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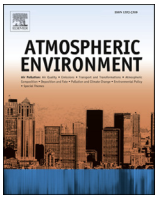

Implementation of plume rise and its impacts on emissions and air quality modelling

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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 SO_2 concentrations.
- When applying plume-rise models the use of real-world stack parameters is mandatory.

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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 SO_2 concentrations, thus increasing modelled concentrations ($1.4\ \mu g\ m^{-3}$ increase in February, $1.5\ \mu g\ m^{-3}$ increase in June) and reducing the model biases for both months (31.1% in February, 73.7% in June). The increase of SO_2 concentrations leads to an increase of SO_4^{-2} 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 NO_2 and PM_{10} concentrations is less significant, leading to average changes of a few $\mu g \cdot m^3$ at most ($0.4\ \mu g\ m^{-3}$ for NO_2 and $0.2\ \mu g\ m^{-3}$ for PM_{10}). In order to maximize the precision of plume rise calculations, the use of stack parameters based on real-world data 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

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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 (SO_2) 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, NO_x) 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 emission heights depending on source and air pollutant 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 × 54 km) and with emissions using the 73 point–source profiles and the EMEP profiles. Results showed that predicted SO_2 and sulphate (SO_4^{-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 SO_2, NO_2 and O_3 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° × 0.5°). Results showed that applying the Bieser et al. (2011) profiles leads to significantly improved simulations of background NO_2 and SO_2 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-ARW/HERMESv2.0/CMAQ/BSC-DREAM8b) 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 × 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 SO_2, SO_4^{-2}, NO_2 and PM_{10} 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 × 399 points with 12 km × 12 km horizontal resolution and centered at 6.073 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, centred in Spain (SW of Europe), is approximately 1596 km * 1596 km and centred at −3.164 Lon and 39.971 Lat, with a 4 km × 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

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