An improved system for modelling Spanish emissions: HERMESv2.0

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Highlights

- An updated high-resolution emission model of 2009 for Spain is presented.
- Emissions are estimated using mainly bottom-up approaches and Spanish local data.
- It represents an updated and improved version of the previous HERMES model.
- Comparison of emissions results shows new model version is more accurate.
- The model will be the emission core of the Spanish air quality system CALIOPE.

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Abstract

Emission models play a key role in the development of high-resolution air quality modelling systems (AQMS). To minimise the uncertainty presented by these models, it is essential to match the high-resolution requirements of chemical transport models (CTMs) and to use up-to-date information and emission methodologies. During 2005 and 2006, the Barcelona Supercomputing Center – Centro Nacional de Supercomputación (BSC-CNS) developed the High-Elective Resolution Modelling Emissions System (HERMES04), which is a model that estimates anthropogenic and biogenic emissions for Spain with a temporal and spatial resolution of 1 h and 1 km², taking 2004 as the reference period. Due to both the changes in Spanish emissions patterns and the age of the activity data and methodologies used, it has become necessary to update and improve the whole system. Hence, a new high-resolution emission model for Spain (HERMESv2.0) has been developed. This work introduces the improved emission estimation methodologies and data on which the model is based, as well as an analysis of the results obtained. The annual emissions estimated by HERMESv2.0 for Spain in 2009 are: NOx, 924 kt; NMVOCs, 2331 kt; SO2, 278 kt; CO, 2178 kt; NH3, 339 kt; PM10, 139 kt; and PM2.5, 105 kt. Compared with HERMES04, major differences are found in NMVOCs (+1172 kt) and SO2 (−870 kt). Important changes in emission patterns are also observed in terms of spatial and temporal distributions. A numerical comparison of both models with the Spanish National Emission Inventory indicates that previous underestimations have been heavily reduced in HERMESv2.0, especially for NOx (from −669 kt·year⁻¹ to −176 kt·year⁻¹), CO (from −761 kt·year⁻¹ to 271 kt·year⁻¹) and NMVOCs (from −1217 kt·year⁻¹ to 135 kt·year⁻¹). The new model substitutes HERMES04 as the emission core of the operational air quality forecasting system for Spain CALIOPE.

1. Introduction

Emission models play a key role in the development of high-resolution air quality modelling systems (AQMS). The primary objective of these models is to provide comprehensive information on emission sources and emission fluxes in the area under consideration (Baldasano, 1998). One of the basic aspects of emission models is representativeness (Borge et al., 2011), which implies that emission results match the requirements of the Chemical Transport Model (CTM) in terms of spatial resolution (Mensink et al., 2008), temporal resolution (Menut et al., 2012) and chemical disaggregation (Heo et al., 2012). During the last several years,
many authors have clearly shown that emission preparation is one of the most critical stages and therefore must be accurately developed, tested and verified (Bieser et al., 2011).

Currently, there are a remarkable number of emission inventories and models for both regulatory and scientific purposes (Table 1). The majority are based either on top-down approaches or downscaling methodologies applied to national emission inventories. For global and regional applications, these emission databases are useful as input data. However, in regard to high-resolution air quality modelling, the use of local information combined with bottom-up approaches is necessary to accurately characterise the emission sources and obtain more realistic results (Kannari et al., 2007).

Butler et al. (2008) studied the representation of emissions from megacities in three global anthropogenic emission inventories (EDGARv3.2, IPCC-AR4 and RETRO) and found large differences between them. The work concluded that local emission inventories at the megacity scale should be integrated in a consistent manner into global emission inventories. Pozzoli et al. (2012) performed a set of air quality simulations to quantify the effect of three different anthropogenic emission inventories (EMEP, EMEP/INERIS and TNO/MACC) on gas and aerosol concentrations over the eastern Mediterranean region (Turkey and the Balkan Peninsula). The results obtained for NO₂ were compared with OMI satellite observations, showing important differences in terms of concentrations and geographical distributions. Other significant works in this direction have been developed during the last years (Maes et al., 2009; Timmermans et al., 2012).

In the case of Spain, with its complex topographic conditions, land use heterogeneity and influence from a great number of climatic patterns, it is even more necessary to use specifically developed emission models (Jiménez et al., 2006). Under this framework, during 2005 and 2006, the Barcelona Supercomputing Center – Centro Nacional de Supercomputación (BSC-CNS) developed the High-Selective Resolution Modelling Emissions System (HERMES04), which generates an anthropogenic and biogenic emission inventory for Europe and Spain with a temporal and spatial resolution of 1 h and 1 km², according to the European

