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A model-based analysis of SO$_2$ and NO$_2$ dynamics from coal-fired power plants under representative synoptic circulation types over the Iberian Peninsula

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HIGHLIGHTS

• Synoptic circulation controls coal-fired power plants plumes from Atlantic facilities.
• A combination of synoptic and mesoscale circulations drives Mediterranean plumes.
• Under Atlantic dominated circulation types, pollution plumes can reach up to 250 km.
• Emission injection within the PBL favours fumigation close to the source (<20 km).
• Hourly contribution to SO$_2$ and NO$_2$ ranges 2–25 μg m$^{-3}$ and 1–15 μg m$^{-3}$, respectively.

GRAPHICAL ABSTRACT

ABSTRACT

Emissions of SO$_2$ and NO$_2$ from coal-fired power plants are a significant source of air pollution. In order to typify the power plants’ plumes dynamics and quantify their contribution to air quality, a comprehensive characterisation of seven coal-fired power plant plumes has been performed under six representative circulation types (CTs) identified by means of a synoptic classification over the Iberian Peninsula. The emission and the transport of SO$_2$ and NO$_2$ have been simulated with the CALIOPE air quality forecasting system that couples the HERMES emission model for Spain and WRF and CMAQ models. For the facilities located in continental and Atlantic areas (As Pontes, Aboño, and Compostilla), the synoptic advection controls pollutant transport, however, for power plants located along the Mediterranean or over complex-terrains (Guardo, Andorra, Carboneras, and Los Barrios), plume dynamics are driven by a combination of synoptic and mesoscale mountain-valley and sea-land breezes. The contribution of power plants to surface concentration occurs mainly close to the source (<20 km) related to a fumigation process when the emission injection takes place within the planetary boundary layer reaching up to 55 μg SO$_2$ m$^{-3}$ and 32 μg NO$_2$ m$^{-3}$. However, the SO$_2$ and NO$_2$ plumes can reach long distances (>250 km from the sources) especially for CTs characterised by Atlantic advection.

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1. Introduction

Air pollution is a threat for human health (Brunkreft and Holgate, 2002; Gurjar et al., 2010; WHO, 2013) and the environment (de Vries et al., 2014). In the last years, the European Union has established limits to the emission of air pollutants (National Emission Ceilings Directive 2001/81/EC) and legally binding limit values for air quality (Directive 2008/50/EC) in order to reduce the exposition of people and ecosystems to harmful concentrations of pollutants. Partially due to these policy actions, during the 2001–2012 period, the SO2 and NO2 concentration at the remote rural European Monitoring and Evaluation Programme (EMEP) stations in Spain declined 3.6 to 7.7%/year and 2.8 to 3.7%/year, respectively (Querol et al., 2014).

Despite the increase in renewable electricity production in Spain by 50% in the 2008–2012 period (REE, 2013) and the use of cleaner technologies and fuels (ORDEN PRE/77/2008), the contribution of coal-fired power plants to the electricity generation pool was 19.3% in 2012, 54,721 GWh for the Spanish Iberian Peninsula (REE, 2013), being the second technology in electricity generation in 2012 after nuclear (22.1%) and before wind power (18.1%). Although SO2 and NO2 emissions from energy industry (SNAP01) have been reduced by 29.7% and 12.5% during the period 2008–2012, respectively (MAGRAMA, 2014); they were still significant in 2012 corresponding to a 41% and 22% of the SO2 and NO2 total Spanish emissions, respectively. The emissions from coal-fired power plants were the main contributor within the SNAP01 with 79% and 55% of SO2 and NO2 emissions, respectively.

The synoptic scale circulation is considered to play a significant role in air pollution both transporting primary and secondary pollutants through long distances (Vivanco et al., 2012; Putero et al., 2014) and controlling the effect on the local meteorological conditions (Kassomenos et al., 1998; Menut et al., 1999, Segura et al., 2013). Several studies relate how different circulation types (CTs) establish dissimilar effects on health for respiratory (Jamason et al., 1997; de Pablo et al., 2006, 2008) and cardiovascular, and digestive diseases (Morabito et al., 2008).

Circulation-type classification summarises a complex series of synoptic conditions into a catalogue containing a small number of predominant modes of atmospheric circulation or CTs (Barry and Perry, 1974; Beck and Philipp, 2010; Philipp et al., 2014). Each CT is associated with a number of distinctive meteorological behaviours and predominant flow characteristics (Shahgedanova et al., 1998). Several CT classifications have been performed over the Iberian Peninsula (IP) for different applications. Recently, an objective classification based on a climatic database has been developed for air quality purposes and could be used to analyse the plume dynamics under representative CTs (Valverde et al., 2014).

The dispersion of the pollutants emitted at high stacks relies on combination of meteorological fields and is affected by topography (Palau et al., 2005, 2009). Plume dispersion at power plants, refineries and incinerators has been analysed in impact assessments studies under particular pollution episodes over Spain (Salvador et al., 1992; Hernández et al., 1995, 1997; Puig et al., 2008; Baldasano et al., 2014). However, there is no a comprehensive characterisation of the plume dynamics from coal-fired power plants considering the influence of (1) the facilities characteristics, (2) the topography, and (3) the synoptic conditions affecting the IP. Understanding the plume dynamics and the specific contribution of power plants to air pollution under representative CTs can improve the power grid management in the context of environmental sustainability.

