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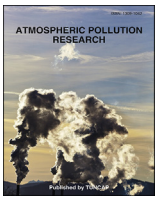

Original article

Particulate matter concentrations originating from industrial and urban sources: Validation of atmospheric dispersion modeling results

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ABSTRACT

This study presents the analysis of the emission, transport, dispersion, and concentration of particulate matter emitted from a large industrial complex dedicated to the manufacture of cement in the town of Malagueño, province of Córdoba (Argentina), using the USEPA's (Environmental Protection Agency) AERMOD model. The model was applied for 224 industrial and background emission sources (8 stacks, 3 limestone quarries, 13 material storage piles, 18 agricultural fields and 182 paved and unpaved segment roads). The application of the model was validated with Total Suspended Particulate matter (TSP) measured at two monitoring sites, where samples were collected for 62 consecutive days in winter. The maximum TSP values obtained at both monitoring sites (748 and 1100 μg m^{-3}) were well above the suggested WHO guidelines. The results obtained showed the impact of this industrial activity on local particulate matter concentrations, from which unpaved industrial roads and stockpiles were the most influential emission sources, directly affecting two of the closest neighborhoods in the area. Future studies will include the accumulation of heavy metals and Polycyclic Aromatic Hydrocarbons in TSP samples, the environmental risk assessment for exposure of the Malagueño population and the source apportionment of these pollutants.

1. Introduction

The increasing demand of society for compliance with air quality standards that protect residents living in the vicinity of industrial centers, involves developing methodologies and reliable calculation tools that contribute to decision making with full transparency (Stein et al., 2007; Donnelly et al., 2009). In this sense, atmospheric dispersion models are a useful tool, given that they incorporate the latest knowledge regarding atmospheric dynamics and can predict, with some degree of confidence, dispersal patterns, chemical transformations and deposition of pollutants, thereby obtaining an estimate of the concentrations of pollutants in the atmosphere over a certain period of time (Holmes and Morawska, 2006).

Atmospheric dispersion models have become an essential tool in decision making about ways to protect human health (Seinfeld, 1986). However, it is imperative that these dispersion models be properly evaluated with observational data before their predictions can be used with confidence, given that model estimates often influence decisions that have large public-health and economic consequences (Chang and Hanna, 2004). The purpose of this type of evaluation is to check whether the model applied represents the real system accurately, by comparing the data obtained experimentally with the model's prediction, in order to make appropriate

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adjustments to the input data in the model, given that a simulation can be run, but fail to produce similar results (Neshuku, 2012).

Globally, there have been several previous studies related to the experimental and predicted dispersion through numerical modeling of particulate matter (PM) emitted from different industrial atmospheric emission sources using the AERMOD steady-state plume model for example, the assessment of the emission and dispersion of PM in the cement manufacturing process from stacks, limestone quarries and active stockpiles (Abu-Allaban and Abu-Qudais, 2011; Kakosimos et al., 2011; Seangkiatiyuth et al., 2011; Neshuku, 2012; Tartakovsky et al., 2013). Since cement plants are among the most important sources of PM emissions, they have generally been investigated in terms of their environmental impacts (Schuhmacher et al., 2004; Abdul-Wahab, 2006; Baroutian et al., 2006; Al-Khashman and Shawabkeh, 2006). PM emissions from cement manufacturing process include particle sizes ranging from 0 to 5 μm (approximately clay size) to greater than 50 μm (silt size) (EPA, 1993), however considering its size, the most important particles with regard to their physicochemical properties and health effects are those that are in the range of 0.1–10 μm (WHO, 2005; Pope and Dockery, 2006).

Previous studies of air quality in Argentina were focused in major cities. The metropolitan area of Buenos Aires (AMBA) a mega city of 12.8 million inhabitants, was the main case study involving: (i) emission inventories (Weaver and Balam, 1999; Mazzeo and Venegas, 2003; Puliafito, 2009; D'Angiola et al., 2010; Allende et al., 2012); (ii) atmospheric dispersion models (Bogo et al., 2001; Venegas and Mazzeo, 2006; Allende et al., 2012); (iii) field measurement campaigns (Reich et al., 2006; Bogo et al., 1999; Jasan et al., 2009; Gallardo et al., 2012); (iv) the study of the boundary layer height (Ulke and Mazzeo, 1998) and the observation and analysis of aerosol optical depth (AOD) (Ristori et al., 2003; Ipiña et al., 2012). Recently, García Ferreyra (2014) obtained air quality maps through the implementation and first application of the chemical transport model CHIMERE over Argentina.

In Argentina, there are currently seventeen cement plants, of which two are located in the town of Malagueño (province of Cordoba) and generate 2 700 000 t/year (metric tonnes), thereby representing 16.3% of the national cement production (Schvarzer and Petelski, 2005). Both cement plants (Yocsina Plant and Malagueño Plant) provide a significant contribution to the concentration of PM, due to the operations of crushing and milling, transportation of rock material, loading and unloading, and the wind drag on stowage and storage. These operations generate PM that can affect human health, which is primarily related to respiratory problems (WHO, 2005). Furthermore, these cement plants were considered to be the main anthropogenic sources of atmospheric pollutants, taking into account the results of previous studies undertaken in the area (Rodríguez et al., 2010; Bermudez et al., 2012; Abril et al., 2014a, 2014b). Abril et al. (2014a) assessed the environmental impact of Yocsina cement plant (which uses industrial wastes as alternative fuels in the cement manufacturing process) by means of biomonitoring studies, and detected significant levels of Cd, Pb, Co, Ni and Ca in the vicinities of this industry. However, despite the fact that potentially polluting industrial activities are taking place, there are currently no stations that monitor the air quality in Malagueño.

Therefore, given the limited government environmental control programs, the difficulties in gaining access to the so called "public information" (i.e. local public health data, environmental compliance of industrial plants and industrial emissions), the scarcity of weather and monitoring stations of air quality, and the existing obsolete air quality legislation (the National Air Quality Act dates back to 1973 and Córdoba province lacks legislation concerning this topic), then there is a need for using predicting tools for decision making.

