The Influence of Emissions from Maritime Transport on Air Quality in the Strait of Gibraltar (Spain)

Supplementary Materials

Methodology

The methodology followed to perform the study includes the following stages:

- Assessment of current air quality levels based on data recorded at RVCCAA (Monitoring and Control Network of Air Quality of Andalusia) [28] emission stations located in the province of Cádiz.

An assessment of the emission levels of those pollutants under study, measured at air quality stations during the period 2017-2019.

- Presentation of the emission inventory to be considered as the basis for the subsequent simulations.

  The emissions considered correspond to the maritime and port traffic in the area of the Bay of Algeciras and the Strait of Gibraltar for the year 2017.

- Development of a dispersion model:

  For the design of an air quality model, the following steps are followed:

  - Selection of the study area to consider in the pollutant dispersion model;
  - Characterization of the most representative meteorological conditions in the area;
  - Selection of topography and land use;
  - Characterization of emission sources;
  - Definition of points of interest (discrete receptors).

- Analysis of the results of the model in the study area, comparing the resulting contributions with the limit values for pollutants set out in Royal Decree 102/2011 of 28 January on improving air quality).

The RVCCAA stations present in the province of Cádiz are distributed mainly in two evaluation zones, which are called "Algeciras Bay Industrial Zone (ES0104)" and "Cádiz Bay Area (ES0124)". Additionally, there are also stations located in the so-called "Rural Areas (ES0123)" (Figure S1).

Figure S1. RVCCAA stations present in the province of Cádiz.

Tables S1, S2, and S3 show the characteristics of the RVCCAA stations:

Table S1. Characteristics of the air quality monitoring stations "Algeciras Bay industrial area".

Air Quality Monitoring Stations	UTM coordinates (WGS-84, HUSO 30)		Town	Measured parameters
	X (m)	Y (m)		
Algeciras EPS	279,239	4,001,847	Algeciras	SO_2, CO, NO, NO_2, NO_x, O_3, PM_{10}, BCN, TOL, PXY, Met.
E4: Rinconcillo	280,289	4,004,653		SO_2, NO, NO_2, NO_x, PM_{10}, $PM_{2.5}$
E1: Colegio Los Barrios	276,184	4.007,408	Los Barrios	SO_2, NO, NO_2, NO_x, PM_{10}
Los Barrios	276,884	4.006,254		SO_2, NO, NO_2, NO_x, O_3, PM_{10}, $PM_{2.5}$
Cortijillos	280,890	4,007,896		SO_2, CO, NO, NO_2, NO_x, O_3, $PM_{2.5}$, SH_2, BCN, TOL, PXY, EBCN
E5: Palmones	281,205	4,006,069		SO_2, NO, NO_2, NO_x, PM_{10}, $PM_{2.5}$
E6: Estación FFCC San Roque	281,534	4,010,206	San Roque	SO_2, NO, NO_2, NO_x, $PM_{2.5}$
Madrevieja	283,811	4,009,303		SO_2, NO, NO_2, NO_x, $PM_{2.5}$
Guadarranque	283,147	4,006,841		SO_2, CO, NO, NO_2, NO_x, O_3, $PM_{2.5}$, SH_2, BCN, TOL, PXY, EBCN
E3: Colegio Carteya	285,021	4,009,758		SO_2, NO, NO_2, NO_x, O_3, PM_{10},

				$PM_{2.5}$
Escuela de Hostelería	285,698	4,009,196		SO_2, CO, NO, NO_2, NO_x, $PM_{2.5}$
Economato	285,910	4,007,229		SO_2, NO, NO_2, NO_x, $PM_{2.5}$
Puente Mayorga	285,741	4,006,559		SO_2, PM_{10}, $PM_{2.5}$, SH_2, BCN, TOL, PXY
Campamento	286,237	4,006,469		SO_2, CO, NO, NO_2, NO_x, O_3, $PM_{2.5}$, SH_2, BCN, TOL, PXY
E7: El Zabal	289,371	4,005,695	La Línea de la Concepción	SO_2, NO, NO_2, NO_x, PM_{10}, $PM_{2.5}$
La Línea	288,757	4,004,181		SO_2, NO, NO_2, NO_x, O_3, PM_{10}, Met.

Table S2. Characteristics of the air quality monitoring stations "Cádiz Bay area".