### Table 1: Summary of emission models and inventories.

<table>
<thead>
<tr>
<th>Model/inventory</th>
<th>Source</th>
<th>Emission sources</th>
<th>Temporal resolution/coverage</th>
<th>Spatial resolution/coverage</th>
<th>Use</th>
<th>Approach used</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Global</strong></td>
<td></td>
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<td></td>
</tr>
<tr>
<td>ACCMIP</td>
<td>Lamarque et al. (2010)</td>
<td>Anthropogenic Biomass burning</td>
<td>Decadal, 1850–2000</td>
<td>0.5° × 0.5°, Global</td>
<td>Scientific</td>
<td>Combination of other models and inventories (RETRO, GAINS, EMEP)</td>
</tr>
<tr>
<td>EDGARv4.2</td>
<td>Janssens-Maenhout et al. (2011)</td>
<td>Anthropogenic Biomass burning</td>
<td>Annual, 1970–2008</td>
<td>0.1° × 0.1°, Global</td>
<td>Regulatory</td>
<td>Combination of national activity data with specific EF, disaggregated using different spatial proxies</td>
</tr>
<tr>
<td>EDGAR-HTAPv1</td>
<td>G. Janssens-Maenhout et al. (2012)</td>
<td>Anthropogenic Biomass burning</td>
<td>Annual, 2000–2005</td>
<td>0.1° × 0.1°, Global</td>
<td>Regulatory</td>
<td>Combination of official/scientific inventories at the national/regional scale with EDGARv4.2 spatial proxies</td>
</tr>
<tr>
<td>RETRO</td>
<td>Schultz et al. (2007)</td>
<td>Anthropogenic Biomass burning</td>
<td>Monthly, 1960–2000</td>
<td>0.5° × 0.5°, Global</td>
<td>Regulatory</td>
<td>Combination of other models and inventories (TNO, VERITAS, GFED)</td>
</tr>
<tr>
<td>POET</td>
<td>Olivier et al. (2003)</td>
<td>Anthropogenic Biomass burning Biogenic</td>
<td>Annual, 1990–2000</td>
<td>1° × 1°, Global</td>
<td>Scientific</td>
<td>Based on national activity data, EF and grid maps</td>
</tr>
<tr>
<td>MEGANv2.04</td>
<td>Guenther et al. (2006)</td>
<td>Biogenic</td>
<td>Hourly, 2003–2009</td>
<td>1 km × 1 km, Global</td>
<td>Scientific</td>
<td>Bottom-Up approach, using gridded EF and land cover information</td>
</tr>
<tr>
<td><strong>European</strong></td>
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<tr>
<td>EMEP</td>
<td>Mareckova et al. (2012)</td>
<td>Anthropogenic</td>
<td>Annual, 1980–2020</td>
<td>50 km × 50 km, All European countries</td>
<td>Regulatory</td>
<td>National emission inventories reported by parties and assigned to the EMEP grid</td>
</tr>
<tr>
<td>GAINS</td>
<td>Amann et al. (2011)</td>
<td>Anthropogenic</td>
<td>Annual, 1990–2005</td>
<td>50 km × 50 km, All European countries</td>
<td>Regulatory</td>
<td>Combination of national activity data with specific EF and grid maps</td>
</tr>
<tr>
<td>SMOKE-Europe</td>
<td>Bieser et al. (2011)</td>
<td>Anthropogenic Biogenic</td>
<td>Hourly, 1970–2010</td>
<td>1 km × 1 km, EU-27 + other countries</td>
<td>Scientific</td>
<td>Downsampling of the EMEP/EPER emissions through the use of specific spatial and temporal proxies</td>
</tr>
<tr>
<td>E-PRTR diffuse air emissions</td>
<td>Theloke et al. (2010)</td>
<td>Anthopogenous</td>
<td>Annual, 2008</td>
<td>5 km × 5 km, EU-27</td>
<td>Scientific</td>
<td>Downsampling of National emission inventories through the use of specific spatial proxies</td>
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<tr>
<td><strong>Spanish</strong></td>
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<tr>
<td>EMIMO</td>
<td>San José et al. (2003)</td>
<td>Anthropogenic Biogenic</td>
<td>Hourly, 2003–2007</td>
<td>1 km × 1 km, Iberian Peninsula (IP) + Balearic Islands</td>
<td>Scientific</td>
<td>Downsampling of the EMEP &amp; EDGAR emissions through the use of specific spatial and temporal proxies</td>
</tr>
<tr>
<td>SMOKE-Spain</td>
<td>Borge et al. (2008)</td>
<td>Anthropogenic Biogenic</td>
<td>Hourly, 1980–2020</td>
<td>5 km × 5 km, IP + Balearic Islands</td>
<td>Scientific</td>
<td>Downsampling of the National Spanish emission inventory through the use of specific spatial and temporal proxies</td>
</tr>
<tr>
<td>MNEQA</td>
<td>Ortega et al. (2009)</td>
<td>Anthropogenic Biogenic</td>
<td>Hourly, 2005</td>
<td>27 km × 27 km, Spain (D1) 9 km × 9 km and 3 km × 3 km, Catalonia (D2/D3)</td>
<td>Scientific</td>
<td>Downsampling of the EMEP emissions (D1) Combination of Bottom-Up and Top-Down approaches (D2 &amp; D3)</td>
</tr>
<tr>
<td>HERMES04</td>
<td>Baldasano et al. (2008)</td>
<td>Anthropogenic Biogenic</td>
<td>Hourly, 2004</td>
<td>1 km × 1 km, IP + Balearic &amp; Canary Islands</td>
<td>Scientific</td>
<td>Combination of Bottom-Up and Top-Down approaches</td>
</tr>
<tr>
<td>HERMEX2.0</td>
<td>This work</td>
<td>Anthropogenic Biogenic</td>
<td>Hourly, 2009 (updateable)</td>
<td>1 km × 1 km, IP + Balearic &amp; Canary Islands</td>
<td>Scientific</td>
<td>Combination of Bottom-Up and Top-Down approaches</td>
</tr>
</tbody>
</table>
Environmental Agency’s (EEA) Selected Nomenclature for Air Pollution (SNAP) and using 2004 as the reference period (Baldasano et al., 2008). The model constitutes the emission core of the operational air quality forecasting system CALIOPE (http://www.bsc.es/caliophe), implemented in the MareNostrum supercomputer (Baldasano et al., 2011; Pay et al., 2010). HERMES04 has also been used in air quality management studies (Baldasano et al., 2010; Soret et al., 2011).