The present investigation aims to: (1) acquire quantitative information through in situ measurements of the current conditions of PM pollution caused by the several sources present in the study area; (2) characterize and estimate the PM emission rates of the main industrial and non-industrial sources; (3) evaluate the performance of the atmospheric dispersion model applied and (4) preliminarily assess the PM impact of the cement plants on the nearest residential areas.

2. Materials and methods

2.1. Study area description

The area under study is the town of Malagueño, located 18 km SW of Cordoba City, Argentina (Fig. 1). In this town, a private enterprise specializes in the production of cement and ready-mixed concrete. It possesses two cement plants separated by approximately 4 km, with the main raw material of these being the limestone extracted from two quarries (the cement manufacturing process is described in Section S1- Supporting Material). According to the latest census, Malagueño has 14 364 inhabitants over 13 neighborhoods, which are far from the central urban core and have relatively large distances between each other. In this research work we specifically focused on the neighborhoods of Central Malagueño (henceforth referred as "Malagueño") and Yocsina, which are the closest to both cement plants.

2.2. Field TSP monitoring

Daily samples were collected over 24-h periods, using two Handi-Vol medium volume samplers (Coêlho Dias, 2007) with flows reaching 0.2 m^3/min (HDC model, "Energética-Qualidade do Ar", Rio de Janeiro) equipped with glass fiber filters (0.6 mm pore size and 10 cm diameter acquired from "Alka Filter", Argentina) for Total Suspended Particulate Matter (TSP). The daily sampling was performed in winter (from June 25th to August 25th, 2012) at two monitoring stations (Fig. S1) close to the two cement plants and 2 km away from each other: one in Yocsina neighborhood (Site 1: 31°26′40″S, 64°22′01″W, close to Cement Plant 1) and the second in Malagueño neighborhood (Site 2: 31°27′34″S, 64°21′14″W, close to Cement Plant 2). Winter season in the province of Cordoba, Argentina, represents the worst case scenario for exposure to atmospheric pollutants, such as dust, since: (i) practically no rain episodes are registered; and (ii) there are cases of strong radiative inversion starting at early morning hours and ending at noon (the long lasting inversion has to do with the almost complete lack of wind during most of the day, and low levels of humidity and irradiance) (Stein and Toselli, 1996).

A total of 124 samples were collected during this study, and the mass content was obtained by weighing the filters before and after exposure with a semi-micro balance (0.0001 g). The filters were lyophilized for 1 h, and the relative humidity and ambient temperature were recorded before weighing (with the range of the conditions being 25–35% RH and T 21–25 °C). Blank and control filters were used for tare weighing, in order to assess the accuracy of the gravimetric TSP analysis. The information gathered through this sampling was used as one of the inputs in order to validate AERMOD results, with the other input information used being the source emission rates, and the meteorological and topographic data.

2.3. TSP atmospheric dispersion modeling

2.3.1. Emission factors

Data from source-specific emission tests or continuous emission monitors are usually preferred for estimating a source's emissions,

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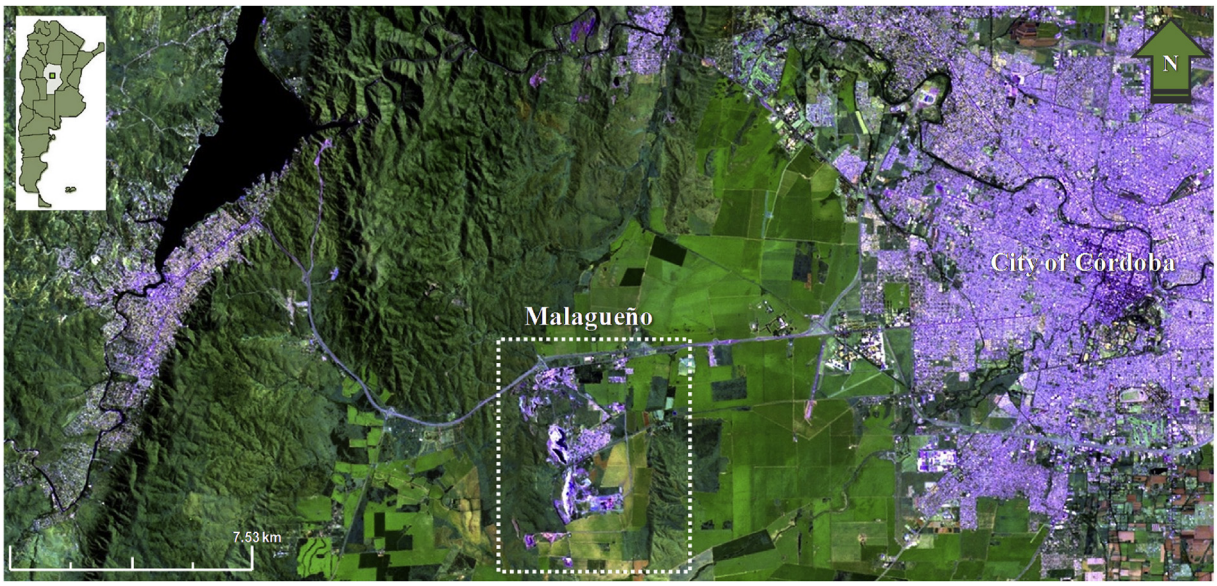

Fig. 1. Location of the town of Malagueño in the province of Cordoba, Argentina.

since those data provide the best representation of the tested source. However, in our case, there was no monitoring data or access to the material balance, so emission factors were estimated. Emission factors have long been fundamental for developing emission control strategies, determining applicability of permitting and control programs, ascertaining the effects of sources and appropriate mitigation strategies (USEPA, 1997). An emission factor is a representative value that attempts to relate the quantity of a pollutant released to the atmosphere with an activity associated with the release of that pollutant. These factors are usually expressed as the weight of pollutant divided by a unit weight, volume, distance, or duration of the activity emitting the pollutant. In most cases, these factors are simply averages of all available data of acceptable quality, and are generally assumed to be representative of long-term averages for all facilities in the source category. The general equation for emission calculation is:

$$E_{TSP} = EF_{TSP} \cdot A \cdot \left(1 - \frac{ER}{100}\right)$$

Where, E_{TSP} is TSP emission rate [kg/yr], EF_{TSP} is TSP emission factor [kg/t], A is the activity rate [t/yr], and ER the emission reduction. The emission reduction only applies if there are any controlling methods to abate the emissions (e.g. enclosure of material storage piles).

