



Impact of forest fires on particulate matter and ozone levels during the 2003, 2004 and 2005 fire seasons in Portugal

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ABSTRACT

The main purpose of this work is to estimate the impact of forest fires on air pollution applying the LOTOS-EUROS air quality modeling system in Portugal for three consecutive years, 2003–2005. Forest fire emissions have been included in the modeling system through the development of a numerical module, which takes into account the most suitable parameters for Portuguese forest fire characteristics and the burnt area by large forest fires. To better evaluate the influence of forest fires on air quality the LOTOS-EUROS system has been applied with and without forest fire emissions. Hourly concentration results have been compared to measure data at several monitoring locations with better modeling quality parameters when forest fire emissions were considered. Moreover, hourly estimates, with and without fire emissions, can reach differences in the order of 20%, showing the importance and the influence of this type of emissions on air quality.

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1. Introduction

Smoke is one of the most disturbing consequences of forest fires. Its impact on air quality and human health can be significant as individuals and populations are exposed to hazardous air pollutants (Coghlan, 2004). During fires large amounts of chemical compounds, such as carbon monoxide (CO) and dioxide (CO₂), methane (CH₄), nitrogen oxides (NO_x), ammonia (NH₃), particulate matter (PM), and non-methane hydrocarbons (NMHC), are emitted into the air and interfere in several atmospheric processes (Ward et al., 1993; Reinhardt et al., 2001). As gaseous and aerosol emissions from fires are transported through the atmosphere, they impair air quality by reducing the visibility (Valente et al., 2007), producing unhealthy levels of PM (Wu et al., 2006), and reacting to form harmful secondary pollutants, such as ozone (O₃) and secondary organic aerosols (SOA) (Wiedinmyer et al., 2006). The effects of these emissions span across scales, from occurrence of local atmospheric episodes, to high O₃ concentrations at medium distances (regional scale) from the emission source (Miranda et al., 2009), to the contribution to greenhouse effect (Miranda et al., 1994; Simmonds et al., 2005).

To understand and evaluate forest fire effects on air quality, several factors should be analyzed and comprehensively integrated into models. Those include the description of fire emissions, atmospheric dispersion of smoke, and the chemical transformations of smoke. There are several air quality numerical tools in development, some

of them already available, aiming to integrate all these factors and to estimate the dispersion and transformation of smoke from forest fires. For instance, Wang et al. (2006) present the first results of the model and examine the ability of the Regional Atmospheric Modeling System (RAMS) – Assimilation and Radiation Online Modeling of Aerosols (AROMA) and examine its ability to simulate the smoke transport considering the smoke radiative impacts on surface energetics, boundary layer, and other atmospheric processes. Hodzic et al. (2007) assessed the 2003 European fire season and the resulting changes in aerosol optical properties, atmospheric radiative forcing and photochemistry using an improved version of the meso-scale chemistry-transport model CHIMERE taking into account the MODIS daily smoke emissions inventory and the injection altitude of smoke particles. The authors suggest that wildfire emissions can exert, at least episodically, an important effect on atmospheric stability, photolysis rates and ozone concentrations in polluted urban areas far away from their source regions.

Crucial in all systems is the quality of the forest fire emission estimates. In this sense, recently, a quite few works used the fire emission inventories derived from satellite data to examine the impacts of specific fire events on regional and urban air quality (Wu et al., 2006; Wiedinmyer et al., 2006; Hodzic et al., 2007; Sofiev et al., 2009). Wu et al. (2006) highlighted that during the 2003 southern California wildfires the forest fire emissions increased the PM₁₀ concentrations by 160 µg.m⁻³. Hodzic et al. (2007) concluded that the modeled 2003 European wildfire emissions caused an increase in averaged PM₁₀ ground concentrations from 20 to 200%.

Remote sensing techniques have been used to identify fire events and estimate burnt area and fuel consumption, mostly through the

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use of the fire radiative power, and emissions by combination with emission factors. Alternatively or as a complement, a bottom-up approach can be used, which is based on local and more detailed information on burnt area, fuel loads, vegetation type, burning efficiency and emission factors to estimate forest fire emissions (e.g. van der Werf et al., 2006; Ottmar et al., 2009). Both approaches are an ongoing research topic.

In Portugal, detailed data is available to estimate forest fire emissions and assess the impact on air quality. Previous Portuguese studies (Miranda, 2004; Miranda et al., 2009) analyzed the impacts of forest fire emissions on air quality for few episodic situations. For a better assessment of the influence of fires on air pollutant concentrations it is essential to investigate in the long term. In this sense, the main purpose of this work is to assess, by numerical modeling, the impact of forest fire emissions on the air quality in Portugal between June and September, 2003, 2004 and 2005.

2. The 2003, 2004 and 2005 fire seasons

Each summer, wildland forest fires burn a considerable area of the southern European landscape, due to persistent, extreme fire conditions. In Portugal, the 2003, 2004 and 2005 fire seasons were especially serious in terms of forest fire activity, with an annual burnt area of 421 835 (San-Miguel-Ayanz et al., 2004), 129 652 (Schmuck et al., 2005) and 338 259 (Schmuck et al., 2006) hectares, respectively.

In 2003, Portugal faced the most severe fire season ever recorded, 4645 fires burned 8.6% of the Portuguese forested area (San-Miguel-Ayanz et al., 2004). The 2003 fire season was characterized by extreme fire weather conditions (Viegas et al., 2006; Trigo et al., 2006) which, associated with physical and structural conditions, led to a disastrous fire season in Portugal. Trigo et al. (2006) analyzed the atmospheric conditions related to this devastating 2003 fire season. Synoptic conditions associated with wildfire occurrences were characterized. The authors concluded that the observed anomalies for temperature daily values at 850 hPa surpassed historical maxima in southern and central Portugal on August 1 and 2, respectively. Additionally, the days with the highest amounts of daily burnt area were characterized by large anomalies of surface meteorological variables that favored wildfire activity, namely surface maximum and minimum temperature, relative humidity, and wind speed and direction. This extreme situation contributed to the highest number of fire ignitions since 1980. When compared with the ten-year average this represents an increase of 32% in the number of fires and a 77% increase for the burnt area.

In 2004 the burnt area and the number of fires were much smaller than in 2003, and also smaller than the ten-year average (Schmuck et al., 2005). By the end of 2004, and during 2005, Portugal suffered an intensive drought. During eleven consecutive months the rainfall amounts were almost insignificant or below the normal values (Schmuck et al., 2006). Not surprisingly, during 2005, the burnt area was also significant.

