Modelling dust emissions and transport within Europe: the Ukraine March 2007 event

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Abstract.

A huge dust event was observed in Central Europe during three consecutive days of March 2007 (23-24-25). Surface observations and lidar profiles showed transport of a dust plume from Ukraine to Germany, with huge concentrations at the surface, [*Birmili et al.*, 2007]. The emissions were assumed to be due to high wind speed and soil surface specificity, mostly arable surfaces, in southern Ukraine where the most important concentrations were observed. Since this event is important, with a well-known location and more or less alone over Europe during this period, it corresponds to an ideal case for the improvement of dust emissions modelling. Being under specific meteorological conditions, this episode also corresponds to an unique long-range transport dust event within Europe. The impact on air quality of such emissions and transport to remote areas over western Europe is adressed and quantified.

1. Introduction

The mineral dust is a natural compound in the atmosphere known to affect air quality in several regions of the world. Mineral dust emissions, transport, deposition and their impact on radiative budget has been particularly studied [*Andreae and Crutzen*, 1997; *Sokolik et al.*, 2001] over the two main emittors in the world, the western Africa and China, using worldwide measurements network, such as AERONET [*Holben et al.*, 2001] and global models such as GOCART [*Ginoux et al.*, 2004]. However open scientific questions remain. Except in China where desert areas such as the Taklamakan desert directly impacts on megacities ([*Sun et al.*, 2001; *Zhang et al.*, 2003]), the direct transport of mineral dust towards large industrialized and urbanized areas is rarely studied. Over Europe, the main impact of dust transport so far considered is that due to Western Africa dust outbreaks originated from Sahara. But the net contribution remains relatively low and, for the daily particulate matter (PM) budget, it is rare to record a dust contribution of more than 20% of the total particulate matter mass, as an averaged value over the year even if sporadic episodes led to a massive contribution of dust during short periods [*Simpson et al.*, 1999; *Querol et al.*, 2004a, b; *Moreno et al.*, 2005; *Escudero et al.*, 2007a].

This natural contribution of mineral dust to European air quality has been particularly studied where the concern is largest, Spain [*Artinano et al.*, 2003; *Alastuey et al.*, 2005; *Escudero et al.*, 2007b; *Querol et al.*, 2007], Portugal [*Petzold et al.*, 2006], Italy ([*Kishcha et al.*, 2005]) and more generally over Europe for comparative studies ([*Simpson et al.*, 1999; *Querol et al.*, 2004b; *Viana et al.*, 2007]). In these countries, a large number of experimental

field campaigns [*Clairborn et al.*, 1998; *Gomes et al.*, 2003], routine measurements of particles concentrations and speciation analyses [*Putaud et al.*, 2004; *Van Dingenen et al.*, 2004], are carried out to provide important answers about the relative amount of these particles in the total budget of air quality over cities. This contribution has to be taken into account because the EU standards fix an annual mean lower than 20 μg.m^{-3} and a maximum allowance for the number of days exceeding the daily limit value (50 μg.m^{-3}) of 7 days per year by the first of January 2010.

On the modelling side, dust emission and transport has been mainly adressed at the global scale with a low horizontal resolution. These studies are often dedicated to climate changes and dust concentrations trends over the world. The model results are compared to measurements such as aerosol optical thicknesses and satellite data, using monthly averaged values. Moreover, since the most important sources areas are located in Africa, China and Australia, the most important efforts are done to better describe the emissions in these areas but more rarely out.

In Europe, a recent study, [*Vautard et al.*, 2005], showed that, with a very simplified approach of wind blown dust emissions inside Europe, a chemistry-transport model, such as CHIMERE, is able to partly explain the lack of modelled particulate matter compared to the background values generally observed. This approach was validated statistically, meaning that using meteorological parameters such as surface humidity and wind speed, a simplified approach allows to give a more realistic view of aerosols over Europe. But this study was not designed to predict sudden and huge dust events as the one observed in Ukraine at the end of March 2007 and extensively described in [*Birmili et al.*, 2007].

