High Resolution Air Quality Forecasts in the Western Mediterranean area within the MACC, MACC-III and MACC-III European projects

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Abstract

The European Earth observation programme Copernicus, formerly known as GMES (Global Monitoring for Environment and Security) is establishing a core global and regional environmental atmospheric service as a component of the Europe's Copernicus/GMES initiative through successive R&D projects led by ECMWF (European Center for Medium-range Weather Forecasting) and funded by the 6th and 7th European Framework Programme for Research and Horizon 2020 Programme: GEMS, MACC, MACC-II and MACC-III. AEMET (Spanish State Meteorological Agency) has participated in the MACC and MACC-II and continues participating projects (http://atmosphere.copernicus.eu). AEMET has contributed to those projects by generating highresolution (0.05 degrees) daily air-quality forecasts for the Western Mediterranean up to 48 hours aiming to analyse the dependence of the quality of forecasts on resolution. We monitor the evolution of different chemical species such as NO2, O3, CO v SO2 at surface and different vertical levels using the global model MOCAGE and the MACC Regional Ensemble forecasts as chemical boundary conditions. We will show different case-studies, where the considered chemical species present high values and will show a validation of the air-quality by comparing to some of the available air-quality observations (EMEP/GAW, regional -autonomous communities- and local -city councils- air-quality monitoring networks) over the forecast domain. The aim of our participation in these projects is helping to improve the understanding of the processes involved in the air-quality forecast in the Mediterranean where special factors such as highly populated areas together with an intense solar radiation make air-quality forecasting particularly challenging.

Key words: Air-quality, MACC European projects, air-quality modelling, Mediterranean.

Predicciones de calidad del aire a alta resolución en el Mediterráneo occidental para los proyectos MACC, MACC-II y MACC-III

Resumen

El Programa Europeo para la observación de la Tierra Copernicus anteriormente conocido como GMES (Global Monitoring for Environment and Security) ha estado desarrollando el núcleo de un servicio atmosférico medioambiental global y regional como parte de la iniciativa europea Copernicus/GMES a través de sucesivos proyectos de I+D liderados por el Centro Europeo de Predicción a Plazo Medio (ECMWF) y financiados por el 6º y 7º Programas Marco de la Unión Europea para la Investigación Científica y el Programa Horizon 2020: GEMS, MACC, MACC-II y MACC-III. La Agencia Estatal de Meteorología (AEMET) ha participado en los proyectos MACC y MACC-II y lo sigue haciendo en el siguiente proyecto MACC-III (http://atmosphere.copernicus.eu).

AEMET ha contribuido en estos proyectos integrando una predicción de calidad del aire diaria de alta resolución a 0,05° con alcance de 48 horas sobre la cuenca mediterránea occidental con la finalidad de

analizar la dependencia de la calidad de las predicciones de la resolución. Para ello se monitoriza la evolución de determinadas especies químicas NO2, O3, CO y SO2 en superficie y en diferentes niveles verticales utilizando el modelo de transporte químico MOCAGE utilizando como condiciones de contorno para las especies químicas información de las predicciones del ensemble regional generado por los diferentes proyectos MACC. Se mostrarán diferentes casos de estudios, donde las especies químicas tengan valores elevados, y se realizará una validación de calidad de aire frente a las observaciones superficiales disponibles (Red de Observación de Contaminación de Fondo EMEP/VAG y redes autonómicas y locales de observación sobre el territorio considerado). El objetivo de nuestra participación en estos proyectos es ayudar a mejorar la comprensión de los procesos que intervienen en la predicción de la composición química en el Mediterráneo donde factores como la elevada población y la intensa radiación solar hacen de la predicción de la calidad del aire un desafío particularmente dificil **Palabras clave**: Calidad del aire, proyectos europeos MACC, modelización calidad del aire, Mediterráneo.

Summary: 1. Introduction. 2. Description of the MOCAGE configuration in AEMET for the MACC project. 2.1. General description. 2.2. Usage of MACC Regional ENS as chemical lateral boundary conditions. 2.3. Meteorological forcings. 2.4. Emissions. 3. Case studies. 3.1. Case study 1: 28th January 2013 - 1st February 2013. 3.2 Case study 2: 15th July 2013 - 18th July 2013. 4. Conclusions. References.

Normalized reference

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1. Introduction

The air-quality forecasting is a complex task involving physical and chemical processes at very different time and space scales occurring simultaneously and depending on natural and anthropogenic emissions and meteorological conditions. This task is particularly challenging in the Mediterranean basin due to the special characteristics of the zone such as high natural and anthropogenic emissions due to the many densely populated areas, especially in the coast, and the intensity of the solar radiation that boost natural emissions and induces the formation of secondary pollutants. An understanding of individual processes (chemistry, transport, removal, etc) does not imply an understanding of the system as a whole (Seinfeld and Pandis, 2006). Air-quality modelling is today one of the most important tools in the research and applications for atmospheric chemistry and physics (Delmas et al, 2005) and in air-quality forecasting in particular.

