Implementation of plume rise and its impacts on emissions and air quality modelling

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HIGHLIGHTS

- Stack emissions are vertically allocated using fixed profiles or plume-rise models.
- Effective emission heights show large differences depending on source and pollutant.
- Currently used vertical fixed emission profiles do not represent this variability.
- Plume-rise models lead to improved simulation of industrial SO2 concentrations.
- When applying plume-rise models the use of real-world stack parameters is mandatory.

ABSTRACT

This work analyses the impact of implementing hourly plume rise calculations over Spain in terms of: i) vertical emission allocations and ii) modelled air quality concentrations. Two air quality simulations (4 km × 4 km, 1 h) were performed for February and June 2009, using the CALIOPE-AQFS system (WRF-ARW/HERMESv2.0/CMAQ/BSC-DREAM8b) differing only by the vertical allocation of point source emissions: i) using fixed vertical profiles based on the stack height of each facility and ii) using an hourly bottom-up calculations of effective emission heights. When using plume rise calculations, emissions are generally allocated to lower altitudes than when using the fixed vertical profiles, showing significant differences depending on source sector and air pollutant (up to 75% between estimated average effective emission heights). In terms of air quality, it is shown that hourly plume rise calculations lead to improved simulation of industrial SO2 concentrations, thus increasing modelled concentrations (1.4 μg m⁻³ increase in February, 1.5 μg m⁻³ increase in June) and reducing the model biases for both months (31.1% in February, 73.7% in June). The increase of SO2 concentrations leads to an increase of SO₄²⁻ surface levels that varies according to the season and location (4.3% in February and 0.4% in June, on average). On the other hand, the impact on NO2 and PM10 concentrations is less significant, leading to average changes of a few μg m⁻³ at most (0.4 μg m⁻³ for NO2 and 0.2 μg m⁻³ for PM10). In order to maximize the precision of plume rise calculations, the use of real-world stack parameters is mandatory.

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1. Introduction

Emission models play a key role in the development of air quality modelling systems (Russell and Dennis, 2000). One of the key aspects in regards to emission inputs is their representativeness in terms of spatial (horizontal and vertical) distribution, temporal distribution and chemical speciation. The horizontal allocation of emissions is usually performed using proxy parameters such as land use, population density or transportation networks (e.g. Kuenen et al., 2014); temporal distribution is commonly described according to average daily, weekly and seasonal time profiles per pollutant sector (e.g. Mues et al., 2014), whereas the chemical speciation is performed according to existing databases that link speciation profiles with air pollution sources (e.g. Simon et al., 2010). In the case of vertical allocation, which mainly affects point sources, emissions can be allocated to different model
layers according to two methodologies: (i) using fixed vertical profiles based on estimates (e.g. the EMEP profiles; Vidic, 2002) or (ii) using plume-rise models that consider stack characteristics as well as meteorological data to estimate effective emission heights (e.g. Bieser et al., 2011).

Plume rise modelling received much attention during the late 1960’s/early 1970’s when power and industrial sulphur dioxide (SO2) emissions peaked, largely contributing to acidic deposition (Briggs, 1971; Linkens and Borman, 1974). The subsequent reduction of these emissions increased the relative significance of other atmospheric pollutants (nitrogen oxides, NOx) and pollutant sources (road transport), shifting the focus of atmospheric dispersion modelling to urban areas. However, recently the role that vertical emission injection plays in modelling has started being discussed again with more interest. Pregger and Friederich (2009) provided typical default values for the driving parameters stack height, flue gas temperature, flue gas velocity and flue gas flow rate for 34 categorized power and industrial sources. All these parameters, derived from a database of real-world stack information (Wickert, 2001), were used to calculate effective emission heights for each source type by applying equations of the Association of German Engineers (VDI, 1985). Results showed significant differences in effective heights depending on source and air pollution, compared to other approaches commonly used (e.g. De Meij et al., 2006). Similarly, Bieser et al. (2011) used the SMOKE-EU model and the stack database provided by Pregger and Friederich (2009) to calculate 44,976 vertical profiles for Europe depending on the Selected Nomenclature for Air Pollution (SNAP) sector, country, climate zone, season, day and night and pollutant. From all these profiles, 73 were selected by means of hierarchical cluster analysis. The CMAQ chemical transport model was run over Europe (54 km x 54 km) and with emissions using the 73 point–source profiles and the EMEP profiles. Results showed that predicted SO2 and sulphate (SO4^2-) concentrations in the surface layer were higher when using the new vertical profiles. However, a comparison against observations was not performed. In terms of air quality, Mailler et al. (2013) studied and evaluated the impact of using different vertical profiles on SO2, NOx and O3 surface concentrations. Five one-year air quality simulations over Europe were performed using the CHIMERE chemistry transport model and the EMEP emission database. The simulations differed only by the vertical profiles used: one using the EMEP profiles only, and three using the EMEP with multiplication of injection height by 0.75, 0.5 and 0.25, and lastly a third one which used vertical profiles derived from Bieser et al. (2011). The evaluation of the impact of these updated vertical profiles in surface layer concentrations was only performed in background stations due to the coarse resolution used (0.5° x 0.5°). Results showed that applying the Bieser et al. (2011) profiles leads to significantly improved simulations of background NOx and SO2 levels.

