AIR QUALITY FORECASTS ON A KILOMETER-SCALE GRID OVER COMPLEX SPANISH TERRAINS

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Abstract

CALIOPE-AQFS represents the current state-of-the-art in air quality forecasting systems of high-resolution running on high-performance computing platforms. It provides a 48-h forecast of the main pollutants ($\text{NO}_2$, $\text{O}_3$, $\text{SO}_2$, $\text{PM}_{10}$, $\text{PM}_{2.5}$, $\text{CO}$, and $\text{C}_6\text{H}_6$) found at a 4-km horizontal resolution over all of Spain, and at 1 km over the most populated areas in Spain with complex terrains (Barcelona, Madrid and Andalucia domains). Increased horizontal resolution from 4 km to 1 km over the aforementioned domains leads to finer texture and more realistic concentration maps, which is justified by the increase in $\text{NO}_2$/$\text{O}_3$ spatial correlation coefficients from 0.79/0.69 (4 km) to 0.81/0.73 (1 km). High resolution emissions using the bottom-up HERMESv2.0 model are essential for improving model performance when increasing resolution on an urban scale, but it is still insufficient. Decreasing grid spacing does not reveal the expected improvement in hourly statistics, i.e., decreasing $\text{NO}_2$ bias only by $\sim$2 $\mu$g m$^{-3}$ and increasing $\text{O}_3$ bias by $\sim$1 $\mu$g m$^{-3}$. The grid effect is less pronounced for $\text{PM}_{10}$ because part of its mass consists of secondary aerosols, which are less affected than the locally emitted primary components by a decreasing grid size. The resolution increase has the highest impact over Barcelona, where air flow is controlled mainly by mesoscale phenomena and a lower PBL. Despite the merits and potential uses of the 1-km simulation, the limitations of current model formulations do not allow confirming their expected superiority close to highly urbanized areas and large sources. Future work should combine high grid resolution with techniques that decrease subgrid variability and also include models that consider urban morphology and thermal parameters.
1 Introduction

The World Health Organization (WHO) has recently shown that there is sufficient evidence supporting the belief that particulate matter (PM), ozone (O\textsubscript{3}) and nitrogen dioxide (NO\textsubscript{2}) affect human health (WHO, 2013). Although NO\textsubscript{2} and PM concentrations improved from 2002 to 2011 in Europe, the situation is still far from matching the WHO air quality guidelines (AQG). The European annual limit values for NO\textsubscript{2} (annual) and PM\textsubscript{10} (daily) had been exceeded at 42-43\% of the traffic stations in 2011. For the same year, about 33\% of the European urban population was exposed to PM\textsubscript{10} concentration above the daily limit value, and nearly 88\% was exposed to the respective WHO AQG (EEA, 2013). Air pollution legislation for the protection of the increasing city population has recently increased the demand for urban air pollution forecasting systems that can assess and understand its dynamics, alert the population when health-related issues occur, and develop emission abatement plans (EEA, 2011).

When applying an air quality modeling system, defining the grid resolution is an important consideration; the potential benefits of higher-resolution modeling should be weighed against: the increased complexity of the inputs, CPU time, and disk space requirements. In theory, higher resolution modeling is expected to yield more accurate forecasts because of better resolved model input fields (topography, land cover and emissions), and better mathematical characterization of physical and chemical processes. Furthermore, high resolutions (ranging from 1 to 5 km) are essential to reproduce mesoscale phenomena, e.g., those controlling O\textsubscript{3} transport along the mountainous northeastern Mediterranean coast (Fay and Neunhäuserer, 2006; Jiménez et al., 2006). Even at the finest scale, the modeled concentrations are not necessarily the best (Mass et al., 2002; Gego et al., 2005; Valari and Menut, 2008), because increasing emission and meteorology spatial resolution can also increase uncertainties, at the risk of reduced model performance. Nowadays, fine horizontal resolution is a persistent challenge when assessing health impact and population exposure studies (Thompson et al., 2013).

Several studies have evaluated the impact of increasing horizontal resolution on different scales over the eastern and southeastern USA using the Community Multiscale Air Quality (CMAQ) model and the Comprehensive Air Quality Model with Extensions (CAMx), which range from 32 km – 12 km – 4 km (Cohan et al., 2006; Tesche et al., 2006; Queen and Zhang, 2008). They found no significant changes for O\textsubscript{3} and PM (<5\%...
on average), and those changes were even lower at resolutions of between 12 km and 4 km (<3%). Concerning PM components, Fountoukis et al. (2013) found that increased resolution provides differences mostly for primary PM rather than secondary PM. Recently, a model intercomparison exercise, named ScaleDep, was performed to determine the effect of grid resolution on air quality modeling performance over Europe at a regional and urban scale (Cuvelier et al., 2013). The exercise, involving five Chemical Transport Models (CTMs) (EMEP, CHIMERE, CMAQ, LOTOS-EUROS and RCGC) running under the same conditions over the full year 2009 and at four resolutions (56, 28, 14, and 7 km), showed that it is difficult to define a grid size that is adequate for resolving the urban signal under all conditions affecting Europe. Still, a 14-km resolution seems to be a good compromise between background applications and those reproducing most of the urban signals (7 km resolution). However, the ScaleDep exercise did not distinguish between the different topographies or complex meteorological patterns which are characteristic of the Iberian Peninsula.

Few studies have been performed over selected areas in Spain; and of those, the focus has been mainly on O₃ and NO₂. Vivanco et al. (2008) evaluated the annual impact by increasing the resolution (to 36, 19, and 7 km) over Madrid for NO₂ and O₃. They used the WRF-CHIMERE model, disaggregating the EMEP emission inventory based on land use information. Their evaluation showed that the model improves greatly for NO₂ than O₃, and the most significant improvement is achieved when resolution increases from 36 to 19 km rather than to 7 km, which is linked to increased uncertainty in the emission data introduced with the disaggregation techniques. Jiménez et al. (2006) used the MM5-CMAQ model along with a bottom-up emission model (EMICAT2000) to assess the influence of grid resolution on O₃ (at 8, 4, and 2 km) over the complex terrain of the northeastern Iberian Peninsula (Catalonia) during a summer pollution episode. They indicate that, due to improved performance of the mesoscale phenomena and a better allocation of emissions, a 2-km resolution improves the capability of the model to simulate exceedances of European limit values. An important issue in both studies is the emission modeling approach (top-down vs. bottom-up) when applying high resolution at the local scale (<10 km). As Fountoukis et al. (2013) and Timmermans et al. (2013) demonstrate, in the range of local scale (e.g., the greater Paris area), the grid resolution is not currently the major source of discrepancies in model performance; but in fact, the predicted concentrations and corresponding gradients are more consistent with observed
concentrations when provided by bottom-up emission inventories rather than down-
scaled inventories. If local variation in input data (e.g., emission patterns or land use)
cannot be properly characterized, modeling with a finer grid resolution may not provide
any great advantages.

Increasing resolution is a technical challenge, since computational cost markedly
increases in inverse proportion to grid spacing. The current progress in computation
allows increased model resolution and for multiple spatial scales to be investigated with
the aim of establishing adequate grid size for forecasting air quality at the local scale.

Recently, Colette et al. (2014) evaluated the impact of increasing resolution up to 2 km
over the European continent by using the CHIMERE model for an episode of air
pollution in 2009. They used 2 million grid cells at over 2000 CPUs of a high
performance computing system, which was hosted by the French Computing Centre for
Research and Technology (CCRT/CEA).

