

Short communication

Implementation of resuspension from paved roads for the improvement of CALIOPE air quality system in Spain

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ABSTRACT

Emissions of resuspended particles from paved roads (RPR) have received an increasing attention in air quality modeling, since chemistry transport models are generally unable to reproduce particulate levels near road traffic. However, among the models operated in the forecasting mode in Europe, there is a lack of systems including the contribution of RPR on an hourly basis. Therefore, this contribution describes the inclusion of the resuspension of particulate matter within HERMES emission model (operationally implemented in the CALIOPE forecasting system) and the improvements obtained in the simulations of the PM₁₀ mass over a domain covering Spain for a whole year of simulation (2004). The results indicate a remarkable improvement of the PM₁₀ predictions, reducing the biases and errors around 15–18% and 2.6 $\mu\text{g m}^{-3}$ for the average bias in all Spain. The RPR emission have strong local effects on the modeled particle concentration in or near the largest urban zones (up to 7 $\mu\text{g m}^{-3}$ as the annual average), albeit those positive effects are more limited in background areas, since the deposition mechanism was found to be a significant sink for RPR in the chemistry transport model. This approach is accurate and effective in order to improve the prediction of the PM₁₀ mass and becomes essential to achieve the standards set in the regulations for modeling applications.

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1. Introduction

Field measurements in urban areas or megacities showed elevated levels of PM₁₀ in the vicinity of roads coming from the contribution from resuspended particles from paved roads (Lenschow et al., 2001; Querol et al., 2004; Ketzler et al., 2007; Querol et al., 2008). However, it is widely known that chemistry transport models are generally unable to reproduce particulate levels near road traffic. For instance, Vautard et al. (2005) and Pay et al. (2010) showed that simulated aerosol loadings, using the current knowledge on aerosol mechanisms, show a pervasive trend towards an underestimation by up to 30–50%.

Hence, since a decade, emissions of resuspended particles from paved roads (later referred to as RPR) have received an increasing attention in air quality modeling and forecasting. The first model attempts to quantify particle emissions resulting from abrasion of

vehicle components and road surface were presented by Rauterberg-Wulff (2000), LUA (2000), Venkatram (2000) following a preliminary formulation by US-EPA (1997), later improved by US-EPA (2006). Empirical emission factors are presently retrieved from various field campaigns at different locations and road types of central and western Europe (Düring et al., 2002, 2004; Thorpe et al., 2007; Ketzler et al., 2007; Gehrig et al., 2004). More recently, the resuspendable amount of particles was experimentally evaluated in Barcelona (Spain) by Amato et al. (2009a,b, 2010), demonstrating that the contribution from road dust resuspension in the coarse fraction is comparable or even higher than the exhaust emissions in this urban environment. In this sense, Spain represents a paradigmatic case of study, since regions with low precipitation climates favour the accumulation of paved road dust and the resuspension of loose material on the road surface (Abu-Allaban et al., 2003; Thorpe and Harrison, 2008).

Nowadays, a total of 23 model systems routinely simulate the air quality over Europe, with seven systems also operated in the forecasting mode (Menut and Bessagnet, 2010). Because of the complexity of the physical processes leading to resuspension and the difficulties in measuring and/or modeling their effects on ambient PM levels, CALIOPE (Baldasano et al., 2008a) is the unique system including the contribution of resuspension of particulate

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matter on an hourly basis, to the authors' knowledge. Emission inventories of non-exhaust emissions are scarce despite their suitability for researchers and regulating agencies for managing air quality and PM reduction measures (Oxley et al., 2009). Therefore, this contribution intends to describe the inclusion of RPR within HERMES (the emission model in CALIOPE) and the improvements obtained in the simulations of the PM10 mass over a domain covering Spain for a whole year of simulation (2004).

