

On the contribution of natural Aeolian sources to particulate matter concentrations in Europe: Testing hypotheses with a modelling approach

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Abstract

The significance of natural sources in the concentrations of particles smaller than 10 µm (PM10) in Europe is addressed. When considering only European anthropogenic emissions, chemistry-transport model simulations underestimate the PM10 concentrations by 30–50%, using the current knowledge about aerosol physics and chemistry. Along this article we hypothesize that the missing mass originates from natural sources like erosion dust entrainment and resuspension. The methodology consists in testing these hypotheses in the CHIMERE regional chemistry-transport model, and comparing the results with long series of PM10 measurements in Europe.

It is demonstrated that the introduction of background Saharan dust boundary conditions greatly improves the model simulation over Southern Europe, and to a smaller extent also over northern Europe. However to accurately simulate acute episodes of Saharan dust transport time-resolved boundary conditions need to be used. Local erosion occurring over European regions is also considered. The introduction of a simplified bulk scheme for online calculation of mineral dust emission makes the model skill improve everywhere in Europe, indicating that this process can be significant in Europe.

We finally assume that resuspension of material available on the ground, explains most of the remaining part of the missing part of PM10 average load. A simplified scheme for the corresponding emission, which depends on turbulence near the ground, is proposed. It also makes the model improve substantially, especially over Northern Europe. However sensitivity tests show that such an improvement can also be obtained by simply increasing the model secondary organic matter in aerosols by a factor of 3. We conclude that our results are consistent with the existence of a strong biogenic resuspension aerosol source, but more experimental work is required to ascertain this hypothesis.

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1. Introduction

The faithful simulation of aerosols and all their components is one of the most challenging exercises for air quality physicists. There are many obstacles to successful particle simulation: The chemistry of secondary organic aerosols is poorly known, the deposition and scavenging of aerosols are very sensitive to rain and clouds, which are poorly simulated variables. Above all the emissions, which are feeding the atmosphere with aerosols, are uncertain.

Primary aerosols come from a large variety of anthropogenic (industries, mobile sources, etc) and natural (natural forest fires, sea salt, wind blown dust, etc...) sources. Sources due to anthropogenic combustion of fossil fuel are probably the least uncertain, since the corresponding inventories are set up using the same classical methodologies as for gas species. Anthropogenic biomass burning is largely unknown. Often dominant in tropical countries, forest fires in Europe, mainly due to arsons (Simpson et al., 1999) probably contribute less significantly. The other primary particle sources are fugitive sources and come from a variety of anthropogenic and natural processes (see e.g. Countess et al., 2001), explaining the difficulty to inventory them.

The objective of this work is to study the impact of emissions that result from natural suspension or resuspension of particles available on or in the ground. Such fugitive emissions are due to Aeolian soil erosion or suspension into the atmosphere of particles deposited on the ground due to mechanical abrasion or biological degradation.

Erosion Emissions result from the *saltation* process (Shao, 2000), where small particles are extracted from the soil by the “bombing” from coarser soil particles due to wind transport. In the United States, the fraction of mineral dust found in PM_{2.5} exceeds 10% in most areas (Malm et al., 2004), and reaches 50% in dry areas. The contribution to total PM₁₀ is probably higher. In Europe, the contribution of mineral dust to PM₁₀ concentrations varies from 10% to more than 30% depending on location and season (Putaud et al., 2004).

Resuspension of natural material available on the surface is a potentially significant process. There are evidences of natural origin for part of the organic matter mass found in continental aerosols, found in the form of cellulose (Kunit and Puxbaum, 1996) or other substances originating from vegetation debris, insects, plant waxes, bacteria, spores, pollens, etc (Simoneit and Mazurek, 1982).

Little is known about natural fugitive dust emissions in Europe. In the EMEP emission inventory (Vestreng, 2003), suspension of anthropogenic particles is generally accounted for, in an average manner, but the suspension of natural particles is not yet considered.

One of the purposes of this article is to discuss whether natural erosion and resuspension could be important source processes in the PM₁₀ mass over Europe. Another objective is to propose methods for introducing these processes in regional air-quality models. The need for the development of such methods is clear while considering the underestimation of aerosol chemistry transport models (CTMs). In the EMEP model intercomparison study (van Loon, 2004), all models involved underestimate the PM₁₀ average concentrations by a factor ranging from about 30% to 50%. In this article, we make the strong assumption that this “missing mass” is entirely due to natural erosion and resuspension, and test this hypothesis by using a regional chemistry-transport model, CHIMERE (Schmidt et al., 2001; Bessagnet et al., 2004), by comparing the results, with and without the associated emission processes, with long series of measurements of PM₁₀ throughout Europe.

There is undeniably a lack of data to support and constrain the development of complex schemes for resuspension and erosion for all types of surfaces encountered in Europe. The bulk approaches proposed here stand for evaluation of the processes relevance. They will have to be made more realistic when data will become more abundant to support their validation.

Section 2 contains a description of the base model, without parameterizations and its performance for the simulation of aerosols. The impact of adding side boundary conditions for dust is discussed. Section 3 describes the simplified erosion scheme and its impact on the simulation quality. Section 4 describes the resuspension scheme and the model results obtained by using it. In Section 5 a critical discussion is given about the assumptions underlying this work. Section 5 also contains a conclusion.

2. The base model and simulations

2.1. The CHIMERE chemistry-transport model

In this study, we use the regional version of the CHIMERE CTM over Europe (Schmidt et al., 2001; Bessagnet et al., 2004, hereafter referred to as B2004). The model grid is almost identical to that used in B2004, running from 10.5W to 22.5E and from 35N to 57.5N with a resolution of 1/2 degree both in latitude and longitude. The domain covers most of Western Europe and the Western Mediterranean basin. The vertical grid contains 8 layers from surface to 500 hPa. The dynamics and gas-phase parts of the model are described in Schmidt et al. (2001), and improvements have successively been brought, some being described in Vautard et al. (2003) and in B2004. The model documentation can

be found on the web server <http://euler.lmd.polytechnique.fr/chimere>.