Several industrial and non industrial emissions sources were considered in this study: point sources (stacks), and area sources (limestone quarries, material storage piles, agricultural fields and paved and unpaved roads). The emission factors were estimated using the AP-42 guidelines "Compilation of Pollutant Emission Factors" (USEPA, 1997) for cement manufacturing and paved and unpaved roads; and California Air Resources Board (CARB, 2003) and the "Procedures Document for National Emission Inventory Criteria Air Pollutants 1985–1999" (USEPA, 2001) for farming operations in agricultural fields.

2.3.2. Meteorological and terrain data

To validate the application of AERMOD at the study area, two sources of meteorological data were used: i) hourly measured surface data from the National Weather Service (NWS, 2012) – Córdoba International Airport station and ii) modeled hourly surface and upper air data from the application of the Weather Research and Forecasting model (WRF, 2012). The NWS data set comes from the nearest met station, located 15 km away from the study area. The WRF model was implemented due to it being a significant distance between the met station and the study area, and because the NWS performs only once-a-day upper air soundings (upper air data are usually measured by twice daily radionsonde soundings, taken at 00 and 12Z (Greenwich time), and this is the method usually employed for local-scale dispersion modeling) (USEPA, 2011). These two sources of meteorological data (NWS and WRF) were used in order to compare and analyze their impact in the validation study.

The NWS provided surface measured met data for: temperature, relative humidity, pressure, wind speed, wind direction and cloud coverage and ceiling. The surface and upper air modeled met data were obtained from the application of WRF by an environmental consultant agency (Quality Environmental Consulting). The pseudo-met station was located at Yocsina at longitude −64.3713 and latitude −31.4453 coordinates. The WRF reanalysis data were provided by the National Centers for Environmental Prediction and the National Center for Atmospheric Research (NCEP/NCAR) Global Tropospheric Analyses with 1° × 1° spatial resolution (~111 km × 111 km) and temporal resolution of 6 h. The vertical structure of the model includes 27 layers (sigma levels) covering the whole troposphere. The selected period for the weather simulation was from June 25th to August 25th, 2012. The model runs were performed for three one-way nested domains. The spatial resolution of the outer domain (D1) was 27 km (with 40 x 40 grid points), the middle one (D2) 9 km (with 46 x 46 grid points) and the innermost (D3) 3 km (with 46 x 46 grid points). The land use data for the pre-processing was based on the interpolation of the 24 categories of land use of the U.S. Geological Survey (USGS) at 30 arcsec, and 16 category top-layer soil types, both with spatial resolutions of 10′, 5′ and 2′, for the domains 1, 2 and 3, respectively. The physics options used in the simulations include: (a) microphysics: 27 km (New Thompson et al., 2004), 9 km (WRF Single-Moment 6-class), 3 km (WRF Single-Moment 6-class); (b) cumulus scheme: 27 km (Grell 3D), 9 km (Betts-Miller-Janjic), 3 km (Betts-Miller-Janjic); (c) Planetary Boundary Layer parameterization: Yonsei University; (d)

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surface physics: Noah Land Surface Model (LSM); (e) longwave radiation: Rapid Radiative Transfer Model (RRTM) scheme and (f) shortwave radiation: Dudhia scheme.

As for terrain data, UTM - NAD27 (North American Datum of 1927) nodes spaced every 3 arc-seconds (approximately 90 m) were preprocessed in AERMAP.

2.3.3. AERMOD input data

AERMOD is a steady-state plume model which calculates atmospheric dispersion based on the planetary boundary layer turbulence structure and scaling concepts, including treatment of both surface and elevated sources over both simple and complex terrain. It is able to model multiple sources of different types including point, area and volume sources (USEPA, 2009). In the stable boundary layer, the dispersion is assumed to be Gaussian in both the vertical and the horizontal directions. In the convective boundary layer, the horizontal distribution is assumed to be Gaussian whereas the vertical distribution is described by a bi-Gaussian probability density function. AERMOD uses surface and profile meteorological data obtained from a single meteorological station. It incorporates a new approach to account for airflow and dispersion in complex terrain. AERMOD's terrain preprocessor, AERMAP, uses gridded elevation data to calculate a representative terrain-influence height, also referred to as the terrain height scale. The terrain height scale, which is uniquely defined for each receptor location, is used to calculate the dividing streamline height (USEPA, 2002; Neshuku, 2012; Tartakovsky et al., 2013).

In the present study, AERMOD was used to assess the concentration of TSP emitted from 224 cement and background sources which included: 8 stacks, 3 limestone quarries, 13 material storage piles, 18 agricultural fields and 182 paved and unpaved segment roads (public and industrial), where also the influence of 32 industrial building was analyzed (Fig. 2). The total modeled area was 40 km^2 and the rural area option was chosen. Terrain data every 90 m were preprocessed in AERMAP and the complex terrain option for receptors was chosen. Hourly modeled (WRF) and measured meteorological data (NWS) were preprocessed in AERMET for the validation period (2 months) and NWS data only for the year 2012. For the model validation, 2 receptors (corresponding to the TSP monitoring stations) were located, and for iso-concentration maps, 1491 receptors were located 1.5 m above ground level every 150 m (main grid) and 50 m (subgrid, surrounding the stacks). The output files obtained were for 24 h TSP concentration values.