The March 2007 Ukraine event is a rare and invaluable opportunity to better understand and describe a natural phenomenon having a strong impact on air quality over Europe. The first goal of this study is to evaluate whether a chemistry-transport model is able to catch the conditions leading to the observed emitted mass of dust over Ukraine. We demonstrate that this is possible by providing new informations on the landuse in the model, and more accurate informations on Chernozemic soils for specific arable lands as highlighted by [*Birmili et al.*, 2007] (and references therein). The second goal is to evaluate the transport of such dust outbreaks. Since the rest of the Europe was under 'relatively low' aerosols background conditions, the dust plume exhibits a clear and important signal all along its way. This event is also a rare opportunity to estimate the impact of dust events on air quality over remote areas far from the emissions area (Ukraine). Surface measurements recorded in United Kingdom, Belgium and North of France are compared to model simulations. The relative part of Ukrainian soil dust particles in the aerosols budget over these lands is quantified and discussed.

2. The event of dust concentrations over Ukraine in March 2007

Figure 1.

As shown in [*Birmili et al.*, 2007], many aerosol measurements are available over Europe for this period, clearly demonstrating the large-scale, European extent of this huge dust event. These authors presented an exhaustive description of the real events, recorded both with meteorological and particles measurements. Briefly, from the 21 March 00:00 to the 23 March 12:00, two low pressure systems located in the mediterranean basin and in northern Europe merged and give a sole system centered over Romania on 23 March 12:00. At the same time,

an anticyclone in Russia drives cold air mass towards central Europe. The combination of these two systems creates a East-West corridor with high speed winds the 23 and 24 March. The 23 March [12:00], wind speeds are the highest in the south steepes of Ukraine, they exceed 15 m.s^{-1} with friction velocites allowing dusts to be blown up. This time corresponds to the maximum of emissions.

A first step for the present study was to evaluate the meteorological model compared to the observations described by [*Birmili et al.*, 2007]. As displayed in Figure 1, the model is able to calculate 10m wind speed values around 15 m.s^{-1}, with a large boundary layer height (BLH). In addition, the associated parameterized friction velocity u_* reflects these high wind speed and presents values potentially able to start saltation processes. Even if the mean BLH remains below 1000m on 24 March [00:00], a ”channel” of higher height is observed and modelled. From south-east (Ukraine) to north-west (north of Germany), heights of more than 1500m (corresponding to high values for a nighttime period) are simulated. This system persists during the next day and exhibits increasing BLH values until noon (Figure 1).

3. Dust modelling with the CHIMERE chemistry-transport model

3.1. Meteorology, aerosols and resuspension

In this study, the V200709A version of the CHIMERE CTM over Europe ([*Schmidt et al.*, 2001] is used; [*Bessagnet et al.*, 2004]. The model is running from 10W to 40E and from 36N to 61.5N with a resolution of 1/2 degree both in latitude and longitude. The domain covers a large part of Europe from Irland up to Ukraine. The vertical grid contains 20 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 [*Vautard et al.*, 2005]. The model documentation can be found on the web server *http://euler.lmd.polytechnique.fr/chimere*. For both ozone and PM10, the model has undergone extensive intercomparisons at European and city scale ([*Vautard et al.*, 2007]; [*Van Loon et al.*, 2007]) and aerosols ([*Schaap et al.*, 2007]). However, for mineral dust such an intercomparison has not been carried out yet.

For meteorological inputs, the model is forced by the MM5 non hydrostatic model ([*Dudhia*, 1993], version 3.6, as in [*Vautard et al.*, 2005]. MM5 is configured with an horizontal resolution of 60km over a domain encompassing the CHIMERE domain, with 32 vertical levels. Classical options are chosen for parameterizations, such as the MRF scheme for the boundary layer. MM5 is itself forced, as in previous studies, by the Global Forecasting System (GFS) model, using the nudging option.

The CHIMERE aerosol module is that described in [*Bessagnet et al.*, 2004]. Anthropogenic emissions are taken from EMEP ([*Vestreng*, 2003]). Aerosol emissions feed the model species denoted as ”primary particle material” (PPM), which may contain several compounds (such as black and organic carbon) coming from various anthropogenic sources. The other modelled species are sulfates, nitrates, ammonium, secondary organic aerosols, sea-salt (considered as inert here) and dust. The particle size distribution ranges from about 10 nm to 40 μm and are distributed into 12 bins. For small particles, and in addition to saltation and sandblasting processes (described in §3.2) CHIMERE also accounts for resuspension [*Vautard et al.*, 2005; *Loosmore*, 2003].