2. Methods

2.1. The CALIOPE modeling and forecasting system

The CALIOPE project, funded by the Spanish Ministry of the Environment and Rural and Marine Affairs, aims at establishing an air quality forecasting system for Spain (Baldasano et al., 2008a). CALIOPE encompasses a high-resolution air quality forecasting system being applied to Europe as a mother domain (12 km × 12 km, 1 h) (Pay et al., 2010) as well as to Spain as the nested domain (4 km × 4 km, 1 h) (Baldasano et al., 2009). Such high resolution of the model system is made possible by its implementation on the MareNostrum super-computer hosted by the Barcelona Supercomputing Center-Centro Nacional de Supercomputación (BSC-CNS). CALIOPE is a complex ensemble of models that integrates a meteorological model (WRF-ARWv3.0.1.1), an emission processing model (HERMES), a mineral dust dynamic model (BSC-DREAM8b), and a chemical transport model (CMAQv4.5) together in an air quality modeling and forecasting system. Current forecasts are available through <http://www.bsc.es/caliope>.

The HERMES model uses information and state-of-the-art methodologies for emission estimations. It calculates emissions by sector-specific sources or by individual installations and stacks following a bottom-up approach. Raw emission data are processed by HERMES in order to provide a comprehensive description of the emissions to the air quality model. Emissions used for the Iberian Peninsula are derived from the aggregation in space from 1 km × 1 km dataset to 4 km × 4 km. A more detailed description can be found in Baldasano et al. (2008b).

Specifically, the on-road traffic emissions module takes into account 72 diesel and gasoline vehicles categories according to COPERT III-CORINAIR methodology (Ntziachristos and Samaras, 2000; EEA, 2006); divided by fuel type, vehicle weight, age of the vehicle and cubic capacity; each of them with its specific emissions factors, defined as a function of the circulation speed. The model includes the definition of the road network, dividing it in stretches with specific temporary disaggregating profiles (distinguishing day-type: weekday-holiday, and month), specific average speed, daily average traffic (number of vehicles per day), stretch length, route type (highway, road or urban) and circulation zones. The emissions account in the on-road traffic module considers hot exhaust, cold exhaust and evaporative emissions. It also estimates particulate matter produced by brakes abrasion, tire wear and pavement erosion. The methodology described below presents a new approach to the inclusion of non-exhaust particulate matter originated by the resuspension from paved roads within the on-road traffic emission module of HERMES. The calculation of the emissions of RPR is proportional to the daily average traffic (DAT) measured in 20,934 observation points throughout Spain. Hourly emissions are based on coefficients calculated with the hourly distribution of the DAT (Baldasano et al., 2008b). The RPR emissions in the road section r of k -cell per hour are calculated with Eq. (1):

$$E_r^{ires}(k, \text{hourly}) = Crh \sum_{j=1}^n Clf Crd DAT_{rj}(k) L_r(k) F_r^{ires} \quad (1)$$

where Crh is the proportion of DAT at h -hour; Clf , the coefficient for daily traffic (labour day or weekend); Crd is the ratio between daily traffic for a specific month and DAT; $DAT_{rj}(k)$ is the daily average traffic for r -road section and j -vehicle category in k -cell (number of vehicles); $L_r(k)$ represents the length of the road section r in k -cell (km); n is the number of vehicle categories considered in HERMES emission model (72 in the present version); and F_r^{ires} is the road dust emission factor.

As a first approach, the estimated PM10 emission factor measured in Düring et al. (2002) for Berlin has been selected. Taking into account the car type, the applied F_r^{ires} is 88 milligrams per vehicle kilometre travelled (mg vkt^{-1}) for light duty vehicle (LDV) and 217 mg vkt^{-1} for high duty vehicles (HDV), respectively. In urban environments resuspension by road traffic tends to be significantly higher than on highways (Gehrig et al., 2004). However due to its complex dependency on vehicle speed, traffic intensity and behaviour (Kupiainen et al., 2005) the road type is not taken into account in the selection of the PM10 emission factor used in this study. The selected F_r^{ires} resulted higher than those obtained for LDV in UK (14–23 mg vkt^{-1} , Thorpe et al., 2007), Germany (57–67 mg vkt^{-1} , Ketzler et al., 2007), Switzerland (17–92 mg vkt^{-1} , Gehrig et al., 2004) but lower than those in Scandinavian countries (121–198 mg vkt^{-1} , Ketzler et al., 2007) and those estimated in the United States (63–780 mg vkt^{-1} , Abu-Allaban et al., 2003). The selected F_r^{ires} for LDV can be compared to the estimates for Spain by Schaap et al. (2009). In Spain mineral ambient PM10 concentrations in city backgrounds and near busy roads are larger than at central Europe (Pataud et al., 2004; Querol et al., 2004). Due to climatic conditions, resuspension flux is higher in Spain than in central Europe.