Fig. 1 shows the burnt area and the number of large fires by month in 2003, 2004 and 2005. Large forest fires (defined by the Portuguese Authorities as fires greater than 100 ha) are responsible for the majority of the burnt area in Portugal. In 2003, large fires burned 96% of total burnt area, 73% in 2004 and 85% in 2005. Notwithstanding the total burnt area by these large fires they only represent about 1% of the total occurrences.

Typically, in Portugal, large forest fires occur during the summer season between June and September (JJAS). Meteorological conditions during the summer largely impact the amount of burnt area and the number of fires (Carvalho et al., 2008; Hoinka et al., 2009). In 2003 and 2005, August presented the highest values of burnt area and number of fires. By contrast, in 2004, large fires occurred uniformly during the summer months. Due to the specific meteorological

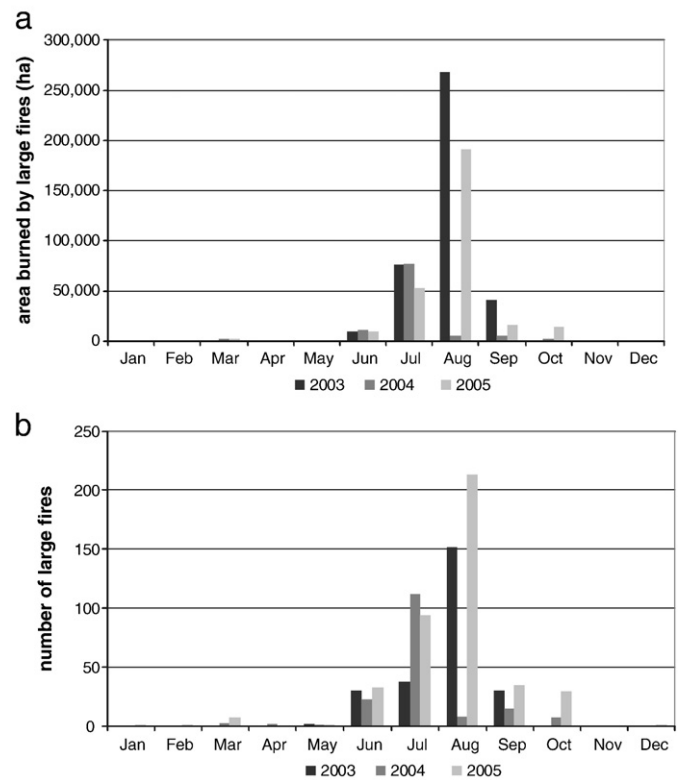


Fig. 1. Area burned (a) and number of large fires (b) that occurred in 2003, 2004 and 2005, by month.

conditions, namely some precipitation events, August registered a reduced number of large forest fires in that year. Forest fire events were also recorded in May and October, which is in accordance with a very recent trend of lengthening in the forest fire season in Portugal (Carvalho et al., 2008).

3. Methodology

The methodology adopted in this study can be divided in two main components: estimation of forest fire emissions and the application of the air quality model.

3.1. Forest fire emissions estimation

Forest fire emissions depend on multiple and interdependent factors, such as: forest fuels' characteristics, burning efficiency, burning phase, fire type, meteorology and geographical location. They are frequently estimated through an equation as shown below, which includes emission factors, burning efficiency, fuel loads and burnt area (e.g. Wiedinmyer et al., 2006; Miranda et al., 2005; van der Werf et al., 2006):

$$E_i = EF_i \times \beta \times B \times A$$

where, E_i is the emission of compound i (g), EF_i the compound i emission factor (g.kg^{-1}), β the burning efficiency, B is the fuel load (kg.m^{-2}), and A is the burnt area (m^2).

According to Miranda et al. (2005) and taking into consideration the data usually available, fuel load and emission factors can be expressed by forest type, namely resinous, deciduous and eucalyptus. Specific values for Portugal were selected based on data from the National Forest Inventory (DGF, 2001) about the characteristics of the consumed forest type and shrubs. Furthermore, fire data such as the starting location and ignition time and burnt area per fire were

collected from the National Forest Fires Inventory (DGRF, 2006). Table 1 summarizes the selected data, in terms of covered area by forest specie. Eucalyptus is the most representative forest type in Portugal, reaching almost 60% and the deciduous species have a reduced coverage in the order of 5%.

Burning efficiency, which is a fundamental parameter for the estimation of emissions, is usually defined as the ratio of carbon released as CO₂ to the total carbon present in the fuel. In laboratory and field experiments, the combustion efficiency can be expressed as the fraction burned related to the total available biomass. There is a wide range of burning efficiency data (Seiler and Crutzen, 1980; Levine et al., 1989; Simpson et al., 1999; Ward, 1999; Battye and Battye, 2002; PNAC, 2002; EEA, 2004) for shrubs and forest types (data on burnt area are discriminated into shrubs and forest). In southern Europe burning efficiency for shrubs is very high due to the low shrub moisture. The 0.8 burning efficiency value from PNAC (2002) was selected for this study because it represents Portuguese conditions for shrub as understory vegetation, as well as fine fuel from other vegetation species. Regarding forest, and taking into account the available information, 0.25 efficiency was taken from EEA (2004) as representing the southern European forest types, namely communities of resinous, eucalyptus and deciduous trees, without understory vegetation.

Fuel load is another important factor affecting fire emissions. It represents the amount of fuel available by unit of area. Several studies (Viegas, 1989; Trabaud et al., 1993; PNAC, 2002; EEA, 2004; Xanthopoulos et al., 2004; Cruz, 2005) propose fuel load values for the forest types as well as for shrubs. However, the EEA (2004), Viegas (1989) and Xanthopoulos et al. (2004) studies don't have the required disaggregation. Cruz (2005), PNAC (2002) and Trabaud et al. (1993) fuel load values could be an acceptable choice for this study, but the first two specifically represent the Portuguese fuel characteristics; Trabaud et al. (1993) fuel load values mainly concern Mediterranean type fuel. Finally, taking into account the above comments and comparing the different values, we selected the PNAC (2002) fuel load values.

For the adequate application of the methodology, which implies the spatial distribution of the forest fire emissions, it was first necessary to distinguish the burnt area by classes of forest type, through two approaches: (i) distribution of forest types by Portuguese district; and (ii) characterization of types of vegetation affected by forest fires in each year. Based on the distribution of forest types by district, the average fuel load by district was estimated, and presented in Table 2 (the location of the different districts is shown in Fig. 7), which also includes the burning efficiency chosen values for shrubs and forest.