The objective of this work is twofold. First, to characterise the plume dynamics for selected Spanish coal-fired power plants under representative CTs over the IP, describing the role of emissions, meteorology, and topography. Second, to determine the contribution of SO2 and NO2 surface concentration of each power plant under each CT.

The paper is organised as follows. Section 2 describes the methods and data used. Section 3 characterises the power plants’ plume dynamics and analyses their contribution to surface SO2 and NO2 concentration. Finally, Section 4 discusses the synoptic circulation role on plume dynamics over the IP.
Table 1

<table>
<thead>
<tr>
<th>Power plant/acronym/PRTR register</th>
<th>Installed capacity/production</th>
<th># stacks and stack height</th>
<th>Type of fuel</th>
<th>SO$_2$ emissions$^a$ (Gg year$^{-1}$)</th>
<th>NO$_x$ emissions$^a$ (Gg year$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>As Pontes/ASP/3536/360</td>
<td>1468 MW/5815 GWh</td>
<td>4 × 356</td>
<td>Brown lignite</td>
<td>4.99</td>
<td>7.46</td>
</tr>
<tr>
<td>Aboño/ABO/2928/42</td>
<td>921 MW/4876 GWh</td>
<td>1 × 175 + 1 × 225</td>
<td>Coal + anthracite</td>
<td>5.83</td>
<td>8.13</td>
</tr>
<tr>
<td>Compostilla II/COM/8246/590</td>
<td>1341 MW/2819 GWh</td>
<td>2 × 270 + 2 × 290</td>
<td>Coal + anthracite</td>
<td>3.79</td>
<td>8.38</td>
</tr>
<tr>
<td>Guardo/Velilla/GUA/3590/1120</td>
<td>498 MW/580 GWh</td>
<td>1 × 70 + 1 × 176</td>
<td>Coal + anthracite</td>
<td>0.82</td>
<td>3.07</td>
</tr>
<tr>
<td>Andorra/AND/3530/612</td>
<td>1101 MW/2717 GWh</td>
<td>3 × 343</td>
<td>Black lignite</td>
<td>11.71</td>
<td>10.00</td>
</tr>
<tr>
<td>Litoral de Almería, Carboneras/CAR/3537/13</td>
<td>1159 MW/5804 GWh</td>
<td>2 × 200</td>
<td>Imported coal</td>
<td>13.99</td>
<td>9.80</td>
</tr>
<tr>
<td>Los Barrios/LBB/3531/11</td>
<td>568 MW/3219 GWh</td>
<td>1 × 230</td>
<td>Imported coal</td>
<td>2.53</td>
<td>5.39</td>
</tr>
</tbody>
</table>

* Based on year 2009.

and evaluated in detail elsewhere (Baldasano et al., 2008, 2011; Pay et al., 2011, 2012, 2014). It integrates the Weather Research and Forecasting model which uses the advanced research dynamical solver, WRF-ARWv3.5 (Skamarock and Klemp, 2008), a specific emission model (HERMESv2; Guevara et al., 2013, 2014), the Eulerian area-limited Community Multi-scale Air Quality model (CMAQv5.0.2; Byun and Schere, 2006), and an off-line mineral dust atmospheric model (BSC-DREAM8bv2 (Pérez et al., 2006a,b; Basart et al., 2012).

WRF-ARW runs over Europe (the mother domain) at 12 km × 12 km horizontal resolution using initial and boundary conditions from the Global Forecast System (GFS/FNL) dataset with 6-h temporal resolution, 0.5° × 0.5° horizontal resolution and 12-h spin-up. Over the IP, WRF runs at 4 km × 4 km using a one-way nesting over the mother domain.

Vertically, WRF-ARW is configured with 38 vertical levels from the surface up to 50 hPa, with 11 levels characterising the planetary boundary layer (PBL). The Yonsei University scheme is used to parametrize the PBL.

The CMAQ model uses MOZART4-GEOS5 forecasts as chemical boundary conditions (Lat 1.9° × Lon 2.5°, 6 h). A time-independent vertical concentration profile is used to settle the initial conditions, and a 7-day spin-up is used to heat the model before each simulation. The Meteorology-Chemistry Interface Processor of CMAQ (MCIP) is used to collapse the 38 σ levels of WRF into 15 vertical layers, 12 of which are below 1500 m above ground level (magl). The vertical diffusion module used is the Asymmetric Convective Model Scheme2 whereas the horizontal and vertical advection schemes are the Yamartino mass-conserving and the Piecewise Parabolic Method, respectively.

The HERMESv2 emission model is used for calculating Spanish emissions (Guevara et al., 2013, 2014). For Spanish power plants, HERMESv2 calculates emissions using real facility activity and measured emissions factors from 2009 (OCEM-CIEMAT, personal communication).

Regarding the CALIOPE-AQFS performance, the system is evaluated in near-real time against hourly air quality observations from more than 400 ground-level stations from the Spanish monitoring network. There are also annual evaluations of the system that provide robustness to the results (Baldasano et al., 2011 and its supplementary material; Pay and Baldasano, 2012; Arévalo et al., 2014).

2.4. Plume dynamics characterisation: methods and data

In order to understand the relationship between each CT and the plume dynamics the present analysis takes into account the location, the stack height and the emission rates of each facility, meteorological fields, and SO$_2$ and NO$_x$ concentration profiles.