Air Quality Monitoring Stations	UTM coordinates (WGS-84, HUSO 30)		Town	Measured parameters
	X (m)	Y (m)		
Jerez-Chapín	221,473	4,064,853	Jerez de la Frontera	SO_2, CO, NO, NO_2, NO_x, O_3, PM_{10}
Cartuja	221,619	4,061,772		SO_2, CO, NO, NO_2, NO_x, O_3, PM_{10}, Met.
Avda. Marconi	207,258	4,045,046	Cádiz	SO_2, CO, NO, NO_2, NO_x, O_3, PM_{10}, Met.
Río San Pedro	211,209	4,046,780	Puerto Real	NO, NO_2, NO_x, O_3, PM_{10}, BCN, TOL, PXY, Met.
San Fernando	212,959	4,039,808	San Fernando	SO_2, CO, NO, NO_2, NO_x, O_3, PM_{10}, Met.

Table S3. Characteristics of the air quality monitoring stations "rural area".

Air Quality Monitoring Stations	UTM coordinates (WGS-84, HUSO 30)		Town	Measured parameters
	X (m)	Y (m)		
Arcos	255,018	4,069,513	Arcos de la Frontera	SO_2, CO, NO, NO_2, NO_x, O_3, PM_{10}, $PM_{2.5}$
E2: Alcornocales	260,630	4,013,178	Los Barrios	SO_2, NO, NO_2, NO_x, O_3, PM_{10}, $PM_{2.5}$
Prado Rey	274,120	4,075,065	Prado del Rey	SO_2, NO, NO_2, NO_x, O_3, PM_{10}, $PM_{2.5}$

1. SENEM MODEL EMISSION

$$P_{transient} = \frac{P_{ref}\left(\frac{t_{transient}}{t_{ref}}\right)^{\left(\frac{2}{3}\right)}\left[\left(\frac{V_{transient}}{V_{ref}}\right)^{n}\right]}{\eta_j \eta_w} \quad \text{(S1)}$$

where

$\eta_w = \text{Speed loss due to wind and waves}$

In the case of fast ferries only, the efficiency of the propulsion system (η_j) was applied by using data from the manufacturers. The remaining parameters and complete procedure are defined in the SENEM model published by the authors of this paper.

The total emissions were calculated using the general equation

Emissions per pollutant(g)= E delivered (kWh) *EF (g/kWh) (S2)

$\text{Total Energy delivered}_{\text{transient}}(\text{kWh}) = (P_{tran.ME} + P_{tran.AE} + P_{boiler}) * t$ (S3)

E delivered = delivered energy for each machine

EF = emission factors

$P_{tran.ME} = Power\ from\ Main\ Engines\ (ME)$, Equation S1

$P_{tran.AE} = Power\ from\ Auxiliary\ Engines (AE)$, Table S6

$P_{boiler} = Power\ from\ boilers$ (Because at main engine loads over 50%, the auxiliary boilers run on exhaust gases, the power delivered by boilers are not considered in this case).

t = time in the study area.

For pollutant emission factor (EF) values, data from ENTEC 2002 [35], Matthias et al., 2010 [20], U.S. Environmental Protection Agency, 2017 [36], and US EPA 2007 [37] were used. For greenhouse gas (GHG) emission factor values, data were used from Starcrest Consulting Group, 2019 [38], Sarvi et al. 2008 [18], IVL 2004 [39], US EPA 2014 [40], and ENTEC 2002 [35].

The types of fuel used by the ships were marine diesel oil (MDO) (30%), residual oil (RO) (62%), and liquified natural gas (LNG) (8%). Emission factors value was used (Table S4).

Table S4. Baseline Emission Factors (g/kWh) for main and auxiliary engines .

