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# SOURCE APPORTIONMENT USING THE BRUTE FORCE METHOD TO ESTIMATE SECTOR IMPACTS ON AIR POLLUTION EPISODES IN SPAIN

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**Abstract**: Source apportionment for pollution episodes in Spain were estimated using the brute force method applied to simulations carried out with the CHIMERE atmospheric chemistry transport model. Emission reductions were applied by sector and emitted species to calculate their impacts on air quality individually. Additional simulations were carried out to quantify the impacts of anthropogenic emissions from neighbouring countries and international shipping, model boundary concentrations and biogenic emissions. The results indicate that Spanish anthropogenic emissions have the largest impact on $NO_2$ and $PM_{2.5}$ concentrations, whereas background concentrations of $O_3$ have the largest impact on $O_3$ concentrations. Interestingly, background $O_3$ concentrations are also estimated to have notable impacts on $PM_{2.5}$ and $NO_2$ concentrations, especially in large cites. A sectoral analysis highlights the importance of impacts from road traffic and residential combustion on $PM_{2.5}$ and $NO_2$ concentrations, although the relative impact of these varies from city to city. Notable impacts of emissions from international shipping, industrial combustion, solvent use, agriculture and waste treatment on $O_3$ concentrations are also estimated. The results of the study have implications for the development of national air quality plans, as well as identify issues that require an international response, such as the reduction of global ozone concentrations or international shipping emissions.

***Key words:*** *Source apportionment; Brute force; Pollution episodes*

## INTRODUCTION

Despite emission reductions in recent decades, there are still frequent and widespread exceedances of tropospheric ozone target values in Spain, as well as high concentrations of nitrogen dioxide and particulate matter. Pollution episodes for the year 2021 (the reference year used in the latest Spanish National Air Pollution Control Programme; MITERD, 2023) were identified using a spatio-temporal analysis of the concentrations measured at the background air quality monitoring stations in Spain. The brute force source apportionment method was applied to simulations carried out with the CHIMERE atmospheric chemistry transport model for the year 2021. Emission reductions of 10% and 25% were applied by sector and emitted species to calculate their impacts on air quality individually, as well as check the linearity of the concentration responses to the changes in precursor emissions. The study was carried out at three levels of detail using simulations covering SW Europe (SWEU) to estimate impacts from neighbouring countries, international shipping and boundary concentrations and simulations covering the Iberian Peninsula and Balearic Islands (IP) to estimate impacts from sectoral emissions (annual simulations) and sectoral-species emissions (episodes).

## METHODS

Pollutant episodes in 2021 were defined as periods of more than one day when the observed concentrations at more then two background monitoring stations were above the daily mean limit value for $NO_2$ or $PM_{2.5}$ (50 and 25 µg $m^{-3}$, respectively) or the $O_3$ target value (maximum daily 8-hour mean (MDA8) of 120 µg $m^{-3}$) as defined in the new European ambient air quality directive (AAQD 2024). Nested simulations were carried out using the CHIMERE (v2013) chemistry and transport model (Menut et al., 2013) at spatial resolutions of 0.15° (SWEU) and 0.1° (IP) driven by meterological data from the IFS model of the ECMWF (obtained from the MARS archive through access provided by AEMET for research projects). Boundary conditions for the SWEU domain were taken from LMDZ-INCA (Hauglustaine et al., 2004) and GOCART (Ginoux et al., 2001) global model climatologies. Emissions for the base case (BC) simulations were taken from the Spanish National Emissions Inventory (Spanish territory) and the EMEP emission inventory (other countries within the domain). The emission reduction scenarios that were used to estimate the impacts relative to the base case are shown in Table 1.