Our work within the MACC, MACC-III and MACC-III ensemble subprojects consists of running a high resolution chemical transport model (CTM) using MACC regional ensemble forecasts (ENS) as chemical lateral boundary conditions for some species (O3, SO2, CO and NO2) at four levels (SFC, 500m, 1000m and 3000m) which are interpolated to the MOCAGE model levels up to 5000 m, and investigating if high resolution forecasting can add value to air quality forecasts. The CTM used in AEMET is MOCAGE (Josse et al, 2004), a global model developed by Météo-France. MOCAGE has been adapted in AEMET to be run using boundary conditions. This model will be referred to as MOCAGE-AEM in this paper. The

model runs up to H+48 daily at 0.05° horizontal resolution and 47 vertical levels. On the other side, the different MACC projects are providing forecasts of seven state-of-the-art atmospheric chemistry models and generating an air-quality ensemble (Marécal et al, 2015) combining the information from the individual models. The seven models are: CHIMERE (CHI), EMEP (EMP), SILAM (FMI), LOTUS-EUROS (KNM), MOCAGE (MFM), EURAD-IM (RIU) and MATCH (SMH). A description of the main characteristics of the models can be obtained in (Kukkonen et al, 2012). The regional forecasting system provides 4-days forecasts (hourly) of the main chemical species (including ozone, NO2 and PM10) and experimental biological species such as birch pollen during the season. All the individual models are run in the same geographical domain and emission data. They use MOZART-IFS forecasts as chemical boundary conditions. Horizontal resolutions of the individual models are different but all are interpolated to the ENS horizontal resolution of 0.1 degrees.

Section 2 of this article contains a description of the MOCAGE configuration used in AEMET in the MACC projects, including information about the domain and the modifications carried out in the model to include information from the regional ensemble boundary conditions for several chemical species. Section 3 discusses a selection of the case studies collected during the project in which ENS and MOCAGE-AEM are compared to observations from different air-quality observation networks as well as to the seven individual models used to generate ENS. Finally, section 4 concludes this article.

2. Description of the MOCAGE configuration in AEMET for the MACC project

2.1 General Description

MOCAGE (Josse et al., 2004) is a 3D Global Chemical Transport Model (CTM) developed by Météo-France with 2-way nesting capabilities (up to three additional domains nested to the global one) used mainly for operational "chemical weather" forecasting. This model has 47 vertical hybrid (σ, P) levels going from the ground surface up to the stratosphere. Seven levels fall within the planetary boundary layer (PBL). MOCAGE uses the semi-Lagrangian advection scheme from Williamson and Rasch (1989) for the grid-scale transport, the parameterization of convective transport from Bechtold et al (2001) and the turbulent diffusion parameterization from Louis (1979). Dry deposition is based on the Wesely approach (1989). The wet deposition by the convective and stratiform precipitation from Mari et al. (2000) and Giorgi and Chameides (1986). MOCAGE includes the RACM scheme (Stockwell et al., 1997) for the tropospheric chemistry and REPROBUS scheme for the stratospheric scheme (Lefèvre et al., 1994). Biogenic emissions are fixed monthly biogenic emissions from Guenther et al. (1995).

In Spain, MOCAGE is used for operational air quality forecasting in virtue of an agreement between Météo-France and AEMET allowing it the usage of MOCAGE for research and official duties. Within the successive MACC projects, AEMET is running MOCAGE on a daily basis providing hourly forecasts up to H+48 for species

such as nitrogen oxides (NO and NO2), carbon monoxide (CO), sulphur dioxide (SO₂) and ozone (O₃). The model version used in AEMET does not include particulate matter (PM).

The AEMET MACC configuration consists of a global domain at 2.0 deg. horizontal resolution and nested to it a 0.05 deg. domain (MACCH3 domain, Fig.1) covering the Western Mediterranean. The model, being a global model, has been modified to allow the usage of MACC ENS chemical forecasts as chemical lateral boundary conditions. The link between both domains in the original model has been switched off to allow the values of the ENS Boundary Conditions prevailing in the borders of the MACCH3 domain. Only this domain is used for MACC project so, to all intents and purposes, MOCAGE-AEM is acting as a limited area model with ENS as chemical lateral boundary conditions.

Figure 1 shows MACCH3 domain. It covers the geographical area from 5°W to 5°E in longitude and from 36°N to 44°N in latitude. The horizontal resolution is 0.05 degrees making a domain with 200 gridpoints in longitude and 160 in latitude. Vertical resolution is 47 hybrid levels from surface to the stratosphere.

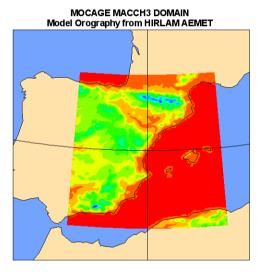


Figure 1. MACCH3 geographical domain

2.2 Usage of MACC Regional ENS as chemical lateral boundary conditions

As mentioned above, MOCAGE, being a global model, has been modified at AEMET for the MACC configuration to allow the model to act as a limited area model (LAM) that makes use of lateral boundary conditions. In our case, the boundary conditions come from the MACC regional ensemble (ENS).