Under precipitation conditions, emissions decrease proportionally to the frequency of measurable precipitation. The precipitation effect is included by the application of a correction factor (f_{prep}) on an hourly basis following the equation proposed by US-EPA (2006). This factor is calculated with Eq. (2):

$$f_{\text{prep}} = \left(1 - 1.2 \times \frac{P}{N} \right) \quad (2)$$

where P is the number of hours with at least 0.254 mm of precipitation during the averaging period, and N is the number of hours in the averaging period. For a precipitation situation occurring continuously for over 48 h, the emissions of RPR are switched off in HERMES. The accumulated precipitation is hourly provided by the WRF-ARW model.

Despite the diameter of RPR spans several orders of magnitude, the peak in the mass distribution is usually in the coarse size range (aerodynamic diameter greater than 2.5 μm) and therefore the emissions of resuspended particles are speciated into this fraction in the HERMES emission model. In the CMAQ chemistry transport model, coarse particles are described with a unimodal lognormal distribution (Byun and Schere, 2006). The coarse particle mode is treated as dry and chemically inert with a fixed geometric standard deviation of 2.2 (Kelly et al., 2010).

For the present contribution two air quality simulations were performed over a domain covering Spain for a whole year of simulation (2004): (1) taking into account the RPR module within HERMES and (2) without the aforementioned module, here in after referred to as CALIOPE-IP-RPR and CALIOPE-IP, respectively.

3. Results

In order to evaluate the implementation of resuspended particles within HERMES emission model, the discussion focuses on a whole year of simulations (year 2004) over the Iberian Peninsula.

Fig. 1a and d displays the impact of the inclusion of RPR for PM10 emissions in Spain. This contribution is calculated by averaging the emitted loadings from the RPR module for each grid cell on the 24 h and is expressed in $\text{kg h}^{-1} \text{km}^{-2}$. The impact of this module is highest in cities of dense population such as Madrid, Barcelona or Valencia, with maximum average values increasing up to $10\text{--}18 \text{ kg h}^{-1} \text{km}^{-2}$ downtown the largest conurbations, doubling the emissions when no resuspension is included in HERMES. The radial highways with the largest daily average traffic undergo a mean increase of $1 \text{ kg h}^{-1} \text{km}^{-2}$ while the rest of the territory is mostly not affected by the RPR module.

Fig. 1b and e represents the average contribution of the RPR module in terms of PM10 air concentrations at the surface layer for the same period. Both emission and air concentration patterns display similar behaviors. The RPR module mainly affects PM10 concentrations in Madrid with an average annual increase of around 4 to $10 \mu\text{g m}^{-3}$ depending on the area of the city observed (with hourly maxima of $26 \mu\text{g m}^{-3}$ at rush hours, Fig. 2, up). This increase is similar in the city of Barcelona, with average increases ranging from 3 to $7 \mu\text{g m}^{-3}$ as the annual average. In other major Spanish particulate levels rather increase by 2 to $5 \mu\text{g m}^{-3}$. PM10 concentrations in more remote areas are not substantially impacted upon by the inclusion of the resuspension module. Fig. 1b and f shows that such a module may have strong local effects on the modeled particle concentrations in or in the vicinity of the largest urban zones, but not in the downwind areas from the local emitting sources.