In terms of computational resources, horizontal resolution is critical to an operational air
quality forecast. In Europe, operational air quality systems use resolutions of between
12-25 km; meanwhile application to a single country can reach resolutions of between
4-10 km (Zhang et al., 2012). Over Spain, there are three systems providing air quality
forecasts running at different horizontal resolutions. The lowest resolution system is the
Technical University of Madrid’s OPANA (OPerational Atmospheric Numerical model
for urban and regional Areas), running at 27 km x 27 km and based on the
MM5/CMAQ/EMIMO models (San José et al., 2009). It is followed by the Spanish
meteorological office’s system (AEMET, http://www.aemet.es/es/eltiempo/prediccion/calidad_del_aire), which forecasts at 10 km
x 10 km using the HIRLAM-HRN/MOCAGE/GEMS-TNO models. The CALIOPE Air
Quality Forecast System (CALIOPE-AQFS; Baldasano et al., 2011; Pay et al., 2012a;
and references therein), of the Barcelona Supercomputing Center–Centro Nacional de
Supercomputación (BSC-CNS), runs at the highest resolution, 4 km x 4 km, and it is
based on the WRFv3.5/CMAQv5.0.1/HERMESv2.0/BSC-DREAM8bv2 models.
Moreover, CALIOPE-AQFS provides 1-km x 1-km resolution forecasts for the Madrid
and Barcelona metropolitan areas (since 2009), and the Andalusian region (since 2013).
Such resolution has been possible thanks to both the high performance computing
resources at the BSC-CNS and the availability of detailed emission data covering Spain.
The previous works demonstrate there is not a single answer which explains the merits of high-resolution modeling for all applications. The present work aims to assess the impact of increasing the horizontal resolution from 4 km to 1 km, specifically over areas affected by heterogeneous emission patterns and complex terrains such as the Barcelona and Madrid metropolitan areas (BCN and MAD) together with the Andalusian region (AND). For that purpose, CALIOPE-AQFS forecasts pollutant concentrations (O$_3$, NO$_2$, and PM$_{10}$) at two horizontal resolutions: first at a 4-km resolution covering Spain (IP4), and second at a 1-km resolution covering the AND, BCN, and MAD domains. The study is performed for the period April 2013, which presented seven days of an air pollution episode. We use observations from routine air quality monitoring networks to evaluate both resolutions.

Section 2 describes the configuration and computational setup of CALIOPE-AQFS; it analyses the domains and the period under study and it defines the methodology used to evaluate the resolution increase. Section 3 quantifies the impact of resolution increase on forecasting hourly concentrations (and exceedances) in terms of pollutant, domain, building density and major emission sources. Section 4 concludes with the main results and some recommendations.

2 Methodology

2.1 Domain and period under study

Figure 1 shows the main NO$_2$ emission patterns and topographic characteristics of the domains: the Barcelona and Madrid metropolitan areas (BCN and MAD) and the Andalusian region (AND). BCN is a coastal area characterized by several valleys perpendicular to the coastline and two main mountain ranges, one coastal (500 m height) and one pre-coastal (1000-1700 m height). These features induce mesoscale phenomena such as sea-breeze and mountain-valley winds. On the other hand, MAD is a continental region with a much simpler topography that includes the Tajo valley in the southern of MAD and the mountain range of the Central System located in the northwestern MAD, with summits reaching 2500 m height. These features bring different locally-driven flows.

The urban contribution in BCN (3.1 million inhabitants) is accompanied by industrial and power generation emissions, the road network and the harbor; meanwhile, the
Spanish capital of MAD is mainly influenced by emission from the urban area (5.8 million inhabitants) and the road network that connects MAD with the surrounding commercial and industrial zones as well as the urban areas.

AND is the southern-most region in Spain, with complex topography characterized by the large depression of the Guadalquivir Basin (delimited by the Iberian Massif and the Betic Range), which crosses the region from NE to SW over a 60–km stretch. About three quarters of AND has a mountainous orography, including the Sierra Nevada (3481 m). AND includes one of the five biggest cities in Spain, Seville (~700 000 inhabitants), which hosts industrial and electric generation activities around the Algeciras bay, and it is affected by dense maritime traffic through the Strait of Gibraltar.

The study is performed over April 2013. At the beginning and end of the month, the synoptic circulation was controlled by a low pressure system displaced over the south of the British Isles and which affected Western Europe by leading to atmospheric instability over the IP. This pattern is typical of transitional months such as April and November (García-Valero et al., 2012; Valverde et al., 2014), which produce precipitation and decreased temperatures because of cold and humid winds entering from the Atlantic Ocean. In contrast, from 12-18 April there was a high pressure system crossing the Iberian Peninsula in a SW-NE direction, transporting dust from the Sahara Desert and increased temperatures of up to 25-28°C. During the latter episode, available air quality stations at the study domains displayed several exceedances of the European limit values (8 exceedances of the NO\textsubscript{2} hourly limit value, 25 exceedances of the O\textsubscript{3} information threshold, and 31 exceedances of the PM\textsubscript{10} daily limit value).

2.2 CALIOPE-AQFS

CALIOPE-AQFS has provided 48-h air quality forecasts for Europe and Spain since October 2006 (www.bsc.es/caliope) and has been described and evaluated in detail elsewhere (Baldasano et al., 2008, 2011; Pay et al., 2011, 2012a). Briefly, it integrates a meteorological model (WRF-ARW v3.5; Skamarock and Klemp, 2008), an emission model (HERMESv2; Guevara et al., 2013), a chemical transport model (CMAQv5.0.1; Byun and Schere, 2006; Appel et al., 2013), and a mineral dust atmospheric model (BSC-DREAM8bv2; Pérez et al., 2006; Basart et al., 2012); together, all of these comprise an air quality forecast system.
Figure 1 shows the working domains of CALIOPE-AQFS. First, CALIOPE-AQFS was run over Europe at a 12-km x 12-km horizontal resolution using initial/boundary conditions from the Final Analyses of the National Centers of Environmental Prediction (FNL/NCEP). The analyses began at 12 h UTC, at intervals of 6 h (0.5ºx0.5º) for meteorology. The global model LMDz-INCA2 (3.75ºx2.5º, Szopa et al., 2009) was used for chemistry. Then, CALIOPE-AQFS was run at a higher horizontal resolution (4 km x 4 km (IP4)) over the Iberian Peninsula using one-way nesting. In the present work, CALIOPE-AQFS runs at 1 km x 1 km over the domains at hand (AND, BCN and MAD), with nesting of over IP4 throughout. HERMESv2.0 forecasts anthropogenic emissions for the year 2009 by following a bottom-up methodology (point, linear and area), and biogenic emissions using the MEGANv2.0.4 model (Guenther et al., 2006). Emissions are aggregated into 1-km grids for AND, BCN and MAD 1-km simulations, and into 4 km for IP4.

Vertically, WRF-ARW is configured with 38 sigma layers up to 50 hPa, with 11 characterizing the planetary boundary layer (PBL); meanwhile CMAQ vertical levels are obtained by collapsing from the 38 WRF levels to a total of 15 layers that steadily increase from the surface up to 50 hPa. Six layers are within the PBL, and the first layer depth is 39 m.

The present WRF setup uses: the Rapid Radiation Transfer Model (RRTM) and Dudhia for long- and short-wave radiation, respectively; the Kain-Fritsch cumulus parameterization (Kain and Fritsch, 1990); the single-moment 3-class (WSM3) microphysics scheme; and the Yonsei University PBL scheme (YSU). The Noah land-surface model (NoahLSM), based on the U.S. Geological Survey’s (USGS) land-use data, is used by default in the present WRF configuration.

Currently, a new CMAQ version is being tested in the CMAS community, namely CMAQv5.0 (CMAQ, 2012). It includes substantial scientific improvements over Version 4.5 and is especially devoted to improving SOA formation as well as the dynamic interactions of fine and coarse aerosols. Based on the evaluation results from the previous CMAQ version within CALIOPE-AQFS (4.5 vs. 5.0) (Pay et al. 2012b), CMAQ has been updated to Version 5.0.1 using the CB05 chemical mechanism (Yarwood et al., 2005), the AERO5 for aerosol modeling, and the in-line photolysis calculation.
CALIOPE-AQFS considers desert dust contribution by means of the BSC-DREAM8bv2, which runs off-line at a 0.5º x 0.5º resolution covering Europe, North Africa and the Middle East. Its outputs are mass conservative interpolated to the CMAQ's Lambert conformal conic grids and at the required resolution and domain. After interpolating, the modeled PM$_{10}$ concentration is the sum of Aitken, accumulation and coarse-mode modes from CMAQ, and the corresponding BSC-DREAM8bv2 bins with a diameter of ≤ 10 µm (Pay et al., 2012a).

2.3 Computational strategy

Running CALIOPE-AQFS at 4 and 1 km is a technical challenge. The simulations are run on MareNostrum supercomputer (Intel Xeon E5-2670, 16 CPUs and 64 GB RAM memory per node) at BSC-CNS. Table 1 depicts the computational requirements for forecasting air quality at 48 h for each domain. The number of CPUs was chosen to maximize CPU efficiency. Thanks to the parallelization of meteorological and air quality models, MareNostrum uses up to 256 CPUs. Due to the variable nature and complex dependencies, the computational time for forecasting 48 h of air quality fields for the 4 domains is 8-9 hours. The most computationally demanding domain is the AND, at 1-km resolution (366x358 cells, 256 CPU max., and 300 min). For the April 2013 simulation, times add up to 2880 CPU hours/day, or 86400 CPU hours in one CPU (9.86 years). The storage for the April 2013 output files was 6.13 TB (~200 GB/day).