The changes that have occurred in human activity during the last few years, mainly due to air quality policies, technological improvements and the economic recession, have resulted in changes to Spain’s emission patterns (MAGRAMA, 2012a). For instance, during the period between 2004 and 2009, the use of coal in Spanish power stations fell by more than 55%, while production in combined cycle plants has increased by 170% (REE, 2009). Moreover, the percentage of diesel passenger cars in road transport has changed from 38% to 51% (DGT, 2012). Along the same lines, the number of Spanish tile and brake industries has fallen by more than 30% (HISPALYT, 2010). These facts, among others, imply changes in emission results and, consequently, in air quality estimations. Hence, emission models need to work with up-to-date activity data (Granier et al., 2011).

In the case of HERMES04, due to both the age of the activity data and the estimating methodologies on which the model is based, it has become necessary to update and improve the whole system. Hence, a new version of this emission model has been developed (HERMESv2.0) with the following main features: (i) updating of the activity data, taking 2009 as the reference year; (ii) improvement and updating of the emission estimation methodologies; (iii) introduction of pollutant activities not considered in the previous version (iv) inclusion of the chemical mechanism Carbon Bond-05 (CB05) in addition to Carbon Bond-IV (CBIV); (v) calculation fully developed in the C++ programming language; (vi) flexible implementation to facilitate revisions and updates.

The present work describes and discusses the methodologies, approaches and emission results obtained with HERMESv2.0. Section 2 of this paper introduces the data, methods and procedures applied to estimate the emissions for each SNAP sector. In Section 3, a thorough analysis of the emission results is conducted along with a comparison of the results obtained with HERMES04 and the Spanish National Emission Inventory (INESP hereinafter). Finally, Section 4 summarises the work.

2. Methodology

HERMESv2.0 is divided into two modules: (1) HERMES-DIS and (2) HERMES-BOUP, which can work either combined or separately depending on the working domain (Figs. 1 and 2).

2.1. HERMES-DIS

The HERMES-DIS module processes the annual EMEP emissions (50 km × 50 km) (Mareckova et al., 2012), performing an SNAP sector-dependent spatial (12 km × 12 km and up to 1 km²) and temporal (1 h) disaggregation as well as a speciation treatment. The module is used to (i) generate emission input files for all European countries except Spain and to (ii) incorporate the EMEP ship traffic emissions in HERMESv2.0. A detailed description of this module is not within the scope of this work; further information can be found in Ferreira et al. (2013) and Guevara et al. (2012).

2.2. HERMES-BOUP

HERMES-BOUP is a high-resolution emission model specifically developed for Spain. The model uses a combination of local information and state-of-the-art methodologies to estimate anthropogenic and biogenic emissions at a high spatial (1 km × 1 km) and temporal (1 h) resolution over the whole Spanish territory, taking 2009 as the reference year. The model estimates emissions for the following primary air pollutants: nitrogen oxides (NOx), non-methane volatile organic compounds (NMVOCs), sulphur dioxide (SO2), carbon monoxide (CO), ammonia (NH3), total suspended particles (TSP) and PM10 and PM2.5 fractions. The model also estimates emissions for the three main greenhouse gases (GHGs): carbon dioxide (CO2), methane (CH4) and nitrous oxide (N2O).

HERMES-BOUP uses a bottom-up approach for the majority of SNAP sectors (SNAP01, 03, 04, 07, 08, 09 and 11), while for the others a combination of top-down approaches (SNAP02; using general Emission Factors (EFs) and national statistics) and downscaling methodologies (SNAP05, 06 and 10; adapting spatially and temporally an existing emission dataset) has been adopted. The pre-processing of the data and cartography edition is performed through ESRI’s ArcGIS 10 Desktop. Once the hourly and gridded emissions are estimated, the model carries out a selective chemical speciation, which includes CB-IV (Gery et al., 1989) and CB05 (Yarwood et al., 2005) mechanisms. In terms of emission file management, the system generates two types of outputs: (i) emission data for air quality modelling (NetCDF emission files) and (ii) emission data for analysis and visualisation (e.g., csv and kmz emission files). Considering the complexity of certain estimation methodologies, the model has been designed to be as flexible as possible so that revisions and updates can be performed simply.

