Using Air Quality Modelling to Improve Air Emission Inventories

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ABSTRACT

Today emission inventories are one of the key components of air quality management. For instance, inventories are supportive for: the modelling of atmospheric pollutants; the process of compliance verification of national emission obligations; and the establishment of baseline scenarios for the development and monitoring of the policies and measures. Therefore, emission inventories should be developed accordingly to high reliable standards and should be validated using appropriate methodologies, guaranteeing that they appropriately may be used to predict air quality when coupled to models.

In Europe, one of the main goals of national inventories has been to verify that countries are complying with their international commitments at national level, such as those set by UNFCCC and the Kyoto Protocol; CLRTAP and UE-NEC Directive; and the Stockholm Convention. In Portugal, efforts were also been made to integrate the total emission estimates at national level, with estimates made for a detailed level of spatial allocation of emissions, and using a suitable temporal disaggregation of emissions. In order to conclude for the best disaggregation level to be used to the Portuguese national system, an air quality modelling system was used to tested several emission inventories resolution schemes available for Portugal (INERPA, EMEP and LOTOS) and further disaggregation schema at sub-municipal level. Evaluations were done using the CHIMERE model, forced by the MM5 meteorological fields, over the Portuguese domain. Results from this exercise show the existence of discrepancies between different disaggregation instances for emission inventories. Different temporal profiles were also tested, indicating that there this has strong influence on results, and confirming that the use of average European profiles may not be appropriate at national nor regional level.

The results of this analysis lead to the improvement of emission inventories in Portugal and the quality of results from air quality models.

INTRODUCTION

Inventories of air emissions, providing estimates for emissions by sources and removals by sinks, are one of the vital components of the environmental decision-making process that must be used to improve air quality. Saying how much pollutants or green-house gases are emitted, when and how, is delivering information suitable to:

- Verify the compliance of national and international obligations. Presently this is one very relevant application of the inventories, given the various submissions with which countries are usually involved: (1) CLRTAP, the Convention on Long-Range Trans-boundary Air Pollution; (2) the UNFCCC, the United Nations Framework Convention on Climate Change and its subsidiary Kyoto Protocol; (3) the Stockholm Convention on Persistent Organic Pollutants (POP) and the Directive of National Ceilings of the European Union (EU);
- Atmospheric modelling of pollutant dispersal and deposition, including air quality assessment;
- Establishment of baseline scenarios for the identification and definition of policies and measures.

Hence, emission inventories must be as accurate as possible in order to provide a solid base for air quality decision-making. They must also be complete, covering all emissions. In the case of air quality modelling, the definition of appropriate spatial allocation of emissions and time variation patterns is also a fundamental factor. Notwithstanding these principles, the air quality modelling community is generally faced with considerable uncertainties in the emission inventories used for modelling, and these are responsible for part of the errors in the model results. This situation is of very significant relevance, because these deficiencies will be reflected in the wrong choice of policies and measures and, in last instance, into misuse of resources.

Looking to this situation in a different angle, the analysis of the use of certain inventory instances in combination with air quality models, may be very helpful to diagnose the uncertainties on emission estimates.

In Portugal the National Inventory, referenced as INERPA, is under the responsibility of the Portuguese Agency for the Environment (*Agência Portuguesa do Ambiente*, APA). APA is in charge of the annual national emission inventory, which is sent to the UNFCCC, to the UN-ECE and to the European Union (EU) according to international commitments. The obligations under CLRTAP require Portugal, like other parties to that convention, to submit estimates allocated spatially and using a large grid (50x50 km), suitable for an European scale. Besides, after being routinely asked to deliver spatial data for national requirements, namely the request of information for the Evaluation of Impacts in the Environment, APA has decided to produce a spatial disaggregation of emissions at more detail, i.e. municipal level[1]. Since late 2005 efforts have been made, jointly by APA and *Universidade de Aveiro* (UA), with the objective to analyse the national inventory (INERPA), and possibly improve it through the development of a better emission model with a more suitable spatial allocation of emissions and the development of a suitable temporal disaggregation of emissions per sector. Apart from the information available at national level, other sources of inventory estimates and disaggregation are available in Europe for Portugal, like EMEP[2] and LOTOS[3].

Therefore, the main goal set in this paper was to verify what differences in accuracy result from the different disaggregation schema, identifying the key factors of uncertainty and, finally, trying to conclude ways to improve the Portuguese inventory and the advantage it may provide, together with air models.

BODY

Methodology

Air quality modelling applications were performed using the MM5-CHIMERE modelling system[4], over mainland Portugal, with a 10 x 10 km^2 grid. The simulations were carried out using meteorological information data for the summer of 2004 (1 June - 31 September), regarding gaseous and particulate pollutants. Comparative testing was performed for different available emission inventory disaggregations (INERPA, EMEP and LOTOS).

A brief description of the modelling system, together with the each emission inventories and the respective modelling application is presented in the following chapters.

<u>The modelling system</u>

The air quality modelling system is composed by the chemistry-transport model CHIMERE, forced by the MM5 meteorological fields[4].