Substance emitted		Slow speed		Medium speed		High speed		LNG	
		Main(RO)	Aux(RO)	Main(RO)	Aux(MDO)	Main (MDO	Aux (MDO)	Main	Aux
CO_2 [33]		607	NA	670	707	NA	707	457	457
NO_x	Tier 0 [27]	18.1	NA	14	14.7	NA	11.6	1.3 [34]	1.3 [34]
	Tier I [32]	17	NA	13	13	NA	8.4		
	Tier II [32]	15.3	NA	11.2	11.2	NA	0,2		
SOx *		10.29	NA	11.35	11.98	NA	11.98	0.00269 [35]	0.00269 [35]
PM [29]		1.42	NA	1.43	1.44	NA	1.44	0.03 [38]	0.03 [38]
CO [21]		0.54	NA	0.54	0.54	NA	0.54	1.3 [34]	1.3 [34]
CH_4 [31]		0.012	NA	0.01	0.008	NA	0.008	8.5 [36]	8.5 [36]
N_2O [32]		0.031	NA	0.034	0.036	NA	0.036	0.018 [35]	0.018 [35]
NMVOC [27]		0.6	NA	0.5	0.4	NA	0.4	0.5 [34]	0.5 [34]

Sources: [41] MEPC 63/23 Annex 8, [27] ENTEC 2002, [40] USEPA [2014], [42] Kristensen 2012, [39] IVL 2004, [43] Kunz & Corse, [37] US EPA 2007, [18] Sarvi et al 2008, [44] MARINTEK 2010; * Mass balance.

More information about the SENEM model configuration used in this study can be found in Moreno and Duran (2021) [19].

2. Air Quality Model CALPUFF

The field of study selected for the simulation of the dispersion of the emissions of pollutants from the marine traffic comprises the entire extension of the Strait of Gibraltar, defined by a 146(N-S) x 124 (E-W) km grid (Figure 1).

The CALPUFF modelling system [45] consists of three main components: CALMET, CALPUFF, and CALPOST. CALMET is a meteorological model that develops clockwise wind fields in a three-dimensional mesh that covers the entire simulation domain. CALPUFF is a transport and dispersion model that models puffs of pollutants emitted from the sources considered. CALPOST is a post-processing program that compiles the results obtained by CALPUFF resulting in concentration outflow fields and pollutant deposition flows (Figure 3).

The CALPUFF atmospheric dispersion model is composed of a series of modules that must be completed to develop the model.

2.1. Topography of the Terrain

To reproduce the effect of the topography of the terrain on the behaviour of the plumes, the sea-level elevations of each of the receiving nodes are used. To this end, a digital mesh has been developed with the dimensions of the study area (171 km * 171 km) obtained from a 30 m resolution Digital Terrain Elevation Model (Shuttle Radar Topography Mission -SRTM- 1 Arc-Second Global).

The following meteorological data sources in the study area have been analysed: (a) stations with data available at NOAA (National Oceanic and Atmospheric Administration, U.S.); (b) RVCCA (stations measuring meteorology (Table S5, Figure S2)).

Table S5. Air quality stations that record ozone values.

Air Quality Monitoring Stations	X coord. (m)	Y coord. (m)	Number of valid data	Min.	Max.	Average
Algeciras EPS	279,239	4,001,847	8,598	0.2	134.7	51.4
Avda. Marconi	207,258	4,045,046	8,440	0.8	158.8	73.1
Cartuja	221,619	4,061,772	8,612	0.2	139.0	63.6
Cortijillos	280,890	4,007,896	8,470	4.2	146.3	57.4
E2: Alcornocales	260,630	4,013,178	8,148	12.7	176.8	74.2
E3: Colegio Carteya	285,021	4,009,758	8,525	6.5	154.2	70.2
Guadarranque	283,147	4,006,841	7,906	5.5	222.8	62.6
La Línea	288,757	4,004,181	8,630	1.2	134.8	64.5
Prado Rey	274,120	4,075,065	8,221	30.8	174.2	86.0
San Fernando	212,959	4,039,808	8,322	2.5	139.7	70.9

Table S6. Main and auxiliary engines power and load factors by ship type in cruising mode.

Type	ME Power (kW)	AE Power (kW)	Relation AE/ME	Load Factor (%)
Car	10,700	2,850	0.266	24%
Bulk Carrier	8,000	1,776	0.222	22%
Containers	30,900	6,800	0.220	17%
Passenger	39,600	11,000	0.278	64%
General Cargo	9,300	1,776	0.191	22%
Miscellaneous	6,250	1,680	0.269	22%
RORO	11,000	2,850	0.259	30%
Reefers	9,600	3,900	0.406	34%
Liquid Cargo	9,400	1,985	0.211	67%

Source: Shore Power Technology Assessment at U.S. Ports.