The only significant difference with previous studies is that, along this article, the model is forced by the 5th generation Penn State University model, MM5 (Dudhia, 1993), version 2.3.6, instead of using the ECMWF analyses as in B2004. In order to have low computational cost, MM5 is configured with a relatively low resolution (36 km) over a domain encompassing the CHIMERE domain, with 25 vertical levels. Classical options are chosen for parameterizations, such as the MRF PBL scheme for the boundary layer. We only made one significant change in the boundary layer scheme in order to have better accuracy on the friction velocity. As shown by Liu et al. (2004), in the V3.6.2 version, friction velocity can be largely overestimated, due to a too simplified estimation of the convective velocity scale. Here, we use the more classical formula proposed by Beljaars (1994), with a fixed height scale $Z_i = 1500$ m.

The aerosol module is that given in B2004. It describes the evolution of 7 chemical species: ASOA and BSOA (Anthropogenic and Biogenic secondary organic aerosol), PPM (Primary Particle Material), sulfate, nitrate, ammonium and water. Particle diameters range over 6 bins from 10 nm to 40 μ m in this version. The model accounts for the coagulation process as described in Gelbard and Seinfeld (1980). The dynamic of the absorption process of organic and inorganic semivolatile species is modelled with a first order equation. For the ternary system, Sulfate/Nitrate/Ammonium, the thermodynamic equilibrium is computed with the ISORROPIA model (Nenes et al., 1998). Heterogeneous chemical processes onto particles (nitrate production) and a simplified sulfur aqueous chemistry (sulfate production) have been implemented. Moreover, a preliminary chemical module to form secondary organic aerosols was introduced.

Anthropogenic emissions are taken from EMEP (Vestreng, 2003), as in B2004. Aerosol emissions feed the model species denoted as PPM, which may contain several substances (black and organic carbon, matter resulting from abrasion, ...) coming from various anthropogenic origin.

2.2. Boundary conditions

In order to take into account aerosols transported from the boundaries of the model domain, we use, for aerosols only, values obtained from the GOCART model (Ginoux et al., 2001; Ginoux et al., 2004) simulation, while for the gas-phase species, the MOZART (Horowitz et al., 2003) climatologies are used as in previous CHIMERE uses. As GOCART runs for Year 2003 were not available at the time of the present study, and as hourly or even daily boundary conditions

were quite difficult to process, we use the average of monthly mean values taken from GOCART runs over Years 2000 and 2001.

The species available from GOCART are: mineral dust, hydrophobic and hydrophilic organic carbon (OC), hydrophobic and hydrophilic black carbon (BC), and sulfates. Sea salt has not been introduced, although it is an important contribution to total PM₁₀ in coastal areas. This should not affect our results since we perform model/observation comparisons mostly with continental sites (see site distributions in Fig. 7). For dust, GOCART provides a 7-bin representation from 0.2 to 12 μ m in diameter. For sulfate and carbonaceous species, we assume a classical distribution (Seinfeld and Pandis, 1998). These spectral distributions proper to GOCART are interpolated to the CHIMERE aerosol size distribution. At the boundaries, the CHIMERE PPM species is made of several GOCART model species: dust, BC and a part of OC.

For dust boundary conditions, a major problem comes from the fact that episodes of Saharan wind-blown dust high concentrations are very sporadic but intense. Mahowald et al. (2003) present a high intramonthly variability over 22 years observation data. The intensity of these phenomena is such that the monthly average is usually much higher than the actual median “background” values encountered most of the time over North Africa. Following the results of Prospero (1999), the average Saharan dust concentration over the Caribbean was found to be about 3–4 times higher than the median value. Therefore, we use, as dust boundary forcing, concentrations three times smaller than the average GOCART values. In order to investigate the effect of the Saharan dust, we perform simulations with and without dust boundary conditions.

Our purpose in this article is to study the *mean* effect of Aeolian sources, and the introduction of background boundary conditions for dust as weakly time-dependent (only seasonal dependence) does not allow the simulation of the transport of acute, massive events of Saharan dust due to desert storms. In order to simulate these events daily or even hourly dust boundary conditions would have to be considered. Here, we only consider transport of background dust concentrations and examine the sensitivity of the average model’s results to this process.

2.3. The model simulations and the observations

Along this article, hypotheses are tested in the CHIMERE model by running it over two distinct seasons of Year 2003, summer and fall. Summer is considered as the season between 1st of May and 2nd of September, fall between the 3rd of September and the 31st of December. These long-term simulations are carried out by 5-day pieces, each new period being

initialized by concentrations at the end of the previous simulation.

For comparison with observed data, we use daily mean observations of PM₁₀ over 163 ground stations obtained from air quality monitoring organizations throughout Western Europe in the following countries from which data were graciously provided: Belgium, Germany, Switzerland, Spain, France, Italy and United Kingdom (see the acknowledgement section for contact persons/organizations). All 163 stations locations are displayed in a late figure of this paper (Fig. 7). Due to the coarse resolution of the model, only stations representative of a large-scale (~50 km) environment should be used. We kept only rural and suburban stations, except where large areas remain uncovered, in which case we use urban stations located in small towns. In the analysis, we distinguish the “rural” stations (the stations really classified as rural) and the “urban” stations (classified as urban and suburban). Also, in order to distinguish the model behaviour over southern and northern Europe, we separate two groups of stations: northern stations are those located north of 46 degree, and southern stations are those located south of 46 degree. As we shall see, as far as PM₁₀ is

concerned, the behavior in these two groups is markedly different.