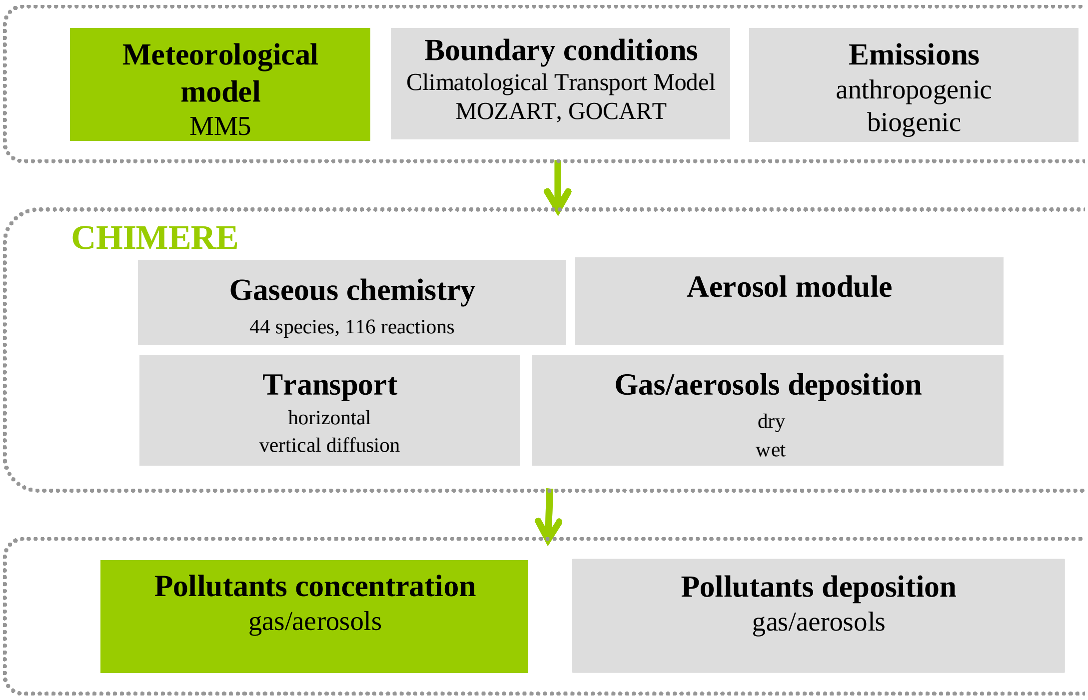
CHIMERE, the 3D chemistry transport model, based on the integration of the continuity equation, was developed specifically for the simulation of gas-phase chemistry, aerosol formation, transport and deposition at European and urban scales. CHIMERE has been used extensively in several research applications, including: sensitivity studies concerning anthropogenic or biogenic emissions[5,6]; emission diagnostics[7], and photo-oxidant forecasting over Europe and the Paris region[8]. This modelling system has been also used for real-time air quality forecasting over Portugal[9]. The model version that was used in this work was originally described in Schmidt et al. (2001)[10] while further updates that were applied especially to smaller-scale versions[7].

Meteorological input variables given by the MM5 model are: 3D fields of horizontal wind, temperature, specific humidity, cloud liquid water content, and 2D fields of surface pressure, heat fluxes, 2 m temperature and cloud cover. They were linearly interpolated to the CHIMERE grid. Linear time interpolation was also used to obtain hourly values.

Besides meteorological input data, CHIMERE model requires definition of boundary and initial conditions, emission data and the land-use and topography characterization.

Figure 1 presents a simplified scheme of the MM5-CHIMERE modelling system and its inputs/outputs.

Figure 1. The MM5-CHIMERE air quality modelling system



Emission Inventories in Portugal

The Portuguese Agency for the Environment has been responsible for the elaboration of the national inventory (INERPA)[11], relying on activity provided by several official institutions, according to the legal bindings institutionalised by the National System. In that sense, emission values from these inventories are considered official data and are used to verify the accomplishment of international obligations such as:

(1) UN-ECE's Convention on Long-Range Trans-boundary Air Pollution (CLRTAP) is preoccupied with several atmospheric pollutants such as SO_x, NO_x, VOC, NH_3, POP and Heavy Metals, and provides information for the EMEP model;

(2) UNFCCC, the United Nations Framework Convention on Climate Change and its subsidiary Kyoto Protocol, dealing with all obligations that developed countries are committed to tackle global warming, is one of the most salient international forum in what concerns the environment. It is related with carbon dioxide (CO_2), Methane (CH_4), Nitroux Oxide (N_2O) and fluoride gases not covered by the Montreal Protocol (PFCs, HFCs and SF_6);

(3) Stockholm Convention on Persistent Organic Pollutants (POP), concerning unintentional emissions such as Dioxins and Furans;

(4) the European Union's National Ceilings Directive, concerning SO_x, NO_x, NMVOCs and NH_3.

At European level other obligations require the compilation of emission data, but these refer mostly to individual units. These are the Large Combustion Plants Directive (LCPD), the European Carbon Trading Scheme (EU-ETS) and the EPER/E-PRTR.

Of all international obligations only CRLTAP requests submission of data with spatially disaggregated, requiring emissions estimates to be provided at 50x50 km grid and individual emissions to be provided to Large Point Sources. However, following frequent requests by teams involved in Environmental Impact Assessment studies, APA has decided in 2006 to elaborate detailed disaggregation of emissions. Accordingly, total emissions per source category were allocated to small territorial units (municipalities) using surrogate indicators such as fuel sales, human population, agricultural area and livestock population.

