**
	Biogenic emissions	Offline	Online	Online
		Parra et al. (2004)	(Simpson et al., 1999)	(Guenther et al. 1995).
<b>Chemistry</b>	Model, version	CMAQ v4.5	CHIMERE 2006	EURAD v4.2
	Horizontal resolution	4 km x 4 km	10 km x 10 km	5 km x 5 km
	Nx, Ny, Nz	397, 397, 15	29, 58, 8	64, 121, 23
	Chemical mechanism	CBM-IV	Reduced MELCHIOR	RACM-MIM
	Aerosol size distribution	(Gery et al., 1989) Three modes	(Bessagnet et al., 2004) Eight bins	(Geiger et al., 2003) Three modes
	Inorganic aerosol	Thermodynamic ISORROPIA	Thermodynamic ISORROPIA	Thermodynamic
	Organic aerosol	Simplified SOA Scheme	Simplified SOA scheme	APC SORGAM model
			(Bessagnet et al., 2005)	
	Initialization and boundary condition	(Schell et al., 2001) Nested from Europe (LMDz-INCA)	Nested from Europe	Nested from Iberian Peninsula
<b>Natural dust transport</b>	Model, version	BSC-DREAM8b	BSC-DREAM8b	BSC-DREAM8b

\*Emissions for Portugal and France are estimated following a top-down methodology from EMEP database. Emissions in Spain are calculated with a bottom-up approach (Balasano et al., 2008b).

\*\* Emissions for Portugal are estimated with a top-down desegregation methodology (Monteiro et al., 2007a)

**Table**[Click here to download Table: Table 2.doc](#)

Table 2: Estimated optimal error ratios for Kalman filter technique for O<sub>3</sub>, NO<sub>2</sub>, SO<sub>2</sub>, PM10 and PM2.5 for the selected stations in the Portuguese mainland domain for 2010.

<b>Pollutant</b>	<b>Period</b>	<b>CALIOPE</b>	<b>MM5-EURAD- IM</b>	<b>MM5- CHIMERE</b>
<b>O<sub>3</sub></b>	Winter	0.07	0.03	0.05
	Spring	0.12	0.03	0.13
	Summer	0.12	0.03	0.09
	Autumn	0.12	0.05	0.09
<b>NO<sub>2</sub></b>	Annual	0.04	0.04	0.04
<b>SO<sub>2</sub></b>	Annual	0.20	0.14	0.13
<b>PM10</b>	Annual	0.08	0.04	0.17
<b>PM2.5</b>	Annual	0.07	0.02	0.08

Table 3: Annual categorical statistics for the three modelling system (CALIOPE, MM5-CHIMERE and MM5-EURAD-IM) (raw models) and for the two bias correction techniques, Kalman filter (KF) and multiplicative ratio (RAT04). The calculated statistics are the accuracy (A), the critical success index (CSI), the probability of detection (POD), the bias (B) and the false alarm ratio (FAR). The number in parentheses next to the statistic indicates the perfect score. Note that A, CSI, POD and FAR are in %. The thresholds (T) used to compute the statistics are chosen from the current European directive (2008/50/EC).

		<b>Raw models</b>	<b>KF</b>	<b>RAT04</b>
<b>O<sub>3</sub> max-1h (T = 180 µg.m<sup>-3</sup>)</b>				
(13 stations)	b(hits)	5	16	48
	a(false alarm)	24	28	36
	d(misses)	148	137	105
	c(correct negative)	13326	13322	13314
	A (100%)	99	99	99
	CSI (100%)	3.0	9.0	25
	POD (100%)	3.0	10	31
	B (1)	0.2	0.3	0.6
	FAR (0%)	83	64	43
<b>O<sub>3</sub> max-8h (T = 120 µg.m<sup>-3</sup>)</b>				
(13 stations)	b(hits)	240	425	479
	a(false alarm)	371	254	266
	d(misses)	651	466	412
	c(correct negative)	12562	12679	12667
	A (100%)	93	95	95
	CSI (100%)	19	37	41
	POD (100%)	27	48	54
	B (1)	0.7	0.8	0.8
	FAR (0%)	61	37	36
<b>PM10 daily mean (T = 50 µg.m<sup>-3</sup>)</b>				
(6 stations)	b(hits)	65	70	92
	a(false alarm)	388	283	214
	d(misses)	139	134	112
	c(correct negative)	5282	5387	5456
	A (100%)	91	93	94
	CSI (100%)	11	14	22
	POD (100%)	32	34	45
	B (1)	2.2	1.7	1.5
	FAR (0%)	86	80	70