3. The impact of Saharan dust

Fig. 1 shows the surface seasonal average of PM₁₀ concentration obtained from the model simulations, with or without dust boundary conditions, and for the two seasons. Without dust boundary conditions, the concentrations are slightly higher than in B2004, mostly due to the addition of aerosol boundary conditions other than dust. In summer mean concentrations remain underestimated by more than 50% for Southern Europe and 40% for Northern Europe, when considering only the rural stations (see Table 1a). These underestimations are consistent with the intercomparison of several European models in van Loon (2004). The introduction of Saharan dust has a very significant effect, particularly over the Mediterranean Sea, where it reaches 5–10 $\mu\text{g m}^{-3}$, and a more moderate effect in Northern Europe, 0–3 $\mu\text{g m}^{-3}$ (Fig. 1c). The PM₁₀ bias decreases to about 30% both in southern and northern areas. In fall (Figs. 1b and d and Table 1b), the concentrations are

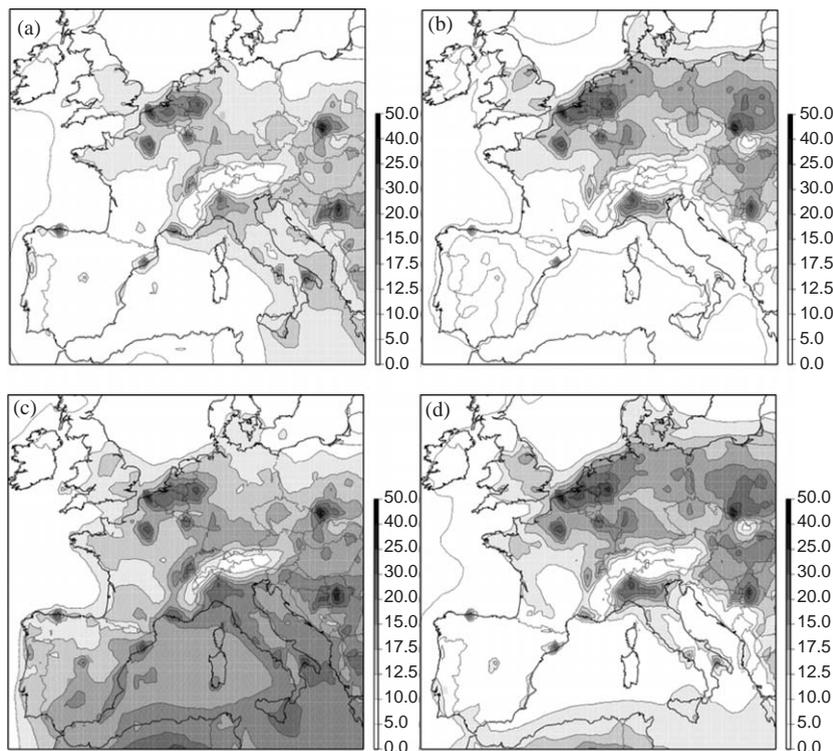


Fig. 1. Seasonal averages of simulated PM₁₀ surface concentrations, for the summer cases (left panels (a) and (c)) and the fall cases (right panels (b) and (d)), and for the experiments with boundary dust (bottom panels (c) and (d)) and the experiments without boundary dust (top, panels (a) and (b)). All concentrations are in $\mu\text{g m}^{-3}$.

Table 1

Model skill PM10 daily averages statistics: rows correspond to various statistics and columns to the categories of sites we distinguish (S = South, N = North)

	Rural S	Urban S	Rural N	Urban N	All
<i>(a) For the summer seasons</i>					
OBS aver.	23.4	23.7	19.8	21.1	20.9
MOD aver.	9.9/15.5	10.5/15.3	12.1/14.3	14.1/16.5	12.7/15.6
RMS	16.7/11.7	15.3/11.0	10.9/9.4	10.9/9.5	11.8/9.8
Correlation	0.53/0.66	0.52/0.68	0.51/0.53	0.60/0.62	0.56/0.61
<i>(b) For the fall seasons</i>					
OBS aver.	17.9	21.6	17.3	20.6	19.3
MOD aver.	8.5/10.7	8.4/10.2	14.6/16.2	16.0/17.6	14.0/15.7
RMS	13.7/12.1	16.1/14.4	9.3/9.3	11.2/ 10.9	11.5/11.0
Correlation	0.51/0.59	0.46/0.53	0.54/0.55	0.57/0.58	0.54/0.56

Averages and root mean square (RMS) errors are in $\mu\text{g m}^{-3}$. The first number in each row corresponds to the simulation without Saharan dust boundary conditions and the second number to the simulation with Saharan dust. Local erosion and resuspension are not included in this analysis.

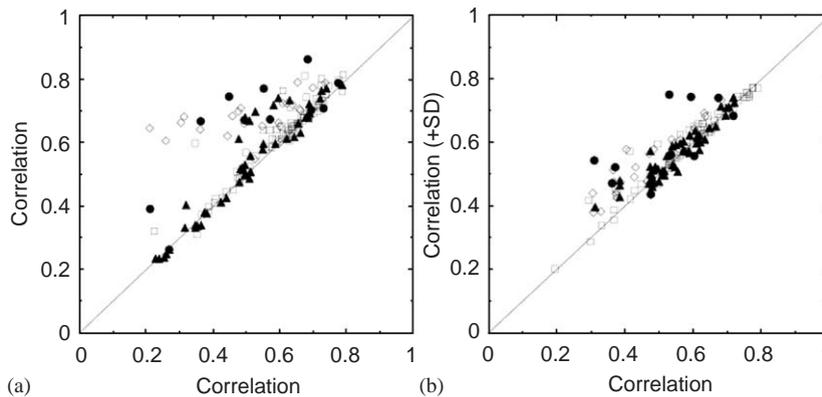


Fig. 2. Individual correlations, for each station, of PM10, daily averages for the experiment with boundary dust versus those of the experiment without, for the summer months (left panel) and the fall months (right panel). Triangle = Rural N, Circle = Rural S, Square = Urban N, Diamond = Urban S.

higher than in summer over Northern Europe and lower over southern Europe. This difference results from a complex combination of dispersion and higher emissions. The bias is about 50% in southern areas and less than 20% in northern Europe. The introduction of Saharan dust reduces the southern bias to 40% and the northern bias becomes insignificant. Saharan dust has a much less pronounced impact over Northern Europe than during summer, due to the fact that (i) dust boundary conditions are weaker and (ii) Europe is more influenced by wet westerly winds during this period.