At each power plant, meteorological hourly fields are extracted from the MCIP module. Wind speed, wind direction and vertical vorticity at different vertical layers enable an analysis of horizontal and vertical plume transport close to the power plant. Positive vertical vorticity causes counter clockwise circulation and upward movement of the air mass, whereas negative values cause clockwise circulation and downward movement.

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Fig. 1. Topographic map of the Iberian Peninsula and location of the studied coal-fired power plants according to Table 1.
As different plume behaviours are expected when emission occurs under and above the PBL, the PBL height (PBLH, magl) was also analysed using data from the MCIP module. SO2 and NO2 hourly concentrations are estimated by the CMAQ model for the representative day of each CT. A zero-out approach (running two equal simulations but setting the emissions from the seven selected facilities to zero in one of them) is used to obtain the contribution of the power plants in terms of air quality concentrations. Two complimentary approaches are considered to analyse these data. On the one hand, N-S and W-E cross-sections up to 3000 m above sea level (masl) passing by the power plant are obtained (daily maximum and hourly concentration are used). Cross-sections plots help characterising the plumes diffusion and their relationship to topography. On the other hand, SO2 and NO2 surface concentration maps and total column mass maps allow describing the horizontal plume transport. The plume length (km) is calculated as the maximum distance from the facility that has a surface contribution higher than the remote background concentration (RBC) in Spain. RBC is calculated as the 2003–2012 average of 10 Spanish EMEP stations corresponding to 4.3 μg NO2 m⁻³ and 1.2 μg SO2 m⁻³.

3. Results

3.1. Evaluation of CALIOPE

To analyse the performance of the modelling system, the three most widely used statistical indicators according to Dennis et al. (2010) have been computed: the Mean Bias (MB), the Root Mean Squared Error (RMSE) and the correlation coefficient (r). The CALIOPE-AQFS properly reproduces SO2 and NO2 concentrations as shown by statistics (Table 3) calculated on an hourly basis for a 4-day episode finishing on the representative day of each CT against validated observations from the Spanish monitoring network (a map with the location of stations used is presented in the Supplementary Material, Fig. S1). The SO2 and NO2 concentrations tend to be underestimated, on average for the six CTs, the MB are −2.2 μg SO2 m⁻³ and −5.6 μg NO2 m⁻³. However, the general temporal variability is well captured, with a r-SO2 = 0.19 and r-NO2 = 0.58. For SO2, the lowest error is found for the WSAdv (RMSE = 5.3 μg m⁻³) and are alike for the other CTs (RMSE = 9.5 μg m⁻³). For NO2, the highest error is found for the AtlHI (RMSE = 20.1 μg m⁻³) and lowest is also minimum for the WSAdv episode (RMSE = 15.2 μg m⁻³). The DELTA TOOL (Thunis and Cuvelier, 2014) is a harmonised tool that is useful to assess the model quality objectives that indicate the level of accuracy considered to be acceptable for regulatory applications according to the Ambient Air Quality Directive 2008/50/EC (AADQ). The DELTA TOOL version 4.0 has been used to evaluate the CALIOPE-AQFS performance for each 4-day episode considering all the available air quality stations regardless their typology. In this tool’s version, the SO2 Target plot is not available because there is no parametrization to estimate SO2 measurements uncertainty and thus only results for NO2 are discussed (Fig. 3). For both plots, stations located in the green area fulfil the model quality objectives based on bias, correlation, error, and observation uncertainty. The number of stations fulfilling the target criteria, indicated as a percentage in the plots, ranges from 82% to 94% on all CTs. According to the AADQ, the minimum expected performance of a model is 90%. The worst results are obtained for the ENAdv (~87%) and IBl (~88%) probably related to a combination of high NO2 emissions together with stagnant conditions over large parts of the IP that make it difficult to properly model the transport dynamics. On the contrary, the best results (>90%) are recorded under Atlantic advection conditions (ZonAdv, WSAdv nd NWAdv). Overall, there is a negative bias for surface concentration, especially for low values (<10 μg m⁻³). Moreover, the target plots highlight that correlation errors dominate over the standard deviation error.

3.2. Emissions characterisation

To explain the power plants’ plume dynamics it is necessary to characterise the temporal emissions variability. The power plants emissions depend on the electricity demand and therefore change throughout the year and along the day (the emission rates for each facility along the representative day of each CT are presented as Supplementary Material, Fig. S2). In general terms, the emission rates are the highest during winter CTs AtlHi and ZonAdv, both with a representative day in January 2004. For the seven power plants, the daily emission rate ranges 250–3400 kg SO2 day⁻¹ and 900–2900 kg NO2 day⁻¹. In ASP however, the maximum rates are reached in NWAdv (1310 kg SO2 day⁻¹ and 1960 kg NO2 day⁻¹). In August, during the IBl, the emissions are minimal in the seven power plants ranging, 50–2000 kg SO2 day⁻¹ and 190–900 kg NO2 day⁻¹. Superimposed to the seasonal variations, the emission rates change following a daily cycle. Emission rates are the lowest at night and rise in the early morning showing their maximum at 12:00. A second maximum is reached at 20:00–21:00. There are larger variations on emission rates along the daily cycle during winter CTs than during summer, especially for power plants with an emission rate lower than 20 kg h⁻¹ (GUA, LBB).