In the domain of Spain, RPR emissions were included in the coarse fraction of the HERMES emission model. In urban areas of large cities coarse particulate matter by resuspension contributes by 40% to the total PM10 emitted, meanwhile PM coarse by road vehicle tyre and surface wear contributes by 6%. In this case, the main contributor to the PM coarse is resuspension (87%) followed by road vehicle tyre and surface wear (13%). The deposition mechanism was found to be an important sink for resuspended

particles from paved roads (Fig. 1c and f). Dry and wet depositions largely contribute to the air cleaning. Dry and wet depositions amounted $6\text{--}15 \text{ kg h}^{-1} \text{km}^{-2}$ depending of the urban area, that is, from 60% to 90% of the total coarse particles emitted in large cities and on the main roads. The high local contribution and the lack of transport of coarse particulate matter is associated to the short lifetime of this kind of particles in the atmosphere as a consequence of the deposition implemented in CMAQ chemistry transport model. In this sense, Zhang et al. (2006) and Jiang et al. (2006) report an over-prediction of geometric mean diameter by CMAQ and Elleman and Covert (2009) also reported that CMAQ aerosol size distributions are shifted to a larger size compared with observations. Therefore, the deposition of coarse-mode particles is too rapid in CMAQ (Kelly et al., 2010). This fact may importantly affect to PM10 predictions that will increase if over-predictions of the geometric mean diameter were corrected, because a large fraction of the coarse mode would fail to the accumulation mode and hence deposition velocities would become much smaller.

Fig. 2 represents the time series of observed and modeled concentrations of PM10 at the urban traffic station of Madrid-Recoletos (as a paradigmatic study case since at this station as the measuring instrument is mainly affected by road activities from the intense traffic nearby) and the average behaviour of all the urban traffic stations considered in the domain of study for which the PM10 increases are over $1 \mu\text{g m}^{-3}$. All of these stations are located in the large metropolitan areas of Madrid (Recoletos and Alcobendas), Castellón (Castellón) and Malaga (Ps. Martiricos) with over 500,000 inhabitants where urban streets are specifically described in the HERMES model. Madrid-Recoletos is located in a wide avenue in central Madrid. The RPR module induces a mean annual increase of $2.8 \mu\text{g m}^{-3}$ in PM10 air concentrations in the coarse fraction compared to the annual simulation including no RPR module, which can achieve $4.2 \mu\text{g m}^{-3}$ in several stations and areas (Fig. 1).