2.4 Evaluating the increase in resolution

Comparing CALIOPE-AQFS grid resolutions and measurements was done in terms of gas-phase and aerosol concentrations (O$_3$, NO$_2$, and PM$_{10}$). Representativeness continue to be a challenge when comparing gridded simulations to observational data at a point in time and space, as modeled concentrations represent a volumetric average over an entire grid cell. Furthermore, the stochastic compound embedded in the observations is not accounted for. Concerning temporal representativeness in the present comparison, both modeled and measured concentrations are averaged hourly. CALIOPE-AQFS operationally receives air quality measurements from Spanish administrative networks in near real time (NRT) without any quality data or quality control. For the present study, NRT measurements are filtered by removing data before and after measurement interruptions or calibrations. Also, a minimum cut-off threshold of 1 µgm$^{-3}$ is applied to
the observed concentrations in order to avoid unrealistic observations. After filtering, the number of stations is 48/30/36 for O₃, 51/42/42 for NO₂, and 52/15/33 for PM₁₀ at AND/BCN/MAD.

The meteorological fields are evaluated for wind speed at 10 m (U₁₀), and wind direction (WD₁₀) and temperature at 2 m (T₂M), all of them at 10 METAR stations located at airports (6/2/2 stations in AND/BCN/MAD). They are discussed in Sect. S1.

Figure 2 shows the location of the air quality and METAR (METeorological Aerodrome Report) stations over the respective domains. The spatial representativeness of the air quality network is highly variable. The influence of the station type is based on two classifications of air quality monitoring stations, the environment type (rural, R; suburban, S; and urban, U), and the dominant emission source (traffic, T; industrial, I; and background, B). These were derived from the Council decision 97/100/EC (Garber et al. 2002).

The evaluation is based on discrete statistics performed on an hourly basis. We consider the correlation coefficient (r), mean (absolute, relative, and fractional) biases (MB, MNBE, and MFB), and error (MAE, MNGE, and MFE). Root Mean Square Error (RMSE) is also calculated because it intensifies large differences between measured and observed concentrations (Table A1).

In order to evaluate the effect of resolution increase on forecasted exceedances and non-exceedances of limit values established by the European legislation, we calculate categorical statistics based on comparisons with fixed concentration thresholds (T). The calculated statistics are accuracy (A), bias (B), probability of detection (POD), critical success index (CSI), and false alarm ratio (FAR), whose formulas and descriptions are explained in Table A2 and elsewhere in Kang et al. (2005) and Eder et al. (2006). The 2008/50/EC directive sets an information threshold of 180 µgm⁻³ for maximum daily O₃ concentrations (Max 1h O₃) and a target value of 120 µgm⁻³ for the maximum daily 8-h running O₃ mean (Max 8h O₃), which should not be exceeded more than 25 days per year. It establishes a limit value of 200 µgm⁻³ for maximum daily NO₂ concentrations (Max 1h NO₂), and 50 µgm⁻³ for the daily PM₁₀ mean (Mean 24h PM₁₀), which should not be exceeded more than 35 times per year. Therefore, categorical evaluation will be performed for Max 1h NO₂, Max 1h and Max 8h O₃, and Mean 24h PM₁₀. Note that mean and maximum concentrations are calculated by considering at least 75% of the...
data in the corresponding time base, i.e., values of at least 18 hours per day for Mean 24 h, Max 1h, and Max 8h; and 6 hours for 8 h values, as established by 2008/50/EC.

3 Concentration maps and spatial representativeness

To analyze the spatial differences between resolutions, Figs. 3, 4, and 5 show the monthly mean concentration maps for April 2013 over MAD, BCN and AND domains at 4 km (left panels) and 1 km (right panels) for NO$_2$, O$_3$, and PM$_{10}$, respectively.

The maps of NO$_2$ and PM$_{10}$ at both resolutions display similar distribution along the MAD and BCN urban plumes. On-road traffic constitutes the main source of primary pollutants in MAD and BCN. HERMESv2.0 estimates that 75% and 59% of NO$_x$ emissions are produced by on-road traffic in both domains, respectively. Consequently, when the resolution increases, the monthly mean O$_3$ concentration maps are almost identical, although the NO$_x$ titration effect on O$_3$ is significant along highways and major point sources. In AND, NO$_2$ and O$_3$ concentrations are also conserved between resolutions along the shipping route crossing the Strait of Gibraltar towards the Mediterranean Sea.

However, the definition of NO$_2$ concentrations along highways connecting the biggest cities with the rest of the country and industrial sectors are more easily identified at 1-km simulations than at 4 km, especially along those roads from/to Barcelona (e.g., the AP7 Mediterranean highway and C32, which connects the harbor and the airport) and Madrid (the A-2 and A-6 in the north, and A-3, A-4 and A-5 in the south). In the same way, 1-km O$_3$ maps are more textured than those at 4 km along highways, because the titration effect is more significant at 1 km, due to less dilution within grid cells. The titration effect of NO$_x$ on O$_3$ over the main sources is more forceful in BCN than in MAD, given that BCN has a larger concentration gradient resulting from complex topography and recirculation flows that accumulate pollutants.

The improvement of the definition along roads in AND is lower than that observed in the MAD and BCN domains, due to the fact that the AND domain is bigger and displays lower traffic emission sources than the MAD or BCN domains. Regarding PM$_{10}$, the main component in AND is the desert dust (~40% in both resolutions) from North Africa. This is because there were two episodes on 14-19th and 25-26th April that affected the IP, as shown by the S-N PM$_{10}$ gradient (Fig.5e and f). The desert dust is transported from long-range simulation with BSC-DREAM8bv2.
Over complex terrains, the 1-km simulation allows reproducing more realistic NO$_2$ concentration maps because of its more detailed topographic information. For instance, the BCN 1-km simulation displays the lowest NO$_2$ concentrations (< 10 µgm$^{-3}$) along the coastal chain (500 m height) and pre-coastal chain (1000-1700 m height), except for the city’s urban hill, where concentrations reach 20-40 µgm$^{-3}$. In contrast, the 4-km simulation provides smoother NO$_2$ concentrations without any concentration gradient. Thus, the 1-km simulation generates slightly higher O$_3$ background concentrations than the 4-km simulation along the BCN pre-coastal chain (66-70 vs. 70-74 µgm$^{-3}$), as well as across the Iberian Massif (AND), where the O$_3$ map displays significant structure due to the higher resolution topography that shapes the basin and the on-road traffic.

Figures 3, 4, and 5 include dots corresponding to mean concentrations at air quality stations that help to qualitatively evaluate the modeled spatial representativeness at both resolutions. Note the high concordance between NO$_2$ observations and the 1-km simulation near the primary suburban traffic roads in BCN (e.g., Vilafranca, Igualada, Manresa, and Mataró). Regarding O$_3$, although observed concentrations depict an overall tendency of the model to underestimate concentrations at both resolutions, the 1-km simulation displays a higher accord with measurements at rural background stations (e.g., El Atazar, San Martín, Villa de Prado, Villarejo and Orosco stations in MAD), and at suburban traffic stations (e.g., Manresa, Igualada and Vilafranca in BCN, with modeled O$_3$ concentrations of around 54-58 µgm$^{-3}$ at 1 km, and 60-66 µgm$^{-3}$ at 4 km). For PM$_{10}$, comparisons with measurements show that modeled concentrations are underestimated over background areas, mainly outside the urban/suburban area, as already discussed in Pay et al. (2012a). However, PM$_{10}$ measurements at the urban/suburban stations of Vilafranca, Sant Celoni, and Mataró in BCN (14-16 µgm$^{-3}$) show a higher concordance at 1 km than at 4 km (12-14 µgm$^{-3}$ vs. 8-10 µgm$^{-3}$).