At district level the forest composition is variable; each district presents a particular distribution as a consequence of forest planning based on different strategies and policies. Therefore, fuel load by district is variable too, ranging from 4.05 kg.m⁻² in Beja to 7.03 kg.m⁻² in Leiria.

Emission factors are defined as the mass of emitted pollutant per mass of dry burnt fuel (g.kg⁻¹) or per burnt area (g or kg.ha⁻¹).

Table 1
Area distribution for each forest specie in Portugal (DGF, 2001).

Forest specie	Forest type	Area (ha)	Area (%)
<i>Pinus pinaster</i>	Resinous	976,069	33.8
<i>Pinus pinea</i>		77,650	
Other resinous		27,358	
<i>Quercus spp.</i>	Deciduous	130,899	5.4
<i>Castanea sativa</i>		40,579	
<i>Eucalyptus spp.</i>	Eucalyptus	672,149	60.9
<i>Quercus suber</i>		712,813	
<i>Quercus rotundifolia</i>		461,577	
Other		102,037	

Table 2

Summary of the selected values concerning forest fire burning efficiency, forest fuel load and forest fire emission factors for different fuel types in Portugal, based on literature review.

Fuel type				
Shrubs	Resinous (R)	Deciduous (D)	Eucalyptus (E)	
Burning efficiency				
0.80	0.25			
Fuel load (kg.m ⁻²)				
5.84	Aveiro			
4.10	Beja			
5.49	Braga			
4.86	Bragança			
6.39	Castelo Branco			
6.82	Coimbra			
4.05	Évora			
4.61	Faro			
6.16	Guarda			
7.03	Leiria			
5.02	Lisboa			
4.21	Portalegre			
5.39	Porto			
5.05	Santarém			
5.43	Setúbal			
6.36	Viana do Castelo			
6.18	Vila Real			
6.69	Viseu			
Pollutant				
Emission factors (g.kg ⁻¹)				
CO ₂	1,477	1,627	1,393	1,414
CO	82	75	128	117
CH ₄	4	6	6	6
NMHC	9	5	6	7
PM2.5	9	10	11	11
PM10	10	10	13	13
NO _x	7	4	3	4
SO ₂	0.8	0.8	0.8	0.8
NH ₃	0.6	0.8	0.6	0.6

Emission factors for CO₂, CO, CH₄, NMHC, PM with aerodynamic diameter below 10 μm (PM10) and 2.5 μm (PM2.5), NO_x, SO₂ and NH₃ were based on literature reviews that selected the most suitable values for southern European ecosystems, namely for the Portuguese land use types (Miranda, 2004). These emission factors are summarized in Table 2 and were used to estimate forest fire emissions by pollutant and year, which are shown in Fig. 2.

In 2003 the forest fire emissions were higher due to the larger burnt area. The total forest fire emissions in 2005 represent 50% of that of 2003. As already stated the year 2003 was the most severe in terms of forest fire activity in Portugal. Table 3 allows a comparison of the estimated forest fire emissions for the year 2003 and the anthropogenic emissions from the national emissions inventory (IA, 2006), for the same year.

Forest fires can represent a significant percentage of the total annual anthropogenic emissions, reaching 40% for CO and CH₄, 30% for PM10 and 12% for CO₂. Forest fire emissions can be higher than those emitted by specific activity sectors; for instance, forest fires emitted more particulate matter (PM10 and PM2.5) than the Portuguese transport sector in 2003. In contrast to the anthropogenic emissions the forest fire emissions are released into the atmosphere few times during short periods and in certain parts of the country. Consequently, forest fire emissions overshadow anthropogenic emissions a period with severe forest fire intensity.

A comparison between the forest fire emission values estimated here, and values reported by other available inventories is shown in Table 4, which presents the total forest fire emissions for Portugal (2003–2005) for CO₂, CO, CH₄, NMHC, PM2.5, PM10, total particulate matter (TPM) and NO_x, based on three different inventories: the

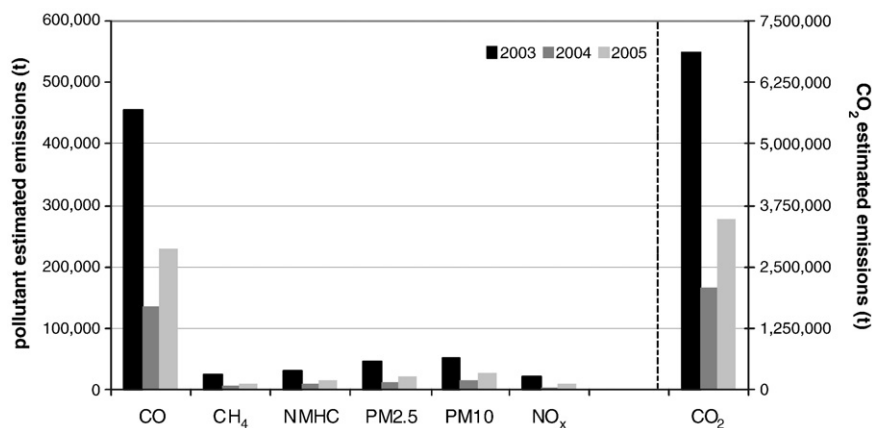


Fig. 2. Forest fires emissions for 2003–2005 in Portugal (only large forest fires were considered).

forementioned Portuguese inventory, the Global Fire Emissions Database (GFED) inventory (van der Werf et al., 2010), and the European Forest Fire Information System (EFFIS) inventory (San-Miguel-Ayanz and Steinbrecher, 2009; Barbosa et al., 2009).

In general, the emission data presents the same order of magnitude, but in some cases it is possible to verify a larger difference, namely: for CH₄ in 2004 and 2005 in the GFED inventory; for NMHC in EFFIS inventory in 2004; for TPM for both GFED and EFFIS inventories. The differences were in some way expected because different methodologies were applied. Another study is foreseen to better explore the uncertainty associated with forest fire emissions estimated within different available inventories, but the values obtained in this paper are quite acceptable and don't significantly differ from other values.

3.2. Air quality modeling description

The analysis of the impact of forest fire emissions on the air quality was based on the application of the air quality model LOTOS-EUROS v1.3 (Schaap et al., 2008). The LOTOS-EUROS system is an operational 3D chemistry transport model aimed to simulate air pollution in the lower troposphere. The LOTOS-EUROS model simulates the O₃ chemistry using a modified Carbon-Bond Mechanism 4 (CBM4) (Whitten et al., 1980) that includes 28 species and 66 reactions, with 12 of them photolytic reactions. The model incorporates primary (combustion) particles (elemental carbon (EC), organic carbon (OC)), sea salt and secondary inorganic aerosols (SIA: sulfate (SO₄), nitrate (NO₃) and ammonia (NH₃)).