3.3. Plume dynamics from Spanish power plants

In this section, the SO2 and NO2 plume dynamics are described on the representative day of each CT using i) maps for the daily maximum surface concentration (Fig. 4), ii) maps for the daily maximum total column mass (Fig. 5), iii) cross-sections at the power plants for the daily maximum concentration (Supplementary Material S5) and, iv) daily cycle at the power plant for the hourly PBLH, wind speed and direction and, the vertical vorticity (Supplementary Material S7). The same analysis is shown for NO2 in the supplementary materials S3, S4, S6 and S7.

3.3.1. NWAdv

ASP and ABO, located the Atlantic coast, are reached by N/NW winds along the day (Fig. 2). The plume from ASP reaches 1250 m above injection height (maih, computed as altitude of the facility plus stack height) and it is oriented towards the SE/E (Figs. 4, 5, S3, S4) reaching 125 km at noon is related to i) the injection is done within the PBL which height is maximum at midday (ASP ~ 1000 maih, ABO ~ 600 maih) due to thermal heating of the ground (Fig. S7a, b); and ii) between 13:00 and 17:00 the vertical velocity is negative, favouring a vertical transport of the source (10 km) of 7.3 μg SO2 m⁻³ and 6.5 μg NO2 m⁻³ in ASP and 14.3 μg SO2 m⁻³ and 16.4 μg NO2 m⁻³ in ABO. There is a fogging process occurring at both power plants at midday leading to a maximum increase in surface concentration close to the source (10–15 km) of 7.3 μg SO2 m⁻³ and 6.5 μg NO2 m⁻³ in ASP and 14.3 μg SO2 m⁻³ and 16.4 μg NO2 m⁻³ in ABO. This fogging process at noon is related to i) the injection is done within the PBL which height is maximum at midday (ASP – 1000 maih, ABO – 600 maih) due to thermal heating of the ground (Fig. S7a, b); and ii) between 13:00 and 17:00 the vertical velocity is negative, favouring a vertical transport of the emissions towards the ground. The main difference between ASP and ABO is that in the former, although the emission rates for SO2 and NO2 are higher than in ABO, the emissions are injected at higher altitude (ASP injection_height = 716 masl > ABO injection_height = 267 masl) favouring a larger pollutants dispersion (in ASP there is little plume formation for NO2, Fig. S3).

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Table 2

<table>
<thead>
<tr>
<th>CT</th>
<th>1983–2012 frequency</th>
<th>2012 frequency</th>
<th>Representative day</th>
</tr>
</thead>
<tbody>
<tr>
<td>NW adv (NWAdv)</td>
<td>23.9%</td>
<td>21.9%</td>
<td>Sunday 29/07/2012</td>
</tr>
<tr>
<td>Iberian thermal low (IBtl)</td>
<td>22.4%</td>
<td>21.6%</td>
<td>Sunday 19/08/2012</td>
</tr>
<tr>
<td>E/NE adv (ENEAdv)</td>
<td>21.3%</td>
<td>8.8%</td>
<td>Thursday 24/05/2012</td>
</tr>
<tr>
<td>Atlantic high (AtlHI)</td>
<td>12.0%</td>
<td>17.8%</td>
<td>Tuesday 24/01/2012</td>
</tr>
<tr>
<td>W/SW adv (WSAdv)</td>
<td>10.4%</td>
<td>20.5%</td>
<td>Tuesday 16/10/2012</td>
</tr>
<tr>
<td>Zonal Western adv (ZonWAdv)</td>
<td>10.1%</td>
<td>9.3%</td>
<td>Tuesday 03/01/2012</td>
</tr>
</tbody>
</table>
Fig. 2. Mean sea-level pressure (hPa) and 10-m wind (m s\(^{-1}\)) of the representative day of each CT in 2012 (12:00 UTC) over the Iberian Peninsula.
COM and GUA are located in Northern Spain in areas with complex topography (Fig. 1). The Cantabrian Mountains (2000 m asl with W–E orientation) situated to the North of COM and GUA behave as an orographic barrier reducing the influence of the Atlantic Ocean over these areas. Synoptic NW winds towards COM and GUA are transformed into westerlies that blow parallel to the Cantabrian Mountains along the day (Fig. S7c and d) transporting the SO2 and NO2 plumes to the E/NE. COM plumes reach ~850 m asl and ~75 km for SO2 and ~55 km for NO2. The surface contributions are registered at 10 km from the source and are 6.4 μg SO2 m⁻³ and 8.6 μg NO2 m⁻³. In GUA, the plume of both pollutants attains 400 m asl and 25 km. The maximum contribution is disclosed at 7 km from the source (3.7 μg SO2 m⁻³ and 10.5 μg NO2 m⁻³). As in ASP and ABO, in COM and GUA, there is also a fumigation process from 10:00 that explains the pollutants diffusion towards the ground although the PBLH is higher (~1400 m) than in the Atlantic power plants. The length and width of GUA plumes is lower than those of COM (Figs. 5, S4) due to a combination of factors: lower SO2 and NO2 emission rates at GUA, lower plume altitude (the emissions are less dispersed because they are exposed to weaker winds) and because GUA is under the influence of mountain-valley winds which favour recirculations close to the source (Fig. S7d).