The process to introduce the chemical lateral boundary conditions is made by a procedure called relaxation. This procedure is well known in NWP Limited Area Modelling and essentially consists of defining a zone where a blending between the

information of the model (MOCAGE-AEM) and the host model (ENS) is made using coefficients assigning weight to both, model and host model in order to avoid sharp differences between the lateral boundary condition and the LAM.

But before carrying out a relaxation it is necessary to have boundary conditions at every single hybrid model level and at the same horizontal resolution that the original LAM. To do so, we need to do horizontal and vertical interpolations previously. The scheme to provide lateral boundary conditions to our MACC configuration of MOCAGE has in consequence three stages:

Horizontal Interpolation

Hourly ENS forecasts are downloaded daily for the species NO, NO2, O3 and SO2 at 4 vertical levels (at surface, z=500 m, z=1000 m and z=3000 m) up to H+48. The horizontal resolution of ENS forecasts is 0.1 degrees. We extract the area of our geographical domain and interpolate them to our horizontal resolution (0.05 degrees) using a linear interpolation approach.

Vertical Interpolation

In the vertical, we also extend the information from the four vertical levels to the 47 hybrid (σ, P) model levels. We assign the surface ENS values to the last level of the model (47) directly. For the rest we carry out a linear interpolation in four steps:

- 1. At every model level we evaluate the height for every single grid point by using both the hydrostatic equation and the ideal gases law.
- 2. If the height lays between two different ENS levels (sfc, 500 m, 1000 m and 3000 m), we simply interpolate linearly from the two adjacent levels to calculate the value at the gridpoint we are considering.
- 3. If the height of the gridpoint is above 3000 m, we assign a weight equal to 1 at z=3000 m that progressively decays to zero at z=5000 m. The weight is defined as (1-p), H=750, z is the height and p as

$$p = \exp\frac{-(z - 3000)^2}{H^2}$$

where p ~ 0.001 at z = 5000 m

4. Above 5000 m no vertical interpolation is done. The value for the resultant chemical lateral boundary condition will be zero.

At the same time, we transform from density ρ (mg.m⁻³) to mixing ratio r (ppv)

$$r = \rho \frac{RT}{M_i P}$$

where M_i is the molecular weight of the considered species i

After the horizontal and the vertical interpolations have been made, we would have extended the information from the 4 vertical boundary condition levels to the 47

model levels, and we will have information from the boundaries at the 47 hybrid model levels at a horizontal resolution of 0.05 deg.

Relaxation

Once the semi-Lagrangian transport has been applied at every single time step (30 minutes in our configuration for SL transport), we force a relaxation process within the relaxation zone shown in figure 2. The relaxation zone of the boundaries with the MACCH3 domains consist of a frame 2 degrees width, enclosed within the MACCH3 domain.

MACCH3 BLENDING ZONE WITH MACC RAO ENSEMBLE MEAN B.C.

Figure 2. MACCH3 2 degrees width relaxation zone used in the relaxation process using ENS chemical lateral boundary conditions as host model for MOCAGE. In the inner part of the domain (in white) no relaxation is done.

We assign weight coefficients α_i to the host model (ENS) values depending on the distance to the border of the domain. We assign a weight $\alpha_i = 1$ for the outer five gridpoints of the domain in the latitudinal and longitudinal axis. The weight of the boundaries decays to zero at a distance equal to 2 degrees from the borders (40 gridpoints). In the middle, the value is a linear interpolation between the MOCAGE-AEM value with a weight $(1 - \alpha_i)$ and the host model weighting α_i . Outside the blending zone, in the inner part of the MACCH3 domain, the weight for the boundary conditions is set to zero $\alpha_i = 0$, and the model is not modified by the boundary information so it evolves freely.

We have tested two different options for the weight function depending on the distance to the limits of the domain i is the number of gridpoints counting from the limit of the domain and n is set to 40 (2 degrees width with 0.05 degrees resolution)

$$\alpha_i = \frac{1}{2} \left\{ 1 + \cos\left(\frac{\pi(i-1)}{n}\right) \right\}$$

$$\alpha_i = 1 - \tanh \frac{2(i-1)}{(n-4)}$$

Both methods produced quite similar results. Finally the first option (cosine) was selected, following the default function in HIRLAM (Undén et al, 2002), which offers a smoother decay for the coefficients.

The relaxed chemical value at a distance of i gridpoints:

$$new_i = \alpha_i field I_i + (1 - \alpha_i) field 2_i$$

new stands for the field after the relaxation is done, *field1* the value of the host model (ENS) and *field2* the value of the MOCAGE-AEM after the semi-Lagrangian transport has been called in the model. The semi-Lagrangian transport is called every 30 minutes in our MOCAGE MACC configuration and then it does the relaxation to the boundary conditions. However, as we only have boundary data every hour, a linear time interpolation is carried out between two hourly boundary conditions for the relaxation to be done every 30 minutes.