Concerning SIA, the thermodynamic equilibrium module ISORROPIA (Nenes et al., 1998) was used to describe the equilibrium among gaseous nitric acid, ammonia and particulate ammonium nitrate, and between ammonium sulfate and aerosol water. As LOTOS-EUROS does not currently incorporate the reaction of nitric acid with sea salt, the results of equilibrium calculations in marine and arid regions should be interpreted with care (Zhang et al., 2000). Dust and secondary organic aerosols are not included in the present simulation as the knowledge on the formation routes of SOA and the sources of dust is considered to be too limited. As a

consequence, LOTOS-EUROS underestimates the observed total PM₁₀ mass as do all current Chemical Transport Models – CTM (e.g. Stern et al., 2008).

In the vertical direction there are three dynamic layers and a surface layer. The model extends in the vertical direction 3.5 km above sea level following the dynamic mixing layer approach. The lowest dynamic layer is the mixing layer, followed by two reservoir layers. The height of the reservoir layers is determined by the difference between ceiling (3.5 km) and mixing layer height. The surface layer has a fixed depth of 25 m.

The LOTOS-EUROS system has been used in several applications showing good agreement between the observed and the modeled data (Schaap et al., 2004, 2008; van Loon et al., 2007).

The system was first applied at the continental scale (with 35 × 25 km²) resolution and then in Portugal, using the same physics and a one-way nesting technique, with 17.5 × 12.5 km² horizontal resolution. Boundary conditions for ozone are obtained from the 3D climatological datasets by Logan and for other components the EMEP method was used (Schaap et al., 2008).

Anthropogenic emissions were obtained from the 2000 European-wide emission inventory performed at the Netherlands Organization for Applied Scientific Research (TNO) (Visschedijk and Denier van der Gon, 2005) with a grid resolution of 0.25° × 0.125°. In this study, we have coupled the forest fire emission data with the LOTOS-EUROS model. The hourly smoke emissions were estimated using the Western Regional Air Partnership – WRAP diurnal profiles (WRAP, 2005) that are based on the fuel consumption data registered for forest fires events in the USA. This type of information is still absent for Portugal, but data gathered for forest fire events indicate peak ignitions between 14 and 17 LST (Local Standard Time) (DGRF, 2007), which agree with the pronounced diurnal cycle with peak emissions during the afternoon and very low emissions during the night suggested by the WRAP study (Eck et al., 2003; WRAP, 2005). According to the WRAP (2005) analysis the daily emissions peak is attained at 16 LST and its minimum values are registered during the night.

In CTMs forest fire emission injection heights have often been represented using empirical or arbitrary procedures (Sessions et al.,

Table 3
Anthropogenic and forest fire emissions in Portugal for the year 2003.

Source	Atmospheric pollutant emissions (t)						
	CO ₂	CO	CH ₄	NMHC	PM _{2.5}	PM ₁₀	NO _x
Forest fires	6,842,000	457,000	26,000	32,000	26,000	53,000	21,000
Transports	19,472,820	315,265	35,660	62,847	9849	9877	130,109
Industry and services	30,919,120	357,701	2760	120,887	80,372	106,365	140,371
Forest fires/total emissions (%)	12.0	40.4	40.4	14.8	22.4	31.3	7.2

Table 4

Total forest fire emissions (t) in Portugal for 2003, 2004 and 2005 fire seasons based on different methodologies.

Source	Forest fire emissions (t)							
	CO ₂	CO	CH ₄	NMHC	PM2.5	PM10	TPM	NO _x
Year	CO ₂	CO	CH ₄	NMHC	PM2.5	PM10	TPM	NO _x
2003								
Portuguese inventory	6,842,000	456,000	26,000	32,000	26,000	53,000	-	21,000
GFED	8,000,000	500,000	70,000	30,000	20,000	-	140,000	14,000
EFFIS	10,510,119	411,945	21,475	17,834	41,406	48,913	68,749	28,745
2004								
Portuguese inventory	2,096,000	137,000	8000	10,000	14,000	16,000	-	7000
GFED	3,000,000	200,000	20,000	10,000	10,000	-	50,000	5000
EFFIS	3,312,543	129,112	6735	5599	13,001	15,361	21,597	9013
2005								
Portuguese inventory	3,470,000	230,000	13,000	16,000	24,000	27,000	-	11,000
GFED	14,000,000	800,000	120,000	50,000	40,000	-	250,000	25,000
EFFIS	7,866,267	325,628	16,843	13,833	32,206	38,034	53,180	22,719

- Not available.

2010). These methods have included linearly filling estimated injection columns, restricting emissions to surface layers, and assumed turbulent mixing by filling the planetary boundary layer. Large forest fires (burnt area larger than 100 ha) in Portugal mainly occur under specific synoptic conditions that favor dry and warm conditions in combination with strong (south easterly) winds (e.g. Trigo et al., 2006; Hoinka et al., 2009). Hodzic et al. (2006, 2007) estimated injection heights between 3 and 5 km for the large Portuguese fires in 2003, based on an adaptation of the WRAP method accounting for fire brightness temperature from satellite data. These authors, however, did not account for the dependency on wind intensity. Under high wind conditions injection heights are significantly lower and not expected to rise into the free troposphere (Freitas et al., 2009). Strada and Mari (2010) illustrated the impact of strong winds for a large fire in southern France under Mistral conditions with an online air quality model and estimated injection height values in the order of 1 km, well within the Planetary Boundary Layer (PBL). Applying the empirical (and “old”) formulae to calculate forest fires plume rise by

Chandler et al. (1991) also yields values within the PBL. On the other hand, small fires don't release enough energy to produce plumes rising higher than some hundred meters. Hence, we consider that for Portuguese and south-European forest fires in general injection heights are below the PBL. Thus, we inject forest fire emissions into the mixing layer. This approach connects well to the vertical structure of LOTOS-EUROS in which the PBL is the lowest dynamic layer and assumed to be well mixed.

4. Results

Simulations were performed from June to September for the 2003, 2004 and 2005 years, regarding gaseous and particulate matter pollutants. A baseline simulation (BS) was performed, including “conventional” emissions and a forest fire simulation (FS), which also considered emissions from large forest fires. Fig. 3 shows the spatial distribution of forest fire PM10 emissions for 2003, 2004 and 2005.