The Ebro valley, where AND is located, connects the Iberian Peninsula Mediterranean and Atlantic coasts. Under NWadv, northwesterlies are channelled by the Ebro Valley. At night, the wind is orientated down valley towards the Mediterranean Sea (Fig. S7e). However, during the day, the wind is up valley (SE) driven by strong sea–land breezes. Moreover, orographic mountain-valley winds originated in the close Gúdar valley towards the Mediterranean Sea (Fig. S7e). However, during the morning until mesoscale sea wind complexity in the region. The SO2 and NO2 plumes from AND drift over, orographic mountain-valley winds originated in the close Gúdar valley towards the Mediterranean Sea (Fig. S7e). However, during the day (Fig. S7c and d) transporting the SO2 and NO2 plumes to the eastern façade of the Algeciras Mountains (Fig. 1, 15 km from the source) is reached by the plumes of LBB contributing to 10.6 μg SO2 m⁻³ and 15.0 μg NO2 m⁻³ to surface concentration. It is also noteworthy that from 20:00 the PBLH is ~35 mgl. This nocturnal layer enhances the concentration of pollutants at surface level.

### Table 3

<table>
<thead>
<tr>
<th>NWadv SO2</th>
<th>NWadv NO2</th>
<th>IBtl SO2</th>
<th>IBtl NO2</th>
<th>ENadv SO2</th>
<th>ENadv NO2</th>
<th>ATH SO2</th>
<th>ATH NO2</th>
<th>WSWadv SO2</th>
<th>WSWadv NO2</th>
<th>ZonSO2</th>
<th>ZonNO2</th>
</tr>
</thead>
<tbody>
<tr>
<td>395</td>
<td>454</td>
<td>402</td>
<td>455</td>
<td>407</td>
<td>458</td>
<td>405</td>
<td>464</td>
<td>394</td>
<td>446</td>
<td>404</td>
<td>465</td>
</tr>
<tr>
<td>MB (μg m⁻³)</td>
<td>-1.6</td>
<td>-3.6</td>
<td>-0.9</td>
<td>-2.8</td>
<td>-2.6</td>
<td>-6.9</td>
<td>-2.5</td>
<td>-7.3</td>
<td>-2.2</td>
<td>-4.7</td>
<td>-3.3</td>
</tr>
<tr>
<td>RMSE (μg m⁻³)</td>
<td>9.8</td>
<td>15.4</td>
<td>10.0</td>
<td>18.6</td>
<td>9.1</td>
<td>16.7</td>
<td>9.4</td>
<td>20.1</td>
<td>5.3</td>
<td>15.2</td>
<td>9.1</td>
</tr>
<tr>
<td>r</td>
<td>0.15</td>
<td>0.47</td>
<td>0.22</td>
<td>0.48</td>
<td>0.26</td>
<td>0.59</td>
<td>0.16</td>
<td>0.63</td>
<td>0.19</td>
<td>0.65</td>
<td>0.17</td>
</tr>
<tr>
<td>OBS (μg m⁻³)</td>
<td>4.4</td>
<td>14.3</td>
<td>4.8</td>
<td>16.3</td>
<td>5.0</td>
<td>16.8</td>
<td>5.6</td>
<td>24.1</td>
<td>3.9</td>
<td>16.3</td>
<td>5.3</td>
</tr>
<tr>
<td>MOD (μg m⁻³)</td>
<td>2.8</td>
<td>16.7</td>
<td>3.9</td>
<td>13.5</td>
<td>2.4</td>
<td>9.9</td>
<td>3.2</td>
<td>16.8</td>
<td>1.8</td>
<td>11.6</td>
<td>2.0</td>
</tr>
</tbody>
</table>

#### 3.3.2. IBtl

Under IBtl, the North-Western Iberian Peninsula is under the cyclonic influence of the Icelandic Low (Fig. 2). The emissions at ASP are the lowest of all the representative days (30 kg SO2 h⁻¹ and 40 kg NO2 h⁻¹), which hinders the formation of distinctive plumes. Although the PBLH at midday is ~1000 mgl (Fig. S7a) and the injection is done within the PBL, there is positive vertical vorticity during this time-span (11:00–14:00), which prevents fumigation towards the ground. The Cantabrian Mountains disconnect ABO from the southwards synoptic advection affecting the IP. Plume dynamics in ABO are driven by a mesoscale vortex that favours sea–land dispersion during the day and land–sea during the night at 500 m asl (Figs. S5b and S6b). The emissions are injected above the PBL (~400 mgl) which favours its horizontal transport. The SO2 and NO2 plumes reach the ground at ~95 and 25 km from the source, respectively. The contribution of the power plant in terms of surface concentration is maximal at 18 km south of the facility (~5 μg m⁻³ for SO2 and NO2).

Inland the IP, during the summer IBtl, the PBLH reaches its maximum in COM (~1900 mgl), GUA (~2000 mgl) and AND (~2100 mgl). Although the emissions at midday are injected within the PBL in these three facilities, there is little transport towards the ground. This is due to i) the SO2 and NO2 emission rates are the lowest of all the CTs (Fig. S2) due to lower electricity demand and ii) the vertical vorticity at midday is mainly positive due to soil heating. The maximum surface contribution occurs in COM at 7 km from the source: 4.7 μg SO2 m⁻³ and 6 μg NO2 m⁻³. Considering the total column, the SO2 and NO2 plumes from COM and GUA are driven by southerly northwards towards the Cantabrian Mountains that are a topographic block that avoids longer dispersion (Figs. 5 and S4). At AND, the winds are weak without dominant direction enabling that AND plumes remain close to the point source and reach 1500 mgl for SO2 and 1000 mgl for NO2.