Regarding the previous model version, in HERMESv2.0 major efforts were made to improve the representativeness and accuracy of the data and emission methodologies implemented (Table 2). The next subsections explain the methodologies followed to estimate the high-resolution emissions by SNAP sector. A detailed summary of the information sources (EFs values, temporal profiles, spatial proxies, vertical distributions and chemical speciation) and mathematical expressions used in HERMESv2.0 to calculate hourly and gridded emissions are available in the Supplementary material.

2.2.1. SNAP01 – combustion in energy industries

This sector includes emissions from combustion processes in power plants, refineries and coke industries. A total of 333 point sources were considered, compiling for each of them the following information: facility name, SNAP elemental activity code (SNAP third level) associated with its activity, geographical location (X and Y coordinates), stack height (in meters), activity factor (e.g. annual production or fuel consumption), EFs for each pollutant, temporal profiles (monthly, weekly and hourly) and speciation profiles (for NOx, NMVOCs and PM2.5).

In the case of power plants (SNAP0101), the annual electricity generated in each facility was obtained from REE (2009a) and UNESA (2009). EFs related to NOx, SO2 and TSP were estimated from measured emission data provided by the Spanish National Office of Emission Control for Large Combustion Facilities (OCEM-CIEMAT, personal communication), while for the rest of pollutants and facilities without measurements (mainly diesel generator groups) information was obtained from EEA (2007) (Chapter B11, Tables 24–30), EEA (2009) (Chapter 1.A.1, Tables 3-11 to 3-23) and TNO (1995). Temporal profiles were derived from the power demand data reported by REE (2009b).

For the refineries (SNAP0103), EFs were chosen from EEA (2007) (Chapter B11, Tables 24–30) and EEA (2009) (Chapter 1.A.1, Tables 4-5 to 4-10). The annual fuel consumed in each plant was obtained combining the information of MINECO (2003) and the Oilgas (2010) encyclopaedia. Regarding the temporal allocation, is has been assumed a constant profile during the whole year.
Information on the total amount of coke manufactured in coke industries (SNAP0104) was obtained from MINETUR (2009a). The total value was assigned among each facility taking into account nominal production capacities obtained from Integrated Environmental Authorisations (IEAs) and Environmental Reports (ERs) reported by governmental authorities. EFs for this activity were obtained from EEA (2007) (chapter B146, Tables 8.2a and 8.2b) and EEA (2009) (chapter 1.A.1, Table 5-3). As in the refineries, the activity was supposed constant during the whole year.

2.2.2. SNAP02 — non-industrial combustion plants

This sector includes all the small combustion installations (mainly boilers) intended for heating and provision of hot water in residential and commercial sectors. Due to the impossibility of estimating installation-specific emissions the sources of this sector are considered collectively as an area source, applying a top-down approach.

In this sense, emissions were estimated using annual energy consumption statistics at NUTS 2 level (Autonomous Communities) and NUTS 3 level (Provinces) obtained from energetic statistics and balances reported by the Corporation of Strategic Reserves of Oil-based Products (CORES, 2010), the Energy Saving and Diversification Institute (IDAE, 2010) and the Spanish Ministry of Industry, Tourism and Trade (MINETUR, 2009b). The fuels considered were natural gas, liquefied petroleum gas (LPG), heating diesel, diesel oil, biomass and coal. A specific EF was assigned for each fuel and subsector (residential or commercial) according to EEA (2009) (Chapter 1.A.4, Tables 3-3 to 3-10). The spatial allocation was made using a population density map based on Tele Atlas Multinet (2011) geographical information. For biomass and coal combustion, the emissions were assigned by establishing a maximum number of inhabitants per grid cell (which varies according to the domain’s resolution). On the other hand, the temporal allocation followed different profiles depending on the fuel used. In the case of natural gas, LPG and heating diesel, the hourly profiles were distinguished according to the type of day and type of month using the data reported by ENAGAS (2010). For diesel oil, biomass and coal the profiles reported by USEPA (2002) were applied.

2.2.3. SNAP03/04 — combustion and production process in industries

A local point source inventory was developed (1321 stacks) that included the following industrial sectors: cement, lime, paper, fine ceramics, bricks and tiles, glass, iron and steel, non-ferrous metallurgy and inorganic chemistry, as well as cogeneration installations. For each of these point sources, the same information as in the SNAP01 sector was compiled (see Section 2.2.1).
For some industrial facilities, specific information on the amount of annual manufactured product (i.e. tonnes of cement) was available in their corresponding IEAs and ERs or in the E-PRTR (2010) database. However, in the majority of the cases the annual production data was only available for the whole industrial sector. In order to estimate the activity factor at a facility-level, an assignment of the total production among each point source was performed using distributional factors such as nominal production capacities (obtained from IEAs and ERs), invoice data (ARDAN, 2009) which indicates the annual turnover associated to each facility, or the National emissions allocation plan 2008–2012 (MAGRAMA, 2007a). EFs were all obtained from the EEA guidelines (see Supplementary material) and the temporal distribution was considered constant in all the cases.