Not surprisingly, with added Saharan dust, the general mean PM10 model concentrations increase. However, it does not show that day-to-day variations improve, especially as a result from the contrast between days when air masses are transported from the model Southern boundary and the other days. Such model's

fluctuations improvements can be assessed by the other two statistical measures of model skill: correlation and root mean square (RMS) errors of daily PM10 averages (Tables 1a and b).

In the summer case, correlations over groups of stations are relatively low, in the range 0.5–0.6 in all cases without Saharan dust, and increase significantly in all cases when it is added. The increase of correlations, at individual stations, by the introduction of Saharan dust can be seen in Fig. 2. The model daily variations improvement is clear for Southern Europe in the two seasons and less pronounced for Northern Europe. RMS errors also decrease a lot due both to the reduction of bias and the improvement of the model daily variability.

As explained in Section 2 the model cannot capture acute episode of massive Saharan dust transport due to

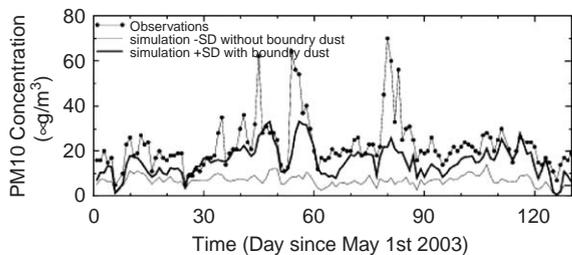


Fig. 3. Time series of daily averages observed (thin line with circles) and simulated, with (heavy line) and without (light line) boundary dust, at the Zarra (South of Spain) EMEP site. Only the summer season is shown. Time is in day since 1st of May.

desert storms, because only GOCART monthly averages are considered at the domains boundaries. This is clear from Fig. 3, showing the time series of PM10 daily averages as observed and simulated with and without dust during Summer 2003 at the south Spanish EMEP site of Zarra. During low to moderate PM10 periods the model with dust reproduces quite well the observed fluctuations with some underestimation. Highest maxima, probably caused by massive imports of Saharan dust, are underestimated by a factor 2–3, but are still simulated as moderate peaks. The simulation without dust boundary conditions very poorly simulates the observed fluctuations.

Our results show that taking into account nonzero aerosol boundary conditions improves the model simulations, but nothing actually proves that these boundary conditions should be made of Saharan dust. However the fact that results improvements are largest over southern areas strongly indicates that the southern boundary (where most of the GOCART dust boundary conditions lie) has a special impact. Along this boundary the aerosol mass is probably made essentially of Saharan dust. There are also other Aeolian processes that could be missing in the model to contribute to the model negative PM10 bias. The fact that previous simulations are not strongly biased during fall over the wet areas (Northern Europe), except along coastal areas (sea salt is not accounted for in the model) indicates that local erosion and resuspension processes could also play a significant role. In the following sections, we perform sensitivity studies in order to evaluate their potential role.

4. Dust emission from local erosion

4.1. Formulation of a simplified bulk scheme

Modelling dust entrainment into the atmosphere has been the subject of many studies since the work of Bagnold (1941). Several detailed models have been

proposed, which take into account the soil composition, moisture and several subtle effects such as the Owen effect on friction velocity (Gilette, 1988; Marticorena and Bergametti, 1995; Nickovic et al., 2001; Zender et al., 2003). However most of the modelling work was motivated by the need, for climate studies, to represent aerosol concentrations and their feedback onto radiation at the global scale. Only a few studies address the problem of dust erosion in the perspective of air quality modelling, where soil other than arid or semi-arid must be considered. Such is the case of the Columbia Plateau case study of Clairborn et al. (1998). A common characteristic of all these studies has been to focus on major dust entrainment events, like those associated to wind storms in desert areas or agricultural areas with highly erodible soils, where PM10 concentrations reach several hundreds or thousands of $\mu\text{g m}^{-3}$. We are not aware of any model application for “background dust” typical concentrations like those observed in Europe (typically a few $\mu\text{g m}^{-3}$).

Here, we examine the impact of the introduction of local erosion on PM10 model skill. Since our goal is not to develop a sophisticated erosion scheme for temperate regions, we follow the classical approach of dust emission modelling in a simplified manner. The dust vertical flux (in $\text{g m}^{-2} \text{s}^{-1}$) F is modelled with a functional approach similar to that of Zender et al. (2003), which itself is based on the Marticorena and Bergametti (1995) approach and the White (1979) formula

$$F = \alpha C u_{*s} (u_{*s}^2 - u_{*t}^2), \quad (1)$$

where u_{*s} denotes the saltation friction velocity, u_{*t} the threshold friction velocity and C is a coefficient that may depend on several surface factors (see below). The sandblasting efficiency, α , describes the ratio of the vertical flux to the horizontal saltation flux. The saltation friction velocity u_{*s} corresponds to that encountered on erodible parcels of the model grid cell, usually smoother than typical vegetated surfaces found in Europe. It is calculated by using the 10 m wind field, a saltation roughness length of $5 \times 10^{-4} \text{ m}$ and the assumption of neutral stability as in most previous studies.