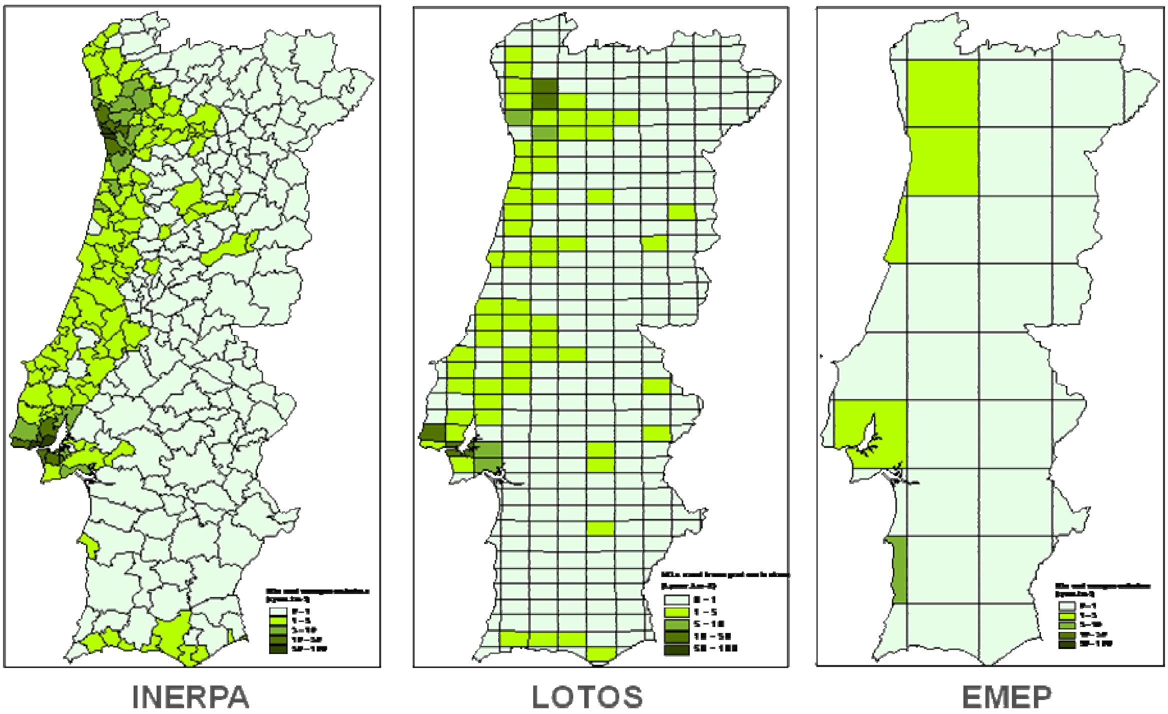
The EMEP emission inventory that was used was obtained through the interactive EXPERT database available via Internet, last updated in June 2006. EMEP provides emission estimates for total emissions as well as data for each specific sector. The gridded emissions that are provided at a regular grid covering all Europe (0.5º x 0.5º long-lat), include partial substitution of national data made by the EMEP Centres (Meteorological Synthesizing Centres - West and East).

The LOTOS emission inventory results from a combination of the TNO emission database (with high resolution: 0.25º x 0.125º long-lat) and the CAFE baseline emissions for 2000. For each source category and each country, the country totals of the TNO emission database are scaled to those of the CAFE baseline emissions. PM emissions for 2000 are assumed to result from CEPMEIP project (derived for 1995), considering that the uncertainty in the emission estimate is much larger than the trend in the PM emissions. This database does not specify the composition of the emitted particles. Therefore, black carbon emissions were derived from the primary PM2.5 emissions and it was assume 2 per cent of the SO_2 emissions to be emitted as particulate sulphate.

All these emission inventories refer the same pollutants species: NOx, VOC, SO_2, NH_3, CO, PM2.5 and PM10. Large point source emissions were not included in modelling applications, because not all the inventories integrated in this study had data individualized for these units.

Figure 2 shows, as an example, NOx emission data from road transport provided by each available emission inventory.