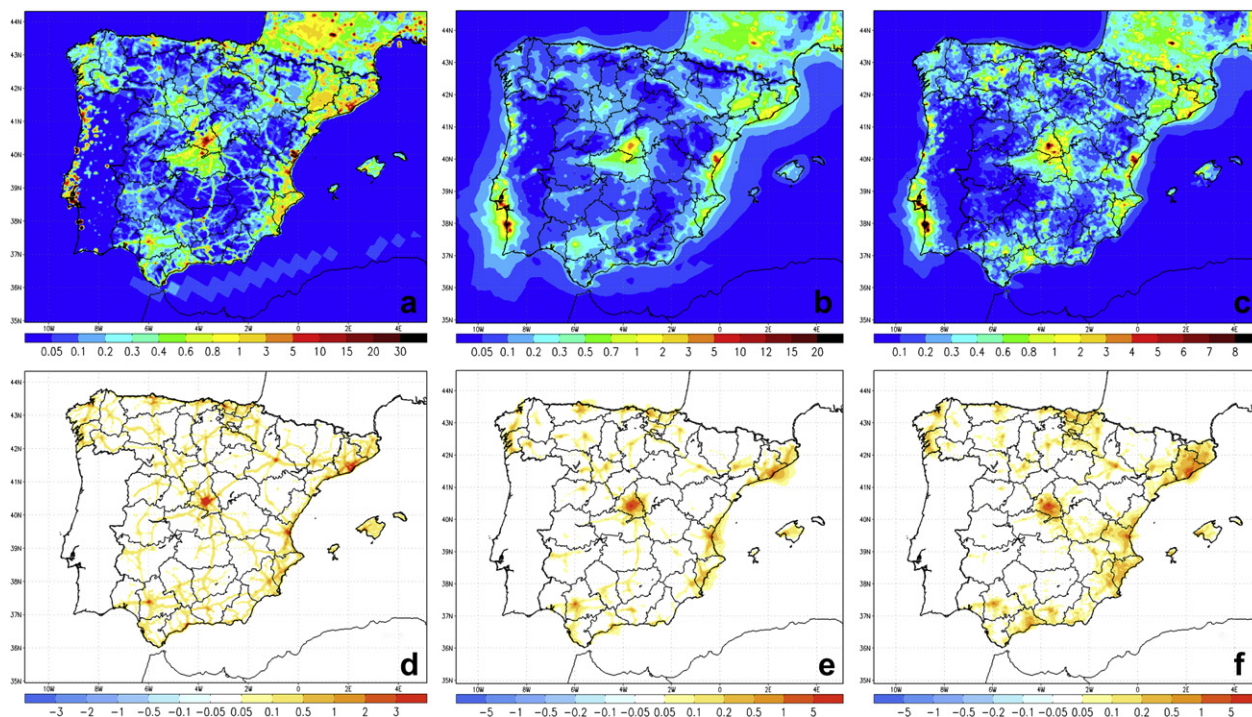


Fig. 1. (a) Emission loading ($\text{kg day}^{-1} \text{km}^{-2}$), (b) air quality contribution ($\mu\text{g m}^{-3}$) and (c) total deposition ($\text{kg day}^{-1} \text{km}^{-2}$) of the coarse particulate matter fraction modeled with CALIOPE modeling system over 2004 including RPR (CALIOPE-IP-RPR) with a resolution of $4 \text{ km} \times 4 \text{ km}$. Annual coarse particulate matter differences with simulation not including RPR (CALIOPE-IP) in (d) emission loading ($\text{kg day}^{-1} \text{km}^{-2}$), (e) air quality concentration ($\mu\text{g m}^{-3}$); and (f) total deposition ($\text{kg day}^{-1} \text{km}^{-2}$).