Fig. 2. Emissions of NO$_2$ [kg h$^{-1}$] for the 08UTC 12 Feb 2013, estimated with HERMESv2.0 and implemented within the CALIOPE air quality forecasting system for the different domains: (D1) Europe (12 km × 12 km, emissions derived from HERMES-DIS); (D2) Iberian Peninsula (4 km × 4 km, emissions for Spain estimated with HERMES-BOUP and derived from HERMES-DIS for the rest of countries and ship traffic); (D3) Canary Islands (2 km × 2 km, emissions estimated with HERMES-BOUP) and (D4) Andalucía, (D5) Barcelona and (D6) Madrid Greater Areas (all 1 km × 1 km, emissions same as D2).
In the case of cogeneration installations, the information on annual electricity produced by each one of them was directly obtained from MINETUR (2009c). EFs and temporal profiles were derived from the same sources of information used in power plants (see Section 2.2.1).

2.2.4. SNAP05 — extraction & distribution of fossils fuels

Due to the lack of specific information on activity data and EFs, emissions from this sector were estimated performing a downscaling methodology of the original Spanish National Emission Inventory version 2009 (INESP09). This database is developed by the Spanish Ministry of the Agriculture, Food and Environment (MAGRAMA, personal communication) and represents the official Spanish contribution to the EMEP emission inventory. It reports total annual emissions of primary pollutants by NUTS 2 level and SNAP elemental activity. A specific spatial proxy and temporal profile has been assigned to each pollutant activity after defining it as point, lineal or area source. This information allows HERMESv2.0 to disaggregate the original INESP data to each one of the grid cells of the domain and for each one of the hourly time steps, obtaining a high-resolution emission database. Table 3 summarizes the different surrogates used for each SNAP activity.

2.2.5. SNAP06 — solvent and other products uses

As in SNAP05, and also due to difficulties in finding accurate information, a downscaling methodology was also applied to the original INESP09 database (Table 3).

2.2.6. SNAP07 — road transport

Exhaust emissions from road transport were estimated according to the Tier 3 method described in chapter 1.A.3.b.v of EEA (2009), which is fully incorporated in version 5.1 of the COPERT 4 software. For gasoline evaporation and road vehicle tyre and brake wear, the Tier 2 methods presented in chapters 1.A.3.b.v and 1.A.3.b.vi (EEA, 2009) were used. The model also considers the particle emissions due to resuspension from paved roads (Pay et al., 2011).

A digitised traffic network (over 111,000 km) with specific information about road stretches on daily average traffic (DAT, [veh day$^{-1}$]), mean speed circulation [km h$^{-1}$], temporal profiles and vehicular park profiles adapted to COPERT 4 categories was obtained combining (i) traffic information from over 24,000 observation stations managed by the governmental authorities; (ii) statistics on vehicular compositions at the NUTS 3 level (Spanish Department of Transportation, personal communication) and (iii) the Spanish digital road network (TeleAtlas® Multinet®, 2011). The database includes information on the largest cities’ urban streets (Barcelona, Madrid, Hospitalet, Valencia, Zaragoza and Bilbao). For Barcelona and Madrid, 19 park composition profiles are considered (12 and 7), considering information based on real circulation data (AB, 2010, AM, 2009).

Although the digitised traffic network covers the territory in high detail, most of the cities are not included due to the lack of traffic flow information. Hence, a specific module known as Small cities was developed to estimate their emissions. The methodology is based on a top-down approach and combines EFs [t of pollutant h$^{-1}$] calculated from the results obtained in the cities with the available data (mentioned above) with a population density map (Section 2.2.2).

2.2.7. SNAP0804 — maritime activities in ports

HERMESv2.0 considers the 46 harbours administered by the Spanish Port System and estimates the emissions based on Entec UK Limited (2007, 2010). Emissions from vessel’s main engines (ME) and auxiliary engines (AE) are estimated according to the type of vessel (i.e. tanker, general cargo), operation (manoeuvring or hoteling), fuel consumed (marine distillate, MD, or residual oil, RO) and engine (slow speed diesel, SSP, medium speed diesel, MSD, or
high speed diesel, HSD). Information on annual operations and gross tonnage (GT) by vessel type and port was obtained from the annual reports of each Spanish port authority (AAPP, 2009). The fuel consumption factors and EFs are based on Cooper and Gustafsson (2004).

The spatial dissemination also distinguishes between the two operations. For manoeuvring, emissions are proportionally distributed into the marine surface of the cells intersected by a circle with a 1-nautical-mile radius at the centre of each port. For hoteling, the emissions were allocated proportionally into the digitised port areas. The annual results were disaggregated monthly with specific port profiles obtained from SPS (2009). For the development of weekly and hourly profiles, real time traffic data reported by www.localizatodo.com was analysed. Results suggested that harbour activity can be considered constant during the week. As for the hourly emissions, a unique hourly profile was used obtained from the combination of traffic information from the harbours of Barcelona, Tarragona, Vigo, Santander and Palma.