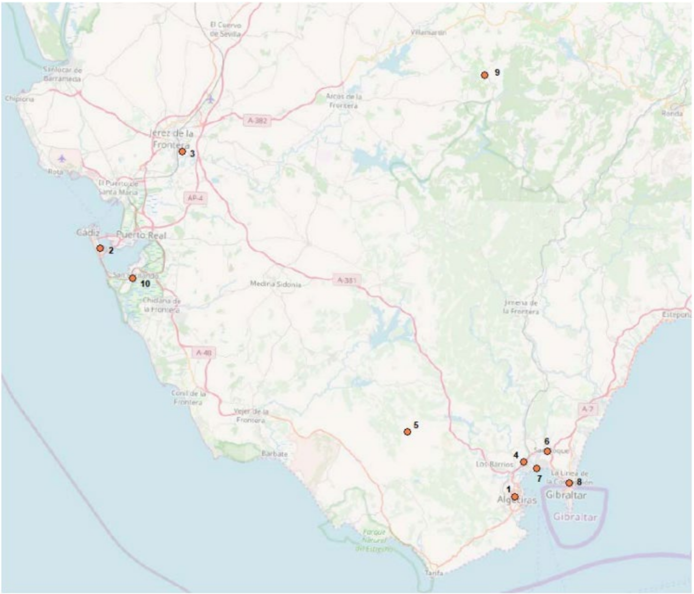

Figure S2. Ozone station locations.

The definition of the area/line sources in the CALPUFF model requires the following parameters to be determined:

- Dimensions of the area source/line-area.

- Emission rate (g/s m²).

- Height of emission focus (m).

- Initial height of plume (m).

To determine the initial height of the tuft, the Holland equation is used (equation normally used to determine the plume rise from a given stack height).

$$\Delta h = \frac{V_s}{u} * d \left[1.5 + 2.68 * 10^{-3} * p * \frac{T_s - T_a}{T_s} * d\right] \quad \text{(S4)}$$

Δh: elevation of the plume above the emitting source (m)
Vs: gases emission rate (m/s)
d: inside diameter of the emission line (m)
u: wind speed (m/s)
p: air pressure (mbar)
Ts: outlet gas temperature (K)
Ta: ambient temperature (K)

The values of Δh obtained with the formula should be corrected by multiplying by a factor, established by Pasquill-Gifford-Turner [46], which is a function of the meteorological conditions.

The Δh correction factor varies from 0.85 to 1.15 depending on levels of stability.

In summary, the Holland Formula was applied for every hour of 2017, considering the wind speed and ambient temperature corresponding to each hour and correcting in each case according to the class of hourly stability. Finally, an average annual plume rise value is calculated.

On the other hand, for point sources, the parameters to consider in the dispersion model are:

Emission source location coordinates (X, Y in m)

- Ground level at that point (m)

- Chimney height (m)

- Chimney diameter (m)

- Gas outlet speed (m/s)

- Gas outlet temperature (K)

- Pollutant emission rates (g/s)

The starting parameters for the definition of the above sources are as follows:

Characteristics of the vessels	Cruise	Others
Number of chimneys	4	1
Chimney height (m)	35-50	20

Diameter(m)	0.6	0.8
Exhaust gas flow (kg/s)	2.4-2.5	2.2-2.3
Output speed (m/s)	10.7-11.2	25
Temperature (ºC)	175	282
Pressure (kPa)	101.3	101.3

2.2. Chemical Reaction Mechanism

The chemical transformation mechanism RIVAD/ARM3 [48], included in the CALPUFF pollutant dispersion model, is used for the modelling carried out in the present study.

RIVAD/ARM3 is a mechanism of six pollutants: SO_2, SO_4^{2-}, NO, NO_2, HNO_3 and NO_3, which considers the following transformations, among others:

- Conversion from SO_2 to SO_4^{2-} (aerosol) - Conversion from NOx (NO + NO_2) to NO^{3-} (aerosol).

With this mechanism, sulfate and nitrate formation rates are estimated from the concentration of hydroxyl radical (OH^-), which is the first oxidant of SO_2 and NO_2, assuming that it is in a stationary state.