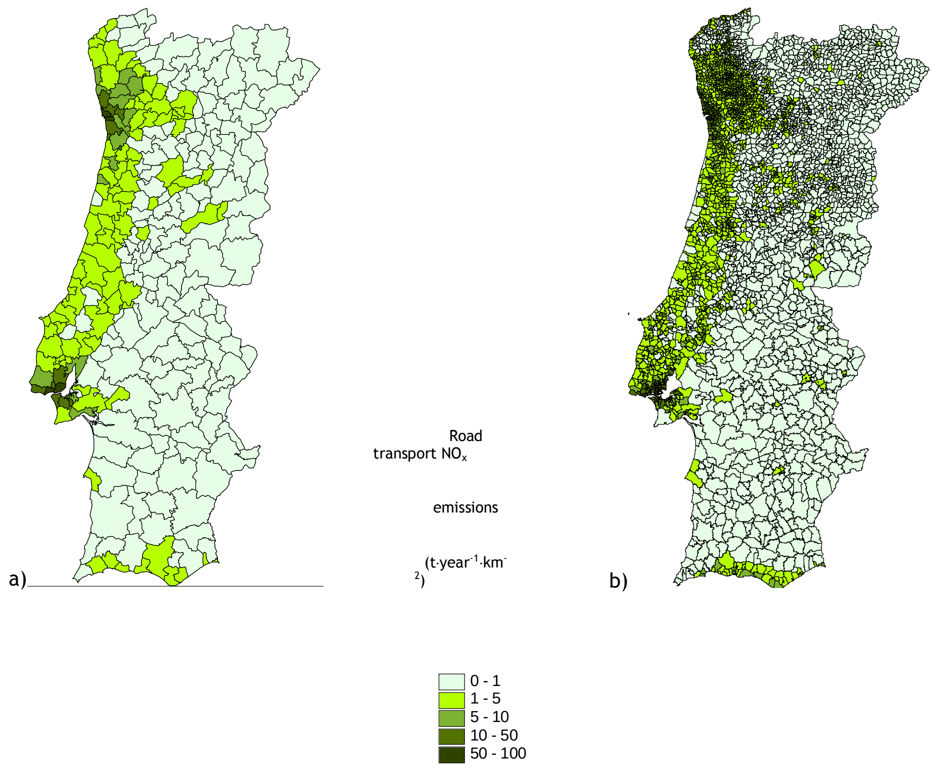
Figure 2. Spatial distribution of NOx emissions from road transport for the three available inventories (INERPA, LOTOS and EMEP).



Spatial disaggregation was applied for each inventory separately, applying an interpolation method and GIS software to the original inventory grid in order to obtain a regular grid of 10 x 10 km^2, according to the simulation domain resolution.

Concerning the INERPA inventory, two different levels of spatial disaggregation were considered: the municipality, available in the original spatialisation, and the sub-municipality. This last one was obtained by further disaggregating the municipality level by population (census) data (see Figure 3).

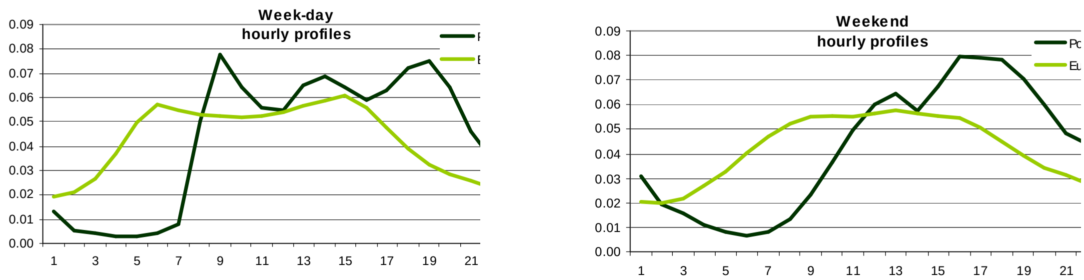
Figure 3. Spatial distribution of NOx emissions (2003 year) from road transport at municipality scale (a) and at sub-municipality scale (parish level) (b).



In all situations a common methodology[12] was used to calculate biogenic emissions with the CHIMERE model.

Two different hourly profiles were applied to the road transport emissions within the INERPA inventory simulation: a European averaged profile and an urban hourly profile obtained in field measurements made in Portugal The European was obtained by application of monthly, weekly and hourly profiles from the University of Stuttgart[13]. A road traffic urban hourly profile measured in a field campaign in Portugal in the scope of the SAPPHIRE European Project (EVK4-2002-00089) was also tested.

Figure 8. Comparison of the European and Portuguese road transport hourly profiles, for weekdays and weekend.

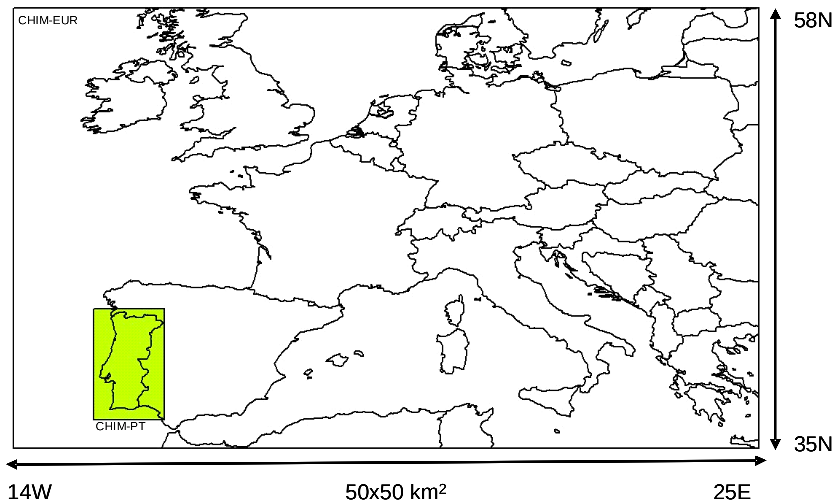


Description of Simulations

The model was applied first to the european continental-scale, using the 50x50 km^2 resolution, and then in detail to Portugal, as shown in Figure 4, using the same physics and a simple one-way technique. The second simulation (Portugal domain) was performed with a horizontal domain of 290 km x 580 km and a 10 km horizontal resolution, the vertical grid consisting of 6 hybrid sigma-pressure layers with a model top at 700 hPa. The top altitudes of the layers evolve in time, but their approximate values are, from bottom to top: 50, 250, 600, 1200, 2000 and 3000 m. Lateral and top boundaries for the large-scale run are obtained by the GOCART climatological model[4].