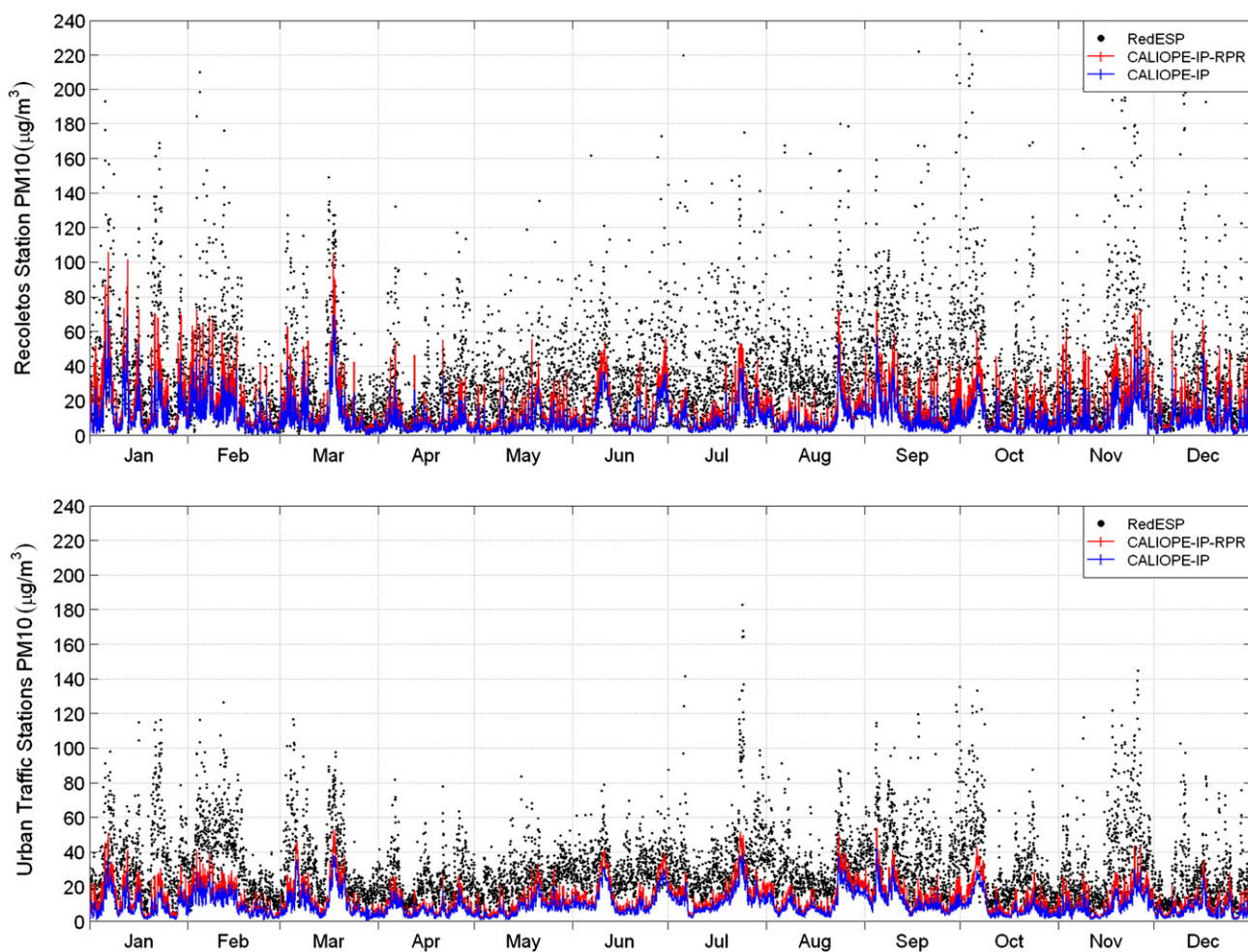


Fig. 2. Annual hourly series of PM₁₀ in the station of Madrid-Recoletos (up) and the average of urban traffic stations in Spain (bottom) over 2004: observations from the Spanish monitoring network, namely RedESP (black dots); model including RPR, namely CALIOPE-IP (red); and without the inclusion of the RPR module, namely CALIOPE-IP (blue).

Table 1 summarizes the performance of the modeling system in the scenarios with or without resuspension, evaluated by means of comparisons with ground-based observations from four urban-traffic stations belonging to the Spanish monitoring network. The selected urban-traffic stations are characterized based on the proposition by Garber et al. (2002) and are representative of pollution levels determined by emission from nearby traffic (roads, motorways, highways) over fully urbanized areas. When the RPR module is implemented the temporal evolution of modeled PM concentration does not change significantly, since resuspension emissions follow the same diurnal profile as exhaust PM emissions.

In this sense, correlation coefficients present a tiny variation from 0.41 to 0.48 globally. However PM concentrations are better reproduced with the inclusion of the RPR module, and the bias is reduced from $-21.5 \mu\text{g m}^{-3}$ to $-18.8 \mu\text{g m}^{-3}$. The root mean squared error also decreases when RPR is included, with improvements in the predictions ranging from $0.3 \mu\text{g m}^{-3}$ in Málaga-Ps. Martiricos to $3.0 \mu\text{g m}^{-3}$ in Madrid-Recoletos. The global improvement of the bias and root mean squared error is $2.6 \mu\text{g m}^{-3}$ and $1.9 \mu\text{g m}^{-3}$. The mean fraction scores are higher for all the stations when RPR emissions are included; the mean fractional bias and error improves by 18.3% and 15.5%, respectively.

Table 1

PM₁₀ annual statistics obtained with the CALIOPE system with and without the inclusion of RPR emissions at several urban traffic stations and global average from the air quality Spanish monitoring network. Statistics parameters are: number of data (N), observed mean (OM), modeled mean (MM), correlation coefficient (r), root mean square error (RMSE), mean bias (MB), mean fractional bias (MFB) and mean fractional error (MFE).