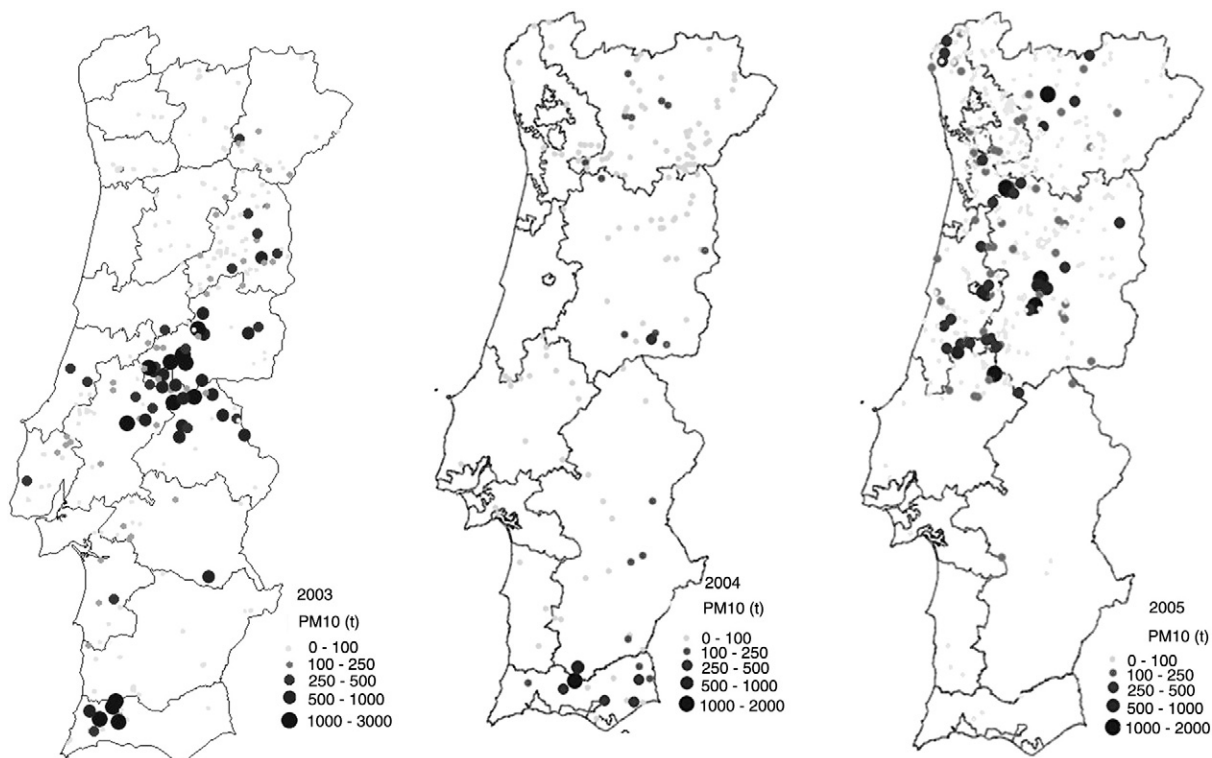


Fig. 3. PM10 forest fire emissions spatial distribution for 2003–2005 fire seasons in Portugal (only large forest fires were considered).

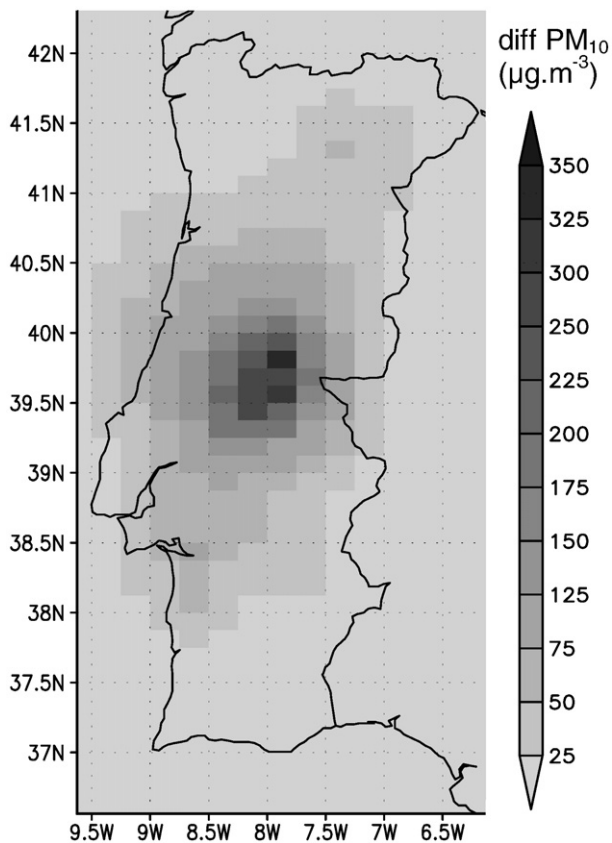


Fig. 4. Spatial differences ($\mu\text{g.m}^{-3}$) between simulation results with (FS) and without (BS) forest fire emissions, for PM₁₀ daily averages on August 3, 2003.

To better analyze the spatial impact of forest fire emissions on the air quality, Fig. 4 shows the spatial differences of the PM₁₀ daily values between both simulations (FS-BS), for one of the most critical days (2003, August 3).

For this specific day, the impact of forest fires was highest at the central inland part of Portugal, and the PM₁₀ daily mean difference reached almost $300 \mu\text{g.m}^{-3}$.

Hourly modeling results were compared to monitored air quality data acquired at different background air quality stations. The air quality monitoring stations selection was based on their acquisition efficiency, which should be at least 90% for PM₁₀ (EC, 1999) and 75% for O₃ (EC, 2002). Air quality data was available at 13 of the 18 districts in Portugal (district identification is depicted in Fig. 7). Most of the stations are located near the major urban centers on the west coast of the country, most notably Lisbon and Porto.

From the comparison between Figs. 4 and 7, it is evident that the areas with impaired air quality do not coincide with the location of the majority of the monitoring stations. Hence, the impact of these fires on this particular day has not attained the coastal areas.

To better understand the impacts of forest fire emissions on air quality, Fig. 5 shows the modeling system results at a short time period for both simulation results (FS and BS) and the measured data. This temporal analysis has been performed for the air quality station "Instituto Geofísico de Coimbra (IGC)", for the most critical period in terms of fire activity (August 1–15, 2003), and for PM₁₀ and O₃. The IGC is an urban background air quality station located in the central part of Portugal in one of the most affected districts (Coimbra district) and thus this comparison would highlight the main difficulties that a CTM model would show when explored in this way. For this period and location statistical parameters were estimated to better

assess the simulation results, namely the root mean square error (RMSE), the systematic error (BIAS), and the correlation coefficient (r) (Borrego et al., 2008). Fig. 5 also includes the estimated statistical parameters.