2.2.8. SNAP0805 – LTO cycles in airports

Emissions in each of the 47 airports managed by the Spanish Airports and Air Navigation (AENA) are estimated by taking into account the five operational phases of landing and take-off cycles (LTO): final-approach, taxi-in, taxi-out, take-off and climb-out. The monthly operations by airport and specific aircraft model (AENA, 2009) were assigned to a total of 77 aircraft categories. For each category, specific fuel consumption factors and EFs were assigned using the data reported by Tier 3 method described in chapter 1.A.5.b of EEA (2009), ICAO (2010) and FOCA (2007).

The spatial distribution varies according to the LTO cycle’s phase. Taxi-in, taxi-out and take-off emissions are assigned at the ground level; taking into account digitised airport areas and runways. Emissions from final-approach and climb-out operations are allocated on a 3D basis, considering the trajectory outlined between the origin/end of the runway and 1000 m of altitude. A total of 470 weekly and hourly temporal profiles were used, based on specific hourly operations by airport reported by AENA (personal communication) for the 8th (applied to Jan—Feb—Mar—Nov—Dec) and the 29th (rest of the months) weeks of the year. Profiles distinguish between working days and holidays as well as type of operation (arrival or departure).

2.2.9. SNAP0806 – agricultural machinery

The model calculates emissions derived from two-wheel tractors, agricultural tractors and harvesters using the Tier 3 methodology described in chapter 1.A.4.c ii of EEA (2009). Information on the total fleet number, mean power, load factor and annual working hours by type of machine were compiled at the NUTS 3 level (MAGRAMA, 2007b). Because the EFs used are grouped into EU emission legislation categories, statistics on machinery ageing were also used.

The model performs a spatial disaggregation by taking into account agricultural land uses of the CORINE Land cover dataset (EEA, 2011). In terms of temporal allocation, the model use the profiles reported by the chapter of EEA (2009) mentioned above.
2.2.10. SNAP09 — waste treatment

This sector considers emissions from waste incinerators and oil refinery flares. As in SNAP01, a local point source inventory was developed gathering the same fields of information for each installation (see Section 2.2.1). EFs from waste incinerators were derived from hourly stack measurements provided by the Catalan Atmospheric Emission Network (XEAC, personal communication) and the Spanish association of waste-to-energy plants (AEVERSU, 2009). This last source was also used for obtaining the annual amount of waste incinerated in each plant. On the other hand, EFs from refinery flares were chosen from chapter 1.B.2.c of EEA (2009). In this case, the activity factor (amount of crude treated by facility) was obtained from the Oillas (2010) Encyclopaedia. The data reported by XEAC suggested the use of constant temporal profiles. In the case of refineries the activity was also considered constant for the whole year.

2.2.11. SNAP10 — agriculture

As in SNAP05 and SNAP06, a downscaling methodology of the original INESP09 emissions is carried out. In this case, agricultural land uses (EEA, 2011) are used as proxy data (Table 4). For the temporal variation, the TNO-TROTEP/POET profiles are applied (Olivier et al., 2003).

2.2.12. SNAP11 — biogenic emissions

The Model of Emissions of Gases from Nature (MEGANv2.04) (Guenther et al., 2006) was implemented in the HERMESv2.0 code for the estimation of the biogenic emissions. MEGAN is a global model with a base resolution of 1 km that estimates emissions from terrestrial ecosystems by combining global distributions of land cover variables (EFs, Leaf Area Index (LAI), and Plant Functional Types (PFT)) with meteorological inputs (surface temperature, income solar radiation). The land cover input variables are obtained from the MEGAN Community Data Portal (MCDP) (http://acd.ucar.edu/~guenther/MEGAN/MEGAN.htm), which report global datasets on EF, LAI and PFT at 30s horizontal resolution (~1 km²), while the WRF-ARW 3.0.1.1 model (Michalakes et al., 2004) is used for the generation of the meteorological fields.

3. Results and discussion

This section discusses the results obtained with HERMESv2.0 and contrasts them, when possible, with HERMES04 and the INESP national inventory. The results on vertical distribution and chemical speciation of primary pollutants are summarized in the Supplementary material.

### Table 4

<table>
<thead>
<tr>
<th>SNAP elemental activity</th>
<th>CORINE land cover 2006 land uses assigned</th>
</tr>
</thead>
<tbody>
<tr>
<td>SNAP100101/SNAP100201 — Permanent crops</td>
<td>Vineyards, fruit trees and berry plantations, olive groves and crops associated with permanent crops</td>
</tr>
<tr>
<td>SNAP100102/SNAP100202 — Arable land crops</td>
<td>Non-irrigated arable land and complex cultivation patterns</td>
</tr>
<tr>
<td>SNAP100103/SNAP100203 — Rice field</td>
<td>Rice fields</td>
</tr>
<tr>
<td>SNAP100104/SNAP100204 — Market gardening</td>
<td>Permanently irrigated land and complex cultivation patterns</td>
</tr>
<tr>
<td>SNAP100105/SNAP100205 — Grassland</td>
<td>Pastures, land principally occupied by agriculture and natural grasslands</td>
</tr>
<tr>
<td>SNAP1003 — On-field burning</td>
<td>Non-irrigated arable land</td>
</tr>
<tr>
<td>SNAP1005/SNAP1009 — Manure management</td>
<td>Non-irrigated arable land</td>
</tr>
</tbody>
</table>