**Table 1.** The emission reduction scenarios simulated for each domain and time period. SNAP: Selected Nomenclature for Air Pollution (1: Combustion in the production and transformation of energy; 2: Non-industrial combustion; 3: Industrial combustion; 4: Industrial processes; 5: Extraction and distribution of fossil fuels; 6: Use of solvents; 7: Road Transport; 8: Non-Road Transport; 9: Waste treatment; 10 Agriculture).

| SWEU Simulations (annual) | IP Simulations (annual) | IP Simulations (episodes) |
|---|---|---|
| • Spain (ES)<br>• France (FR)<br>• Portugal (PT)<br>• Other Countries (OTH)<br>• International Shipping (SHIP)<br>• Biogenic emissions (BIO)<br>• Boundary conc. of $O_3$ (BC_O3)<br>• Boundary conc. of other species (BC_OTHER) | • Spain (SNAP1 to SNAP10)<br>• Other Countries (OTH)<br>• International Shipping (SHIP)<br>• Biogenic emissions of VOCs (BIO_VOC)<br>• Biogenic emissions of NOx (BIO_NOx)<br>• Biogenic emissions of paticles (BIO_PM)<br>• Boundary conc. of $O_3$ (BC_O3)<br>• Boundary conc. of other species (BC_OTHER) | • Spain (SNAP1 to SNAP10 by species)<br>• Other Countries (OTH)<br>• International Shipping (SHIP)<br>• Biogenic emissions of VOCs (BIO_VOC)<br>• Biogenic emissions of NOx (BIO_NOx)<br>• Biogenic emissions of paticles (BIO_PM)<br>• Boundary conc. of $O_3$ (BC_O3)<br>• Boundary conc. of other species (BC_OTHER) |

Impacts for a 100% reduction of emissions were estimated by multiplying the difference between the base case and scenario concentrations by 100/α, where α is the emission reduction in % (10 or 25).

## RESULTS AND DISCUSSION

The analysis of the SWEU simulations indicates that the largest impact on $NO_2$ daily mean concentrations comes from the Spanish anthropogenic emissions although notable impacts are estimated from international shipping emissions along the Mediterranean coast as well as from boundary $O_3$ in large cities. Spanish emissions also have a large impact on $PM_{2.5}$ daily mean concentrations, as well as notable impacts from background concentrations. By contrast, the main impact on $O_3$ MDA8 values is from the background concentrations of $O_3$ and (to a lesser extent) its precursors, with small but still notable impacts from other sources, such as international shipping emissions along the Mediterranean coast. The impacts of the Spanish sectoral emissions were estimated from the IP simulations. The main impact on $NO_2$ concentrations is estimated to come from road-traffic emissions, especially in urban areas but there are also considerable impacts from industrial combustion and residential emissions (non-industrial combustion); the latter is especially important during winter pollution episodes. There are also notable seasonal impacts from agricultural NOx emissions. Residential and agricultural emissions are also estimated to have a considerable impact on $PM_{2.5}$ concentrations, especially during winter pollution episodes. Spanish anthropogenic emissions have a small but notable impact on $O_3$ MDA8 concentrations, both positive ($O_3$ formation) and negative (titration of $O_3$ by NO). The largest impacts from Spanish anthropogenic emissions are estimated to come from road traffic, although notable impacts are estimated from industrial combustion, solvent use and NOx emissions from agriculture and waste treatment (burning of agricultural waste). The simulations used to estimate the sectoral-species impacts during pollution episodes provide a detailed analysis of the sectors and emitted species that have the largest impact on these pollution events. For example, the results highlight the importance of NOx emissions from road traffic and industry together with the VOC emissions from the solvents sector in the production of $O_3$ episodes. With regards to the impact of anthropogenic emissions on $PM_{2.5}$ concentrations, the results show that the sector impacts vary spatially. For example, the analysis estimates that most of the impact in Bilbao, in the north of Spain, comes from residential combustion emissions whereas in Madrid (central Spain), the impacts are from multiple sources, including residential combustion, road traffic and agriculture. This has implications for policy since it shows that the success of individual air quality improvement measures will vary geographically.

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