In order to keep the formulation simple, the threshold friction velocity is assumed to depend only on gravimetric soil moisture w . As in Zender et al. (2003) and Nickovic et al. (2001), this dependence uses the Fécán et al. (1999) formulation:

$$u_{*t} = f_w u_{*0}, \quad (2)$$

where u_{*0} is the minimal friction velocity for dust entrainment over dry soil, and is taken to be uniformly

constant. The soil moisture factor f_w is given by

$$\begin{cases} w < w_t : f_w = 1 \\ w > w_t : f_w = \sqrt{1 + 1.21(100(w - w_t))^{0.68}}, \end{cases} \quad (3)$$

where w_t is the gravimetric soil moisture content (in kg kg^{-1}) above which entrainment is inhibited by soil moisture. This threshold is usually calculated as a function of the fraction of clay, in a rather empirical manner. In Europe during the summer season, the volumetric soil moisture provided by the NCEP GFS analyses and MM5 drops below 0.15 (gravimetric equivalent of about 0.10) only after several weeks without precipitation. We take here the uniform constant value $w_t = 0.1 \text{ kg kg}^{-1}$ throughout the model domain, which corresponds to a large clay fraction (Fécan et al., 1999).

The independence of threshold friction velocity on soil type is a strong assumption, since our aim is only to test the impact of introducing erosion processes. Iversen and White (1982) and Shao and Lu (2000) have proposed formulations linking the threshold velocity to the soil particle distribution. In both cases a minimal value of 0.2 m s^{-1} is found, related to soil particles with a mean diameter of 0.1 mm. According to Xuan (2004), threshold friction velocities adjusted to wind tunnel observations are too large as they do not account for subgrid scale turbulence. Following this argument we divided the previous minimal threshold by 2 and take $u_{*0} = 0.1 \text{ m s}^{-1}$.

The sandblasting efficiency is taken as $\alpha = 5 \times 10^{-5} \text{ m}^{-1}$, an order of magnitude obtained from the measurements, over Northern Spain and Niger, of Gomes et al. (2003a, b), and it is again kept uniformly constant, although its dependence can be made explicit as a function of clay content (Marticorena and Bergametti, 1995).

In the ideal case of a bare, non crusted soil, the factor C in Eq. (1) has been taken equal to (Zender et al., 2003):

$$C_i = \frac{2.61 \rho_{air}}{g}. \quad (4)$$

In practice, the soil is covered with vegetation, snow or water over most of Western Europe and is very often crusted. We, therefore, define

$$C = f_b f_c C_i, \quad (5)$$

where f_c is the crustation factor, which has been found from field studies to lie in the 0.001–0.1 range (Goossens, 2003; Gomes et al., 2003a), and f_b is the fraction of bare ground where dust is available for entrainment. In practice, the product $f_b f_c$ is taken here uniformly constant over land and is tuned to optimize the model/observation correlations while keeping the average mineral dust fraction close to that found in the

climatological observations of Putaud et al. (2004). This leads to $f_b f_c = 4 \times 10^{-3}$. This strong assumption of a uniform factor leads to a large uncertainty. However, our results remain valid if other factors of the same order of magnitude are taken. Due to this uncertainty, our results should be considered in a qualitative manner only, which is enough for a sensitivity study.

The Fécan et al. (1999) parameterization of the effect of soil moisture was unable to inhibit dust erosion in sporadic events of fall wind and rain storms over Western Europe under wet conditions. Dust emissions are therefore set to zero when gravimetric soil moisture exceeds 0.2, and kept as the scheme calculates them only when it is below 0.16, a linear interpolation being performed between these two values.

The above emissions are assumed to be total emissions. Since emissions of mineral dust occurs primarily in the coarse mode, one assumes $\frac{1}{2}$ of emissions in the 10–40 μm mode, 45% of emissions in the 2.5–10 μm mode and the remaining 5% are distributed in the fine mode using the same distribution as for anthropogenic.

4.2. The impact of erosion dust emission on model simulation

The introduction of local erosion in our simulations leads to additional mineral dust concentrations of PM10 whose averages are mapped, for the two seasons, in Fig. 4. In summer, the increase in PM10 reaches about 4–5 $\mu\text{g m}^{-3}$ in dry areas, over south-central Spain and Portugal, while it has a small contribution (of less than 1 $\mu\text{g m}^{-3}$) over most of Northern Europe. Due to our scheme tuning the total mineral dust (boundary conditions and local erosion) contribute respectively to about 35% and 15% for the two parts of Europe, which is consistent with the climatology of Putaud (2003). In fall, due to wet soil, this contribution drops down to 1 $\mu\text{g m}^{-3}$ over Spain, and is small elsewhere. The day-to-day variability of simulated PM10 concentrations improves when introducing the erosion dust entrainment. Almost all stations have a slightly higher correlation (not shown). The improvement is quite uniform across Europe, showing that erosion can play a significant role everywhere in Europe during this season. By contrast, in fall, we do not find any systematic improvement. Due to wet soil it remains a marginal process.