### 3.1. Total annual emissions

Fig. 3 represents the total annual emissions for Spain by pollutant [t·year⁻¹] estimated by HERMES04, INESP04, HERMESv2.0 and INESP09. The INESP results are included to have a reference of the estimations made with the corresponding HERMES model (HERMES04/INESP04 and HERMESv2.0/INESP09). The anthropogenic and biogenic total annual emissions in Spain for the year 2009 were estimated by HERMESv2.0 as follows: NOx, 924 kt; NMVOCs, 2332 kt; SO2, 278 kt; CO, 1976 kt; NH3, 339 kt; and TSP, 182 kt. Compared with HERMES04, the major differences are shown in NMVOCs (±1172 kt) and SO2 (±873 kt). In the case of NMVOCs, these differences are mainly due to the change of the biogenic model used (SNAP11). In HERMES04, biogenic emissions were estimated using a methodology based on the work of Parra et al. (2004) (Baldasano et al., 2008). On the other hand, SO2 differences are due to the reduction of coal use in thermal stations, as mentioned in Section 1. For the remaining pollutants, the total amounts changed in a lower degree: NOx (25 kt); CO (−2 kt); NH3 (−23 kt) and TSP (−27 kt).

The absolute emissions per specie and SNAP sector as well as the differences obtained by subtracting INESP04 from HERMES04 and INESP09 from HERMESv2.0 are presented in Table 5. The NH3 emissions are not included because most (~90%) come from SNAP10, for which a downsampling methodology has been applied to the INESP inventory (Section 2.2.11). Although the total amounts of estimated emissions by both HERMES models are similar, the contributions of each SNAP sector to the total amount of pollutants emitted have significantly changed. The results also show that HERMESv2.0 is in better agreement with the INESP inventory, especially for NOx, NMVOCs and CO.

For NOx, the most significant sources are road traffic in both cases, followed by combustion in energy industries. The updating of the road traffic database (SNAP07) and the consideration of agricultural machinery (SNAP08) has entailed a significant underestimation reduction compared to INESP inventory (from −669 kt·year⁻¹ to −176 kt·year⁻¹). In the case of NMVOCs, mainly emitted by terrestrial ecosystems in both cases, it is important to highlight the variation of the contribution from road transport, caused by the increase of diesel-powered vehicles in vehicular park compositions, which present lower hydrocarbon emissions than gasoline-fuelled vehicles according to chapter 1.A.3.b of EEA (2009). Compared to INESP results, a major improvement is also observed for this pollutant (from −1217 kt·year⁻¹ to 135 kt·year⁻¹) due to the introduction of MEGANv2.0.4 (SNAP11) and the industrial solvent use (SNAP06). For SO2, more than 80% of the emissions come from point sources in both model versions. In HERMES04, however, most of the emissions are exclusively from energy industries.
Compared to INESP, the underestimation has not varied, due to the non-consideration of important fuel-oil industrial boilers due to the lack of data. In HERMES04, the CO emissions mainly come from road traffic, a contribution that has decreased in HERMESv2.0 due to the same diesel-gasoline vehicle factor mentioned above for NMVOCs. On the other hand, the consideration in HERMESv2.0 of both residential biomass combustion and biogenic CO emissions has entailed an improvement of the results compared to the INESP inventory (from −761 kt year\(^{-1}\) to 271 kt year\(^{-1}\)). For this pollutant, however, major discrepancies are still observed in SNAP07, which may be caused by the different assumptions and emission factors considered. Regarding TSP, emissions are mostly emitted by road transport. In HERMESv2.0 the contribution of residential/commercial combustion has increased because of the biomass burning. This pollutant presents the lowest differences compared to INESP results (from −11 kt year\(^{-1}\) to 20 kt year\(^{-1}\)).

### 3.2. Spatial distributions

Fig. 4 shows the spatial distribution of the HERMESv2.0 total annual emissions as well as the differences obtained after subtracting the HERMES04 results. The analysis is focused on NO\(_x\), NMVOCs and PM\(_{10}\) because of their key contribution to Spanish air quality exceedances in terms of NO\(_2\), PM\(_{10}\) and O\(_3\) (MAGRAMA, 2012b).

The highest NO\(_x\) values are shown in cells where power plants and industrial sources are located as well as in large urban areas (the Madrid and Barcelona urban areas account for nearly 5% of the total emissions) and highways. The background levels observed in some areas correspond essentially to emissions derived from agricultural machinery. Compared with HERMES04, interurban and background emissions have experienced a slight increase. By contrast, cells located in industrial sites and the greater Madrid and Barcelona metropolitan areas show a decline.