The application of the RIVAD/ARM3 mechanism requires data on existing ozone concentrations in the study area for the year to be modelled. To this end, a file is used with the hourly ozone concentrations recorded at the following air quality measurement stations, distributed throughout the study area (Table S5)

2.3. Procedure for Quantifying Pollutant Emissions

- SO_2:

- Annual average;

  99.73 percentile of hourly average values;

  99.18 Percentile of daily average values.

- NO_x/NO_2:

- Annual average NO_x;
- Annual average NO_2;
- Percentile 99.79 of hourly average values.

- CO:

- Maximum daily moving octohorary averages.

- PM_{10}:

- Annual average;
- Percentile 90.41 of hourly average values.

- $PM_{2,5}$:

- Annual average.

- HC, NMVOC, and CO_2:

- Annual average.

For HC and NMVOC, no limit value is established in Royal Decree 102/2011. Therefore, the limit value established for the annual average of benzene will be used as a reference. For CO_2, as the main references for concentrations in the atmosphere are monthly average values, the annual average shall also be extracted for assessment**.**

3. Meteorological Model

The initial wind field generated by the Weather Research and Forecasting (WRF). The meteorological model is shown in Table S7.

Table S7. Specification WRF model

Model specification - WRF model	
Period	Year 2017
Domain area	171 km * 171 km
Resolution	3 km
Number of vertical levels	18

Digital Elevation Model (DEM)	GTOPO30
Land Use, Land Cover model (LULC)	USGS GLCC (1000m)
Altitude data	Every 6 hours
Start time	00:00, 06:00, 12:00, 18:00 UTC
Lateral boundary and initialization FNL	Operational Global Analysis data (1-degree) NCEP (Final)
Data output frequency	Every hour

The study area selected for the simulation of the dispersion of pollutant emissions consists of an area measuring 146 km N-S and 124 km E-W (Figure 1).

The application of the CALPUFF dispersion model requires the definition of emission sources in the scope of study.

For the simulation of the dispersion of emissions from maritime traffic, the definition of emission sources (ship chimneys) as area or line sources is considered appropriate, as these are emission sources moving along the different established navigation routes. In this case, the model assumes that the emissions generated by the ships are emitted in a uniform quantity throughout the area or line source.

In contrast, for the sources associated with emissions in the Port of Algeciras, the point emission sources are more adapted to reality according to the tests carried out to adjust the dispersion model.

Therefore, for emissions associated with the Strait of Gibraltar and the Port of Ceuta Route, area sources are used, while the Tarifa–Tangier Route is represented with a line-area source and the Port of Algeciras with point sources, as seen in Figure S3.

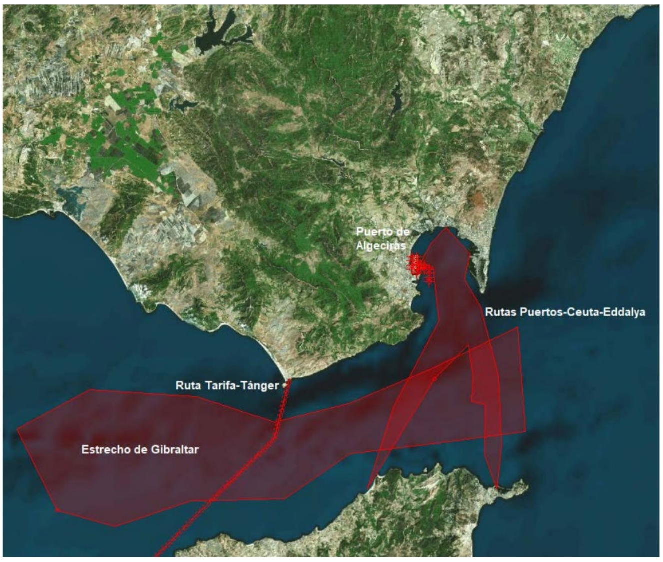

Figure S3. Routes and line points studied.

Data from Table S7 and Figure S4 shows the input data for the air quality (CALPUFF) model. The meteorological data for 2017 were generated by the "Red de Vigilancia y Control de la Calidad del Aire de Andalucía (RVCCAA)". The temperatures in the area varied according to the season. For example, the spring was the only period in which the Strait experienced a prolonged period of high pressure. To support the results of the present study, model output PM_{10}, $PM_{2.5}$, NO_x, and SO_x concentrations were compared with data from the monitoring stations of RVCCAA. Annual mean concentrations were observed at 91 stations for all the pollutants.

Figure S4. Location of the weather stations considered in the dispersion model.