The purpose of the present paper is to analyse and evaluate the impacts of implementing hourly plume rise calculations in terms of: i) vertical emission allocations and ii) modelled air quality concentrations by means of high resolution air quality modelling. In order to perform this task, the CALIOPE-AQFS air quality forecast system (WRF-ARWv3.2.1/CMAQ-ModPlmrs/CNS) was used to run two simulations over Spain for February and June 2009. These two simulations differ only by the vertical allocation of point source emissions: i) using a fixed vertical profile based on the stack height of each facility and ii) using an hourly bottom-up calculation of effective emission heights applying the CMAQ in-line plume rise module. The approach presented in this study has several distinctive features: i) a high spatial (4 km x 4 km) and temporal (1 h) resolution is adopted in order to investigate more in-depth the small-scale effects of plume rise calculations on industrial and urban areas; ii) plume rise calculations are applied using a bottom-up approach through the use of a specific Spanish point source database (1796 facilities considered) developed in the Earth Science department of the Barcelona Supercomputing Center – Centro Nacional de Supercomputación (BSC–CNS) and mainly based on real-world information; iii) the impact of these plume rise calculations is assessed by analysing the results obtained in terms of vertical emission allocation, and comparing them not only against the fixed vertical profiles currently used in the model but also against widely used emission profiles from the relevant literature: The EMEP (Vidic, 2002) and the Bieser et al. (2011) profiles (the latter are based on the average stack data reported by Pregger and Friederich (2009)); iv) results are evaluated for SO2, SO4^2-, NO2 and PM10 surface concentrations by comparison with measurements in order to analyse how the additional information improve the model results.

Section 2 describes the model setup and configuration, including the plume rise algorithms and point source database used. Section 3 analyses the model results against available observational data. Finally, Section 4 summarizes and discusses the results.

2. Methodology

The CALIOPE-AQFS modelling framework (Baldasano et al., 2011, 2014) was set up to perform the simulations for February and June 2009 in the Iberian Peninsula domain (IP-4 km). In order to provide adequate boundary conditions and initial conditions to the IP-4 km domain, CALIOPE-AQFS was initially run on a regional scale to model the European domain (EU-12 km), which consists of a grid of 479 x 399 points with 12 km x 12 km horizontal resolution and centered at 6.074 Lon and 42.546 Lat. Then, a one-way-nesting was performed from one domain to the other in order to retrieve the meteorological and chemical conditions. The study domain, centered in Spain (SW of Europe), is approximately 1596 km x 1596 km and centred at –3.164 Lon and 39.971 Lat, with a 4 km x 4 km horizontal resolution and a Lambert Conformal projection (Fig. 1a). The configurations and parameterizations of the meteorological (WRF-ARWv3.2.1) and chemical transport (CMAQ5.0.1) models used are summarized in the supplementary material (Table S1).

2.1. Plume rise algorithm

The CMAQ in-line plume rise module (CMAQ-ModPlmrs) calculates time-dependent effective plume heights for point sources employing a multi-layer stability-dependent plume rise (Houyoux, 1998). The approach is based on the algorithm first employed in the Regional Acid Deposition Model (RADM; Byun and Binkowski, 1991) with some important improvements. This algorithm distinguishes between three stability regimes (stable, neutral, and unstable) and injects emissions into all layers from the bottom through to the top of the plume. There is a widely applied “rule-of-thumb” (Turner and Schulze, 2007), which assumes that plume depth equals plume rise when distributing mass to multiple model layers in all conditions. This “rule-of-thumb” is adopted within the algorithm to determine the plume bottom and top, assuming their existence at 50% of the plume rise below and above the centreline, respectively. The stack and meteorological variables used to determine the effective emission heights are: i) stack height, stack diameter and stack flue gas exit temperature and flow rate and ii) temperature, pressure, wind speed, water vapour mixing ratio and Planetary Boundary Layer (PBL) height.

The set of equations used by CMAQ-ModPlmrs to determine effective emission heights are provided by Bieser et al. (2011), while a detailed analysis of the performance of the methodology
compared to other approaches to calculate emission heights can be found in Emery et al. (2010).

2.2. HERMESv2.0 — point source database

HERMESv2.0 is a high resolution emission model that estimates atmospheric emissions for Spain with a temporal and spatial resolution of 1 h and up to 1 km², according to the SNAP nomenclature and taking 2009 as the reference period (Guevara et al., 2013, 2014). The model uses a bottom-up approach for the most significant pollutant sources (e.g., point sources, road transport), while for the others a combination of top-down approaches (e.g., residential/commercial combustion) and downscaling methodologies (e.g., use of solvents, agriculture) is adopted. Biogenic emissions are estimated using the MEGAN model (Guenther et al., 2006), which is fully implemented inside the HERMESv2.0 code.