5. Resuspension of particles by wind and turbulence

5.1. Bulk resuspension scheme

Saltation, which is necessary to transfer small mineral particles from the soil into the atmosphere, is not the

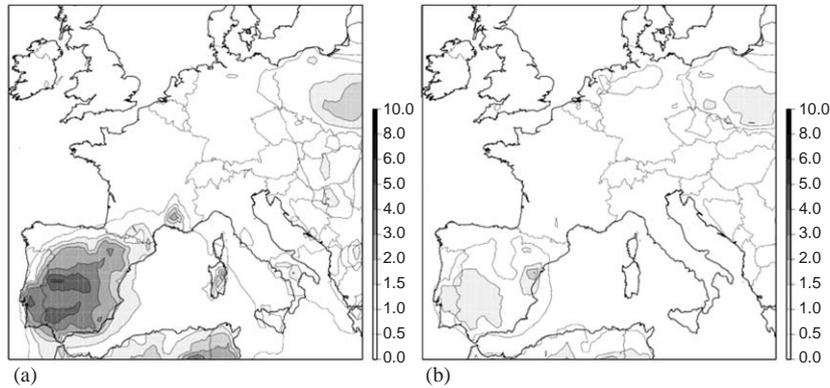


Fig. 4. Spatial distribution of the average difference between the simulation with and the simulation without local erosion (transport from dust boundary conditions excluded), for the summer months (left panel) and the fall months (right panel).

only natural aerosol upward entrainment process. Resuspension of freshly deposited small particles lying at the surface by turbulent wind is a distinct process, their extraction resulting from the imbalance between adhesive and lifting forces (see e.g. Reeks and Hall, 2001). Such particles can originate from the atmosphere or the biosphere, and are particularly easy to extract shortly after deposition (Loosmore, 2003). They can be made of vegetal debris obtained from the mechanical or biological degradation of canopy elements, spores, pollens, etc. (Simoneit and Mazurek, 1982). The evidence of the presence of biogenic non mineral compounds in the continental aerosols has been reported several times (see e.g. Andreae and Crutzen, 1997), for instance in the form of “humic matter”, consisting in carbonaceous polymers (see Gelencsér et al, 2002 and references therein), and cellulose (Kunit and Puxbaum, 1996). As shown by Gelencsér et al (2002), some of these aerosols could be of secondary nature. However, for the sake of simplicity, we assume that this biogenic aerosol component is of primary origin.

In order to represent these processes, we use a bulk formulation based on the simple resuspension rate empirical formula of Loosmore (2003), which was shown to provide a very good fit to the available resuspension measurement data:

$$A = 0.01 \frac{u_*^{1.43}}{\tau^{1.03}}, \quad (6)$$

where A is the resuspension rate (in s^{-1}) and τ is the time after the start of resuspension. In the experiments used by the previous authors, the particles are first deposited then resuspended. In reality, deposition and resuspension are simultaneous, and the available dust concentration on the ground is governed by resuspension, washout by runoff and absorption by soil water, production by deposition and other biological or mechanical processes. The detail of all these processes

is essentially unknown, and we assume here that the available concentration of dust does only depend on the wetness of the surface. In this empirical view, the resuspension flux is governed by

$$F = Pf(w)u_*^{1.43}, \quad (7)$$

where $f(w)$ is a function of the soil water content and P is a constant tuned in order to approximately close the PM10 mass. The soil water modulation factor is vanishing for wet soil (gravimetric soil water content of 0.2 or more, as before, and is equal to 1 for dry soil, w less than 0.1). A linear relation is assumed between these values. The resuspension flux is found to be $F = 1800 \mu\text{g m}^{-2} \text{h}^{-1}$ for a dry soil and for a friction velocity of 1 ms^{-1} . In Northern Europe the typical MM5 value for volumetric soil water content is 25%, and 0.5 ms^{-1} for the friction velocity. This leads to a mass flux of about $170 \mu\text{g m}^{-2} \text{h}^{-1}$ and then about 1.4 ton km^{-2} and per year, which is much larger than the anthropogenic emissions in most remote rural areas, but an order of magnitude smaller than that in urban areas. Finally, in the absence of any information the reentrained PM10 particle mass is supposed to be distributed in a standard atmospheric size distribution: $\frac{2}{3}$ of the mass as PM2.5 and $\frac{1}{3}$ as coarse PM10–PM2.5. Within PM2.5, particles are distributed as for the anthropogenic emissions.

5.2. Model results

Fig. 5 shows the increase of PM10 surface concentration due to resuspension. The main effect is to increase concentrations where the soil is dry. The largest difference in summer is observed, again, over Spain and North Africa, with a $8\text{--}12 \mu\text{g m}^{-3}$ amplitude. A relatively large difference ($5\text{--}10 \mu\text{g m}^{-3}$) is also observed over parts of Eastern Europe, while in rainier areas (Northwestern Europe), the effect does not exceed

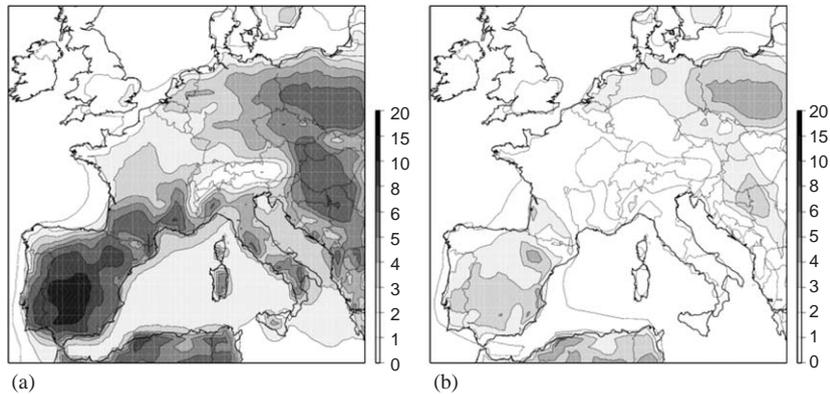


Fig. 5. Spatial distribution of the seasonal averages of the increase of concentration due to the assumed resuspension process only. Left panel: summer months; right panel: fall months.