2.3 Meteorological Forcings

MOCAGE is as an off-line model and needs meteorological forcings. The meteorological forcings (both surface and upper-air) for MACCH3 domain come from the AEMET HIRLAM HNR suite (0.05 degrees), which uses as Boundary Conditions the AEMET HIRLAM ONR suite (0.16 degrees). See figure 3 for details. HIRLAM HNR runs at AEMET 4 times every day at 00, 06, 12 and 18 UTC up to H+36 (Navascues et al., 2013). In MOCAGE-AEM configuration we use the forcings derived from the 00 (D+0) and 12 UTC (D+1) runs.

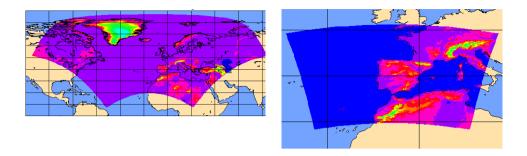


Figure 3. AEMET HIRLAM ONR (left) and HNR (right) domains. Meteorological forcings for MACCH3 MOCAGE domain come from HIRLAM HNR suite.

2.4 Emissions

Emissions are an essential part of a chemical weather forecast. MOCAGE-AEM uses the high resolution TNO-GEMS emission inventory, with a native resolution of 0.125° x 0.0625° longitude-latitude (Visschediijk, et al., 2007) over land. TNO-GEMS emission inventory covers totally the European inland MACCH3 domain. We take into account seasonal and daily variations of the emissions which are time-dependents in our model.

Over the sea and North Africa, we use the EMEP emissions inventory with a resolution of 50 km.

3. Case studies

Every six months a deliverable for the project is done including information about the evolution of the MACC regional ensemble (ENS) and the MOCAGE run for MACC (MOCAGE-AEM) skill scores (Bias and RMSE) compared to data from the EMEP/GAW air-quality monitoring network. ENS is an ensemble of seven individual models, but for convenience, here we will refer to as a model. Besides, case studies are included (usually one per term). The case studies are selected searching situations with high pollution levels in which high resolution could have been an advantage. Then, they are analyzed comparing the forecasts from ENS and MOCAGE-AEM to observations from EMEP and different local and regional Air Quality Monitoring networks. Figure 4 shows the location of the Spanish stations from the EMEP/GAW air-quality monitoring network. We show here two case studies.



Figure 4. EMEP/GAW Air Quality Monitoring Network in Spain. The EMEP/GAW network in Spain is managed by AEMET.

3.1 Case study 1: 28th January 2013 – 1st February 2013

The greatest differences between ENS and MOCAGE-AEM hold usually when an anticyclone persists during several days, causing subsidence, thermal inversions and weak flow (or no flow at all). Pollution episodes, with high concentration levels, are prone to happen in these conditions, because pollutants remains confined in the boundary layer. This is the case of the situation between 28^{th} January and 1^{st} February 2013, with an anticyclone persistent on time, and cause pollution to accumulate during several days. In this case, the parameter that behaved worst was NO2. Ozone kept in relatively low values during the episode as it was expected with low solar radiation. The EMEP GAW stations, being background stations, did not register significant pollution levels of NO2. Nevertheless, NO2 levels were high in some stations. In Madrid some stations reached values above 200 μ g/m3 as it can be observed in the plot of NO2 values for five different Madrid air quality stations (including suburban and urban stations) for the period 28/01/2013 at 00 UTC and 01/02/2013 at 00 UTC (Figure 5).

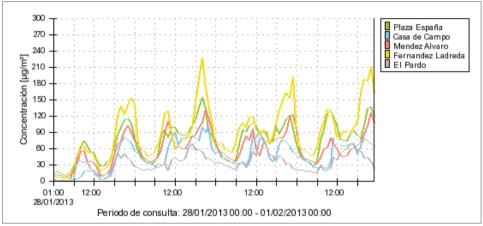


Figure 5. NO2 measured in five air quality stations belonging to the Madrid City Council for the period 28th January at 00 UTC to 1st February at 00 UTC. Source: Madrid City Council

The same was observed in other stations located in the East and South of Spain, with NO2 values above $100 \mu g/m3$ as can be seen in Table 1.

The main differences between ENS and MOCAGE-AEM forecasts deal with the degree of detail reached by the two "models". In general, we see a smoother behaviour of ENS with somewhat higher values in the most important cities such as Barcelona, Valencia or Madrid and a quite uniform distribution of NO2 in the rest of the domain with values that, in general, do not surpass 20 µg/m3. On the other side, we realize that MOCAGE-AEM seems to have a finer structure showing more details in the NO2 distribution. But it seems to overestimate values of NO2 in big cities or industrial areas. This could be due mainly to two factors; firstly, even if the nominal resolution of ENS is 0.1 degrees, some of the individual models are constructed from models using even coarser horizontal resolutions. MOCAGE-AEM have a resolution of 0.05 degrees in latitude and longitude, which represents around 5 km in our latitudes. Secondly, the emissions inventory used by MOCAGE-AEM is GEMS-TNO whereas ENS uses the more recent MACC-TNO. However, the difference between the inventories used would be more important for species such as SO2 because its emissions have very much reduced in Spain in the last decade. We think that, in this case, the most important effect is coming from the resolution side and the averaging effect of ENS with respect to individual models. ENS captures well the background EMEP GAW air quality stations but it does not do it so well with suburban or urban stations. MOCAGE-AEM captures better this kind of stations. Perhaps not exactly in the same location or at the right NO2 level but, looking at the forecasts of both models it can be stated that MOCAGE-AEM (right) forecasts are closer to the reality than the smoother ENS (left) forecasts. ENS provides too low values in some medium

cities such as Granada or Alicante. See for instance the maps of 28th January H+18 or 29th January H+18 (Figs. 6 and 7).