In Spain, a large share of anthropogenic emissions is emitted by point sources (Fig. 2). More than 90% of the SOₓ emissions are emitted from facilities related to SNAP01 (Combustion in energy), SNAP03 (industrial combustion) and SNAP04 (industrial processes) sectors. In the case of NOₓ and PM₁₀ these shares are 40% and 20%, respectively. Taking into account the important contribution of these sources, HERMESv2.0 estimates their emission according to a point source database that compiles the following information per stack: facility name, SNAP elemental activity code, geographical location, stack height, activity factor, emission factors for each pollutant, temporal profiles and speciation profiles. A total of 1796 stacks are considered in the inventory, including the main energy and manufacturing industries (Fig. 1b).

Currently, in HERMESv2.0 the vertical assignment of point source emissions is performed using a constant vertical distribution. Each SNAP sector point source is individually assigned to one of the eleven first CMAQ model layers considering its stack height (Table 1). This assignment is based on the knowledge acquired by the Earth Science department of the Barcelona Supercomputing Center — Centro Nacional de Supercomputación (BSC—CNS).
performing air quality impact studies (http://www.bsc.es/earth-sciences/technology-transferstudies). The rest of the pollutant sectors (SNAP02-05-06-07-08-10) are treated as near-ground emissions (area and line sources), so it is assumed that they are released into the lower layer of the vertical domain.

In order to adapt the HERMESv2.0 point source database to the CMAQ-ModPlms requirements, the following information was added per facility (besides stack height [m]): stack diameter [m], and the stack flue gas exit temperature [K], velocity [m s\(^{-1}\)], and flow rate [m\(^3\) s\(^{-1}\)]. This information was mainly obtained from Integrated Environmental Authorisations (IEAs) and Environmental Reports (ERs) reported by governmental authorities, as well as from stack databases provided by the Galician Registry of Emissions (REGADE, personal communication) and the Andalusia’s Environmental Management Company (EGMASA, personal communication). For those facilities without available local real-world data two different approaches were assumed: i) use of average values from other stacks grouped in the same pollutant sector or ii) use of stack characteristics published by Pregger and Friederich (2009) (the later only used for coke ovens, blast furnaces and basic oxygen furnaces). Stack height and stack diameter parameters are mostly based on real-world data (IEA/ER/Measurements) and averaged values, while information related to temperature and flow rate is mainly obtained from Pregger and Friederich (2009) and Yang et al. (1998) (Fig. S1). The stack height is the parameter that presents higher shares of total NO\(_x\), SO\(_x\) and PM\(_{10}\) point sources emissions released from stacks based on real-world data (IER/ER/Measurements) (59%, 74% and 46%, respectively), while the lowest values are observed in temperature and flow rate (between 9.5% and 18.4%) (Table S2). A comparison between the averaged stack height values based on Spanish real-world data and obtained from the Pregger and Friederich (2009) database showed significant discrepancies. This was found especially for the “Power stations ≥50 MWth” and “Petroleum refining” categories, for which HERMESv2.0 presents stack heights two times higher than the ones reported by Pregger and Friederich (2009) (189 m versus 96 m and 107 m versus 54 m, respectively) (Table S3).

3. Results and discussion

Results obtained using both the fixed vertical allocation (SIM-Fixed simulation hereinafter) and the CMAQ-ModPlms calculations (SIM-Plms simulation hereinafter) are discussed in terms of vertical emission allocation and air quality concentrations. It is important to highlight that in all cases the modelled outputs are presented without any correction factor or post-process applied.

3.1. Vertical emission profiles

The hourly vertical emission profiles calculated in this study (SIM-Plms) were averaged and contrasted not only against the fixed ones currently used in HERMESv2.0 (SIM-Fixed) but also against other emission profiles from the literature: The EMEP profiles (Vidic, 2002) and the Bieser et al. (2011) profiles estimated for Spain. Fig. 3 shows a comparison of the fractions emitted for different SNAP sectors (SNAP01, 03 and 04) and pollutants (SO\(_x\), NO\(_x\) and PM\(_{10}\)) in the different model layers when using each one of the vertical profiles mentioned. On the other hand, Table 2 summarises the average effective emission heights [m] obtained in each case. Note that the EMEP and Bieser et al. (2011) emission profiles used for this comparison do not take into account fugitive emissions.

Generally speaking, it is shown that the seasonal variation (i.e. February versus June) of emission profiles obtained with SIM-Plms is rather low, since the maximum difference between averaged effective emission heights is only 52 m for SNAP01 NO\(_x\) emissions. Similarly, day-time variation of emission profiles is not significant, detecting just minor differences in lower layers during the month of June (i.e. up to 5% for NO\(_x\) in layer 2) (Fig. S3). These

\begin{table}[h]
\centering
\begin{tabular}{|c|c|c|c|}
\hline
Model layer & Range [m] & Layer thickness [m] & Stack height range \\
\hline
1 & 0–39 & 39 & Area/Line sources \\
2 & 39–78 & 39 & h < 25 \\
3 & 78–119 & 41 & 25 ≤ h < 50 \\
4 & 119–157 & 38 & 50 ≤ h < 75 \\
5 & 157–197 & 40 & 75 ≤ h < 100 \\
6 & 197–237 & 40 & — \\
7 & 237–315 & 78 & 100 ≤ h < 125 \\
8 & 315–390 & 75 & 125 ≤ h < 200 \\
9 & 390–560 & 170 & 200 ≤ h < 250 \\
10 & 560–820 & 260 & 250 ≤ h < 300 \\
11 & 820–1250 & 430 & h ≥ 300 \\
\hline
\end{tabular}
\caption{Current vertical assignment of point sources by stack height performed by HERMESv2.0.}
\end{table}
results are in accordance with the clustered profiles obtained by Bieser et al. (2011), which point out the fact that seasonal and daytime variation of emission profiles is low in Mediterranean regions.