2.3.4. Validation of AERMOD results

The performance evaluation of the AERMOD application was carried out by comparing the predicted and measured results during the validation period. There are a number of performance measures used to evaluate dispersion models, which include mean, standard deviation, fractional bias (FB), geometric mean bias (GM), index of agreement (IOA), factor of two (FAC2) and the mean square normalized error (NMSE) (Kumar et al., 2006; Singh et al., 2006). The model performance was evaluated by comparing these

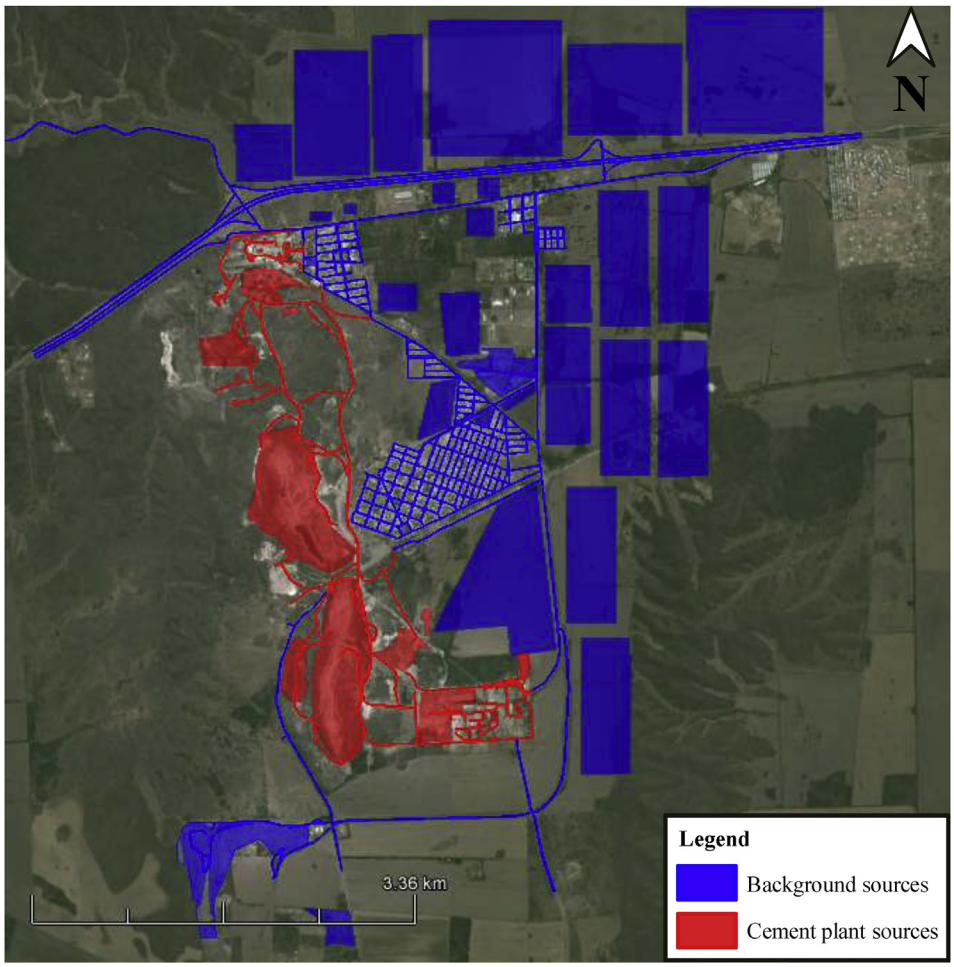

Fig. 2. Industrial and background modeled sources.

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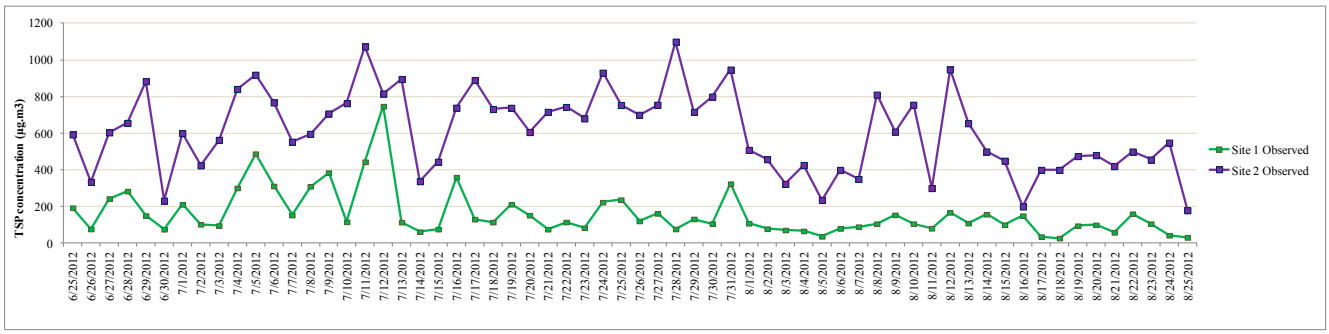

Fig. 3. Time series of TSP concentration values (μg m^{-3}) experimentally obtained at sites 1 and 2 in the validation period (from June 25th to August 25th, 2012).

measures of performance from the observed values with the modeled ones. These performance measures can be found in Section S2.

3. Results and discussion

3.1. TSP measured concentration values

As can be seen in Fig. 3, the observed concentration values showed pronounced variations, with the particularity that both sites had similar increasing/decreasing trends. This suggests that these fluctuations had the same origin and/or were subject to the same meteorological conditions. For Site 1 (Table 1), the measured concentration values varied between 27.4 and 747.6 μg m^{-3} (average 159.6 μg m^{-3}), and for Site 2 these varied between 179.4 and 1099.8 μg m^{-3} (average 612.1 μg m^{-3}). Moreover, 25 of the 62 sampling days (40%) at Site 1 exceeded the World Health Organization recommended guideline value of 150 μg m^{-3} for 24 h (WHO, 2000), while Site 2 exceeded this value for the entire sampling campaign.