3.2. Saltation and sanblasting

Modelling dust entrainment into the atmosphere has been the subject of many studies since the work of [*Bagnold*, 1941]. Several detailed emissions models have been proposed such as [*Marticorena and Bergametti*, 1995]; [*Alfaro and Gomes*, 2001]. They are currently used in transport model for sensitivity studies, analysis or forecast as in [*Nickovic et al.*, 2001], [*Forêt et al.*, 2006], [*Menut et al.*, 2005] and [*Menut et al.*, 2006]. In this study, we use an improved version of the [*Vautard et al.*, 2005] scheme. This scheme starts from that of [*Marticorena and Bergametti*, 1995] and the saltation dust flux is estimated as:

$$F = \alpha C u_{*s} \left(u_{*s}^2 - u_{*t}^2\right) \tag{1}$$

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. 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 10m wind field, a constant saltation roughness length of 5.10^{-4}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 and follows the [*Fecan et al.*, 1999] formulation. The sandblasting efficiency is constant and taken as $\alpha = 5.10^{-5} m^{-1}$ an order of magnitude obtained from the measurements, over Northern Spain and Niger, of [*Gomes et al.*, 2003].

3.3. Model improvements for this study

Figure 2.

For the present study, specific improvements were performed on the soil description and the calculation of the saltation over Europe. In general, the problem of dust emissions estimation is that we need to estimate two mandatory parameters without any measurements: (i) the emissions magnitude, and, (ii) the dust size distribution. For instance, over western Africa, the lack of concentrations measurements is a major limitation. Parameterizations are optimized after long term simulations and using statistical adjustements with comparisons to aerosol optical thicknesses (with the AERONET photometers network for example). This approach is suitable in case of long-term global modelling when only seasonal tendencies are studied.

3.3.1. First model improvement A first improvement to the [*Vautard et al.*, 2005] scheme, is the dependence of the saltation parameters to the model surface characteristics over land, assumed constant in that study. In the present work, we refine this aspect and affect **dust saltation only for cropland** representing the whole contribution of arable surface. This change eliminates unrealistic dust fluxes diagnosed over mountainous areas.

Figure 3.

3.3.2. Second model improvement A second improvement concerns the **soil description**. As described in [*Dolgilevich*, 1997], chernozemic soils cover a large part of Ukraine arable lands. In order to specifically work on this episode, and following analyses of [*Birmili et al.*, 2007], we added additional informations about these soils in the model landuse.

Indeed, these ”black soils” contain a large fraction of organic compounds, and they are considered to be the most fertile in the world. Intensive agriculture, hydric and wind erosion

have destroyed an area of 220.000 km^2 in Ukraine. The number of dust storms (wind gusts above 75 km.h^{-1}) reaches 5 days per year in the Southern steppes. During these storms a large amount of telluric dusts containing heavy metals, radionuclides, pesticides are transferred into the air and can be transported on the whole Europe. Based on [*Zobler*, 1986], [*Webb et al.*, 2000] gives a set of 108 soil types at a 1/2 degree resolution all over the world. Four types of chernozem are identified (glossic, haplic, calcic and luvic). The Figure 2 (bottom) displays the fraction of Chernozems over the CHIMERE domain used in this study and Figure 2 (top) the fraction of crop lands after [*Hansen et al.*, 2000]. The second improvement is to have a real distinction between non-chernozemic and chernozemic crop lands in the model.

3.3.3. Third model improvement Finally, a third improvement concerns the **size distribution of emissions**. In our case, we have two types of input informations to constrain the size distribution all along the plume (from the emissions to the most remote areas):

- The size distribution of emitted dust after measurements campaigns over Europe as described in [*Alfaro and Gomes*, 2001]. This leads to mass median diameters for three emissions modes, as D_p=1.5, 6.7 and 14.2μm.
- The PM2.5/PM10 concentrations ratio after surface measurements recorded in Houtem (Belgium) and Harwell (UK).