For SO$_x$, major differences are observed between SIM-Fixed and SIM-PlmRs results in all SNAP sectors. For all sectors SIM-PlmRs emissions are allocated to lower altitudes than in the SIM-Fixed profiles. These differences are larger when comparing SIM-PlmRs with the EMEP profiles except for SNAP04, for which SIM-PlmRs estimates a larger average effective emission height (~155 m versus ~57 m). On the other hand, the Biesetal11 effective emission height for SNAP01 is in line with the value reported by SIM-PlmRs (~366 m versus ~328 m). For SNAP01, SIM-Fixed allocates almost 40% of the emissions to layer 7 (237–315 m) while in the case of SIM-PlmRs the share of injected emissions in this layer only accounts for 15%. However, in both cases the major share of emissions are allocated between layers 7 and 10 (560–820 m) (~76% for SIM-Fixed; ~70% for SIM-PlmRs). This is due to the fact that SO$_x$ emissions are mainly controlled by power plants and refineries (~70% of total emissions, section 3.1.1), which present high stack heights. Substantial differences are also observed in the highest layer (layer 11, 820–1250 m), where SO$_x$ emissions are mainly emitted by the two largest Spanish coal-fired power plants (in-depth discussion in

Fig. 3. Comparison of SNAP-sectorial average SO$_x$, NO$_x$ and PM$_{10}$ emission vertical profiles obtained with SIM-PlmRs, SIM-Fixed, EMEP and Bieser et al. (2011) (Biesetal11).
The patterns observed for PM$_{10}$ in SNAP01 are very similar to the ones obtained for SO$_x$. This is in accordance with the fact that for this sector coal-fired power plants are also the main contributors to total point source emissions. Regarding SNAP03 vertical distributions, both SIM-Fixed and SIM-PlmRs inject more than 60% of total emissions in layer 2. For this sector, larger contributors are paper and pulp industries (section 3.1.1), which present stack height values between 20 and 30 m. For SNAP04, emissions are again allocated to higher altitudes when using the fixed vertical distribution due to the contribution of electric furnaces (stack height ~ 55 m) to total PM$_{10}$ emissions.

### 3.1.1. Cumulative curves

Fig. 4 represents the cumulative share of NO$_x$, SO$_x$ and PM$_{10}$ emissions over model layers for different point source categories (month of February). The highest values result for power plants (more than 50% of total emissions released above model layer 8, between 315 and 390 m) and refineries (more than 50% of total emissions released above model layer 4, between 119 and 157 m). On the other hand, industrial boilers, paper and pulp plants, bricks and tiles plants and fine ceramic plants release almost 100% of their emissions in the second model layer (between 38 and 78 m). The profiles obtained for each point source category remain quite close from one pollutant to the other. However, large differences are observed between facilities belonging to the same general SNAP category (e.g. cement and brick/tile plants, both belonging to SNAP03). This fact highlights the importance of reporting vertical profiles distinguishing between types of industries, rather than only between general SNAP sectors and pollutants, which is what is usually done (e.g. De Meij et al., 2006).

The vertical distribution of SO$_x$ emissions is mainly controlled by power plants and refineries (more than 70% of total emissions), which implies that this pollutant is mainly allocated in high altitudes (Fig. 4a). In the case of NO$_x$, power plants (41%) and cogeneration installations (23%) are the main contributors to its vertical allocation (Fig. 4b), being the global injection of this pollutant in much lower altitudes than in the case of SO$_x$. Finally, for PM$_{10}$ there is not a clear dominant category; emissions are scattered through the different point source sectors, being the main emission contributor refineries, iron and steel industries and paper and pulp industries (12% for each category) (Fig. 4c).

### 3.2. Air quality concentrations

Next subsections summarize the statistical results and time series obtained for the comparison of the SIM-Fixed and SIM-PlmRs simulations to AirBase station measurements (EEA, 2013) (Fig. S2 and Table S4). The model-to-data statistics correlation coefficient (r), root mean square errors (RMSE) and mean bias (MB) values are calculated on an hourly basis for NO$_x$, SO$_x$ and PM$_{10}$ and for each period of time (i.e. February and June) considering a total of 46 industrial and 2 urban (i.e. traffic and background) stations. Due to the lack of available data (none of the 46 industrial stations considered in the study have information on SO$_x$ emissions) modelled SO$_x$ concentrations are contrasted against daily measurements at 11 EMEP rural stations. Moreover, time series of four specific stations and a 3D representation are presented to analyse in more detail the effect of plume rise calculations on SO$_x$ concentrations. Concentrations simulated in the two urban (traffic and background) stations are discussed in the supplementary material, showing a significant effect of plume rise calculations on the urban NO$_2$ levels, which are increased by ~2 µg m$^{-3}$ (Table S5). Additionally, the difference between monthly mean SO$_x$, NO$_x$ and PM$_{10}$ concentrations [µg m$^{-3}$] at ground level driven by each run are summarized in the supplementary material, showing differences of

### Table 2

Average effective emission heights [m] (avg) of SO$_x$, NO$_x$ and PM$_{10}$ from SNAP01, SNAP03 and SNAP04 calculated using the plume rise calculations (SIM-PlmRs) and the fixed vertical profiles (SIM-Fixed) and estimated from the average profiles reported by Vidic (2002) (EMEP) and Bieser et al. (2011) (Biesetal11). SIM-PlmRs and SIM-Fixed also include the standard deviation (std) values.