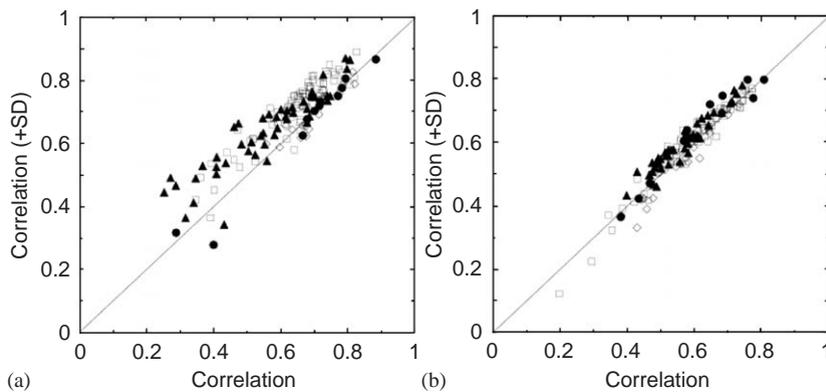


Fig. 6. Correlations between observed and simulated PM₁₀ daily average concentrations for the experiment with assumed resuspension vs. the experiment without resuspension, but all other processes. Symbols as in Fig. 2.

$5 \mu\text{g m}^{-3}$. Although there is obviously no resuspension above the sea, the impact of resuspension around the Western Mediterranean basin is significant above the sea itself (about $2\text{--}3 \mu\text{g m}^{-3}$) due to long-range transport. In fall, due to wet soil, the effect of resuspension is strongly reduced ($5 \mu\text{g m}^{-3}$ at maximum), but occurs over the same areas as in summer.

The most striking result is the improvement of the model simulation in terms of correlations, as shown in Fig. 6. The increase in the correlations is particularly marked for stations in Northern Europe in summer, where many correlations increase by 0.1 or so. In fall correlations also increase in northern Europe but to smaller extent. In southern areas correlations are not modified significantly on average. The RMS errors and the biases (see Table 2) decrease significantly. The adjustment of the P constant, makes the mass budget close to within 10% with the observations in summer, and the assumed resuspension process contributes to about 20–30% of the PM₁₀ mass in southern areas and

10–20% in northern areas. In winter there is still a large underestimation over southern rural stations. In fact this bias is essentially due to the contribution of four rural stations located in northern Italy in mountainous areas where we suspect the model resolution is not appropriate to simulate valley flows. By contrast the bias is small over Spanish rural stations. The simulated PM_{2.5}/PM₁₀ fraction, about 60% on average, is consistent with our current knowledge of the aerosol particles in rural areas in Europe (see e.g. Van Dingenen et al., 2004). The model improvement shows that resuspension, as it is parameterized here, is a potentially important process. The fact that the improvement is large over Northern Europe is a sign that this process needs particular attention in vegetated areas.

Interestingly the spatial distribution of correlations is fairly heterogeneous. Fig. 7 shows the summertime distribution of stations with correlations lower and higher than $r = 0.7$. Over Spain and France most correlations are high, except in mountainous areas while

Table 2

Statistics of the model (with all processes, i.e. transport from the boundaries, local erosion and resuspension) results and the PM10 observations comparison

	Rural S	Urban S	Rural N	Urban N	All
<i>(a) For summer months</i>					
OBS Mean	23.4	23.8	19.8	21.1	21.2
MOD Mean	22.0	20.1	18.0	20.0	19.6
RMS	9.7	8.7	7.7	8.0	8.1
Correlation	0.66	0.70	0.64	0.70	0.68
<i>(b) For the fall months</i>					
OBS aver.	17.9	21.6	17.3	20.6	19.6
MOD aver.	13.0	11.6	18.4	19.5	17.6
RMS	10.7	13.6	9.6	10.9	10.8
Correlation	0.64	0.52	0.58	0.59	0.58

Statistics are averaged over the categories of stations.

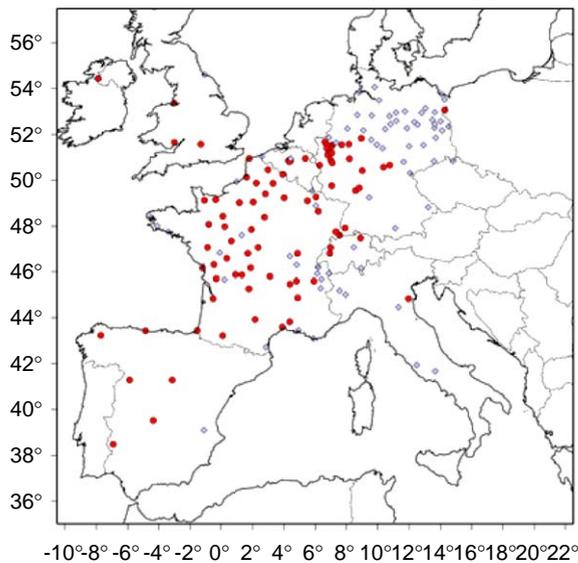


Fig. 7. Locations of the measurement sites, with a different symbol depending on whether the correlation of the full experiment with resuspension is larger (red circles) or smaller (blue diamonds) than 0.7.

there is a group of stations with low correlations over Northern Germany. This latter region is associated with a general negative bias of the model. A careful examination of the corresponding time series (not shown) indicates that low correlation is due to sporadic very high values of observed PM10 missed by the model. The origin of these misses is not clear, and needs further investigation.

Table 3 shows the progression of model skill from the base case with no boundary dusts to the model with all parameterizations, for all groups of stations, in the

summer and fall cases. Clearly the most important skill factor in Southern areas is the introduction of dust transport from model boundaries. Resuspension has almost no effect on skill in these areas while erosion has a moderate but systematic effect. In northern areas, our parameterization of resuspension seems to significantly increase the summer skill, while other processes have a more moderate effect.