In this case, MOCAGE-AEM seems to fit better to observations. Palma de Mallorca is an area where usually differences between both models can be observed, and normally observations support MOCAGE-AEM. Other areas, e.g. the north-east of Aragón, are, in our opinion, better described by MOCAGE-AEM model. Even if not always the observations (that used to be higher than the forecasted values) are in agreement with models, MOCAGE-AEM usually presents nearby areas with values similar to the measurements in the air-quality networks. This did not happen in the case of ENS. In big cities, MOCAGE-AEM provides too high values of NO2, probably due to exaggerated values of emissions, but for instance in Madrid values close to 200 $\mu g/m3$ have been observed in some stations. Probably none of the models considered here are ready to capture correctly the city small scale patterns.

Table 1. Values of NO2 measured in some stations belonging to Spanish Regional Authorities air-quality networks. All of them are included in the ENS and MOCAGE-AEM maps. Data have been gathered from Regional Authorities air-quality webpages

Day	28/01/2013				29/01/2013					30/01	/2013		31/01/2013				
Station / Hour	00	06	12	18	00	06	12	18	00	06	12	18	00	06	12	18	
Palma de Mallorca (39.5725N, 2.6580E)	27	9	43	65	48	-	-	-	43	38	77	49	48	29	-	66	
Benicassim (40.0622N, 0.0728E)	6	3	3	24	19	6	3	39	25	34	17	52	23	25	13	68	
Alicante (38.3586N, 0.4711W)	3	7	23	42	19	96	68	91	73	90	83	132	10	63	47	79	
Monzón (41.9161N, 0.1911E)	20	24	9	18	22	9	30	28	17	15	45	13	25	13	29	34	
Bujaraloz (41.5047N, 0.1531W)	10	10	10	13	51	20	21	14	27	18	16	35	35	33	18	18	
Cuenca (40.0619N, 2.1294W)	2	2	2	5	27	11	17	23	21	14	15	18	9	6	13	13	
Puertollano (38.7036N, 4.1102W)	2	5	14	11	28	17	68	22	52	26	74	16	29	18	51	16	
Granada (37.1953N, 3.6155W)	19	30	51	109	83	46	94	142	73	25	48	22	73	39	63	44	

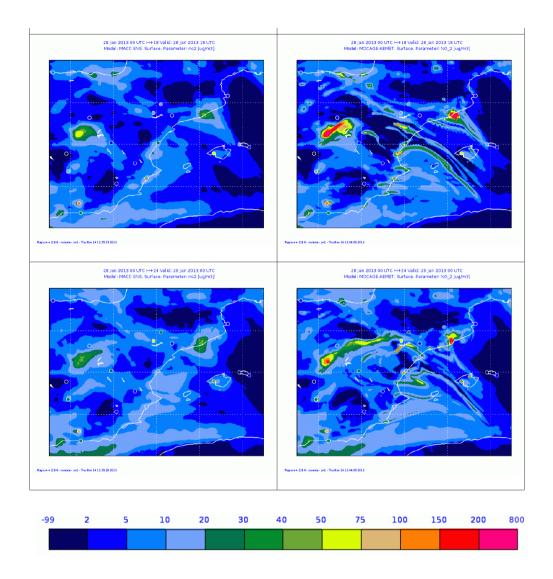


Figure 6. NO2 ENS (left) and MOCAGE-AEM (right) forecasts corresponding to 28th January 2013. H+18 (above) and H+24 (down) are shown.

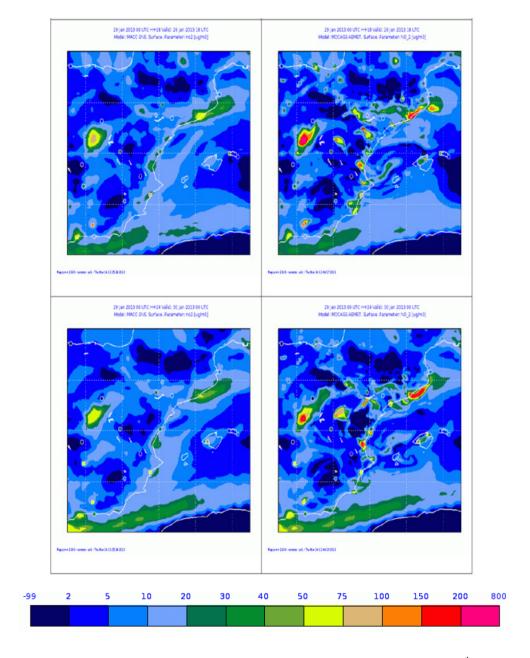


Figure 7. NO2 ENS (left) and MOCAGE-AEM (right) forecasts corresponding to 29^{th} January 2013. H+18 (above) and H+24 (down) are shown.