At CAR, the wind blows from the E/NE along the day (Fig. S7f). In LBB, however, the wind is from the E (Fig. S7g). The plumes of CAR are transported in altitude 680 m asl for SO2, 380 m asl for NO2 (difference due to the large SO2 than NO2 emission rate in CAR) towards the SW (Figs. 5 and S4) whereas in LBB, the plume is oriented to the W reaching 360 m asl for both pollutants. In both cases, the injection is done above the PBL favouring its horizontal transport. The SO2 plumes from CAR and LBB have a maximum length of 97 and 81 km from the source, respectively.

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Fig. 3. Target plot (left) and scatter diagram plot (right) from DELTA TOOL for hourly pairs of observed-modelled NO₂ concentration along a 4-day episode of each CT in 2012. Symbols and colours represent the different stations. Percentage indicates stations fulfilling the model quality objective target criteria.
Fig. 4. Daily maximum SO$_2$ surface concentration (μg/m$^3$) associated to the emissions in the seven analysed power plants on the representative day of each CT.
Fig. 5. Daily maximum SO$_2$ total column mass (μg) associated to the emissions in the seven analysed power plants on the representative day of each CT.
3.3.3. ENEadv

The blocking anticyclone over Central Europe that characterises ENEadv leads to the arrival of easterlies towards ASP, ABO, COM and GUA. However, this general advective pattern is modulated by the Cantabrian Mountains. Winds arriving at ASP are from the SE/E during 00:00–12:00 and from NE/E from 15:00 onwards (Fig. S7a). The plumes from ASP are transported northwards during the morning (at 1200 maith) and after the change in wind direction, the plumes go towards the SW (Fig. 5 and S4). At midday, the injection is done within the PBL favouring fumigation towards the ground (<5 μg m⁻³ for both pollutants).

In ABO, the advection is from the E, parallel to the coastline at surface and at the injection height (Fig. S7b). For both pollutants, the plume is oriented towards the W at 230 maith (Figs. 5 and S4). The injection is done above the PBL along the day. This hinders the fumigation towards the ground and favours the horizontal transport (SO₂ plume length is 59 km whereas the NO₂ does not even reach the ground.

The easterlies arriving at COM drive the SO₂ and NO₂ plumes towards the W/NW. However, there is little horizontal plume advection (Figs. 4 and S3). The plume dynamics are dominated by the vertical vorticity (the maximum plume altitude is ~1500 maith). At midday negative vorticity combined with emission injection within the PBL (PBLH_{max} = 1800 magl) enhances fumigation towards the ground, close to the source. The maximum contributions to surface concentrations occur at 3.5 km from the source and attain 17.3 μg SO₂ m⁻³ and 24.1 μg NO₂ m⁻³. GUA plumes are also transported to the W without affecting the ground at 300 maith (Figs. S5d and S6d).

The advection under ENEadv at AND is from the North channelled by the Ebro valley (Fig. 2). The simulation shows a complex situation with a superposition of the synoptic northerlies, sea–land breezes from the Mediterranean and downslope winds from the Gúdar Mountains (Fig. S7e). The plumes from AND are transported to the SE during the morning (maximum altitude: 1350 maith for SO₂ and 1050 for NO₂). At midday with the development of sea–land breezes, the plumes turn westwards and are oriented towards the SW from the facility (Figs. 5 and S4). The change in wind direction at midday and AND together with negative vertical vorticity and emission injection within the PBL (PBLH_{max} ~ 1800 magl) enables a fumigation process. The maximum contribution to surface concentration is at 9 km from AND (5.6 μg SO₂ m⁻³ and 5.6 μg NO₂ m⁻³).

In Southern Spain, the ENEadv is characterised by easterlies (Fig. 2). The CAR plumes are transported towards the SW and W with a maximum altitude of ~800 maith. As the emission is done above the PBL (PBLH_{max} < 200 maql, Fig. S7f) the horizontal transport is enhanced enabling long plumes up to 200 km away from the stack (Figs. 5 and S4). It is noteworthy that the marine boundary layer is a barrier to the vertical movement of the plume favouring the transport of the pollutants inland. The plume affects the ground when it reaches the nearby Alhamilla Mountains, W of CAR (Figs. S5f and S6f) with up to 14.2 μg SO₂ m⁻³ and 7.5 μg NO₂ m⁻³.

In LBB the plumes are transported westwards (~100 km) close to the ground (~200 maith). The maximum contribution to surface concentration is 2.4 μg SO₂ m⁻³ and 20 μg NO₂ m⁻³ over the Algeciras Mountains, 20 km W of the source. The difference between pollutants is due to the emission rates (10 kg SO₂ h⁻¹ and 20 kg NO₂ h⁻¹). Moreover, during the night hours, there is a nocturnal layer (PBLH ~ <50 maql) that increases surface concentration close the source (Fig. S7g).

3.3.4. AtlHi

Under the winter AtlHi there is a low PBLH at the seven power plants locations (PBLH ~ <500 maql), This fact enables that the emission injection occurs above the PBL favouring their dispersion. On the other hand, AtlHi together with ZonWadv are the CTs characterised by highest emission rates (Fig. S2).