The spatial variability of the increased resolution is quantitatively analyzed by means of concentration maps, shown in Fig. 6 for NO$_2$, O$_3$ and PM$_{10}$ over AND, BCN and MAD. Over all domains, the explained spatial variability improves as a function of the resolution increase for NO$_2$ and O$_3$, which is sustained by the increase in monthly $r$ from 0.79 (4 km) to 0.81 (1 km) for NO$_2$, and from 0.69 to 0.73 for O$_3$. The slopes improve with the resolution increase, from 0.72 (4 km) to 0.77 (1 km) for NO$_2$, and from 0.50 (4 km) to 0.54 (1 km) for O$_3$. This results from the improved model
performance at urban stations, indicating that CALIOPE-AQFS explains better the magnitude of the variability between urban regions at 1 km than at 4 km. In contrast, for PM$_{10}$, the monthly $r$ decreases from 0.67 to 0.58 when the resolution increases. Although the PM$_{10}$ spatial variability over BCN and MAD improves when the resolution increases ($r$ increases by 0.01 and 0.04, respectively), the global correlation coefficient deviates mainly at the AND stations (52/100), where $r$ decreases by 0.1 from 0.36 (4 km) to 0.26 (1 km). Despite the unfavorable effect of the resolution increase in PM$_{10}$ over AND, the NO$_2$ and O$_3$ concentrations show the highest absolute increase in the spatial $r$ over this domain, from 0.62 (4 km) to 0.71 (1 km) for NO$_2$ and from 0.58 (4 km) to 0.64 (1 km) for O$_3$ (increasing $r$ by 0.09 and 0.06, respectively).

4 Temporal evaluation

The present section discusses the temporal evaluation of the resolution increase by pollutant, environment, predominant emission sources, and study domain. Fig. 7 summarizes the statistical evaluation.

4.1 Pollutant

Table 2 depicts the statistical evaluation by pollutant, with a focus on the reproduction of high concentrations established by the European directive (2008/50/EC). Depending on the pollutant’s lifetime and variability, as well as its dependency on precursors, increased resolution shows different impacts. The resolution increase has a positive effect on NO$_2$, decreasing its bias by 2.0 $\mu$gm$^{-3}$ (from -4.5 to -2.5 $\mu$gm$^{-3}$); but it also increases absolute (squared) errors by 0.3 $\mu$gm$^{-3}$ (0.9 $\mu$gm$^{-3}$). This positive effect is sustained by the perceptual variability, where the MB (MFB) is reduced by 42% (19%); whereas MAE (MFE) only increases by 2% (1%). The correlation coefficient does not significantly change, which is obvious because emissions at both resolutions are modeled using the same approach. The bias improvement at 1-km resolution is justified, because the higher resolution leads to better emission allocation from point, linear or area sources; it decreases the artificial dilution of emission compared to the larger grid area; and, due to the decrease of artificial dilution, it treats chemistry more properly near large emission sources.

In contrast, the resolution increase has a negative effect on hourly and Max 8h O$_3$ concentrations because it increases biases and errors by 0.1-0.8 $\mu$gm$^{-3}$. Relative (fractional) biases and errors increase by 8% (15%) and 1% (1%), respectively, for
hourly $O_3$; and 6% and 4% for Max 1h $O_3$. However, the statistical evaluation alone is not enough to explain the impact of the resolution increase on $O_3$ concentrations.

According to the categorical evaluation, only a few exceedances of the European target and limit values were detected for Max 1h $NO_2$ (9), Max 1h $O_3$ (25), and Mean 24 PM$_{10}$ (31) in April 2013. Thus, categorical evaluation is performed on the temporal basis established by the European legislation, but it uses a $T$ based on the 75p of the observed concentrations in each case. $T$ corresponds to 71 µgm$^{-3}$ for Max 1h $NO_2$, 108 (101) µgm$^{-3}$ for Max 1h (Max 8h) $O_3$, and 27 µgm$^{-3}$ for Mean 24h PM$_{10}$.

Overall, CALIOPE-AQFS underestimates exceedances at both resolutions, indicating that errors of missing observed exceedances are not totally resolved by a resolution increase ($a<d$). The best performance is found for Max 1h $NO_2$, where bias (B) improves from 37% (4 km) to 40% (1 km).

For $NO_2$ Max 1h, there are 953 observed exceedances (b+d) of the threshold ($T=47$ µgm$^{-3}$). Increasing the resolution increases the POD from 49% (4 km) to 56% (1 km). As POD, CSI studies the exceedances, but in a more coherent way by considering both false alarms and missing events. Both POD and CSI increase by 14% and 20%, when resolution is increased. The opposite effect appears for $O_3$. Aside from the fact that the $O_3$ POD is relatively low, the POD decreases as the resolution increases. Of the 1306 observed exceedances of the 108 µgm$^{-3}$ Max 1h, CALIOPE-AQFS detected 112 exceedances at 4 km and only 96 at 1 km. Increasing resolution decreases POD and CSI by 22% and 25% for $O_3$ Max 1h, whereas they do not significantly change for Max 8h $O_3$ and Mean 24h PM$_{10}$.

FAR increases for Max 1h $NO_2$ (from 40% to 42%) and decreases for Max 1h $O_3$ (from 27% to 17%) when the resolution increases. In relative terms, this variability is more significant for Max 1h $O_3$ (37%) than for Max 1h $NO_2$ (5%), indicating that, in terms of failures, the resolution has a positive global effect by reducing false exceedances.

For various reasons, accuracy (A) remains almost constant when the resolution increases. Regarding $NO_2$ and $O_3$, it is due to a stable sum of b and c, increasing the b at the cost of c, and vice versa. For $NO_2$, the number of hits (b) to forecast Max 1h at 1 km is higher than 4 km (537 vs. 466), but the number of correct negatives at 1 km is
lower than at 4 km (2439 vs. 2517). The resolution increase has the opposite effect on 
O$_3$ over b and c for both Max 1h and Max 8h.

### 4.2 PM$_{10}$ components

The resolution increase has the lowest effect on PM$_{10}$ hourly concentrations and its 
exceedances (<1%). PM$_{10}$ components are secondary inorganic aerosols (SIA), which 
include sulfate (SO$_4$), nitrate (NO$_3$) ammonium (NH$_4$), secondary organic aerosol 
(SOA), elemental carbon (EC), sea salt (SS), desert dust (DD), and primary PM (PPM).

Pay et al. (2012a) already evaluated the PM components at some Spanish urban and 
rural background stations using the CALIOPE-AQFS based on CMAQv4.5, and they 
showed that the model underestimated the secondary inorganic aerosols by a factor of 2-3. The highest underestimation was found for fine carbonaceous aerosols (a factor of 4), 
in part related to the state-of-the-science concerning secondary organic aerosol 
formation pathways. The updated version of CMAQ, v5.0.1 includes scientific 
improvements concerning SOA formation and aerosol dynamics, which could improve 
the modeled PM$_{10}$ performance for its components.

Figure 8a shows that the resolution increase does not significantly change the PM$_{10}$ 
composition. DD remains the main contributor (~40-41%), followed by PPM (22-24%), 
SIA (~21-22%), SS (9-11%), EC (~4%) and SOA (~0.6%). However, the effect of the 
increased resolution on PM$_{10}$ component concentrations is different (Fig. 8b), 
depending on their origin, atmospheric cycle and the way they are modeled. DD 
concentrations do not change between resolutions, because they are mass conservative 
when interpolated from 0.5°x0.5° till 1 km x 1 km.

Regarding SIA, increasing the resolution increases NO$_3$ and NH$_4$ concentrations by 4 
and ~2%, respectively, and it decreases SO$_4$ by ~2%. The NH$_4$ increase means there are 
more primary precursors (H$_2$SO$_4$ or HNO$_3$/NO$_2$) available to neutralize NH$_3$ (gas) to 
NH$_4$ (aerosol). However, the variability between SO$_4$ and NO$_3$ is more difficult to 
explain, due to the nonlinearity of photochemistry and aerosol formation, which is 
controlled to some extent by the ISORROPIA thermodynamic equilibrium. 
Furthermore, the absence of aerosol measurements for April 2013 does not allow us to 
explain this situation.
The resolution increase displays the highest decrease for SS (~16%). CMAQv5.0.1 simulates SS emission as a function of the wind speed and the relative humidity (Gong, 2003; Zhang et al., 2005). Although not shown here, when the resolution increases, the wind speed increases at the available PM$_{10}$ stations by ~1.4/0.4/0.2 m s$^{-1}$ over AND/BCN/MAD, and also over the open ocean.