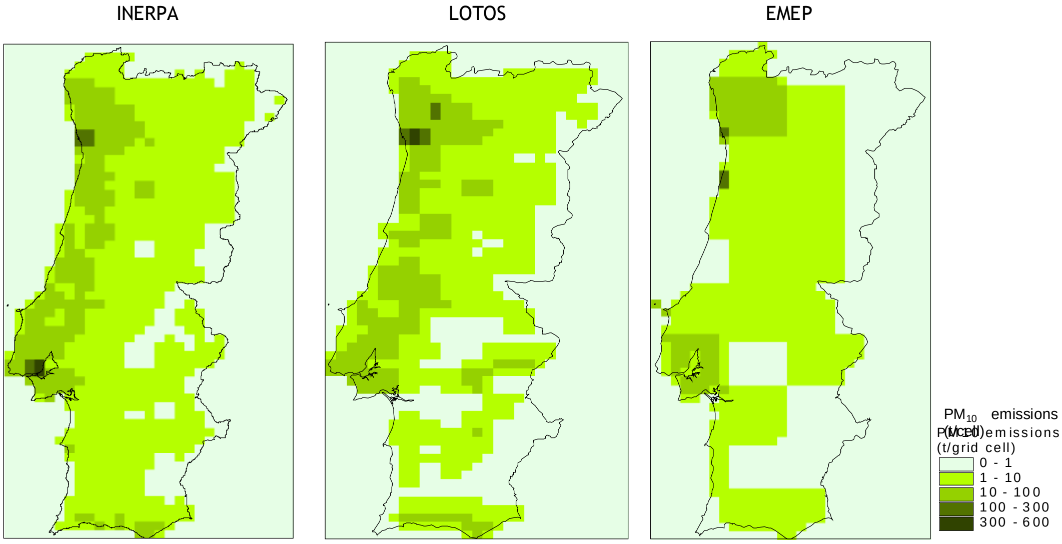
Using the same large-scale simulation (with EMEP emissions) as boundary conditions, several applications were performed for the Portugal scale in order to test the different emission inventories with their distinct spatial disaggregation, and also the temporal profiles used for time disaggregation.

Figure 4. European and Portuguese domains used by the air quality modelling system.



In order to be used by numerical models, emission data from the three sources had to be converted to a regular grid according to the spatial resolution defined. This process was performed using the "Kriging" method and Geographical Information Systems. Figure 6 shows the result of this interpolation process and the obtained grid (with 10x10 km^2) for the PM_{10} emissions for the three inventories (INERPA, EMEP and LOTOS).

Figure 6. Example of the PM_{10} emissions grid conversion (10x10 km^2) for the three available inventories (INERPA, LOTOS and EMEP), necessary for model simulation.