Table S1 (Supplementary Material) presents the TSP concentration values (μg m^{-3}) obtained in this study and in other regions of the world. The maximum TSP value for Site 1 (Yocsina) was higher than values reported in other regions; however, its average value (Table 1) indicates that during the sampling this level of concentration was uncommon. The minimum value obtained for Site 1 was lower than values obtained in other regions, which may be representing background concentrations at Yocsina. On the other hand, the minimum, average and maximum TSP concentrations for Site 2 (Malagueño) were significantly higher than those values reported in other regions, except in the case of the measurements obtained in the vicinity of a cement plant in Nigeria during the dry season (Olaleye and Oluyemi, 2010), in which the values were similar to Site 2.

3.2. TSP atmospheric dispersion modeling

3.2.1. TSP emissions from industrial and non-industrial sources

Tables 2–4 present data referred to sources operations and the TSP emission factors and emission rates for point (stacks) and diffuse sources (quarries, material storage piles, and paved and unpaved roads). The information needed for the emission factor estimation was obtained either from these cement plants; bibliographical research of similar cement plants and/or from the AP-42 guidance. For the dispersion calculations for TSP, emissions were considered to be constant throughout the operational hours, as the cement factories usually only stop functioning for kiln cleaning. Table S2 summarizes the most important data for estimating the TSP emission factors from these cement plants. The activity rates from each source of emission were obtained from personal communication with former employees. Table S3 presents emission rate estimations from agricultural fields (diffuse sources) close to the neighborhoods of Yocsina and Malagueño.

Stacks (Table 2): each cement plant has four active stacks: at the main kiln, the clinker cooler, the cement mill and the raw mill; and bag filters to control these emissions. The annual operating time of the kilns was estimated to be 8040 h, considering that kilns run 24 h a day except during maintenance stops (USEPA, 2008).

Limestone quarries and stockpiles (Table 3): for limestone quarries, primary, secondary and tertiary crushing; screening and truck loading and unloading were considered. For stockpiles areas, these were obtained from the analysis of satellite images. The average wind speed (U) was estimated in an annual base using the meteorological data obtained, and the moisture content from stockpiles (M) was taken from the AP-42 guidance as surrogated data.

Agricultural fields (Table S3): the main crop cultivated in the area is wheat, and the field areas were obtained from the analysis of satellite images.

Paved and unpaved roads (Table 4): the roads activity rate is characterized by a variable called VKT (Vehicle Kilometer Traveled), which represents the kilometers traveled by the vehicle fleet. The vehicle count on public roads was done for 1 h at peak hour on several streets, whereas on industrial roads (a) unpaved roads between quarries and the crusher, it was estimated considering the annual cement production of each cement plant and the truck load capacity in 12 h of daily work. In the case of (b) industrial paved roads, the estimation was similar, but in this case the circulating fleet was estimated counting vehicles entering and leaving each cement plant. The vehicle counts were later extrapolated to the rest of the streets, taking into account similarities in commercial and industrial activity. The vehicle weight (W) was estimated through bibliographic research of industrial trucks used in cement manufacturing; the vehicles average speed (S) was estimated from the vehicle count; the number of days with rainfall above 0.254 mm (P) was estimated in an annual base using the meteorological data; and silt content (s), silt load (sL) and moisture content (M) for roads were obtained from the AP-42 guidance as surrogated data.

Table 1
Descriptive statistics of the TSP concentration values experimentally obtained at monitoring sites 1 (Yocsina) and 2 (Malagueño).

Site	n	Mean	S.D.	S.E.	% C.V.	Min.	Max.
1	62	156.9	126.9	16.1	79.5	27.4	747.6
2	62	612.1	220.4	28.0	36.0	179.4	1099.8

S.D.: Standard deviation; S.E.: Standard error; %C.V.: Coefficient of variation.

Table 2
Data referred to the operation of point sources (stacks) from cement plants 1 (Yocsina) and 2 (Malagueño) and estimated TSP emission rates.

Type		Description	Height [m]	Diameter [m]	Gas temp. [K]	Gas speed [m/s]	EF [kg/t][a]	Activity rate [t/yr][b]	Emission rate [g/s]
Point sources (stacks)	Cement plant 1	Main kiln	31	2.2	455	16.0	0.1 (preheater/precalciner kiln with bag filter)	1 188 000	3.765
		Clinker cooler	32	2.0	368	11.5	0.068 (clinker cooler with bag filter)	1 188 000	2.560
		Cement mill	15	2.5	351	7.0	0.0042 (cement mill with bag filter)	792 000	0.105
		Raw mill	20	2.0	338	13.0	0.0062 (raw mill with bag filter)	1 425 600	0.280
	Cement plant 2	Main kiln	36	4.0	403	12.5	0.1 (preheater/precalciner kiln with bag filter)	1 980 000	6.274
		Clinker cooler	20	3.0	398	9.0	0.068 (clinker cooler with bag filter)	1 980 000	4.267
		Cement mill	28	1.2	368	8.0	0.0042 (cement mill with bag filter)	1 425 600	0.190
		Raw mill	42	2.0	358	6.5	0.0062 (raw mill with bag filter)	2 376 000	0.467

[a] EF: TSP emission factor for point sources [kg/t].
[b] Productivity of the main kiln, clinker cooler and mills from cement plants 1 (Yocsina) and 2 (Malagueño) [t/yr].

With respect to point sources (Table 2), the stacks from the main furnace and the clinker cooler were the ones with the highest emission factors, since pyroprocessing (which includes the main furnace clinker cooler) is the largest generator of particulate matter in the process (USEPA, 1995). Regarding diffuse sources (Tables 3 and 4 and S3), material storage piles and the unpaved industrial roads had the highest emission rates.

Qualitative uncertainties: emission factors have long been fundamental for developing emission control strategies, determining applicability of permitting and control programs, ascertaining the effects of sources and appropriate mitigation strategies (USEPA, 1997). Even though, in general, the emission factor estimation technique is very useful, there are uncertainties involved in emissions estimation related to the input data quality, the use of surrogate data and the use of parameters. Other uncertainties involved include model formulation and its subsequent application outside the validation range (Frey et al., 2006). In the cement manufacturing process, particularly in the case of fugitive sources, dust concentrations released vary considerably depending on the nature of local sources, and other factors such as topography and the general weather conditions (Abdul-Wahab, 2006). In addition, emitting activities (e.g. mobile sources) are time-varying, and the knowledge about this is often incomplete.