Station name	N	OM ($\mu\text{g m}^{-3}$)	CALIOPE-IP					CALIOPE-IP-RPR						
			MM ($\mu\text{g m}^{-3}$)	r	RMSE ($\mu\text{g m}^{-3}$)	MB ($\mu\text{g m}^{-3}$)	MFB (%)	MFE (%)	MM ($\mu\text{g m}^{-3}$)	r	RMSE ($\mu\text{g m}^{-3}$)	MB ($\mu\text{g m}^{-3}$)	MFB (%)	MFE (%)
Madrid-Recoletos	8740	36.3	10.2	0.49	38.1	-26.2	-104	109	14.4	0.50	35.1	-22.3	-80	90
Málaga-Ps. Martiricos	8416	16.2	12.2	0.42	13.0	-3.9	-46	70	13.3	0.43	12.7	-2.8	-34	62
Castellón-Castellón	4420	36.3	10.9	0.46	34.3	-26.8	-113	115	15.1	0.46	31.4	-23.0	-91	95
Madrid-Alcobendas	8598	36.0	7.1	0.55	40.6	-28.9	-133	135	8.8	0.55	39.1	-27.2	-118	120
Global	30171	30.6	9.3	0.38	33.7	-21.6	-101	108	12.8	0.41	31.4	-18.4	-80	92

Hence, the inclusion of RPR importantly contributes to the achievement in several stations of the criteria established by Boylan and Russell (2006) for model performance regarding particulate matter (mean fractional error $\leq 75\%$ and mean fractional bias $\leq \pm 60\%$).

During winter days without rain and weak dispersion (see January and mid-March in Fig. 2), PM₁₀ concentrations are increased by up to $30 \mu\text{g m}^{-3}$. These events are associated with meteorological conditions favoring the accumulation of pollutants. For instance, on the 22 January 2004, low wind velocities were monitored and modeled (aprox. $1\text{--}2 \mu\text{g m}^{-3}$), enhancing the increase of PM₁₀ concentrations up to $26 \mu\text{g m}^{-3}$. Melios et al. (2006) and Flaounas et al. (2009) evaluated PM emission factors in urban areas from the observation of PM₁₀ to NO₂ ratios at urban stations. Following their methodology we observed a linear dependency between the two species for the stations in Table 1 during winter stagnant situations with negligible wind. Observed PM₁₀/NO₂ ratios range about 0.7–0.9 while those calculated with HERMES vary between 0.4–0.6. This indicates that despite RPR module reduces the PM₁₀ concentration bias, PM₁₀ emissions from road traffic continue being underestimated in a factor between 3 and 5 during these episodes. This underestimation of urban PM may be related to the underestimation of real PM exhaust emission factors and the underestimation of urban PM sources like the local natural erosion emissions resulting from saltation processes, especially for the conditions in the Iberian Peninsula.

4. Conclusions

In this study, an emission module including resuspended particles from paved roads has been implemented in the CALIOPE air quality modeling and forecasting system (<http://www.bsc.es/caliope>) and assessed against observations for period covering the whole year 2004. The impact of the newly developed emission module considering RPR inclusion in the CALIOPE air quality modeling system highly improves the PM₁₀ predictions in those areas of dense population in Spain, around 15–18% for the mean fractional scores and $2.6 \mu\text{g m}^{-3}$ for the average bias in all Spain. The RPR emission may have strong local effects on the modeled particle concentration in or near the largest urban zones (up to $7 \mu\text{g m}^{-3}$ as the annual average and a contribution of $30 \mu\text{g m}^{-3}$ to the rush hour peaks). Those positive effects are, however, more limited in background areas, since the deposition mechanism was found to be a significant sink for resuspended particulate matter from paved roads in the chemistry transport model.

However, there are still some development tasks for a better reproduction of particulate matter in the CALIOPE system. Presently, further research is devoted to the inclusion of specific emission factors for the area of Spain (from Amato et al., 2010 instead of those of Düring et al., 2002 which represent the conditions of more northern latitudes). Moreover, the large uncertainties in local natural erosion emissions resulting from saltation processes need further development in emission estimates and modeling, especially for the conditions in the Iberian Peninsula. Despite the aforementioned limitations, the results demonstrate that this first approach is accurate and effective in order to improve the prediction of the PM₁₀ mass and becomes essential to achieve the standards set in the regulations for modeling applications.

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