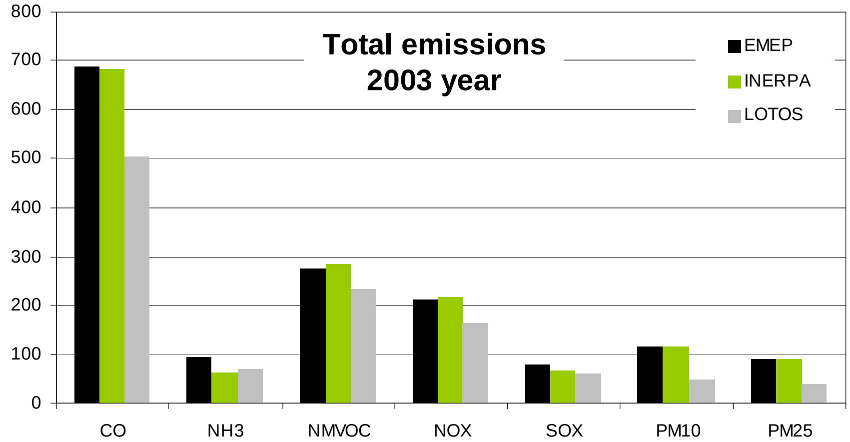


Results and Discussion

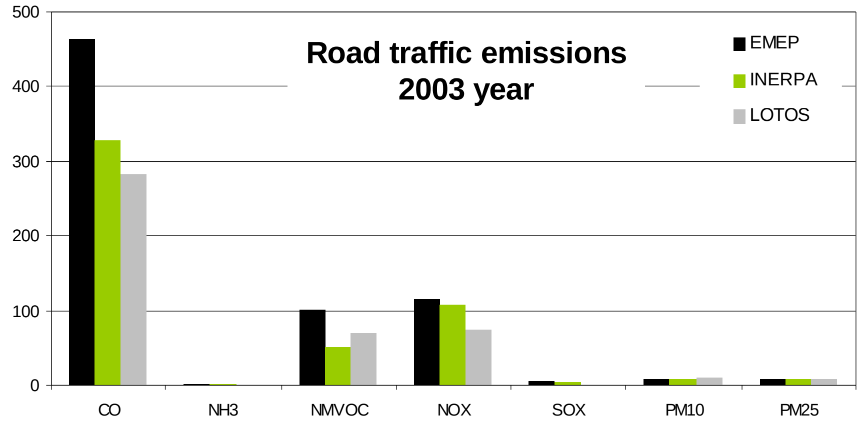
<u>Comparison of Total Emissions</u>

In Figure 5a it is presented for each pollutant the comparison between the total emission values (sum of all area emission source activities) estimated by each inventory, considering only mainland Portugal area. The major differences were obtained for the LOTOS database, which estimates are significantly lower than those referring to EMEP and INERPA (20-30 per cent), mainly regarding PM emissions (these differences reach 50 per cent). The similarity between the EMEP and INERPA totals was expected, given that EMEP results from INERPA, with small corrections and revisions made by MSC-W experts over the officially national data. Individual analysis by sector activity shows that the major discrepancies between inventories are registered for road transport (more than 30 per cent), as shown as example in Figure 5b.

Figure 5. Comparison of the total (a) and road transport (b) emissions for each inventory, by pollutant specie.



a)

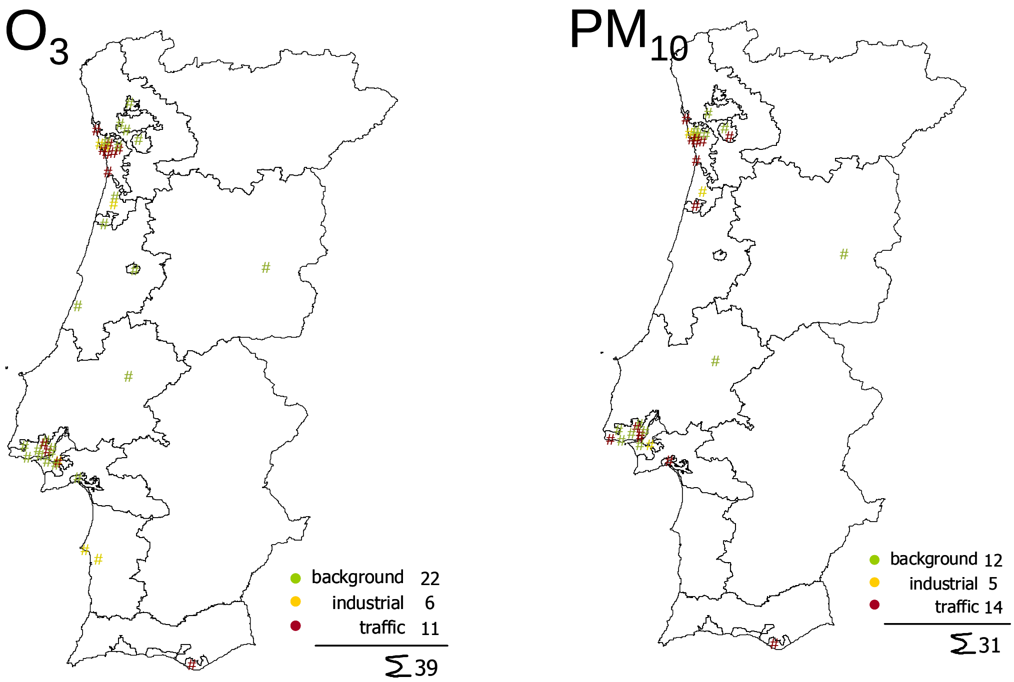


b)

Modelling results

Results from modelling were compared to monitoring data available from the air quality networks, which comprehends 39 stations for O_3 (22 background and 11 traffic) and 31 stations for PM_{10} (12 background and 14 traffic). These stations are located mainly in the urban area of Porto and Lisbon, with a few rural stations outside these areas (Figure 7).

Figure 7. Spatial location of the air quality monitoring stations (background, industrial and traffic) used for model validation, for O_3 and PM10.



Before testing of emissions inventories, a sensitivity test to the spatial disaggregation of INERPA national inventory, using the original municipality values and its disaggregation to sub-municipality degree was performed. The statistical analysis is presented in Table 1, for background stations, which are more representative of the used model grid (10 x 10 km^2). Concerning O_3, the Root Mean Square Error (RMSE) and the systematic error (BIAS) analysis suggests that the further spatial disaggregation performed introduced more errors to the emission inventory, namely in what concerns urban area where the RMSE and BIAS differences are bigger. On the other hand, regarding PM_{10}, there are no significant differences between results from both inventories. The BIAS analysis indicates that when the sub-municipality spatial disaggregation is applied, the rural and suburban PM emissions decrease, in opposition to the increase in urban areas (where more population exists).

Since there is an overall tendency for emissions underestimation (bias negative), it is expectable that model performance could be reduced in rural areas and improved in urban zones, when the INERPA disaggregated inventory is used.

Table 1. Validation of CHIMERE simulations, using different spatial disaggregation of INERPA inventory, and considering the average of each background station type.