The influence of biogenic sources, which are scattered throughout the territory, is observed in the NMVOCs results. The highest emissions correspond to the greater areas of the large urban agglomerations due to both residential solvent use and evaporative road traffic emissions. The map with the differences shows how HERMESv2.0 estimates higher BNMVOCs. The decrease in urban areas could be related to the large fraction of diesel-powered vehicles considered in HERMESv2.0 (evaporative emissions from diesel vehicles are considered to be negligible due to the presence of heavier hydrocarbons and the relatively low vapour pressure of diesel fuel). Differences due to the consideration of offshore extracting activities (SNAP05) are also observed near Catalonia’s coast.

The PM\(_{10}\) spatial distribution follows a pattern similar to the NO\(_x\) results. In this case, however, the background emissions are influenced not only by agricultural machinery but also by biomass combustion in rural sites. The decreasing emissions in most of the industrial sites have been compensated for by an increase in urban and rural areas.

### 3.3. Temporal variations

The total daily emissions performed by HERMES04 and HERMESv2.0 are illustrated in Fig. 5a and b. The daily trend of NO\(_x\) is similar in both cases, with marked weekly cycles because of the road traffic contribution. Nevertheless, HERMESv2.0 presents a slight increase in the summer period due to the incursion of agricultural machinery. In the case of CO, HERMES04 exhibits a similar pattern throughout the year, while the HERMESv2.0 results are strongly influenced by both the contribution of biomass combustion during winter and biogenic emissions during the summer period. For NMVOCs, the results show major peaks in HERMESv2.0, especially during summer, which are mainly due to the influence of the biogenic emissions. The differences for SO\(_2\) are primarily due to
Fig. 4. Spatial distribution of the HERMESv2.0 annual emissions in the Iberian Peninsula domain (4 km × 4 km) and differences with HERMES04 results [t·year⁻¹·cell⁻¹]: (a) NOₓ, (b) NMVOCs (c) PM₁₀.
the decrease in coal combustion, while in PM<sub>10</sub>, both trends are fairly close. The introduction of a seasonal variation for agricultural NH₃ may be important in terms model performance, even if it is a simplified temporal profile (Skjøth et al., 2011).

4. Conclusions

This work presents and describes the main elements and results of the HERMESv2.0 system, a high-resolution model that provides detailed emission information according to the SNAP nomenclature using 2009 as the reference year and using bottom-up approaches for the most significant sources. The model will substitute HERMES04 as the emission core of the European and Spanish operational air quality forecasting system CALIOPE. Several efforts were made to update and improve the quality and representativeness of both the databases and the methodologies used.

The emissions estimated by HERMESv2.0 for Spain in 2009 are as follows: NOₓ, 924 kt; NMVOCs, 2331 kt; SO₂, 278 kt; CO, 2178 kt; NH₃, 339 kt; and TSP, 182 kt, with road transport and energy combustion the most significant pollutant sources. Regarding its predecessor, major changes are only observed in NMVOCs (÷1170 kt) and SO₂ (−873 kt). However, the contributions of each SNAP activity to the total amount of pollutants emitted have significantly changed from one model to another on account of the combination of both changes in Spanish activity patterns and improvements applied to the model.

The effects of all these changes and improvements have also been shown in terms of spatial and temporal distribution. In the first case, it is important to highlight the increase of background NMVOCs emissions as well as NOₓ and PM<sub>10</sub> in highways and large cities, respectively. Considering the temporal variations, the introduction of MEGANv2.04 increased NMVOCs levels during the summer period. Moreover, the inclusion of biomass in residential/commercial sectors and agricultural machinery is also observable in the CO levels during winter and the NOₓ levels in summer time.

The comparison between the two HERMES versions and the INESP emission inventories for the years 2004 and 2009 has shown that the HERMESv2.0 results are in better agreement than those of their predecessor. The underestimations observed with HERMES04 have generally been reduced, especially in NOₓ (from −669 kt·year⁻¹ to −176 kt·year⁻¹), CO (from −761 kt·year⁻¹ to 271 kt·year⁻¹) and NMVOCs (from −1217 kt·year⁻¹ to 135 kt·year⁻¹). This fact proves that the coverage of the datasets used in HERMESv2.0 bottom-up approaches (i.e., point source inventory) is more complete and more capable of capturing the total amount of pollutants estimated using official governmental statistics.

As in most emission models, and despite all of the improvements applied, HERMESv2.0 still presents shortcomings and limitations. Some pollutant sources require more specific data and others that could be significant are not yet considered (e.g., wind-blown dust and forest fire emissions). In this sense, future developments of the system will be related to the inclusion of other potential pollutant activities and revision of methodologies. Future works will also be related to the evaluation and discussion of HERMESv2.0 as the emission core of the CALIOPE air quality system. This work will be useful both to analyse how improvements and the updates applied affect air quality results and to identify errors in the inventory data.

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Appendix A. Supplementary material

The supplementary material summarizes all the information sources used, as well as the EFs, spatial proxies, temporal profiles and estimated emission expressions used in HERMESv2.0 for the calculation of hourly and gridded emissions. It also includes the
methodologies and results on the emission vertical assignment and emission chemical speciation performed with the model.

Supplementary material related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2013.08.053.

References


AB (Barcelona City Council), 2010. Evaluation of the NOx and PM10 Emission Reductions from Traffic in Barcelona City Based on the Characterization of the Vehicle Pool.