3.2 Case study 2: 15th July 2013 - 18th July 2013

In July 2013 high values of ozone were measured in Spain in some air-quality monitoring stations. High temperatures, together with stagnation conditions due to an anticyclone, were responsible for this. There were several episodes with high ozone observed values. We are going to study one of them that took place between the 15th July and the 18th July. The synoptic situation had a high pressure system with centre in the Easternmost North Atlantic, affecting Ireland, Great Britain, the Iberian Peninsula and Western Europe. The high pressure system moved slowly and got into continental Europe, but it still affected the mentioned areas during the whole episode.

Table 2 shows the ozone values at some air-quality monitoring stations. EMEP/GAW monitoring stations are shown in table in light-green. The rest belongs to the air-quality monitoring networks from regional or local authorities. Values above $100~\mu g/m3$ were very common during this episode, and measurements of $140~\mu g/m3$ or more were highlighted in red in this table. Only measurements at 00, 06, 12 and 18 UTC are shown.

Plots with the behaviour of the individual members, ENS and MOCAGE-AEM models compared to observations at all the EMEP/GAW stations within the MACCH3 domain are generated. Figure 8 shows the behaviour of the models at ES10 Cabo de Creus station. The models that are seen in the figures are the seven ones that contribute to generate the regional air-quality ensemble: CHIMERE (CHI), EMEP (EMP), SILAM (FMI), LOTUS-EUROS (KNM), MOCAGE (MFM), RIUUK (RIU) and MATCH (SMH). ENS and MOCAGE-AEM (MACCH3) are included too.

The main characteristic of the figures is a high dispersion between models. This is especially noticeable during the daytime. During the night the dispersion between the model forecasts usually decreases and all the forecasts are more similar. The similarity in the results of the models is not necessarily related to a good agreement with observations. Generally speaking, the observations are situated in the upper side of the forecast values for every forecast range, meaning that the model tendency in this study is to underestimate the ozone levels in the analysed air quality monitoring stations.

ES10-Cap de Creus station (Figure 8) is situated in the North-East corner of the Iberian Peninsula in the coast of the Mediterranean, near the French border (Fig. 1). We find here a mixed behaviour with low dispersion in the models some days (18th July forecasts) and higher dispersion some other days (17th July forecasts). We observe once again that models usually destroy ozone too fast at the beginning of the night (see for instance 15th July H+00 and H+24, 17th July H+24 and H+48 and 18th July H+00 and H+24). During the day measurements usually lie in the upper side of model forecasts.

If we try to understand the qualitative analysis of ENS and MOCAGE-AME behaviour, the obvious result is that ENS provides smoother forecasts that MOCAGE-AEM. It is not a surprise since it was something expected. On the other side, MOCAGE-AEM provides forecasts with much more small scale structures. The level of detail is clearly higher due to higher resolution and the fact that MOCAGE-AEM is not averaged in any sense as ENS is.

In general, we can say that ENS provides too low values at 00 UTC, with values under the measurements at Cabo de Creus, Mahón, Zarra and San Pablo de los Montes on 16th, at San Pablo on 17th and 18th, all EMEP/GAW stations (see figure 9).

Values under the measurements were forecasted as well at other locations of stations from different air quality networks. On the other side, MOCAGE-AEM forecasts present much more small scale patterns. To evaluate if those patterns make sense or not, we would have to have a more dense monitoring network than the one we have at our disposal according to the resolution used and the small scale patterns forecasted. Using the observations at our disposal, we can say that in general MOCAGE-AEM do it well at 00 UTC in areas like the Balearic Islands where it is able to forecast differences between nearby stations in Menorca (differences between Mahón in the Eastern coast and Ciudadela) or Mallorca. Those differences were actually observed. In general forecasts agree with the observations at 00 UTC. However, at 12 UTC (and in general during the day), even if most of the observations agree with the forecast, there are some patterns in the forecasts with values above 200 μ g/m3 in wide areas which have not been confirmed by values at the monitoring stations. We think this could be related to a problem with emissions. Emissions used in MOCAGE-AEM model are too high or perhaps too old.

Table 2. Ozone measurements in air-quality stations from different networks. EMEP-GAW stations are highlighted in green. Values equal to or higher than 140 μ g/m3 are highlighted in red. Measurements are in μ g/m3