Using these informations, we adjust the size distribution at the emissions and we checked after long-range transport the modelled concentrations. The comparison of measurements versus model for the PM2.5/PM10 ratio is displayed in the Figure 3. Two important insights have to be mentioned : (i) the general variability of modelled PM2.5/PM10 ratio is conserved

and consistent with measurements, (ii) the sudden decrease near the hour 96 (corresponding to the 24 march [00:00], time of the dust plume passage over these two remote stations) is observed and predicted by the model. The Figure 3 displays a large contribution of coarse particles (about 75 %) in the PM budget. The resulting distribution is displayed in the Figure 4. This corresponds to the best adjustment for the model in this specific case; this also corresponds to a realistic and previously well known distribution such as values recorded over North America by [*Watson and Chow*, 2000].

Figure 4.

A second validation step is to estimate whether this distribution is realistic, by using measurements in between the emissions area (Ukraine) and the end of the plume over land (i.e Belgium and UK). Model and measurements size distributions are compared at the Melpitz station (Germany), located on the way of the plume. The Figure 5 presents the size distribution of modelled PM concentrations for dust only (grey bars) and all PM (including dust and all other compunds, white bars). These values are extracted from the model cell corresponding to the Melpitz (51.53N and 12.93E) station. For the 22 March 2007, there is no dust arriving in Melpitz and the size distribution has a peak centered around 0.3 μm, representative of usual anthropogenic pollution. On 23 March 2007, a first contribution of dust is modelled but without inducing a change in the shape of the concentration distribution. On 24 and 25 March, dust dominates the total mass of the modelled PM and shifts the peak of the distribution to the coarse particles, around 3 μm, the same value as observed in [*Birmili et al.*, 2007]. The model is thus able to fairly reproduce the complex behaviour of dust during this event.

Figure 5.

3.4. Direct impact of model improvements on concentrations

Accounting for the three improvements described above, surface dust emissions are hourly calculated, an example of results is displayed in the Figure 6. The largest fluxes are limited to the Ukraine area.

ure 6.

Using the dust emissions data with these improvements, two simulations were carried out in order to test sensitivity of the model emission parameterization and the ability of the model to reproduce observed remote surface concentrations. The results are presented in the Figure 7 for the two sites already presented in the Figure 3.

ure 7.

The first simulation used the CHIMERE model with all processes (gas and aerosols) except the dust emissions and transport (dashed lines in Figure 7). The second simulation is the same but adding dust emissions and transport. After 24h, the first dust emissions fluxes, and far from the source (in Houtem, Belgium and Harwell, United Kingdom), the model is able to reproduce the magnitude of the concentrations as well as the time of the dust peak. Comparing the two simulations, the net contribution of the dust event in the local Belgium and United-Kingdom air quality is rather clear. While 'background' particulate matter concentrations of $\approx 30\ \mu g.m^{-3}$ is estimated, the dust event contributes to an additional concentration reaching 120 to 180 $\mu g.m^{-3}$, 90% issued from Ukraine. It is noteworthy that these levels largely exceeded the EU standard of 50 μg.m^{-3} (daily mean value), only due to dust emissions.

4. Results

The previous sections showed that after some adaptations to realistic soils, the model is able to calculate a huge and sporadic dust event over a typical Europe surface type. We now turn to the fact that the corresponding dust plume may have been transported farther than over the Slovakia, Poland and Germany stations as described in [*Birmili et al.*, 2007].

4.1. Surface maps of dust concentrations

The develoment of the dust plume is described with a simulation going until 26 March 2007 [00:00]. The surface concentrations results are presented in the Figure 8, with nine maps, every six hours from 23 [12:00] to 25 [12:00] March. The main dust event starts on 23 March 2007 [12:00]. First, it is spatially limited to the Ukraine area and concentrations reaches observed maximum values ($\approx$ 1000 $\mu g.m^{-3}$), the model slightly understimating the maximum amplitude but delivering the huge emission event at the right place and at the right moment. The plume then moves towards Germany (Figure 8 from 23 March [18:00] to 24 March [06:00]). Starting from 24 at [12:00], the plume splits into two parts: a long-range transported plume travelling towards Belgium, United Kingdom (UK) and north of France. A second spot is observed corresponding to new and moderate emissions in Ukraine. At the end of the episode (25 at 12:00), and after two diurnal cycles, a long plume stretches from Ukraine to Belgium, with two additional high concentrations spots: one over the western part of the UK and France, and one over the emissions area. For this episode, the model is consistent with the surface measurements values presented in [*Birmili et al.*, 2007]. It also adds informations over the whole Europe showing that surface concentrations of more than 120$\mu g.m^{-3}$ are due

to Ukrainian dust and may be observed far from the source over the most western part of Europe and Atlantic Ocean.

ure 8.