<table>
<thead>
<tr>
<th>Sector</th>
<th>Pollutant</th>
<th>SIM-PlmRs</th>
<th>SIM-Fixed</th>
<th>EMEP</th>
<th>Biesetal11</th>
</tr>
</thead>
<tbody>
<tr>
<td>SNAP01</td>
<td>SO$_x$</td>
<td>389.6</td>
<td>168.9</td>
<td>410.2</td>
<td>255.7</td>
</tr>
<tr>
<td></td>
<td>NO$_x$</td>
<td>384.6</td>
<td>192.1</td>
<td>400.6</td>
<td>289.9</td>
</tr>
<tr>
<td></td>
<td>PM$_{10}$</td>
<td>371.9</td>
<td>167.6</td>
<td>396.0</td>
<td>237.0</td>
</tr>
<tr>
<td>SNAP03</td>
<td>SO$_x$</td>
<td>87.7</td>
<td>22.2</td>
<td>104.8</td>
<td>60.0</td>
</tr>
<tr>
<td></td>
<td>NO$_x$</td>
<td>97.1</td>
<td>44.1</td>
<td>99.6</td>
<td>49.7</td>
</tr>
<tr>
<td></td>
<td>PM$_{10}$</td>
<td>75.9</td>
<td>38.8</td>
<td>90.9</td>
<td>56.0</td>
</tr>
<tr>
<td>SNAP04</td>
<td>SO$_x$</td>
<td>159.6</td>
<td>23.4</td>
<td>262.4</td>
<td>116.6</td>
</tr>
<tr>
<td></td>
<td>NO$_x$</td>
<td>100.8</td>
<td>57.1</td>
<td>135.0</td>
<td>104.3</td>
</tr>
<tr>
<td></td>
<td>PM$_{10}$</td>
<td>91.6</td>
<td>41.2</td>
<td>121.4</td>
<td>67.5</td>
</tr>
</tbody>
</table>

Effective emission height [m] for SNAP03 and SNAP04.

#### Effective emission height [m] for SNAP03 and SNAP04.
up to ± 10 μg m⁻³ for SO₂ in some of the main industrialized areas (i.e. NW and SW of Spain) (Fig. S4).

3.2.1. Comparison to observations

Results show that the use of plume rise calculations leads to a better performance of modelled SO₂ concentrations, reducing significantly the model biases for both months (31.1% in February and 73.7% in June) (Table 3, Fig. S5). However, results also show that the correlation factor (r) between modelled and observed SO₂ concentrations slightly decreases when using plume rise calculations. This reduction (of 0.03 in February and 0.06 in June) could be related to the relationship between input accuracy and level of modelling detail. As pointed out by Smit et al. (2010) more complex models have the potential to provide more accurate predictions as they take into account more variables. However, they also require more detailed input data which may not be readily available to the model user (i.e. stack parameters). In the present case, most of the stack parameters related to flue gas exit temperature and velocity were obtained from literature due to the lack of real-world data, and were considered to be constant rather than variable over time. In this sense, it is possible that the low degree of this specific input data is somewhat

![Fig. 4. Cumulative share of (a) SOₓ, (b) NOₓ, and (c) PM₁₀ emissions over model layers for different point source categories (February).](image-url)
offsetting the potential accuracy gains of introducing plume rise calculations.

Considering that SO$_2^2$ is mainly formed by oxidation of SO$_2$, this pollutant was also analysed. Table 4 summarizes the mean hourly concentrations of SO$_2^2$ and SO$_2$ [µg m$^{-3}$] obtained with the two simulations. Values are averages over all the industrial stations as well as two individual industrial stations (i.e. Campo de Fútbol and Meriñán). As results show, the influence of the emission profiles on the vertical concentration distribution of particular SO$_2^2$ is smaller than on SO$_2$. Focussing on the averages over the overall industrial stations, for SO$_2$ concentrations in the surface layer the bias between the two runs (SIM-PlmRs — SIM-Fixed) is 27% in February and 22% in June, while in the case of SO$_2^2$ the increase is 4.3% and 0.4%, respectively. This behaviour varies significantly from one station to the other. For instance, the results observed at Campo de Fútbol station show that a 32% increase in SO$_2$ concentrations in February implies a 22% increase in SO$_2^2$, while in June a SO$_2$ increase of 40% only leads to a SO$_2^2$ increase of 5%. By contrast, results estimated at Meriñán station show that a higher decrease of SO$_2$ concentrations (~46% in February and ~51% in June) also implies a higher decrease of SO$_2^2$ (~6% in February and ~10% in June). All these different behaviours between stations and seasons are related to a combination of different factors affecting atmospheric aerosol processes (e.g. availability of NH$_3$, relative humidity), which influence the sulphur conversion ratio (Khoder, 2002). Modelled SO$_2^2$ concentrations at EMEP stations are practically identical (Table 3), which is in line with the fact that these stations are located such that significant local influences (e.g. point source emissions) are minimized. For both simulations modelled concentrations are slightly overestimated, especially in June (mean bias of 0.5 µg m$^{-3}$).