6. Discussion and conclusion

In this article the question of the aerosol PM10 source apportionment in Europe is addressed. Starting from the fact that several models, forced by anthropogenic emissions and running over Europe miss a large fraction of the aerosol mass (Van Loon, 2004), it is assumed here that this missing mass comes from natural origin, new bulk parameterizations for the associated emission processes are proposed and it is shown that the model skill improves significantly when using them. While this skill increase is an indication that such emission processes are real, only a separate experimental validation of each process would provide a formal proof.

Starting from a model configuration described in Bessagnet et al. (2004), we added three increasingly uncertain processes: transport of dust from the boundaries of the model domain, and in particular from the Saharan region, erosion within the European domain, and resuspension of deposited particles. However there may be other possibilities for completing the missing mass, a possibility which is discussed now.

The base model misses about 30–50% of the total PM10 mass. This could be due to a dynamical problem. For instance a systematic overestimation of the boundary layer height or diffusivity near the ground could lead to such a bias. Many experiments with the model and

Table 3

Correlations for all stations categories, between simulated and observed PM10 daily averages, for the base case model and the model where gradually all processes are included, and with the experiment where resuspension is simply replaced by multiplying secondary organics by a factor of 3 (3xSOA)

	Rural S	Urban S	Rural N	Urban N	All
<i>(a) For the summer months</i>					
Base model	0.53	0.52	0.51	0.60	0.56
With bound dust	0.66	0.68	0.53	0.62	0.61
With erosion	0.67	0.70	0.56	0.63	0.62
With resuspension/3xSOA	0.66/0.68	0.70/0.67	0.64/0.64	0.70/0.70	0.68/0.68
<i>(b) For the fall months</i>					
Base model	0.51	0.46	0.54	0.57	0.54
With bound dust	0.59	0.53	0.55	0.58	0.56
With erosion	0.62	0.54	0.55	0.58	0.57
With resuspension/3xSOA	0.64/0.64	0.52/0.52	0.58/0.60	0.59/0.61	0.58/0.60

the analysis of its skill in ozone and nitrogen oxides simulation (see e.g. Schmidt et al., 2001, and the web air quality forecast server <http://www.prevoir.org> where routine ozone forecasts and their verifications are presented) let us believe that such a dispersion error could not be so large in the model. Moreover, the vertical structure of the model boundary layer was shown to faithfully fit backscattering lidar observations in a site near Paris (Hodzic et al., 2004).

Another possibility is that the EMEP anthropogenic emissions (Vestreng, 2003) are largely underestimated. This should not either be the case since our base model bias occurs everywhere in Europe, especially far from the main source areas, and in southern parts of the continent (pronounced bias in Spain), where the influence of anthropogenic sources is less pronounced.

Sea salt is also missing in the model, but according to Putaud et al. (2004), its counterpart can reach 10% on annual average, mostly near the coasts. Over continental areas, it does not exceed a few percents.

There are also large gaps in our knowledge about secondary aerosol formation (Griffin et al., 2002), and at least part of the base model bias could be due to secondary organic matter underestimation. By comparing the model aerosols with sun photometer and lidar measurements Hodzic et al. (2004) suggested that secondary organic aerosols could be underestimated by a factor of 3. In order to test this hypothesis, we take the results from the model with boundary dust and erosion only (no resuspension), multiply by 3 the daily average contribution of secondary organic matter and recalculate the model skill with the new PM10 concentrations. In other words, we completely replace the resuspension process of Section 4 by an artificial increase of secondary organic matter. In this way the model mass is also closed. Quite surprisingly the model skill, shown in Tables 3a and b is equivalent to that of the model with

resuspension, in terms of correlations, and is even slightly higher in fall. Root mean square error and biases also behave similarly for all types of stations. However the average differences between this experiment and the boundary dust + erosion experiment (not shown) display a fairly different structure than in Fig. 5, with maxima over central Europe, as in Fig. 2 (SOA) of B2004.

The results of this sensitivity experiment show that one cannot claim that resuspension is the only responsible for the missing PM10 mass. The artificial increase of organic matter would lead us to the same conclusion. Most probably the base model bias problem results from a mixture of problems due to the two processes. The only way to provide definite conclusions to this issue is to design measurement campaigns able to distinguish and validate the processes. Accurate measurements of aerosol particle fluxes above vegetated canopies could help quantifying the resuspension process.

In summary, despite the lack of definite conclusions about quantitative aspects of each process, this article showed several points. First, it has been demonstrated that the introduction of boundary conditions, especially for Saharan dust is necessary in order to model correctly the particulate matter mass over Southern Europe. The impact is also significant, albeit less pronounced, over northern Europe. Here boundary conditions come from 2000–2001 monthly averages issued from GOCART model simulations (Ginoux et al., 2001). Strong desert storm dust erosion events and the subsequent transport cannot be represented here. However, the transport of “background dust” present at the southern boundary seems fairly well simulated, as the model/observation PM10 daily average correlations increase significantly. The simulation of massive dust events can only be simulated if better time resolved boundary conditions (daily, hourly) are used.

The simplified scheme proposed here for simulating local erosion and the improvement in the simulation by its introduction indicate that erosion, particularly in southern Europe can contribute to the mass budget of PM₁₀. Finally, we assumed that resuspension of material deposited on the ground, mostly of biological origin, may explain the remaining part of the missing mass in PM₁₀ average load, and provide a simplified scheme for the corresponding emission, which depends on turbulence intensity near the ground through the friction velocity. With this scheme, the model skill improves substantially in northern Europe, but sensitivity tests show that such an improvement can also be obtained by simply increasing the model organic matter by a factor of 3. We conclude that our results are consistent with the existence of a substantial natural resuspension aerosol source, but more experimental work is required to confirm and quantify it.

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