Concerning PM₁₀ and for the selected period, the LOTOS-EUROS system tends to underestimate the measured peak PM₁₀ values, namely for the period August 2–3, when the fire activity was highest. In fact, for a particular hour of the 3rd of August there is a $\sim 200 \mu\text{g.m}^{-3}$ difference between the measurement and the simulated values. But this difference would have been much higher ($\sim 300 \mu\text{g.m}^{-3}$) without the fire emissions contribution. Moreover, these are hourly values and to be able to correctly simulate the hourly trends and to capture the PM₁₀ peaks, most probably originated by the forest fires, should be considered an important achievement. Anyway, the FS simulation was able to capture the effect of forest fires on PM₁₀ levels during these two particular days with results much closer to the measured values than the BS estimates. For the other days, the FS results agree quite well with the measurements and the estimated statistical quality indicators for the entire period improved with the inclusion of fire emissions (e.g. the correlation coefficient increased from 0.4 to 0.7 and the BIAS changed to a value closed to zero).

In terms of O₃, the modeling system results present the same behavior as observation values (correlation coefficients vary from 0.75 without fire emissions to 0.77 with them) but the higher concentration values are underestimated and the minimum ones are overestimated. However, important improvements were achieved with the inclusion of forest fire emissions, namely regarding daily maximum values. During the daytime photochemistry takes over and the additional NO_x and VOC provided by the fire emissions cause additional ozone formation. The ozone night time concentrations can be lowered during the night due to titration by emitted NO.

Table 5 summarizes the averaged statistics obtained (RMSE and BIAS) by district, and for both simulations (with and without forest fire emissions), for PM₁₀ and O₃, for 2003 and 2004.

Regarding PM₁₀, the LOTOS-EUROS model underestimates their values for both fire season years and the estimated BIAS values are positive. The model's performance increases when forest fire emissions are included, with smaller BIAS and RMSE. On average the RMSE decreases 4% and 1.5%, respectively, for 2003 and 2004. Adding fire emissions lowers the amount of missing PM₁₀ and results in model estimates that are closer to the measured ones. This is exactly why the new important source for PM was implemented as described in the model. Still, bias values continue to be positive indicating an under-prediction due to the other missing components and potentially other short comings in the emission and model description.

Regarding O₃, the LOTOS-EUROS model tends to overestimate concentrations, except in the Castelo Branco, Santarém and Lisboa districts in 2003, and in the Vila Real and Lisboa districts in 2004. In general, the inclusion of forest fire emissions did not improve the model performance. This could be explained by the O₃ particularities as a secondary pollutant. Its main effects would have occurred far away from the forest fires and probably from the areas where monitored data was available. Hence, effects of forest fire emissions on photochemical pollution were probably not observed at the monitoring networks.

To complement the analysis, Fig. 6 presents the district-averaged RMSE and BIAS for both simulations (with and without forest fire emissions), for PM₁₀ and O₃ for 2005.

For the year 2005, the model performance increases substantially when forest fire emissions are considered, mainly regarding PM₁₀. On average, the RMSE decreases 20% for PM₁₀ and 15% for O₃. As verified for 2003 and 2004, the modeling system tends to underestimate PM₁₀ concentrations and overestimate O₃ values. The major differences between the BS and FS results for PM₁₀ concentrations

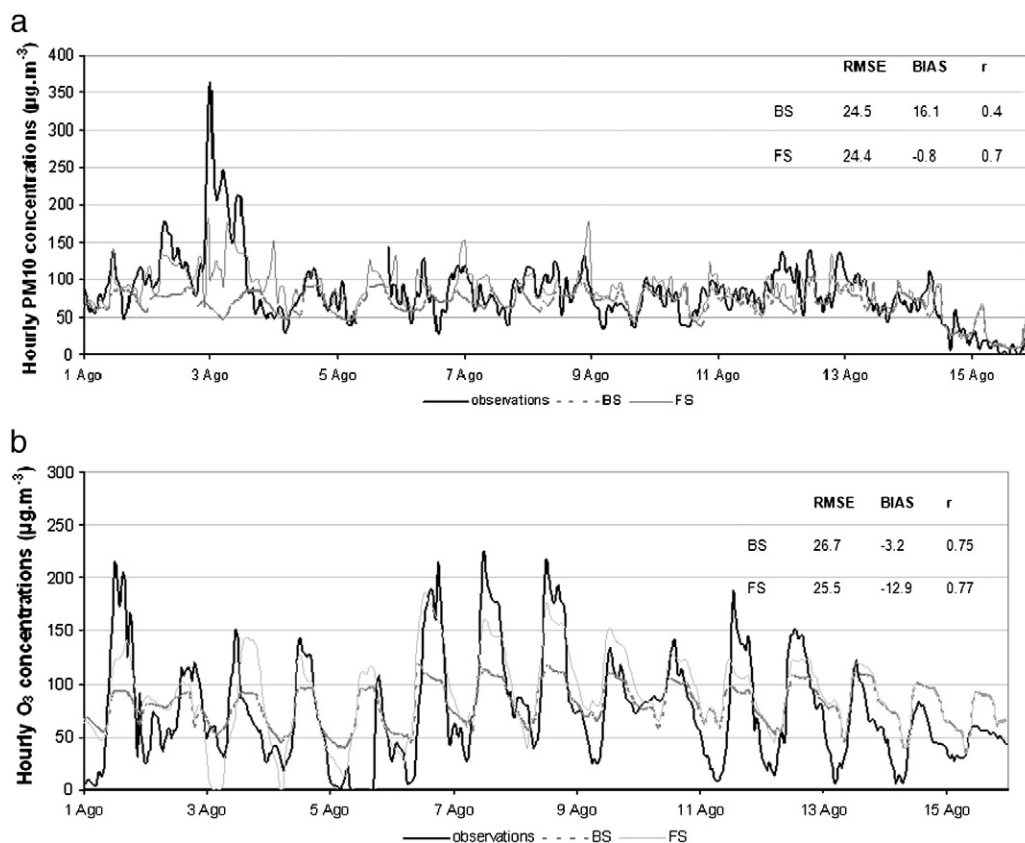


Fig. 5. Observed and simulated hourly concentrations values ($\mu\text{g}\cdot\text{m}^{-3}$) for PM10 (a) and O_3 (b), between August 1 and 15, 2003, at IGC station. Statistical parameters related to the model performance concerning the simulation results with (FS) and without (BS) forest fire emissions are so included.

happen in the districts of Braga, Porto, Vila Real, Aveiro, Castelo Branco and Santarém. In 2005, these districts presented the larger values of burnt area in Portugal.