For NO$_2$, similar results are obtained regardless of the vertical disaggregation methodology used to allocate point source emissions (Fig. S5). The reduction of the model bias in industrial stations is only of 4.9% (February) and 2.5% (June) when using the CMAQ-MdPlmRs module. Correlation coefficients and RMSE also remain at the same values (Table 3). As pointed out by Biwer et al. (2011) the stack properties have the largest influence on the effective emission height (especially stack height and exit velocity). As seen in the supplementary material (Table S2), only 59% of total NO$_x$ point source emissions are released from facilities that have real-world stack height data. This lack of specific information could be a key factor in explaining the poor improvement obtained when compared to SO$_2$ results (the total share of SO$_2$ emissions based on real-world stack height data is ~75%, Table S2).

PM$_{10}$ results show that concentrations for this pollutant are largely underestimated regardless of the vertical injection methodology used (Fig. S5). The maximum reduction of model bias is only by 1.2% (February), the correlation factor being increased by 0.02 points and the RMSE reduced by 0.03 µg m$^{-3}$ (also February) (Table 3). In June, statistical results remain exactly the same for the two simulations. It is well known that PM$_{10}$ concentrations are commonly underestimated by regional air quality models (e.g. Rouil and Bessagnet, 2013). The reasons for this are mainly due to: (i) uncertainties in emissions (e.g. not all possible sources considered) and (ii) uncertainties in some process parameterisations (e.g. formation of secondary organic aerosols, SOA). For the first case, underestimations in industrial areas could be related to the non-consideration of dust emissions from management of bulk solids (storage, handling, and transportation) (Monfort et al., 2011) as well as from piles in open storage yards (Chalvatzaki et al., 2012). Concerning the second reason, several studies have shown how the CMAQ model has had a historical tendency to simulate SOA levels lower than those observed (e.g. Wyat et al., 2008; Carlton et al., 2010). The evaluation studies of the CALIOPE system performed by Baldasano et al. (2011) and Pay et al. (2012) also highlighted the difficulties of the system in reproducing the PM$_{10}$ concentration levels across Spain, and associated them with the non-consideration of fugitive dust emissions from anthropogenic sources and wind erosion events and the uncertainties in SOA formation.

### Table 3

Monthly statistics obtained with the SIM-Fixed and SIM-PlmRs simulations on an hourly basis for February and June 2009, and averaged for the 46 selected industrial stations (SO$_2$, NO$_2$, and PM$_{10}$). SO$_2^2$ statistics are obtained on a daily basis and averaged for 11 EMEP stations. The calculated statistics are measured mean [MeasM, µg m$^{-3}$], modelled mean [ModM, µg m$^{-3}$], mean bias [MB, µg m$^{-3}$], correlation coefficient ($r$), and root mean square error [RMSE, µg m$^{-3}$].

### Table 4

Mean hourly concentrations for February and June 2009 of SO$_2^2$ and SO$_2$ [µg m$^{-3}$] obtained with SIM-Fixed and SIM-PlmRs simulations. Values are averages over all industrial stations as well as the individual stations of Meriñán and Campo de Fútbol and their standard deviations.

3.2.2. Individual stations

Given that SO$_2$ is strongly related to emissions from point sources (Fig. 2) and considering the observed impacts of implementing plume rise calculations on this pollutant (Table 3), four specific industrial stations (yellow dots in Fig. S2) were selected to analyse in more detail the effect of plume rise calculations on SO$_2$ concentrations. Fig. 5 shows observed and modelled time series of hourly concentrations for SO$_2$ [µg m$^{-3}$] at the stations of Congosto and Meriñán (February) and Campo Fútbol and Rábida (June), all of them located near large point sources that release high amounts of SO$_2$. Note that all of them present a lower detection limit of 5–10 µg m$^{-3}$. 