On the Cantabrian coast, AtlHi advection is depicted by northerlies that turn westwards when reaching the coastline. The plumes from ASP are transported westwards reaching ~1100–1200 maith without contacting the surface. The region of ASP is affected by quick easterlies coming from Asturias transporting SO₂ and NO₂ concentrations from ABO along the surface (Figs. S5a and S6a). In ABO, the plumes are parallel to the Atlantic coast (E–W orientation) and reach long distances (268 km for SO₂ and 204 km for NO₂) because there are not topographic barriers that stop them (0–600 maith, Figs. S5b and S6b). The highest contributions to surface concentration from ABO occur within 23 km from the source (~29 μg m⁻³ for both pollutants). The high emission rates together with high dispersive conditions favour the formation of wide multi-source plumes on North-Western IP during AtlHi (Fig. 5 and S4).

The COM plumes are also orientated westwards but the transport is partially stopped by the Galician Mountains (~40 km to the W) affecting the surface concentration in that area (Figs. S5c and S6c). The plumes from COM have a length of ~100 km and a maximum altitude of ~1100 maith. In COM and ABO, the PBL is very close to the ground (PBLH ~ <50 maql) during the night, enhancing the concentration of the pollutants at the surface (Fig. S7b and c).

During the day, the plume from GUA is transported towards the W (27 km, ~500 maith for NO₂ and ~700 maith for SO₂). However, the nocturnal downslope winds from the nearby mountains drive the plume towards the S (Figs. S5d and S6d). At midday (12:00–15:00) the PBLH is high enough (~500 maql) to enable the emission injection within the PBL favouring the increase of surface concentration close to the source (~3 km, 6.8 μg SO₂ m⁻³ and 16.6 μg NO₂ m⁻³).

Under AtlHi, the northern advection towards AND is channelled by the Ebro valley (Fig. 2) driving the plumes down valley towards the LBB.
The differential diffusivity of the pollutants explains this difference in altitude. The plumes reach 130–140 km from the source and the contribution little to surface concentrations (≤4 μg m⁻³).

Westerlies are characteristic of the AtlHi on the Strait of Gibraltar as well over the Alboran Sea (Fig. 2). However, winds blow from the NE in CAR (Fig. S7f) showing a complete disconnection from the main synoptic advection caused by an anticyclonic eddy located on the Mediterranean, 60 km away from the coastline. The plumes from CAR are transported to the SW/W. The Alhamilla Mountains blocks the progress of the plume westwards and enhances the concentration of pollutants on its eastern side 15–20 km from the source (45.1 μg SO₂ m⁻³ and 26.1 μg NO₂ m⁻³; Figs. S4f and S5b). The SO₂ and NO₂ plumes reach 2800 m and 1200 m and up to 250 and 180 km away from the source, respectively.

The plume from LBB is driven by westerlies into the Mediterranean during the morning (00:00–10:00). However, at midday there is a change in wind direction (E advection) and the LBB plume is transported towards the W (~180 km and 500–600 m) when the wind changes its main direction there is a significant diffusion of SO₂ and NO₂ from the stack to the ground at midday that contributes up to 21.6 and 24.5 μg m⁻³, respectively.

33.5. WSWadv and ZonWadv

In ASP the wind fields of WSWadv and ZonWadv are similar with a dominant SW surface advection coming from the Atlantic Ocean. However, at the injection height, ZonWadv presents clear western advection whereas under WSWadv, the advection is from the SW (Fig. S7a). High wind speed at the facility on both days together with emission injection above the PBL favours their horizontal dispersion towards the E/NE (Figs. 5 and 3) without attaining the surface in any moment.

Under WSWadv, the SW winds at ABO lead to a NE/N transport of the plumes at ~400 m towards the Ocean reaching 160 km for SO₂ and 110 for NO₂. The ABO contribution to surface concentration occurs at ~40 km east of the source (6.9 μg SO₂ m⁻³ and 11.0 μg NO₂ m⁻³). On the other hand, under ZonWadv westerlies disperse the plume towards the E at a maximum altitude of ~400 m without affecting the surface (Figs. S5b and S6b).

The synoptic winds that characterise WSWadv reach COM and GUA from the SW. The plumes are driven towards the NE at an altitude that ranges 300–500 m high and are stopped by the southern façade of the Cantabrian Mountains (contributing with ~5 μg m⁻³ for both pollutants, 20–30 km from the sources). Under ZonWadv, W winds are faster than at WSWadv, both at surface and along the column (Fig. S7c and d) driving the plumes of COM and GUA towards the E without affecting the surface (Figs. 4 and S3).

In AND, under WSWadv the horizontal plume dynamics are a combination synoptic advection and mesoscale breezes (Fig. S7e). During the night the plumes are driven by northwesterlies towards the S however during the day sea–land breezes dominate, leading to a N/NE transport at a maximum altitude of 1650 m for SO₂ and 1050 for NO₂. Under ZonWadv the westerlies are channelled by the Ebro Valley driving the plumes down-valley going beyond the Mediterranean coastline without affecting the surface (Figs. 5 and S4).

The simulated wind field at CAR shows a complex behaviour. Under WSWadv the sea–land breezes together with an anticyclonic vortex located over the Alboran Sea drive the CAR plumes towards the NE during the morning, towards the N at midday and towards the W in the afternoon (Fig. S6f). This CT is characterised by atmospheric instability with negative vertical vorticity that enhances the fumigation of the plume. The maximum contribution to surface concentration is reached at 4–6 km from the source (54.7 μg SO₂ m⁻³ and 32.0 μg NO₂ m⁻³). Furthermore, the SO₂ plume is transported up to 290 km from CAR (at an altitude of 1300 m) and the NO₂ plume reaches 50 km (700 m). Under ZonWadv, sea breezes are coupled with synoptic westerlies driving the plumes towards the NE, parallel to the coastline from surface up to 800–1200 m (Figs. S5f and S6f). The strong wind speed at the surface (15–20 m s⁻¹) and over the column that characterise the ZonWadv, favour the formation of long plumes (~250 km). The maximum contribution to surface concentration (22.4 μg SO₂ m⁻³ and 12.9 μg NO₂ m⁻³) is reached far away from the source (66 km for SO₂ and 116 km for NO₂).