Fig. 7 shows the correlation coefficient values for the simulation considering the forest fire emissions for PM10 and O_3 , for 2003, 2004 and 2005.

Table 5

Averaged statistical indicators (RMSE and BIAS) for the LOTOS-EUROS model system performance for BS and FS, concerning PM10 and O_3 for 2003 and 2004.

Year	2003				2004			
	RMSE ($\mu\text{g}\cdot\text{m}^{-3}$)		BIAS ($\mu\text{g}\cdot\text{m}^{-3}$)		RMSE ($\mu\text{g}\cdot\text{m}^{-3}$)		BIAS ($\mu\text{g}\cdot\text{m}^{-3}$)	
District	BS	FS	BS	FS	BS	FS	BS	FS
PM10								
Braga	-	-	-	-	18.3	18.1	25.9	25.5
Vila Real	-	-	-	-	15.2	14.6	14.0	13.0
Porto	21.8	21.4	36.0	34.9	18.4	18.2	24.8	24.5
Aveiro	20.7	20.1	33.9	31.9	-	-	-	-
Coimbra	22.3	20.7	38.5	34.1	-	-	-	-
Castelo Branco	-	-	-	-	-	-	-	-
Leiria	19.9	18.5	30.3	26.8	-	-	-	-
Santarém	-	-	-	-	-	-	-	-
Lisboa	17.6	17.0	22.9	21.3	17.0	16.9	19.8	19.7
Setúbal	19.0	18.4	28.2	26.2	-	-	-	-
Average	20.2	19.3	31.7	29.2	17.2	17.0	21.1	20.7
O₃								
Braga	-	-	-	-	44.8	44.8	-19.1	-19.2
Vila Real	-	-	-	-	44.5	44.6	23.1	23.1
Porto	41.5	41.7	-83.6	-88.9	44.6	44.6	-21.5	-21.6
Aveiro	43.7	43.8	-116.4	-129.5	44.9	45.0	-26.5	-26.7
Coimbra	40.9	40.6	-62.2	-69.3	55.9	56.0	-61.5	-61.7
Castelo Branco	40.3	40.0	3.3	1.1	45.1	45.1	-12.9	-13.3
Leiria	-	-	-	-	42.2	42.2	-22.3	-22.4
Santarém	43.7	44.0	15.3	14.6	40.0	40.0	-2.4	-2.6
Lisboa	43.3	43.3	9.8	8.6	42.7	42.7	1.4	1.4
Setúbal	40.2	40.2	-27.5	-31.2	52.2	52.2	-39.1	-39.2
Average	41.9	41.9	-37.3	-42.1	45.7	45.7	-18.1	-18.2

- Monitored data not available.

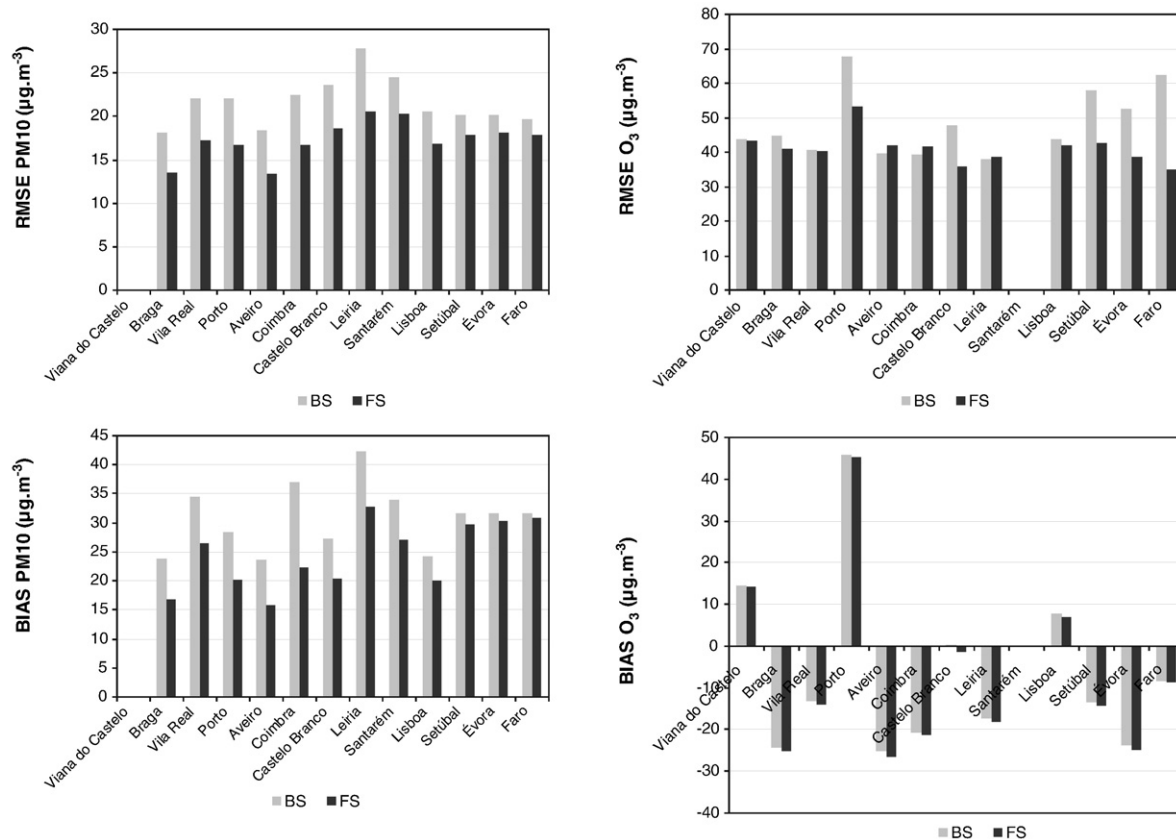


Fig. 6. Averaged statistical indicators (RMSE and BIAS) for the LOTOS-EUROS model system performance for BS and FS, concerning PM10 and O₃ for 2005.