Table 3
Data referred to the operation of area sources (limestone quarries and stockpiles) from cement plants 1 (Yocsina) and 2 (Malagueño) and estimated TSP emission rates.

Type	Description	EF [kg/t][a]	Activity rate [t/yr][b]	Area [m^2]	Emission rate [g/s]	Emission rate [$g/s.m^2$]
Quarries[c]	Quarry 1	0.0027 (primary crushing) 0.0027 secondary crushing)	1 900 800	640 662	1.335	2.08E-06
	Quarry 2	0.0027 (tertiary crushing) 0.0125 (screening)	3 168 000	574 144	2.224	3.87E-06
	Quarry 3	0.0015 (conveyor transfer point) 8E-6 (truck unloading) 5E-5 (truck loading)	3 168 000	110 367	2.224	2.02E-05
Area sources Cement Plant 1	Pile next to the cement mill stack		1 900 800	6125	2.833	4.63E-04
	Pile between clinker hall and limestone cilos	$0.0016\,k\frac{\left(\frac{U}{2.2}\right)^{1.3}}{\left(\frac{M}{2}\right)^{1.4}}$ [1]		7000	2.833	4.05E-04
	Area 1 next to primary crushing			6864	2.833	4.13E-04
	Area 2 next to secondary crushing	U = 4 m/s; M = 0.25%		108 026	2.833	2.62E-05
Area sources Cement Plant 2	Area 1 next to Cement Plant 2		3 168 000	95 035	4.721	4.97E-05
	Area 2 next to Cement Plant 2			68 565	4.721	6.89E-05
	Area 3 next to Cement Plant 2			149 423	4.721	3.16E-05
	Pile 1 next to coke mill	$0.0016\,k\frac{\left(\frac{U}{2.2}\right)^{1.3}}{\left(\frac{M}{2}\right)^{1.4}}$ [1]		13 270	4.721	3.58E-04
	Pile 2 next to coke mill			82 713	4.721	5.71E-05
	Pile 1 next to coke mill			10 806	4.721	4.37E-04
	Area 4 next to Cement Plant 2	U = 4 m/s; M = 0.25%		17 090	4.721	2.76E-04
	Crushing area			446 972	4.721	1.06E-05
	Area 5 next to Cement Plant 2			77 897	4.721	6.06E-05
	Area 3 next to Quarry 2			96 394	4.721	4.90E-05

U: mean wind speed [m/s].
M: material moisture content [%].
k: particle size multiplier (dimensionless) k = 0.74.
[1] TSP emission factor equation for area sources.
[a] EF: TSP emission factor for area sources (limestone quarries and material storage piles) [kg/t].
[b] Productivity of the limestone quarries from cement plants 1 and 2.
[c] Includes: primary, secondary and tertiary crushing; screening and truck loading and unloading.

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Table 4
Data referred to the operation of area sources (public and industrial paved and unpaved roads) from the neighborhoods of Yocsina and Malagueño and estimated TSP emission rates.

Type		Description/location	EF [g/VKT][a]		Activity rate [VKT/h]		Emission rate [g/s m^2]
					Segment road [km][b]	Vehicle count [Veh/h][c]	
Public	Unpaved roads (UR)	At La Perla neighborhood	$281.9\left[\frac{k(s/12)(S/30)^{0.3}}{(M/0.5)^{0.3}} - 0.00047\right]\left(\frac{365-P}{365}\right)$ [1]	498.9	0.32	26	1.8E-05
		At Malagueño neighborhood		498.9	0.23	22	1.6E-05
		At Malagueño neighborhood		498.9	0.74	32	5.0E-08
		At Malagueño neighborhood	$s = 16.6\%$; $S = 15.5$ mph; $M = 6.5\%$; $P = 160$	498.9	0.75	46	7.1E-08
	Paved roads (PR)	Collector Distributor Road (North)		1.074	3.20	166	2.6E-07
		Highway RN 20 (North)		1.074	7.80	834	1.3E-06
		Highway RN 20 (South)		1.074	6.70	1054	1.6E-06
		Collector Distributor Road (South)		1.074	6.10	586	9.1E-07
		Highway RP E81 (segment a)		1.074	2.40	106	1.6E-07
		Entrance to Cement Plant 1		1.074	0.38	38	5.9E-08
		At Yocsina neighborhood	$k\ (sL)^{0.91}(W)^{1.02}\left(1-\frac{P}{4N}\right)$ [2]	1.074	0.43	32	5.0E-08
		At Yocsina neighborhood		1.074	1.15	52	8.1E-08
		At Yocsina neighborhood	$sL = 0.2$ g m^{-2}; $W = 1.6$ t; $N = 365$; $P = 160$	1.074	0.50	96	1.5E-07
		At Malagueño neighborhood		1.074	0.53	104	1.6E-07
		At Malagueño neighborhood		1.074	0.23	128	2.0E-07
		Highway RP E81 (segment b)		1.074	1.50	86	1.3E-07
		Entrance to Cement Plant 2		1.074	3.20	112	1.6E-07
		At Malagueño neighborhood		1.074	0.61	156	2.4E-07
		At Malagueño neighborhood		1.074	0.80	204	3.2E-07
		At Malagueño neighborhood		1.074	0.80	136	2.1E-07
		At Malagueño neighborhood		1.074	0.53	76	1.2E-07
Industrial	UR	Roads from Cement Plant 2 to quarry	$281.9[k(s/12)^{0.7}(W/3)^{0.45}]\left(\frac{365-P}{365}\right)$ [3]	1701.8	4.44	84[d]	4.1E-04
		Roads from Cement Plant 1 to quarry	$s = 8.3\%$; $W = 30.5$ t; $P = 160$	1701.8	3.81	50[e]	2.5E-04
	PR	Internal road from Cement Plant 1	$k\ (sL)^{0.91}(W)^{1.02}\left(1-\frac{P}{4N}\right)$ [4]	158.6	1.67	38[f]	1.7E-05
		Internal road from Cement Plant 2	$sL = 11$ g m^{-2}; $W = 6$ t; $N = 365$; $P = 160$	158.6	2.42	70[g]	3.2E-05