Despite the Atlantic advection that affects the IP under WSWadv and ZonWadv, the area of the Strait of Gibraltar is disconnected from this general pattern and it is dominated by Mediterranean winds under both CTs. The SO₂ plumes from LBB, driven by easterlies at surface and at injection height, reach 170 km and 250 km W of the source at 260 m and 560 m for WSWadv and ZonWadv, respectively (Figs. S5g and S6g). The NO₂ plumes share the same transport pattern but they are shorter (55 and 85 km, respectively). The main difference between WSWadv and ZonWadv at LBB is that the emission rates during the latter (22 kg SO₂ h⁻¹ and 48 kg NO₂ h⁻¹) duplicate those of WSWadv (11 kg SO₂ h⁻¹ and 24 kg NO₂ h⁻¹) leading to a larger contribution at surface level (14.1 μg SO₂ m⁻³ and 19.9 μg NO₂ m⁻³) over the eastern Algeciras Mountains facade.

4. Conclusions

The CALIOPE-AQPS, used with a zero-out approach, is useful to characterise Spanish coal-fired power plants’ SO₂ and NO₂ plume dynamics under representative synoptic circulation types (CTs), and to quantify their contribution to air quality concentrations.

The plume dynamics of the power plants located on the Atlantic Iberian Peninsula (ASP, ABO and COM) are mainly driven by the advection pattern associated to each CT. However, the plumes from AND, CAR and LBB, located over the Mediterranean coast, are driven by a combination of the synoptic advection and mesoscale processes, namely sea–land breezes and wind-channeling by river valleys. The GLA plumes follow the synoptic advection under IBtl, ENEadv, WSWadv and ZonWadv however under NWadv and AtlHi, mesoscale winds related to its complex topographic context (valley drainage) influence the horizontal plume motions.

On average for the seven facilities and six CTs, the maximum plume lengths are 230 km for SO₂ and 112 km for NO₂ (with plumes up to 250–300 km for SO₂ and 200–250 km for NO₂). The average SO₂/NO₂ plume length for CTs characterised by Atlantic advection (NWadv, AtlHi, WSWadv and ZonWadv) is 110/54 km whereas for CTs with non-Atlantic advection (IBtl and ENEadv), the SO₂/NO₂ plume length is 66/6 km. The high coal-fired power plants stack height (70–356 m), which enable an injection of the emissions at high altitude where the atmosphere is more dispersive, explain these remarkable plume lengths.

The power plant contributions to SO₂ and NO₂ surface concentration occur mainly close to the source (~20 km). On average for the six CTs, the contributions range 2–25 μg SO₂ m⁻³ and 1–15 μg NO₂ m⁻³. The largest contributions are registered in CAR (25 μg SO₂ m⁻³, 15 μg NO₂ m⁻³), ABO (9 μg SO₂ m⁻³, 14 μg NO₂ m⁻³), and LBB (9 μg SO₂ m⁻³, 10 μg NO₂ m⁻³) whereas the lowest occur in ASP and AND (~5 μg SO₂ m⁻³, 1 μg NO₂ m⁻³), the power plants with higher stack heights. The stack height also explains why ASP and AND plumes reach higher altitudes than those of the other facilities. The contributions are more significant when the emissions are injected within the PBL enabling fumigation processes, usually at midday. When the injection occurs above the PBL, horizontal plume dispersion is favoured. However, if the emission rates are low, even when the meteorological conditions favour vertical diffusion, the pollutants are dispersed horizontally.

Within Eulerian photochemical models, the proper simulation of surface pollutant concentration has been related to the accurate determination of PBLH and vertical diffusion (Athanassiadis et al. 2002; Byun et al., 2007). The WRF models the PBL following the Yonsei University parametrisation although it is not clear which PBL scheme performs best for the Iberian Peninsula (Banks et al., 2015). Moreover,
the CMAQ models the diffusion using the ACM2 scheme which tends to predict smaller concentrations of primary pollutants at the surface [Plein, 2007]. Considering these modelling uncertainties the described fumigation dynamics have to be taken with caution.

The plume dynamics for each power plant under the six CTS shows similar patterns for SO2 and NO2. However, the absolute values of plume length, altitude and contribution to surface concentration differ depending on the emission rates (that are type-fuel dependent) and on the pollutant diffusivity in the air (higher for NO2 than SO2). Differences in SO2 and NO2 deposition velocity need to be studied to further explain the dissimilarities in their plume dynamics.

The obtained results over the Iberian Peninsula confirm that the pollution dynamics associated to coal-fired power plants depend on a combination of interlinked variables: location and topographic characteristics, stack height, emission rates, and synoptic and mesoscale meteorology.

**Q3 Uncited references**

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**Appendix A. Supplementary data**

Supplementary data to this article can be found at http://dx.doi.org/10.1016/j.stoi.2015.09.111.

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