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**Table 3**

<table>
<thead>
<tr>
<th>Period</th>
<th>MeasM [µg m$^{-3}$]</th>
<th>ModM [µg m$^{-3}$]</th>
<th>MB [µg m$^{-3}$]</th>
<th>r</th>
<th>RMSE [µg m$^{-3}$]</th>
</tr>
</thead>
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<tr>
<td></td>
<td>SIM-Fixed</td>
<td>SIM-PlmRs</td>
<td>SIM-Fixed</td>
<td>SIM-PlmRs</td>
<td>SIM-Fixed</td>
</tr>
<tr>
<td>SO$_2$</td>
<td>Feb 9.4 4.9 6.3</td>
<td>-4.5 -3.1 0.26 0.23</td>
<td>12.6 12.8</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Jun 8.7 6.7 8.2</td>
<td>-1.9 -0.5 0.24 0.18</td>
<td>14.7 15.7</td>
<td></td>
<td></td>
</tr>
<tr>
<td>NO$_2$</td>
<td>Feb 19.9 11.7 12.2</td>
<td>-8.1 -7.7 0.51 0.50</td>
<td>17.7 17.5</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Jun 15.9 12.0 12.1</td>
<td>-4.0 -3.9 0.41 0.41</td>
<td>16.8 16.7</td>
<td></td>
<td></td>
</tr>
<tr>
<td>PM$_{10}$</td>
<td>Feb 25.7 8.6 8.8</td>
<td>-17.1 -16.9 0.22 0.24</td>
<td>27.2 26.9</td>
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<td></td>
</tr>
<tr>
<td></td>
<td>Jun 27.4 14.8 14.9</td>
<td>-12.6 -12.5 0.23 0.23</td>
<td>28.0 28.0</td>
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<td></td>
</tr>
<tr>
<td>SO$_2^2$</td>
<td>Feb 0.6 0.7 0.7</td>
<td>0.1 0.1 0.62 0.62</td>
<td>0.4 0.4</td>
<td></td>
<td></td>
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<tr>
<td></td>
<td>Jun 0.8 1.3 1.3</td>
<td>0.5 0.5 0.51 0.51</td>
<td>0.8 0.8</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

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**Table 4**

Mean hourly concentrations for February and June 2009 of SO$_2^2$ and SO$_2$ [µg m$^{-3}$] obtained with SIM-Fixed and SIM-PlmRs simulations. Values are averages over all industrial stations as well as the individual stations of Meriñán and Campo de Fútbol and their standard deviations.
Results for the station of Congosto show how SO$_2$ peaks are enhanced in the SIM-PlmRs simulation. The two peaks at the beginning and at the end of the period are significantly better reproduced when using plume rise calculations, increasing the maximum modelled concentrations by up to $\sim 20 \text{ mg m}^{-3}$. It is important to highlight that this station is located near a coal-fired power plant (~5 km) for which all the stack characteristics are obtained from real-world data. For Meriñán the positive impact that CMAQ-ModPlmRs has on the model's performance is also observed. Most of the overestimated peaks simulated with SIM-Fixed are reduced when running SIM-PlmRs (up to a factor of 4). However, for this station underestimations are still observed (even increased when using plume rise calculations), especially at the beginning of the month. Campo de Fútbol is a station located in a major industrial region, with two coal-fired power plants and a refinery installation situated within a radius of 10 km. As in the case of Congosto, SIM-PlmRs performs a better reproduction of high concentration peaks, especially the one occurred between the 17th and 18th of June. Nevertheless, there are still some peaks that are not adequately caught by the model. Finally, the station of la Rábida is shown as an example in which the use of CMAQ-ModPlmRs leads to worse results. SO$_2$ overestimation is stronger in the SIM-PlmRs simulation, with differences reaching $\sim 90 \text{ mg m}^{-3}$ between observed and modelled values. As in the case of Campo de Fútbol,

Fig. 5. Observed and modelled time series of hourly concentrations for SO$_2$ [\text{mg m}^{-3}] at the stations of Congosto and Meriñán (February) and Campo de Fútbol and la Rábida (June).
this station is situated in a highly industrialised area (a refinery installation, a primary copper manufacturing plant, two sulphuric acid manufacturing plants and a titanium dioxide manufacturing plant). For all these facilities, real-world stack height and diameter information is available only for the refinery plant and the primary copper plant, while the rest of stack properties are based on average values and other literature.

3.2.3. 3D analysis

A 3D representation of the distribution of average SO$_2$ concentrations in the vicinity of the second largest Spanish coal-fired power plant (Andorra, consisting of three units with stack heights of 343 m) was performed for each simulation for the month of February (Fig. 6). Fig. 6a shows the differences between monthly mean SO$_2$ concentrations [$\mu$g m$^{-3}$] at ground level driven by SIM-...
PlmRs and SIM-Fixed for February in the vicinity of the three Andorra stacks (grupo 1, grupo 2 and grupo 3). It is observed that downwind of the facility (~50 km) SIM-PlmRs leads to higher simulated ground SO2 concentrations (up to 2 μg m$^{-3}$). Note that the negative differences appearing in the map (up to 10 μg m$^{-3}$) correspond to the different vertical injection of the emissions released by the Escucha coal-fired power plant (one unit with stack heights of 120 m); when applying the fixed vertical profiles the SO2 emissions released are 100% injected in the model layer 7, while SIM-PlmRs only allocates 30% of the total emissions in this layer, since almost 50% is injected in higher layers (i.e. 18% in layer 8, 20% in layer 9 and 8% in layer 10). In order to understand the differences in surface concentrations, a 3D representation of the selected area was performed using the Visit visualization tool. Fig. 6b (SIM-Fixed) and Fig. 6c (SIM-PlmRs) present the zenithal view of the average SO2 concentrations [ppm] in the different model layers, while Fig. 6d (SIM-Fixed) and Fig. 6e (SIM-PlmRs) show the results from a slightly raised point of view looking diagonally.