k: particle size multiplier (dimensionless).
[1] Empirical constants for TSP for vehicle traffic at unpaved public roads: k = 6.0; a = 1; c = 0.3; d = 0.3.
[2] Empirical constants for TSP for vehicle traffic at paved public roads: k = 3.23.
[3] Empirical constants for TSP for Terex trucks traffic at unpaved industrial roads: k = 4.90; a = 0.70; b = 0.45.
[4] Empirical constants for TSP for trucks traffic at paved industrial roads: k = 3.23.
U: mean wind speed [m/s].
M: moisture content [%]. When no data on the moisture content is available, default data from USEPA (1997) is used.
s: surface material silt content [%]. Road to/from (average content). When no data on the silt content is available, default data from USEPA (1997) is used.
W: mean vehicle weight [t]. In the case of industrial roads, the average weight of the empty and loaded vehicle is considered.
P: number of "wet" days with at least 0.254 mm (0.01 in) of precipitation during the averaging period.
S: mean vehicle speed [mph].
sL: road surface silt loading [g/m^2]. When no measurements on the surface silt loading are available, default data from USEPA (1997) is used.
N: number of days in the averaging period.
[a] EF: Emission factor [g/VKT]; VKT: Vehicle Kilometers Traveled.
[b] Segment road length [km] where vehicle count was done.
[c] Number of vehicles circulating within the hour count.
[d] Traffic flow calculated from the activity rate of the quarry from Cement Plant 2 of 3 168 000 t/yr working 12 h/day.
[e] Traffic flow calculated from the activity rate of the quarry from Cement Plant 1 of 1 900 800 t/yr working 12 h/day.
[f] Traffic flow estimated from the truck count at the "Entrance to Cement Plant 1" street.
[g] Traffic flow estimated from the truck count at the "Entrance to Cement Plant 2" street.

3.2.2. Meteorological data analysis

The meteorological variable summary from the NWS and WRF data for the validation period (from June 25th to August 25th, 2012) is presented in Table S4. Figs. S2 and S3 show the wind roses (from NWS and WRF data), indicating the wind direction flow vector. Despite sampling being conducted in winter, maximum temperatures were recorded up to 32.9 °C, which are not common for this season (Table S4). In addition, the winter season is normally characterized by moderate relative humidity and a lack of rainfall, but the relative humidity at the time of the study was high and there were also three episodes of rainfall. With respect to wind speed and direction, moderate speeds were detected (average 3.5 m/s), primarily from the NNE, N and S.

3.2.3. Validation of AERMOD results

Since the concentration is a random variable, it must be analyzed statistically (Csanady, 1973; Lewellen and Sykes, 1989). Chang and Hanna (2004) and Kumar et al. (2006) have argued that any model must meet some requirements in order to be acceptable, and these are presented in Table S5. The validation of AERMOD results was done (a) using measured surface met data and modeled profile met data (NWS); and (b) using modeled surface and profile met data (WRF). Table S6 presents the performance indices obtained with the NWS and WRF data, and Fig. S4 shows the scatter plots and the FAC2 between observed and modeled TSP concentration values for Sites 1 and 2, respectively. Only twice the statistical values were not within the ranges proposed by Chang and Hanna (2004) and/or Kumar et al. (2006), indicating that the validation of the model using both meteorological data were acceptable, and that this model can be applied to assess the air quality in the study area (Table S6). Considering then that the standard deviations were lower using the data from NWS and that the Index of Agreement (IOA) values were closer to unity, these met data were used in the model application and diagnosis of air quality in the town of Malagueño (for the elaboration of time series graphs and TSP iso-concentration maps).

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Fig. S5 shows the time series graphs of observed and modeled TSP concentration values in the validation period for both monitoring sites (using NWS data). While the daily values did not agree on certain occasions, the overall modeled values showed a similar pattern to those observed, and this was reflected in the implementation of the validation indices. It is important to underline the difficulty of achieving acceptable PM validation results, considering the amount of emission sources, the variability in the behavior of these sources (most of them fugitive type) and the uncertainties that come from the emission factors estimation technique.

3.3. Assessment of TSP impact at the study area

The TSP iso-concentration maps (μg m^{-3}) obtained from the validation period for all the identified emission sources in the area, are presented in Fig. S6 (a, b and c). From this figure, it can be seen that the industrial emissions from the cement plants contributed to the highest and more widespread TSP concentration values in the area, showing maximum values in the order of 3200 μg m^{-3}. While these maximum values were remarkably high, these were concentrated in the vicinity of the emission sources, with the values at 1 km away falling by up to 800–1000 μg m^{-3}. From these figures, it is noticeable that Site 1 was more affected to the cement plant TSP emission sources, while Site 2 to background TSP emission sources (soil resuspension from agricultural fields and paved and unpaved public roads). In the neighborhoods of Yocsina and Malagueño, the maximum levels were up to 800 and 1250 μg m^{-3}, respectively, which were within the order of magnitude obtained in the TSP samples collected at both monitoring sites and greatly exceeded the WHO recommended guideline (150 μg m^{-3}). The more distant residential areas were not affected by the TSP concentration values emitted from this industrial complex. Although the cement plants were the main TSP emitters (Fig. S6.b), the background emission sources (public paved and unpaved roads, agricultural fields and other area sources) also played an important role.