For primary PM components (EC and PPM), increasing resolution presents the highest increase in concentration (by 10 and ~12%, respectively). As for NO$_2$, the 1-km simulation leads to a reduced effect of artificial dilution of emissions in a grid cell, so concentration gradients are stronger than in the 4-km simulation.

### 4.3 Domain

Due to differences in geographical location and emission patterns over the domains under study, the resolution increase has different impacts (Fig. 7). BCN shows the highest NO$_2$ bias decrease (73%) when the resolution increases, yet with no effect on the correlation (<7%). However, O$_3$ shows significant variability over BCN, increasing r (by 4%) and MB (by 23%). To a lesser extent, MB also increases over AND (by 8%). Meanwhile, the variability over MAD is reduced (<4%). MB decreases for PM$_{10}$ (<1 µgm$^{-3}$) over the urban domains of MAD (3%) and BCN (16%), and increases over AND (7%).

Figure 9 analyzes the impact of the resolution increase on daily cycles. Although PBL measurements are not available, PBL daily cycles are displayed together in order to find some correlations with the daily pollutant variability. Due to the lamination of PBL growth by the Mediterranean sea breezes, the PBL reaches its maximum height at midday, being the highest in MAD (1600 m AGL) followed by AND (1000 m AGL) and BCN (900 m AGL).

As shown in Sect. S1, the pollutant transport at the BCN coastal domain is controlled by mesoscale phenomena such as sea-breezes (day) and land-breezes (night), which are a result of its complex topography and location (Baldasano et al., 1994; Millán et al., 1997; Gonçalves et al., 2009). The NO$_2$ daily cycle is highly influenced by traffic emissions (Fig. 9). Both resolutions show the highest underestimations at the morning peak (5-9 am) (~20 µgm$^{-3}$). Although the afternoon peak is well reproduced, there is excessive variability at both resolutions, which results from problems with wind
direction. During the sea breeze period, the mean simulated wind was more easterly than westerly, as registered by measurements (Sect. S1). Several works indicate that WRF does not faithfully reproduce the morning and evening transition over the urban environment, possibly because it does not model the heat retention in cities (Makar et al., 2006; Appel et al., 2013). Increasing the resolution increases the NO$_2$ concentrations from 14 µgm$^{-3}$ (4 km) to 17 µgm$^{-3}$ (1 km) during the morning hours after sunrise (5-9 am) and in the evening hours after sunset (5-9 pm). This behavior could be explained by PBL variability when increasing the resolution, which decreases PBL height by ~33 m for these hours.

NO$_2$ performance impacts the O$_3$ daily cycles over BCN, showing that 4- and 1-km simulations underestimate maximum O$_3$ concentrations by ~20 µgm$^{-3}$ at midday (1-4 pm), and it overestimates minimum O$_3$ concentration by ~20 µgm$^{-3}$ in the morning hours after sunrise (5-9 am). The resolution increase allows slightly decreasing O$_3$ concentrations at night, which is perhaps controlled by the PBL decreasing at 1 km during the early morning and late afternoon, when PBL reaches the minimum height. During these hours, the titration effect of NO$_2$ on O$_3$ is more effective, improving the O$_3$ overestimation of the daily minimum, which allows a slightly increasing hourly r (2%). However, O$_3$ underestimation increases in the late afternoon, contributing to an increase in the hourly mean bias from ~9 µgm$^{-3}$ (4 km) to ~11 µgm$^{-3}$ (1 km).

In BCN, the PM$_{10}$ underestimation is not systematic throughout the daily cycle (Fig. 9), which shows a bias of ~20/10 µgm$^{-3}$ at day/night time. The higher daytime underestimation as compared to the nighttime cannot be explained by the current results, but it could be a result of missing sources and problems with PBL overestimation and emission dilutions. The resolution increase allows reducing the bias by ~1 µgm$^{-3}$ (16%), especially during early morning and late afternoon, when the highest PBL variability between resolutions is detected. Although the evaluation of T2M, U10 and WD10 indicates that the resolution increase has a low effect over BCN (Sect. S1), the reduction of the artificial dilution of NO$_2$ emissions together with a lower PBL height at 1 km than at 4 km during the night and early morning allows improving NO$_2$, O$_3$ and PM$_{10}$ concentrations, which in turn decreases their biases.

In AND, the model at both resolutions underestimates observed NO$_2$ concentrations throughout the daily cycle (~5 µgm$^{-3}$), with the highest underestimation at the morning
peak (~25 µg m\(^{-3}\)) and the lowest at the afternoon peak (~10 µg m\(^{-3}\)). The resolution increase reduces the bias from 3.5 to 2 µg m\(^{-3}\) (by 43%) and increases \(r\) by 7% (from 0.39 to 0.41). As in BCN, the \(\text{NO}_2\) underestimation directly impacts the \(\text{O}_3\) daily cycle (which the resolution increase cannot resolve), increasing the bias by ~1 µg m\(^{-3}\), a phenomenon that is predominant in the morning hours. In the case of \(\text{PM}_{10}\), the daily cycle indicates that the biases are almost systematic throughout the daily cycle (~22 µg m\(^{-3}\)). Increasing the resolution increases the bias by less than 4% in the late afternoon, which is perhaps dominated by the PBL decrease. When the resolution is increased, \(\text{NO}_2\) performs better because of the improved model performance for the temperature and wind speed (Sect. S1), as well as the lower nocturnal and higher diurnal PBL. Meanwhile, the \(\text{O}_3\) and \(\text{PM}_{10}\) performance do not significantly change.

During April 2013, the main flow over MAD was controlled by S-SW synoptic winds channeled by orographic barriers in the NW domain and the Tajo valley (Valverde et al., 2014). The \(\text{NO}_2\) daily cycle depicts a high influence of traffic emissions (Fig. 9), showing significant model underestimation at both resolutions for the morning/afternoon peaks (~15/10 µg m\(^{-3}\)). Note that, in terms of mean and variability resulting from southeastern winds, the model performs well at the afternoon peak. \(\text{NO}_2\) performance leads to more accurate \(\text{O}_3\) daily cycles than in AND and BCN, especially in the early morning, when the titration effect of \(\text{NO}_2\) is more efficient, because the \(\text{NO}_2\) morning peak underestimation is lower when compared to the other domains. Meanwhile, the modeled \(\text{PM}_{10}\) at both resolutions presents a profile controlled by traffic emissions. Observed concentrations display a flatter daily cycle, in which the model underestimation reaches 40 µg m\(^{-3}\) in the morning. Increasing resolution shows a positive effect for \(\text{NO}_2\), and \(\text{PM}_{10}\) increases \(r\) by 0.01 and reduces MB and RMSE by 0.1-0.2 µg m\(^{-3}\). However, it depicts the lowest variability when compared to the other domains (<5 % for bias, error and \(r\)), which is the result of a relatively simpler topography and meteorological patterns.

### 4.4 Environment and major sources

Figure 7 shows that the resolution impact also depends on the type of area and the dominant emission source. Theoretically, the meteorological fields of urban areas differ from those of surrounding rural areas because of their different morphology (radiation
trapping and wind profiles), surface materials (heat storage) and variable energy consumption (heat release).

Increasing resolution reduces the NO₂ bias at suburban and urban stations by 1.8-2 µgm⁻³, and, to a lesser extent, by 1.2 µgm⁻³ at rural stations. The correlation coefficients also improve at suburban stations (from 0.48 to 0.52) and rural stations (from 0.34 to 0.35). That is not surprising, because the 1-km grid allows better allocation of land-use categories (urban vs. rural) and of their fraction in a grid cell than does a 4-km grid. The NO₂ biases exhibit a relative 39% (65%) decrease at urban (background) stations, but O₃ biases increase by 9% (5%). For PM₁₀, the resolution increase does not significantly change as a function of area type, and it depicts variation in biases and errors of less than ±4% (<0.5 µgm⁻³).

The low improvement at urban stations is obviously because the NoahLSM land-surface model does not consider the effect of urban morphology or thermal parameters in order to accurately model meteorological fields. Modeling air quality on an urban scale over cities requires a description of the heat/momentum exchange between buildings and the lower atmospheric layers. For instance, the impact of using an urban model on meteorological fields over the greater Paris area was studied by Kim et al. (2013) using WRF with the Urban Canopy Model, demonstrating that, below a 1000-m height, overestimations of wind speed were significantly reduced.