	O$_3$ (μg.m^{-3})				PM$_{10}$ (μg.m^{-3})			
Station zone	RMSE		BIAS		RMSE		BIAS	
	INERPA disaggregated	INERPA original	INERPA disaggregated	INERPA original	INERPA disaggregated	INERPA original	INERPA disaggregated	INERPA original
Rural	28.88	25.32	-13.24	-9.58	16.81	16.71	-13.13	-13.01
Suburban	33.12	29.00	-12.70	-8.93	34.85	37.15	-9.82	-8.80
Urban	37.26	24.71	-23.19	-4.70	25.54	25.84	-7.15	-8.31

$$\text{bias} = \frac{1}{N}\sum_i (M_i - O_i)$$

$$\text{RMSE} = \frac{1}{N}\sum_i |(M_i - O_i)|$$

N is the number of samples, Oi are observations and M_i are model predictions

Tables 2 and 3 show the statistical parameters obtained for the three emission inventory simulations, for O_3 and PM_{10}, respectively.

For both pollutants, there are no significant discrepancies between the RMSE values for each inventory application, indicating that higher resolution in the emission inventory do not necessarily result in better model performance.

The negative values of BIAS suggest that there is an overall tendency for underestimation of ozone precursors and PM emissions. In fact, the more negative bias is attributed, in both Tables, to the LOTOS inventory that presents the lower emission values (see Figure 1), in opposition to INERPA that shows the higher emissions for NO_x, VOC, and PM, and consequently inferior bias. Nevertheless, regarding PM_{10} results, there is a particular overestimation over Porto region (LEC, VNT, ERM stations) when the INERPA (original) inventory is used. Moreover, for this particular area, the absolute errors are also higher and the correlation coefficient lower comparatively to the others emissions inventory, indicating spatial disaggregation problems.

The similar correlation factor (CF) found for the three applications, and regarding O_3 and PM_{10} results, can be explained by the same temporal emission disaggregation (seasonal, diurnal and hourly) applied to each annual inventory.

Table 2. Statistical parameters obtained for each simulation, for O_3 background stations.

				RMSE ($\mu g.m^{-3}$)			BIAS ($\mu g.m^{-3}$)			CF (correlation factor)		
Zone	Long	Lat	Station	EMEP	LOTOS	INERPA*	EMEP	LOTOS	INERPA*	EMEP	LOTOS	INERPA*
Rural	-8.6	40.8	AVA	25.57	29.99	24.13	-7.91	-12.49	-6.04	0.81	0.79	0.82
Rural	-8.5	39.3	CHA	31.77	33.72	30.22	-15.4	-19.71	-14.3	0.65	0.67	0.69
Rural	-8.9	39.9	ERV	18.4	19.48	18.4	2.84	1.43	2.08	0.76	0.73	0.76
Rural	-7.3	40.2	FUN	28.01	29.63	28.54	-19.52	-21.61	-20.06	0.64	0.63	0.63
Suburban	-8.5	41.3	CAL	23.11	23.81	24.45	-10.32	-10.92	-11.93	0.86	0.87	0.86
Suburban	-8.7	40.6	ILH	20.12	21.57	20.19	1.49	-1.12	1.52	0.8	0.78	0.79
Suburban	-9.1	38.6	PP	25.24	23.7	25.81	-8.57	-8.35	-0.48	0.73	0.77	0.72
Suburban	-8.6	41.3	VNT	24.58	25.19	25.72	5.55	7.18	-2.91	0.66	0.66	0.6
Urban	-8.9	38.5	ARC	24.42	22.4	26.18	2.52	-3.57	2.65	0.68	0.69	0.65
Urban	-9.1	38.7	BEA	21.82	21.17	23.1	-2.42	-0.3	2.99	0.72	0.73	0.72
Urban	-8.5	41.2	ERM	24.99	24.53	26.24	-3.59	0.94	-7.1	0.75	0.78	0.74
Urban	-8.4	40.2	IGEO	22.86	26.18	22.47	-8.79	-12.61	-8.53	0.8	0.78	0.81
Urban	-9.2	38.7	LAR	23.09	22.01	23.67	-5.56	-1.12	1.15	0.7	0.71	0.7
Urban	-8.4	41.3	LAT	32.35	34.03	33.45	-20.4	-21.6	-20.98	0.84	0.84	0.83
Urban	-9.2	38.8	LOU	21.72	23.06	22.78	-2.38	-3.81	-1.43	0.74	0.71	0.71
Urban	-9.3	38.7	MARQ	22.9	23.34	22.04	-3.53	-0.13	-1.7	0.71	0.69	0.73
Urban	-9.3	38.8	MEM	19.81	19.88	19.31	-1.19	-0.78	0.08	0.71	0.7	0.73
Urban	-9.1	38.8	OLI	22.75	22.94	24.03	-0.79	-1.86	3.48	0.72	0.73	0.72
Urban	-9.2	38.7	REB	24.12	23.56	23.52	-7.59	-4.45	-6.06	0.71	0.7	0.71
Urban	-9.2	38.7	RES	31.23	28.38	28.62	-18.15	-11.47	-15.1	0.64	0.62	0.68
Urban	-8.5	41.3	STIR	24.86	25.94	25.79	-9.15	-10.13	-10.57	0.87	0.88	0.87
Average				**24.46**	**24.98**	**24.7**	**-6.33**	**-6.5**	**-5.39**	**0.74**	**0.74**	**0.74**

*original

Table 3. Statistical parameters obtained for each simulation, for PM_{10} background stations.