Day		20130715				20130716				2013	0717	20130718				
Station/Hour	0	6	12	18	0	6	12	18	0	6	12	18	0	6	12	18
San Pablo de los Montes (39.55 N, 4.21 W)	120	84	-	125	133	135	134	146	140	130	136	135	136	107	139	13
Mahón (39.87 N, 4.32 E)	111	136	146	-	143	76	144	137	104	21	90	135	137	122	128	13
Campisábalos (41.27 N, 3.13 W)	100	89	109	116	105	51	127	136	95	88	112	115	51	31	125	13
Cabo de Creus (42.32 N, 3.32 E)	112	111	155	150	133	102	127	128	99	110	126	145	109	94	117	12
Zarra (39.09 N, 1.01 W)	52	80	105	120	106	90	116	135	121	124	124	-	115	112	143	14
Palma de Mallorca - Bellver (39.56 N, 2.62 E)	100	96	159	127	108	72	114	135	74	75	140	151	71	11	124	15
Ciudadela (40.01 N, 3.86 E)	72	85	141	138	91	61	109	125	76	29	94	130	90	76	-	-
Palma de Mallorca – Foners (39.57 N, 2.66 E)	53	19	58	69	66	-	52	70	37	27	77	91	53	33	65	8
Vinarós (40.54 N, 0.43 E)	48	31	97	91	53	33	-	95	50	45	106	96	104	43	97	10
Valencia - Av. Francia (39.46 N, 0.34 W)	-	-	89	84	66	17	97	96	75	13	101	111	93	29	96	10
Caudete de las Fuentes (39.56 N, 1.28 W)	63	52	91	119	66	25	108	119	87	79	111	123	104	97	122	13
El Plà – Alicante (38.36 N, 0.48 W)	6	34	78	77	60	16	97	105	73	48	111	103	64	55	120	99
Lorca (37.69 N, 1.70 W)	106	53	109	115	96	43	96	132	123	72	133	147	113	62	142	14
Caravaca (38.11, 1.87 W)		71	102	117	93	73	94	127	107	89	112	138	118	94	122	13
Monzón (41.92 N, 0.19 E)	72	47	110	128	77	50	122	120	94	60	118	134	75	90	107	13
Bujaraloz (41.50 N, 0.15 W)	86	60	132	99	108	65	116	118	80	52	119	136	55	50	119	13
Motril – El Varadero (36.72 N, 3.52 W)	101	66	107	112	102	63	81	89	55	55	95	133	65	74	94	9.
Granada - Palacio de Congresos (37.17 N, 3.60 W)	79	75	99	145	71	83	75	142	79	80	85	125	67	83	27	12
Almería (36.84 N, 2.46 W)	119	56	91	75	54	56	66	88	98	80	87	104	93	26	86	91
Nijar – Agua Amarga (36.95 N, 1.94 W)	72	55	99	93	76	53	84	96	58	59	129	127	64	66	111	11

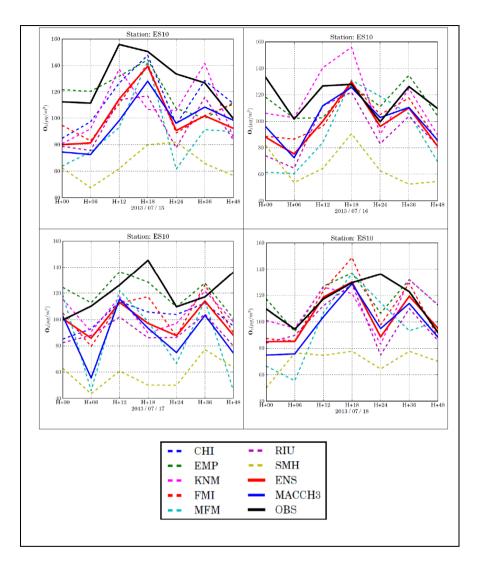


Figure 8. Comparison between observed values and forecasts (up to H+48, for ENS, MOCAGE-AEM (MACCH3) and individual members) in ES10 (Cabo de Creus) EMEP/GAW monitoring station within the domain of MOCAGE-AEM for the O3 case study.

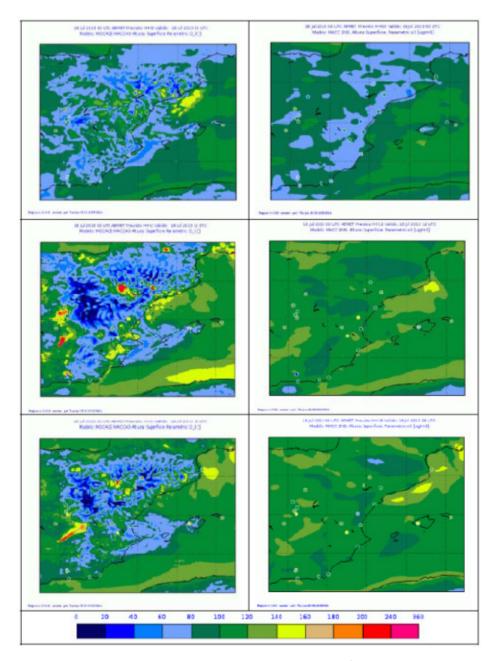


Figure 9. MOCAGE-AEM (left) and ENS (right) outputs for $18^{\rm th}$ July 2013 at 00, 12 and 18 UTC

4. Conclusions

Among the conclusions of our contribution to the different MACC projects it can be mentioned that, provided that they use the same geographical domain, meteorological forcings (ECMWF-IFS), high-resolution emission inventory (TNO-MACC) and chemical lateral boundary conditions (MOZART-IFS), the variability of the different individual models remains very high in the cases we have studied. The daily monitoring of bias and RMSE scores of ENS and MOCAGE-AEM against validated observations from the EMEP/GAW network shows that ENS scores used to be better than MOCAGE-AEM. We think the reason for this could be that ENS provides smoother forecasts and lower pollution levels and this would be advantageous when comparing with background air-quality observations. One additional difference could be the emission inventory used in MOCAGE-AEM (GEMS-TNO) is older and contains higher emissions than MACC-TNO. However, when additional stations from regional and local networks are used in the selected case-studies during this project, it is possible to observe that MOCAGE-AEM seems to have more skill than ENS to detect peaks near medium size cities (e.g. Palma de Mallorca, Alicante or Granada) and present more small scale structure that could be compatible with the observations.