In order to analyze the TSP contributions from each industrial source type, Fig. 4 shows the concentration values (μg m^{-3}) for quarries, material storage piles, industrial roads (paved and unpaved) and stacks. With respect to TSP distribution in quarries, the highest concentration values were centralized in the vicinities of this source, but also affected part of the neighborhoods of Malagueño and Yocsina, and distributed in the directions N, S and E. However, at short distance the concentration values decreased and

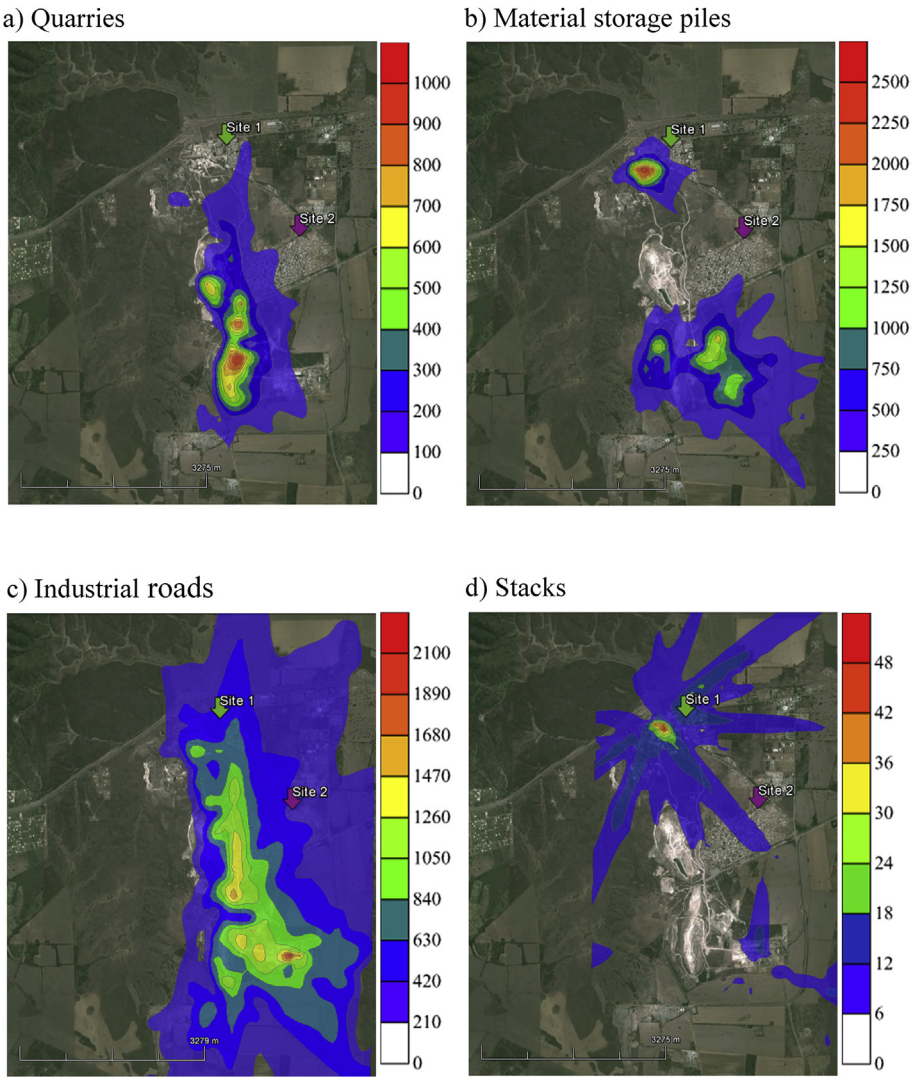

Fig. 4. TSP iso-concentration maps (μg m^{-3}) obtained for the validation period (from June 25th to August 25th, 2012) for the different industrial source groups: (a) quarries; (b) material storage piles; (c) industrial roads (paved and unpaved) and (d) stacks.

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came to be within acceptable values. It is likely that this source was underestimated, since possible emissions from excavation and blasting material were not considered. As for material storage piles, this emission source reached the highest TSP concentration values (up to 2500 μg m^{-3}) being the main PM emitter in the area. Cement Plant 1 is noticeably close to Yocsina neighborhood, and this fugitive dust source largely impacted on the residential area. Regarding industrial roads, the highest concentration of TSP values (up to 2100 μg m^{-3}) were found at the unpaved roads leading to the quarries, where Terex trucks travel daily carrying considerable loads of material. In the case of point sources (stacks), the TSP concentration values were not significant with respect to the total values found in the area (maximum value: 48 μg m^{-3}). However, these emissions contain heavy metals, PAHs and other pollutants adsorbed to the cement dust (IFC, 2007), and therefore need to be taken into account. Furthermore, although higher concentration values were found in the vicinity of the stacks (indicating building downwash effects) dispersion was also observed to the NE, E, SE and SW directions. Moreover, the TSP concentration values from stacks belonging to the Cement Plant 2 were significantly lower compared to the ones from Cement Plant 1.

Fig. S7 presents the distribution map of the maximum TSP concentration values (μg m^{-3}) for all sources for the year 2012. Similarly to the validation period, the annual iso-concentration map shows that the highest TSP levels were found mainly at unpaved industrial roads and active stockpiles, which also noticeably exceeded the WHO guideline.

4. Conclusions

Field TSP measurements showed that the area under study is greatly affected by particulate matter, considering the repeated exceedances of the WHO guideline value, therefore involving potential risks of exposure to the population of the town of Malagueño. The considerable differences found between the two monitoring sites, were mainly due to the fact that nearby Site 2 the streets were mostly gravel, and particulate matter was emitted from the drag of the wind and passing vehicles. As for the modeling outcomes, these suggest that the main sources of PM were industrial roads and stockpiles, with stacks being negligible in comparison. However, it was also found that despite cement plants being the main responsible factor for the PM levels in the area, the background sources were also of consideration, especially near Site 2. With respect to the uncertainties involved in this study, it can be concluded that by obtaining (i) “on-site” meteorological data; and (ii) a higher quality data related to emissions, uncertainties will decrease and the model performance will probably increase, thus allowing more reliable results on the study area.

Being public health an aspect of relevant concern, future studies will include the accumulation of heavy metals and Polycyclic Aromatic Hydrocarbons in TSP samples, the environmental risk assessment for exposure of the of the inhabitants from Malagueño and the source apportionment of these pollutants.

Conflict of interest

There is no conflict of interest.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.apr.2015.08.009.

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