The effect of increasing resolution is positive for primary pollutants near important emission sources. For example, it reduces NO₂ biases at traffic (industrial) stations by ~3 µgm⁻³ (2 µgm⁻³), but it increases O₃ biases by ~2 µgm⁻³ (1 µgm⁻³). However, the resolution increase in the range of 4-1 km does not exhibit the expected improvement on the hourly statistics that are based on the constraints of the current model formulation. In other words, it cannot resolve the subgrid air quality variability merely by increasing resolution. For instance, although on-road traffic emissions are estimated by following a bottom-up approach along highways and routes, heterogeneity is lost in the CTM volume averaging process, which artificially dilutes emission rates over the grid cells. The resolution effect is the lowest at background stations, which are not influenced by any single source, but rather by the integrated contribution from all sources upwind of the stations where variations are less than 1% for O₃ and PM₁₀ (< 1
µgm\(^3\)). However, background NO\(_2\) levels increase by \(-1\) µgm\(^3\) (48\%) from 4 km to 1 km. Figure 10 shows the temporal series and daily cycles for NO\(_2\) and O\(_3\) at traffic and background stations throughout the episode of 12-18\(^{th}\) April, 2013. At traffic stations, the temporal series show a remarkable O\(_3\) daily cycle (observed 25p = 23.2 µgm\(^3\) and 75p = 77.5 µgm\(^3\)), due to O\(_3\) destruction caused by high NO\(_x\) levels (observed 50p = 34.5 µgm\(^3\)). In contrast, the NO\(_2\)–limited regime at background sites (observed 50p = 19 µgm\(^3\)) allows higher O\(_3\) concentrations (observed 25p = 38 µgm\(^3\) and 75p = 89 µgm\(^3\)) than in high NO\(_2\) environments.

During the episode mentioned above, the resolution increase at traffic stations had a positive effect by increasing the correlation coefficient for NO\(_2\) (from 0.73 to 0.76) and O\(_3\) (from 0.83 to 0.86), and also by decreasing the NO\(_2\) mean bias by \(-5\) µgm\(^3\) (from 6 to 1 µgm\(^3\)). The NO\(_2\) daily cycle improves in the morning hours after sunrise, reducing bias by 5-10 µgm\(^3\) and contributing to a reduction in O\(_3\) overestimations (~5 µgm\(^3\)). In contrast, at background stations, where the NO\(_x\)/O\(_3\) chemistry is less dominant, the resolution effect is not significant. Such behavior indicates that finer resolution improves the performance, because horizontal resolution affects the representation of chemical processes near large emission sources, such as the efficient formation of O\(_3\) and nighttime O\(_3\) titration (Mathur et al., 2005). However, the loss of subgrid variability and improved meteorological fields (transport and temperature) are required.

5 Conclusions

The present work shows the effects of increasing the horizontal resolution from 4 km to 1 km using the CALIOPE-AQFS on pollutant concentrations (NO\(_2\), O\(_3\), and PM\(_{10}\)) over three Spanish domains (AND, BCN and MAD) in April 2013.

The global features of concentration maps at both resolutions are quite similar, with zones of high/low concentration identically located, which is obviously because both simulations are based on the same emission dataset. Further comparisons demonstrate that increasing the resolution provides better-defined and more realistic concentration structures over large sources (roads and industries) and complex terrains (more sharply defined orographic hills). The titration effect on O\(_3\) concentrations along highways and major point sources is more evident in 1-km simulations than at 4 km, because the latter...
is affected by higher dilution within the grid cells. This improvement is quantified by an increase in spatial correlation coefficients of 3% (6%) for NO₂ (O₃).

However, the resolution increase in the range of 4-1 km does not exhibit the expected improvement in hourly statistics for any pollutant. Hourly correlation coefficients do not significantly change, and absolute (relative) errors and biases vary < 2 µgm⁻³ (9%). The merit of the resolution increase may be underrated when classical statistics are applied at measurement stations (Mass et al., 2002; Gego et al., 2005). For instance, although the structure of important NO₂ urban plume features (> 40 µgm⁻³) often become more realistic (stronger and more defined plumes) as resolution increases, statistics are deeply degraded by even small timing and spatial errors.

The resolution increase has a significant impact on reducing NO₂ hourly bias (by 42%, 2 µgm⁻³), without any significant change in the error and the r (<2%). But it increases O₃ hourly biases (<1 µgm⁻³). The main differences between resolutions appear at daytime and nighttime traffic peaks, when the mixing height experiences rapid changes, allowing the 1-km simulation to slightly reduce NO₂ underestimation in the morning by ~5-10 µgm⁻³. The O₃ daily cycles at large sources depict a high influence of hourly NO₂ concentrations, increasing the hourly O₃ bias by ~3 µgm⁻³. That behavior is controlled by the daytime O₃ underestimation and, to a lesser extent, by the nighttime overestimation. The resolution increase allows reducing the O₃ overestimations at night (by ~5 µgm⁻³), partly because of higher nocturnal NO₂ concentrations.

Concerning the capability of forecasting 75p exceedances in the observed maximum 1h concentrations, the increased resolution has a positive effect: it increases the number of hits that forecast 75p exceedances in the observed Max 1h NO₂ (537 vs. 466 over 953 exceedances), and it reduces the false alarms for Max 1h O₃ exceedances (FAR improves by 37%).

The grid effect is less pronounced for PM₁₀ than for NO₂ and O₃. When the resolution increases, the low increment of PM₁₀ mean concentrations (<0.1 µgm⁻³) is the result of compensating biases of PM₁₀ components, which is controlled mainly by the PPM and EC increase as well as the SS decrease.

BCN is the domain where the resolution increase has the highest effect, with changes in bias (error) of 16-73% (< 5%), followed by AND with 4-43% (< 5%) and MAD < 3-5%.
(< 1%). In BCN, as in the western Mediterranean Basin, the transport of O\textsubscript{3} and its precursors is governed by mesoscale circulation. In that sense, the resolution increase has a great impact over BCN, where induced mesoscale phenomena control the air flow, meanwhile synoptic transport is more prominent in MAD and AND. The benefits of increasing the resolution to 1 km over rural areas (Mass et al., 2002) are that it increases the accurate representation of mesoscale meteorological structures such as orographic wind and circulation. Over urban areas along the western Mediterranean coast (Toll and Baldasano, 2002; Jiménez et al., 2006; Fay and Neunhäuserer, 2006), further improvements and urbanization steps are required before seeing any benefits in increasing the resolution to 1 km.

In urban areas or near large emission sources (industrial and traffic stations), NO\textsubscript{2} and O\textsubscript{3} concentrations are more sensitive to changes in the grid resolution. The concentration increase in primary anthropogenic pollutants (NO\textsubscript{2}, PPM and EC) is obvious because the high resolution allows better allocation of emissions at point, linear and area sources. What is more, it decreases the artificial dilution of emissions when compared to the larger grid area. However, the 1-km simulation attempt to more accurately describe the chemical formation of O\textsubscript{3} and dilution of NO\textsubscript{2} concentrations over those areas was not generally successful.

This analysis demonstrates weaknesses in the current model formulations that cannot be resolved with only high-resolution modeling. The subgrid air quality variability at 1-km resolution is not reproduced over large emission sources or urban areas, because a finer spatial structure is expected but unresolved. There are some underlying problems. First, there is a loss of subgrid emission heterogeneity. Emission inputs to CTM are an average rate, which accounts for the volume averaged quantity of mass released per unit of time. No other information regarding emission allocation (e.g., point, linear or per area) is considered; for instance a large amount of mass can be emitted by a small portion of the grid surface or by several sources scattered around it (Galmarini et al., 2008; Cassiani et al., 2010; Ching and Majeed, 2012). Despite the fact that emissions are estimated by following a bottom-up approach emission model, emission heterogeneity is lost in the volume averaging process performed within CTM. The loss is even higher when resolution decreases (from 1 km to 4 km). Second, there is a low degree of complexity in flow and dispersion details at urban scales, where most of the pollutants come from street canyons and/or tree canopies, where they are transported
until mixing conditions allow the pollutants to disperse above these urban canopy levels 
(Kim et al., 2013; Ching, 2013). Third, the USGS land-use data used in the WRF model 
is based on 1993 data, and urban changes in MAD and BCN over the last 20 years are 
significant.