When applying the fixed vertical profiles, the SO2 emissions released by the three Andorra stacks are 100% injected in the model layer 11 (820–1250 m). On the other hand, CMAQ-ModPlmRs injects 24% of total SO2 emissions into model layer 9 (390–560 m), 11% into model layer 10 (560–820 m) and 22% into model layer 11 (820–1250 m). These differences in the vertical allocation have a direct impact on the horizontal (x,y) and vertical (z) dispersion of the SO2 plume. As seen in Fig. 6b–e, the injection of SO2 emissions in lower layers with lower layer thickness when using plume rise calculations leads the model not only to increase surface concentrations but also to simulate a plume that reaches a high downwind distance.

4. Conclusions

This work analyses the impact of CMAQ hourly plume rise calculations on allocating point source emissions and predicting air quality surface concentrations over Spain and within the high resolution air quality system CALIOPE-AQFS. When using the CMAQ-ModPlmRs module (SIM-PlmRs), SOx, NOx and PM10 emissions are generally allocated to lower altitudes than when using the fixed vertical profiles used in HERMESv2.0 (SIM-Fixed), especially in the case of SOx and PM10 emissions released from SNAP01 and SNAP04 point sources, finding differences of up to 75% between estimated average effective emission heights. These differences are more prominent when compared to the EMEP vertical profiles (~9 m versus ~485 m effective emission heights for SNAP03 SO2 emissions) except for sector SNAP04, in which case the EMEP profiles lead to the lowest effective emission heights for all the pollutants. On the other hand, the Bieser et al. (2011) profiles exhibit a large peak of SNAP01 emissions at layer 7 while SIM-PlmRs allocates more emissions above and below this layer, leading to a broader vertical distribution of emissions. This result is probably due to the fact that the SIM-PlmRs profiles are based on a multitude of stack datasets (i.e. specific stack parameters by facility) instead of average stack data (i.e. general stack parameters by source categories according to Pregger and Friederich (2009)).

SO2 is the pollutant allocated to higher altitudes, due to the fact that its emissions are mainly controlled by coal-fired power plants and refineries with high stack heights (more than 70% of total emissions). Differences observed in the vertical allocation of facilities belonging to the same general SNAP category highlight the importance of reporting vertical profiles distinguishing between type of industries and not only general SNAP sectors as has usually been done (e.g. De Meij et al., 2006).

In terms of air quality, it is shown that using hourly plume rise calculations leads to an increase in modelled surface SO2 concentrations when compared to industrial station measurements, leading to a significant reduction of the model bias (from ~4.5 μg m$^{-3}$ to ~3.1 μg m$^{-3}$ in February). However, the correlation factor between modelled and observed SO2 concentrations slightly decreases when using the CMAQ-ModPlmRs module. The low specificity of some input data (i.e. stack flue gas exit temperature and velocity) may be somewhat offsetting the potential accuracy gains of introducing plume rise calculations. On the other hand, the averaged increase of SO2 concentrations (27% in February and 22% in June) leads to a lower averaged increase of SO2 surface levels (4.3% in February and 0.4% in June). This impact varies according to the location and the season analyzed (i.e. at Campo de Fútbol station a 32% increase in SO2 concentrations implies a 22% increase in SO2 in February) due to the different factors affecting atmospheric aerosol processes (e.g. availability of NH3, relative humidity).

The influence of using the CMAQ-ModPlmRs module is rather low in terms of NO2 concentrations, since the reduction of the model bias in industrial stations is only by 4.9% (February) and 2.5% (June). On the other hand, PM10 results are largely underestimated regardless of the vertical injection methodology used. This reinforces the idea that the difficulties of the system in reproducing PM10 concentration levels across Spain are associated with uncertainties in secondary organic aerosols formation (CMAQ model) and the non-consideration of fugitive dust emissions from anthropogenic sources and wind erosion events (HERMESv2.0 model). The analysis of SO2 concentration results in four specific industrial stations shows the positive impact of CMAQ-ModPlmRs on the model reproducing SO2 concentration peaks. Maximum modelled concentrations are increased by up to ~20 μg m$^{-3}$ when applying plume rise calculations. However, the bad performance achieved in one of these stations also points out the uncertainties resulting from the limited availability of source specific data on stack properties based on real-world information.

Finally, a 3D representation of the distribution of average SO2 concentrations in the vicinity of the second largest Spanish coal-fired power plant (Andorra) proved the direct impact of vertical allocation on the horizontal (x,y) and vertical (z) plume dispersion. The injection of SO2 emissions in lower layers with lower thickness leads the model not only to increase surface concentrations but also to simulate a plume that reaches a larger downwind distance.

In order to maximize the precision of plume rise calculations the use of stack parameters based on real-world data is mandatory. The current lack of detailed stack data highlights the need for initiatives and projects to build up a substantial point source database for Europe containing source information about stack and flue gas parameters. Considering the overall improvements seen when using plume rise calculation (especially for SO2 concentrations) the CMAQ-ModPlmRs will be implemented within the CALIOPE-AQFS system.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2014.10.029.

References