				RMSE ($\mu g.m^{-3}$)			BIAS ($\mu g.m^{-3}$)			CF (correlation factor)		
Zone	Long	Lat	Station	EMEP	LOTOS	INERPA*	EMEP	LOTOS	INERPA*	EMEP	LOTOS	INERPA*
Rural	-7.3	40.2	FUN	20.48	20.65	20.46	-16.11	-16.28	-16.13	0.56	0.55	0.58
Suburban	-8.5	41.4	CAL	28.29	29.76	26.48	-23.73	-25.28	-21.82	0.65	0.65	0.67
Suburban	-8.6	41.2	LEC	25.65	23.95	40.23	-21.28	-19.61	**33.86**	0.79	0.78	0.75
Suburban	-8.7	41.3	VNT	26.12	25.16	33.74	-23.13	-22.15	**15.86**	0.75	0.75	0.7
Urban	-8.6	41.2	ERM	25.82	24.76	38.37	-21.62	-20.53	**37.31**	0.69	0.69	0.66
Urban	-9.2	38.7	LAR	24.9	29.85	18.96	-18.08	-23.62	-9.38	0.65	0.65	0.66
Urban	-8.4	41.3	LAT	28.67	29.87	24.19	-24.06	-25.25	-19.05	0.76	0.76	0.67
Urban	-9.2	38.8	LOU	21.95	26.91	21.68	-16.77	-22.34	-16.3	0.62	0.62	0.63
Urban	-9.3	38.7	MARQ	25	28.24	22.85	-18.45	-22.24	-15.16	0.48	0.45	0.48
Urban	-9.4	38.8	MEM	15.7	18.15	14.98	-12.51	-15.62	-11.36	0.58	0.62	0.6
Urban	-9.1	38.8	OLI	20.43	20.43	20.43	-15.18	-15.18	-15.18	0.86	0.86	0.86
Urban	-9.2	38.8	REB	29.33	34.68	27.29	-20.36	-26.85	-17.35	0.52	0.52	0.52
Average				**24.36**	**26.03**	**25.81**	**-19.27**	**-21.25**	**-4.56**	**0.66**	**0.66**	**0.65**

*original

Concerning the 2 hourly profiles that were applied to the road transport emissions for the INERPA emission inventory: an European averaged profile and a Portuguese urban hourly profile, Table 4 shows the correlation factor and errors found for each profile model application. There are no differences found concerning background stations, but there is an improvement in model performance, regarding traffic sites, when the Portuguese (measured) road transport profile is used.

Table 4. Validation of CHIMERE simulations, using INERPA inventory, with different Hourly Profiles (HP), considering the average for background and traffic stations.

	O_3				PM_{10}			
	CF		RMSE ($\mu g.m^{-3}$)		CF		RMSE ($\mu g.m^{-3}$)	
Station zone	European HP	Portugal HP	European HP	Portugal HP	Euroopean HP	Portugal HP	European HP	Portugal HP
Background	0.74	0.74	25.93	25.80	0.66	0.66	27.05	27.00
Traffic	0.64	0.65	25.55	25.12	0.63	0.66	29.95	29.48

CONCLUSIONS

Considering the widespread use of air emission inventories for policymaking, planning, and research purposes, it is nowadays important to continuously assess the quality of the inventories, to identify any shortcomings and prioritise for its improvement. The work presented here intends to perform an evaluation of the emission inventories for Portugal, through a validation procedure using air quality modelling. Efforts were done to identify any weakness and strengths in the inventory and to detect the key sources of uncertainty that can be targeted for reduction via additional data collection and research.

The evaluation model exercise, comparing results against observed data, shows that there are no significant discrepancies between the direct applications of the three emission inventories. This similarity, already verified on the total emission values, proves that higher resolution in the emission inventory do not mean necessarily better model performance. However, model results for all cases indicates that there is an overall general tendency for emissions underestimation that could be emphasised by the point sources omission. This underestimation is more notorious with the LOTOS inventory, and less with INERPA, which presents, on average, less systematic errors. Nevertheless, the range of uncertainty varies with locals and pollutants.

Analysis of results provided further clues for improving emission inventories. It was found, for instance, that there is a probable overestimation of the INERPA inventory concerning emissions of particulate matter on Porto region, and the use of a less appropriate spatial disaggregation could be the reason for that situation.

Sensitivity tests with road traffic temporal profiles shows that the exact pattern has a certain influence on the air quality results, in what concerns traffic and urban stations. The Portuguese average profile was shown to be more appropriate for these specific areas.

The spatial disaggregation performed over the original INERPA, performing a further disaggregation, from municipality to the sub-municipality level and using population census data as surrogate variable, shows that model results do not necessarily improve when higher-resolution emission data is developed, indicating that spatial disaggregation of an emission inventory should be performed carefully, searching for representative indicators or, otherwise could this could be an additional source of uncertainty.

Planned future work for this project will involve testing this methodology with other air quality modelling systems and analysing each emission source category. The development and assessment of an emission inventory ensemble will also be the focus of future work.

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KEY WORDS

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