Since temperature and wind speed are very sensitive to the ratio of building width to 
road width, the next improvement should focus on using an urban canopy model that 
considers effects on the transfer of energy and momentum between urban structures and 
the lower atmosphere. This is crucial for modeling meteorology and air quality. 
However, it requires an urban canopy scheme and a canopy parameter database (urban 
fraction, building height and area). Furthermore, in order to gain any benefits from 
increasing resolution, the meteorological modeling should include an improved 
description of the land instead of relying on USGS data from the year 1993. To this end, 
the Coordination of Information on the Environment (CORINE) provides a high 
resolution (100 m) land use database, which was developed by the European 
Environmental Agency and updated to the year 2006 (CLC2006) (EEA, 2007). This 
could be implemented in the WRF model following the methodology described in 
Pineda et al. (2004).
### Appendix A

Table A1. Definition of the discrete statistics used in the evaluation. Where \( C_m(x, t) \) and \( C_o(x,t) \) are the modeled and observed concentrations at a location \( (x) \) and time \( (t) \); \( N \) is the number of pairs of data. \( \bar{C}_m \) and \( \bar{C}_o \) are the modeled and observed mean concentrations over the whole period, respectively.

<table>
<thead>
<tr>
<th>Statistic</th>
<th>Formula</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mean bias</td>
<td>[ MB = \frac{1}{N} \sum_{i=1}^{N} (C_m(x,t) - C_o(x,t)) ] A1</td>
</tr>
<tr>
<td>Mean normalized bias error</td>
<td>[ MNBE = \frac{1}{N} \sum_{i=1}^{N} \frac{(C_m(x,t) - C_o(x,t))}{C_o(x,t)} \cdot 100 ] A2</td>
</tr>
<tr>
<td>Mean fractional bias</td>
<td>[ MFB = \frac{1}{N} \sum_{i=1}^{N} \frac{(C_m(x,t) - C_o(x,t))}{(C_o(x,t) + C_m(x,t))/2} \cdot 100 ] A3</td>
</tr>
<tr>
<td>Mean error</td>
<td>[ ME = \frac{1}{N} \sum_{i=1}^{N}</td>
</tr>
<tr>
<td>Mean normalized gross error</td>
<td>[ MNGE = \frac{1}{N} \sum_{i=1}^{N} \frac{</td>
</tr>
<tr>
<td>Mean fractional error</td>
<td>[ MFE = \frac{1}{N} \sum_{i=1}^{N} \frac{</td>
</tr>
<tr>
<td>Root mean squared error</td>
<td>[ RMSE = \sqrt{\frac{1}{N} \sum_{i=1}^{N} (C_m(x,t) - C_o(x,t))^2} ] A7</td>
</tr>
<tr>
<td>Correlation coefficient</td>
<td>[ r = \frac{\sum_{i=1}^{N} (C_m(x,t) - \bar{C}_m)(C_o(x,t) - \bar{C}<em>o)}{\sqrt{\sum</em>{i=1}^{N} (C_m(x,t) - \bar{C}<em>m)^2} \sqrt{\sum</em>{i=1}^{N} (C_o(x,t) - \bar{C}_o)^2}} ] A8</td>
</tr>
</tbody>
</table>

Table A2. Definition of the categorical statistics used in the evaluation. Exceedance analysis is based on a comparison with a fixed threshold concentration \( (T) \), where \( a \) is the number of false alarms, \( b \) is the number of hits, \( c \) is the number of correct negatives, and \( d \) is the number of misses.

<table>
<thead>
<tr>
<th>Statistic</th>
<th>Formula</th>
</tr>
</thead>
<tbody>
<tr>
<td>Accuracy</td>
<td>[ A = \left( \frac{b+c}{a+b+c+d} \right) \cdot 100 ] A9</td>
</tr>
<tr>
<td>Bias</td>
<td>[ B = \left( \frac{a+b}{b+d} \right) \cdot 100 ] A10</td>
</tr>
</tbody>
</table>
Critical success index

\[ CSI = \left( \frac{b}{a + b + d} \right) \times 100 \]  

A11

Probability of detection

\[ POD = \left( \frac{b}{b + d} \right) \times 100 \]  

A12

False alarm ratio

\[ FAR = \left( \frac{a}{a + b} \right) \times 100 \]  

A13

1

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Table 1. CALIOPE-AQFS computational requirements, in terms of Central Processor Units (CPU) and computational time (in min), for simulating 48h air quality forecasts as a function of the domain: IP-4km (D2), AND-1km (D3), BCN-1km (D5) and MAD-1km (D4), all of which are described in Figure 1. D-domains are described in Figure 1.

<table>
<thead>
<tr>
<th>Domain</th>
<th>Meteorological Modeling</th>
<th>Emission Modeling</th>
<th>Air Quality Modeling</th>
</tr>
</thead>
<tbody>
<tr>
<td>D2-4km</td>
<td>128 CPU/15 min</td>
<td>1 CPU/ 4 min</td>
<td>256 CPU/210 min</td>
</tr>
<tr>
<td>D3-1km</td>
<td>256 CPU/80 min</td>
<td>1 CPU/ 4 min</td>
<td>256 CPU/220 min</td>
</tr>
<tr>
<td>D5-1km</td>
<td>128 CPU/20 min</td>
<td>1 CPU/1 min</td>
<td>128 CPU/150 min</td>
</tr>
<tr>
<td>D4-1km</td>
<td>128 CPU/20 min</td>
<td>1 CPU/1 min</td>
<td>128 CPU/110 min</td>
</tr>
</tbody>
</table>

(399x399 cells)  (366x358 cells)  (146x146 cells)  (146x158 cells)
Table 2. Discrete and categorical statistics for NO$_2$, O$_3$, O$_3$-8h, and PM$_{10}$ for April 2013 as a function of horizontal resolution (4 km and 1 km). n indicates the number of pairs of data used in the discrete evaluation on an hourly basis. OM and MM depict the measured and modeled mean concentrations, respectively. T is the threshold applied in the categorical evaluation. Max 1h and mean 24h concentrations are calculated considering ≥75% of the hours in a day, as established by Directive 2008/50/EC.

<table>
<thead>
<tr>
<th></th>
<th>NO$_2$</th>
<th>O$_3$</th>
<th>O$_3$-8h</th>
<th>PM$_{10}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>n (stations)</td>
<td>90761 (135)</td>
<td>76471 (114)</td>
<td>3248 (114)</td>
<td>66642 (100)</td>
</tr>
<tr>
<td>OM ($\mu$gm$^{-3}$)</td>
<td>22.0</td>
<td>68.4</td>
<td>88.6</td>
<td>20.6</td>
</tr>
<tr>
<td>EU LV/TV ($\mu$gm$^{-3}$) (temp basis)</td>
<td>200 (Max 1h)</td>
<td>180 (Max 1h)</td>
<td>120 (Max 8h)</td>
<td>50 (Mean 24h)</td>
</tr>
<tr>
<td>EU LV/TV exceedances</td>
<td>0</td>
<td>25</td>
<td>0</td>
<td>31</td>
</tr>
<tr>
<td>T* ($\mu$gm$^{-3}$) (temp. basis)</td>
<td>71 (Max 1h)</td>
<td>108 (Max 1h)</td>
<td>101 (Max 8h)</td>
<td>27 (Mean 24h)</td>
</tr>
</tbody>
</table>