From Fig. 7 it is possible to verify that the year 2005 presents the best correlation coefficients between measured and estimated concentrations. The regions in the north and the central part of Portugal show the highest values, reaching 0.88 in Braga district for PM10. It should be noted that the number of available air quality stations which measured PM10 and O₃ concentrations in 2005 is higher than in 2003 and 2004.

The averaged correlation coefficients obtained for each district revealed that, considering the difference between BS and FS, they increased from 0.53 to 0.55 for PM10 in 2003, from 0.36 to 0.50 in 2004 and from 0.39 to 0.72 in 2005. Concerning O₃, the correlation coefficients increased on average from 0.56 to 0.58 in 2003. For 2004 and 2005, there are no relevant differences.

In general, for 2005, the model presents a better performance.

The majority of the air quality monitoring stations is located in the coastal regions and the large fires occurred mainly in the inland districts. This fact may deeply influence the model performance evaluation strategy. In 2003 and 2004, the impact of the forest fire emissions seen on a few days, at a very limited subset of the monitoring stations, is averaged out in the long-term assessment. The situation in 2005 was special in the sense that the areas with the most severe fires coincided with the areas that contain most of the monitoring stations. Hence, the impact registered at each of the station was large. The simulation that considered the forest fires captured these events, leading to significantly improve the model performance statistics for that year.

5. Conclusions

This work investigated the impacts of forest fire emissions on the air quality, namely on PM10 and ozone levels, in Portugal. The numerical modeling approach applied in this work confirms the significant impact of forest fires on atmospheric pollutants' concentrations.

In general, the LOTOS-EUROS model shows a good performance, which improves when forest fire emissions are considered, particularly for the PM10 concentrations. For summer conditions in Portugal the impact of the fires are considered to be of high importance and we included fire emissions aimed to capture their contribution to the higher measured PM10 values. On the other hand, the influence of the forest fire emissions on O₃ formation is not evident and needs more attention. The detailed simulation and analysis of specific episodes should be considered as future work that will allow better understanding of processes that have to be implemented in the model to simulate the chemistry and transport of forest fire emissions.

Future work might also benefit from the extension of the study area to the whole Iberian Peninsula. For this purpose, however, input data from Spain should be harmonized with that available for Portugal. Therefore, future applications will also explore the use of satellite data to estimate the fuel consumption. These additional estimates may prove very valuable in conjunction with the detailed Portuguese data at hand.

As a broad conclusion we can highlight the importance of the inclusion of forest fire emissions in the air quality modeling systems that support the air quality assessment in Portugal during the summer months. This advice can be extended to other southern European areas annually affected by forest fires.

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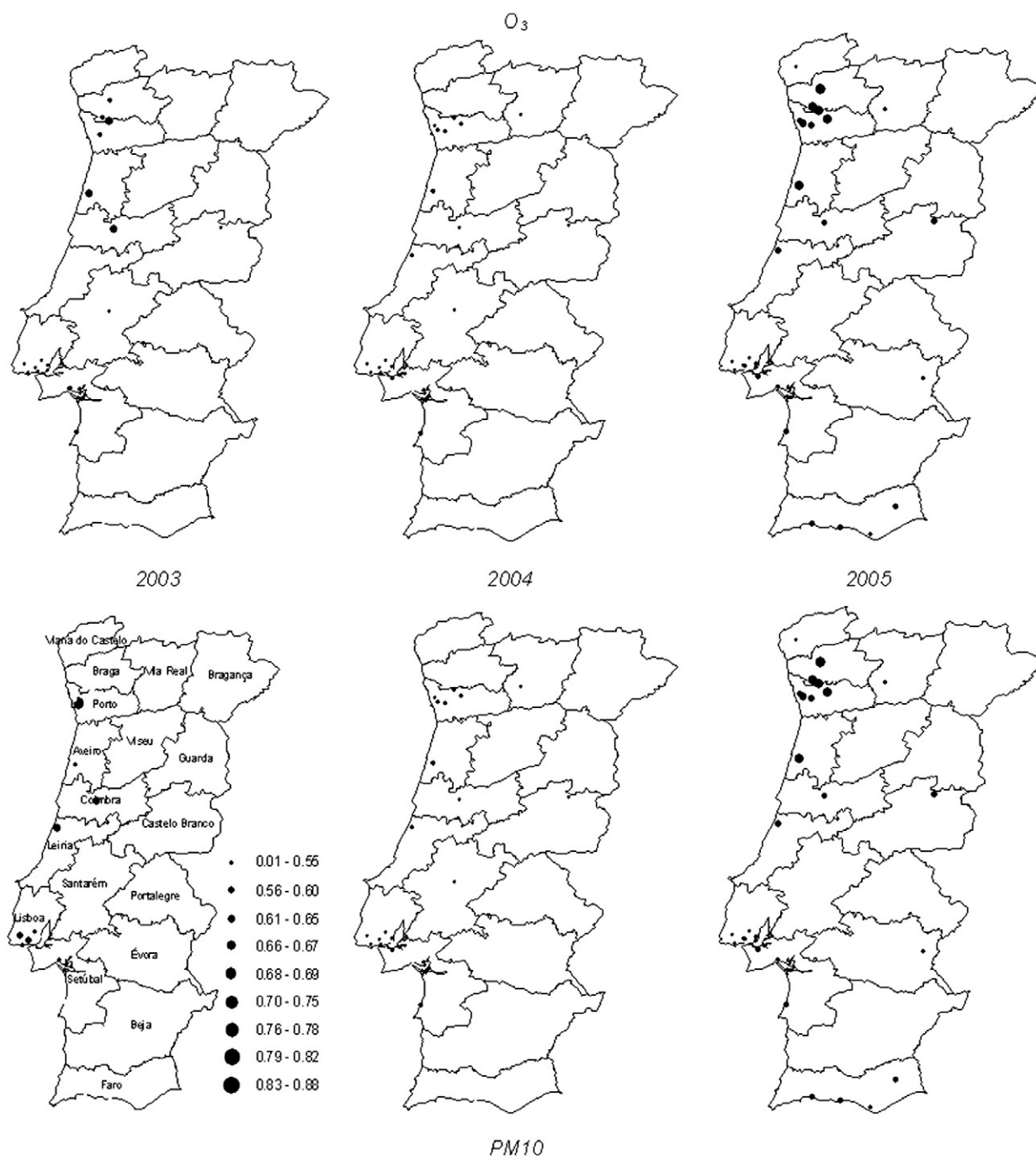


Fig. 7. Correlation coefficient of LOTOS-EUROS model system for FS, concerning PM10 and O_3 for 2003, 2004 and 2005.

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