The weakness of our study is the limited number of observations used to validate models. Improving the quality of the work involves necessarily an increase in the number of stations used to calculate the scores. This remains very necessary when we want to study the behaviour of high resolution models and compare them to models with coarser resolutions. If this could be achieved, it would be possible, in principle, to increase considerably our capacity to objectively validate forecast data from models and will make statistics more robust making possible to draw more definitive conclusions.

References

- BECHTOLD, P., BAZILE, E., GUICHARD, F. ET AL. (2001). A mass flux convection scheme for regional and global models. *O.J.R. Meteorol. Soc* 127, 869-886
- DELMAS, R., MÉGIE, G. & PEUCH, V.H. (2005). Physique et chimie de l'atmosphère. *Collection Échelles, Éditions Belin*
- GIORGI, F. & CHAMEIDES, W.L. (1986). Rainout lifetimes of highly soluble aerosols and gases as inferred from simulations with a general circulation model. *Geophys. Res.-Atmos.*, 91, 14367-14376.
- GUENTHER, A.B., HEWITT, C.N., ERICKSON, D. ET AL (1995). A global model of natural volatile compound emissions. *J. Geophys. Res.*, 100, 8873-8892, doi: 10.1029/94JD02950.
- JOSSE, B., SIMON, P. & PEUCH, V.H. (2004), Radon global simulations with the multiscale chemistry and transport model MOCAGE. *Tellus B*, 56: 339–356. doi: 10.1111/j.1600-0889.2004.00112.x
- KUKKONEN, J., OLSSON, T., SCHULTZ, D. M. ET AL. (2012) A review of operational, regional-scale, chemical weather forecasting models in Europe. *Atmos. Chem. Phys.*, 12, 1-87, doi:10.5194/acp-12-1-2012, 2012.

- LEFÈVRE, F., BRASSEUR, G.P., FOLKINS, I., SMITH, A.K. & SIMON, P. (1994) Chemistry of the 1991-1992 stratospheric winter: three-dimensional model simulations. *J. Geophys. Res.-Atmos.*, 99, 8183-8195.
- LOUIS, J.-F. (1979) A parametric model of vertical eddy fluxes in the atmosphere. *Bound.-Layer Meteorol.* 17, 187-202.
- MARÉCAL, V., PEUCH, V.H., ANDERSSON, C. ET AL. (2015). A regional air quality forecasting system over Europe: the MACC-II daily ensemble production. *Geosci. Model Dev. Discuss.*, 8, 2739–2806. doi:10.5194/gmdd-8-2739-2015.
- MARI, C., JACOB, D.J. & BECHTOLD, P. (2000), Transport and scavenging of soluble gases in a deep convective cloud. *J. Geophys. Res.-Atmos.*, 105, 22255-22267.
- NAVASCUÉS, B., CALVO, J., MORALES, G. ET AL. (2013). Long term verification of HIRLAM and ECMWF forecasts over Southern Europe. History and perspectives of Numerical Weather Prediction at AEMET. *Atmos. Res.* 125-126, pp 20-33.
- SEINFELD, J.H. & PANDIS, S.N. (2006). Atmospheric Chemistry and Physics. From Air pollution to Climate Change, 2nd Edition. *Wiley-Intescience, John Wiley and Sons, Inc.*
- STOCKWELL, W.R., KIRCHNER, F., KUHN M. & SEEFELD, S. (1997). A new mechanism for regional atmospheric chemistry modelling. *J. Geophys. Res.-Atmos.*, 102, 25847-25879.
- UNDEN, P, et al. HIRLAM-5 Scientific Documentation (2002). Norrköping. (http://www.hirlam.org/index.php?option=com_docman&task=doc_download&gid=270&Itemid=70)
- VISSCHEDIJK, A.J.H., ZANDVELD, P.Y.J. & DENIER VAN DER GON, H.A.C. (2007). A High Resolution Gridded European Emission Database for the EU Integrate Project GEMS. *TNO-report 2007-A-R0233/B, Apeldoorn, The Netherlands*.
- WESELY, M. (1989), Parameterization of surface resistances to gaseous dry deposition in regional-scale numerical methods. *Atmos. Environ.*, 23, 1293-1304.
- WILLIAMSON, D.L. & RASCH, P.J. (1989). Two-dimensional semi-lagrangian transport with shape-preserving interpolation. *Monthly Weather Review*, 117, 102-129.
 - doi: http://dx.doi.org/10.1175/1520-0493(1989)117<0102:TDSLTW>2.0.CO;2