Discrete evaluation

<table>
<thead>
<tr>
<th></th>
<th>4 km</th>
<th>1 km</th>
<th>4 km</th>
<th>1 km</th>
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<td>MM ($\mu$gm$^{-3}$)</td>
<td>17.4</td>
<td>19.3</td>
<td>58.0</td>
<td>57.3</td>
<td>72.4</td>
<td>71.5</td>
<td>13.9</td>
<td>14.0</td>
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<tr>
<td>r</td>
<td>0.54</td>
<td>0.54</td>
<td>0.61</td>
<td>0.61</td>
<td>0.54</td>
<td>0.51</td>
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<tr>
<td>MB ($\mu$gm$^{-3}$)</td>
<td>-4.5</td>
<td>-2.6</td>
<td>-10.5</td>
<td>-11.3</td>
<td>-16.3</td>
<td>-17.2</td>
<td>-6.7</td>
<td>-6.6</td>
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<tr>
<td>MAE ($\mu$gm$^{-3}$)</td>
<td>12.9</td>
<td>13.2</td>
<td>19.7</td>
<td>19.8</td>
<td>18.4</td>
<td>19.2</td>
<td>12.6</td>
<td>12.7</td>
</tr>
<tr>
<td>RMSE ($\mu$gm$^{-3}$)</td>
<td>19.8</td>
<td>20.4</td>
<td>24.6</td>
<td>24.7</td>
<td>21.8</td>
<td>22.8</td>
<td>17.2</td>
<td>17.4</td>
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<tr>
<td>MNBE (%)</td>
<td>-20.4</td>
<td>-11.8</td>
<td>-15.3</td>
<td>-16.5</td>
<td>-18.4</td>
<td>-19.4</td>
<td>-32.5</td>
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<tr>
<td>MNGE (%)</td>
<td>58.5</td>
<td>59.9</td>
<td>28.8</td>
<td>28.9</td>
<td>20.8</td>
<td>21.7</td>
<td>61.1</td>
<td>61.6</td>
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<tr>
<td>MFB (%)</td>
<td>-28.5</td>
<td>-23</td>
<td>-13.1</td>
<td>-15.1</td>
<td>-19.3</td>
<td>-20.5</td>
<td>-63.3</td>
<td>-64.1</td>
</tr>
<tr>
<td>MFE (%)</td>
<td>69.2</td>
<td>68.7</td>
<td>37.1</td>
<td>37.4</td>
<td>22.5</td>
<td>23.5</td>
<td>85.7</td>
<td>87.1</td>
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Categorical evaluation (Threshold = T*)

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<tr>
<th></th>
<th>4 km</th>
<th>1 km</th>
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<th>1 km</th>
<th>4 km</th>
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<tbody>
<tr>
<td>a (false alarm)</td>
<td>306</td>
<td>384</td>
<td>41</td>
<td>19</td>
<td>17</td>
<td>6</td>
<td>131</td>
<td>133</td>
</tr>
<tr>
<td>b (hits)</td>
<td>466</td>
<td>537</td>
<td>112</td>
<td>96</td>
<td>6</td>
<td>4</td>
<td>331</td>
<td>334</td>
</tr>
<tr>
<td>c (correct negative)</td>
<td>2517</td>
<td>2439</td>
<td>1846</td>
<td>1868</td>
<td>2826</td>
<td>2837</td>
<td>1978</td>
<td>1976</td>
</tr>
<tr>
<td>d (misses)</td>
<td>487</td>
<td>416</td>
<td>1194</td>
<td>1210</td>
<td>399</td>
<td>401</td>
<td>334</td>
<td>331</td>
</tr>
<tr>
<td>B (%, 100)</td>
<td>37</td>
<td>40</td>
<td>8</td>
<td>7</td>
<td>1</td>
<td>1</td>
<td>42</td>
<td>42</td>
</tr>
<tr>
<td>POD (%, 100)</td>
<td>49</td>
<td>56</td>
<td>9</td>
<td>7</td>
<td>1</td>
<td>1</td>
<td>50</td>
<td>50</td>
</tr>
<tr>
<td>CSI (%, 100)</td>
<td>81</td>
<td>97</td>
<td>12</td>
<td>9</td>
<td>6</td>
<td>2</td>
<td>69</td>
<td>70</td>
</tr>
<tr>
<td>FAR (%, 0)</td>
<td>40</td>
<td>42</td>
<td>27</td>
<td>17</td>
<td>74</td>
<td>60</td>
<td>28</td>
<td>28</td>
</tr>
<tr>
<td>A (%, 100)</td>
<td>79</td>
<td>79</td>
<td>61</td>
<td>62</td>
<td>87</td>
<td>87</td>
<td>83</td>
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</table>

T* is defined as 75p of the observed concentrations estimated temporally, as established by EU Directive 2008/50/EC
Fig. 1. CALIOPE-AQFS nesting strategy (D-domains) and study domains (Andalucia, AND; Madrid, MAD; and Barcelona, BCN). Colour chart at D-domains shows NO2 emission rate (kg h\(^{-1}\)) for 17\(^{th}\) April, 2013 at 18UTC. HERMES-DIS model generates emissions at 12 km x 12 km over Europe (the mother domain, D1) by performing disaggregation from the EMEP database. HERMES-BOUP model estimates emissions at 1 km x 1 km, following a bottom-up approach.
Figure 2. Air quality stations for NO$_2$, O$_3$ and PM$_{10}$ in the three domains under study (AND, BCN and MAD) in April 2013. Different types of stations are shown by symbols and color codes. The various symbols represent the major emission type affecting each station (Traffic: triangle; Industrial: square; and Background: circle), while the colors reflect the environment of each station (Urban: red; Suburban: green; and Rural: orange). Cyan dots represent METAR stations used in Sect. S1.
Figure 3. CALIOPE-AQFS mean NO₂ concentration (µgm⁻³) in April 2013 over (a,b) MAD, (c,d) BCN, and (e,f) AND, as a function of horizontal resolution: 4 km (left column) and 1 km (right column). Dots indicate mean concentration at air quality stations.
Figure 4. CALIOPE-AQFS mean O₃ concentration (µgm⁻³) in April 2013 over (a,b) MAD, (c,d) BCN, and (e,f) AND, as a function of horizontal resolution: 4 km (left column) and 1 km (right column). Dots indicate mean concentration at air quality stations.
Figure 5. CALIOPE-AQFS mean PM$_{10}$ concentration (µg m$^{-3}$) in April 2013 over (a,b) MAD, (c,d) BCN, and (e,f) AND, as a function of horizontal resolution: 4 km (left column) and 1 km (right column). Dots indicate mean concentrations at air quality stations.
Figure 6. Monthly mean scatter plots for CALIOPE-AQFS (y-axis) and observed (x-axis) concentrations for the three study domains (AND in green, BCN in yellow, and MAD in red), as a function of horizontal resolution for (a) NO$_2$, (b) O$_3$ and (c) PM$_{10}$. Equations show the linear adjustment between models and observations at 1 km (light grey) and 4 km (dark grey). Spatial correlation coefficients as a function of resolution and domain are shown for (e) NO$_2$, (f) O$_3$, and (g) PM$_{10}$. 

<table>
<thead>
<tr>
<th></th>
<th>$r_{\text{nea}}$</th>
<th></th>
<th>$r_{\text{obs}}$</th>
<th></th>
<th>$r_{\text{seas}}$</th>
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<tbody>
<tr>
<td>All</td>
<td>0.79 0.81</td>
<td>4km</td>
<td>1km</td>
<td>All</td>
<td>0.69 0.73</td>
</tr>
<tr>
<td>AND</td>
<td>0.62 0.71</td>
<td></td>
<td></td>
<td>AND</td>
<td>0.58 0.64</td>
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<tr>
<td>BCN</td>
<td>0.72 0.73</td>
<td></td>
<td></td>
<td>BCN</td>
<td>0.54 0.57</td>
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<tr>
<td>MAD</td>
<td>0.70 0.74</td>
<td></td>
<td></td>
<td>MAD</td>
<td>0.71 0.73</td>
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Figure 7. Statistics (r, MB, and RMSE in rows) for each pollutant (NO₂, O₃, and PM₁₀ in columns) on an hourly basis as a function of horizontal resolution: 4 km (black) and 1 km (grey). Four categories are considered: all stations (all), domain (AND, BCN and MAD), station environment (R, S, and U), and main sources (B, I, and T).
Figure 8. Resolution effect on PM\textsubscript{10} components in April 2013. (a) Percentage of PM\textsubscript{10} components: sulfate (SO\textsubscript{4}), nitrate (NO\textsubscript{3}), ammonium (NH\textsubscript{4}), secondary organic aerosol (SOA), elemental carbon (EC), sea salt (SS), desert dust (DD), and primary particulate matter (PPM). (b) PM\textsubscript{10} component concentrations in the 1-km simulation (black) and 4-km simulation (grey). Numbers over bars indicate the % of increase when increasing resolution.
Figure 9. Daily cycles for NO$_2$, O$_3$ and PM$_{10}$ for each study domain at available stations as a function of resolution. No observations of PBL are available. Q1, Q2 and Q3 indicate quartiles for the daily cycle. Bars show Q1 and Q3 at each hour.
Figure 10. Temporal series and daily cycles for NO$_2$ and O$_3$ at background (a and b, respectively) and traffic stations (c and d, respectively) for the episode of 12-18$^{th}$ April, 2013. Q1, Q2 and Q3 indicate quartiles for the daily cycle. Bars show Q1 and Q3 at each hour.