4.2. Vertical cross-section

The Figure 10 presents vertical cross-sections of dust concentrations focused on the period of the largest emissions event and the transit of the plume to the western part of the domain. The plume vertical extension reaches 1800m at the beginning of the episode, dust being injected in the boundary layer and well-mixed around 12:00 due to the strong turbulence induced by the strong horizontal pressure gradient. From east to west, the dust plume is transported and the vertical structure becomes less homogeneous: several maxima are modelled within the plume (e.g. 24 March [06:00]). At the end of the period, two distinct plumes are observed, one over Northern Germany with a low thickness on 24 March, [00:00] (1200m height) and a most intense one in Ukraine on 24 March [12:00] (more than 2000m height). The Figure 10 shows that the model is able to reproduce the vertical structure of the plume observed and described in [*Birmili et al.*, 2007] with the correct vertical extension around 1800m at noon for each observed days.

ure 9.

4.3. Modelled time series versus measurements

The Figure 10 shows a more detailed comparison of modelled versus measured surface concentrations for the reference simulation with all particles (solid line) and for the simulation without dust (dashed line). This highlights the strong impact of this episode onto local surface measurements very far from the emissions area.

ure 10.

All time-series displayed in the Figure 10 correspond to Belgian, United Kingdom and France (North) sites as explained in the Table 1. The first part of the time period, i.e hours ranging from 21 March [00:00] to 24 [00:00] showed background PM concentrations fairly well reproduced, with values around 30 $\mu g.m^{-3}$. On 25 March [00:00], the plume is first observed over Belgium, followed by UK and north of France. The maximum values reach 200 $\mu g.m^{-3}$ in the Phare d'Ailly rural site. The arrival time of the plume is a slighty predicted in advance by the model. This may be due to the uncertainties in modelled surface wind speed and by the cumulative effect of two days of concentrations to transport across Europe. Even if the station of Lough Navar is the most remote station of our comparison, the model succeeds in catching the local PM maximum with values twice higher than the background values. **Table 1.**

5. Conclusion

In March 2007, an exceptional dust emission event was observed over Ukraine. Surface observations showed that the episode coming from Ukraine induced high dust concentrations transport until Germany. This episode stands as an opportunity to evaluate a dust emission and transport model. We evaluated the CHIMERE chemistry-transport model, in similar versions as described in previous articles, but with an improved dust emission scheme, taking into account landuse and Chernozemic soils in more details. The dust scheme was adaptated to better represents dust size distributions at the emissions (by comparison with surface data after transport). With some tuning performed for a few model parameters, the model is able to successfully reproduce the PM10 concentrations amplitude, spatial pattern, and aerosol size distribution, even very far (a few thousand kilometres) from the source. At peak over Western

Europe the model reproduces the observed peaks of PM up to 200 $\mu g.m^{-3}$ and we showed the net contribution of the Ukrainian episode is certainly up to 170 $\mu g.m^{-3}$.

Using this new CHIMERE model version, the potential impact of such episode on the surface PM concentrations over the whole Europe was evaluated. The starting time of the dust emission was correctly simulated showing that the MM5 meteorological model catches well the specific wind speed values induced by the synoptic meteorological situation. Additional measurements were used and surface concentrations were compared over Belgium, UK and France. The model correctly predicts the peak values over all stations used. This includes stations where the plume was not very intense proving that the surface extension of the plume is fairly reproduced. The vertical structure was shown to be complex with a well-mixed boundary layer and dust concentrations with high values from surface to 1800m far from the emissions area. Finally, we showed that using a size distribution of twelve bins, the model is able to correctly reproduce the complex evolution of the relative part of soil dust in the particle size distribution, from free to loaded dust atmosphere, the model showed a shift from 0.3 to 3 μm in the aerosol distribution.

This event is a relatively rare event, and it would be interesting to diagnose its frequency. It is not clear whether other important dust events could be initiated from other European regions. However, since they are probably of more minor amplitudes they are hardly detectable from the aerosol background. Improvements in model natural emissions are required to better simulate these phenomena, which are required to better predict PM10 concentrations and understand their role in regional air quality.

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Figure Captions

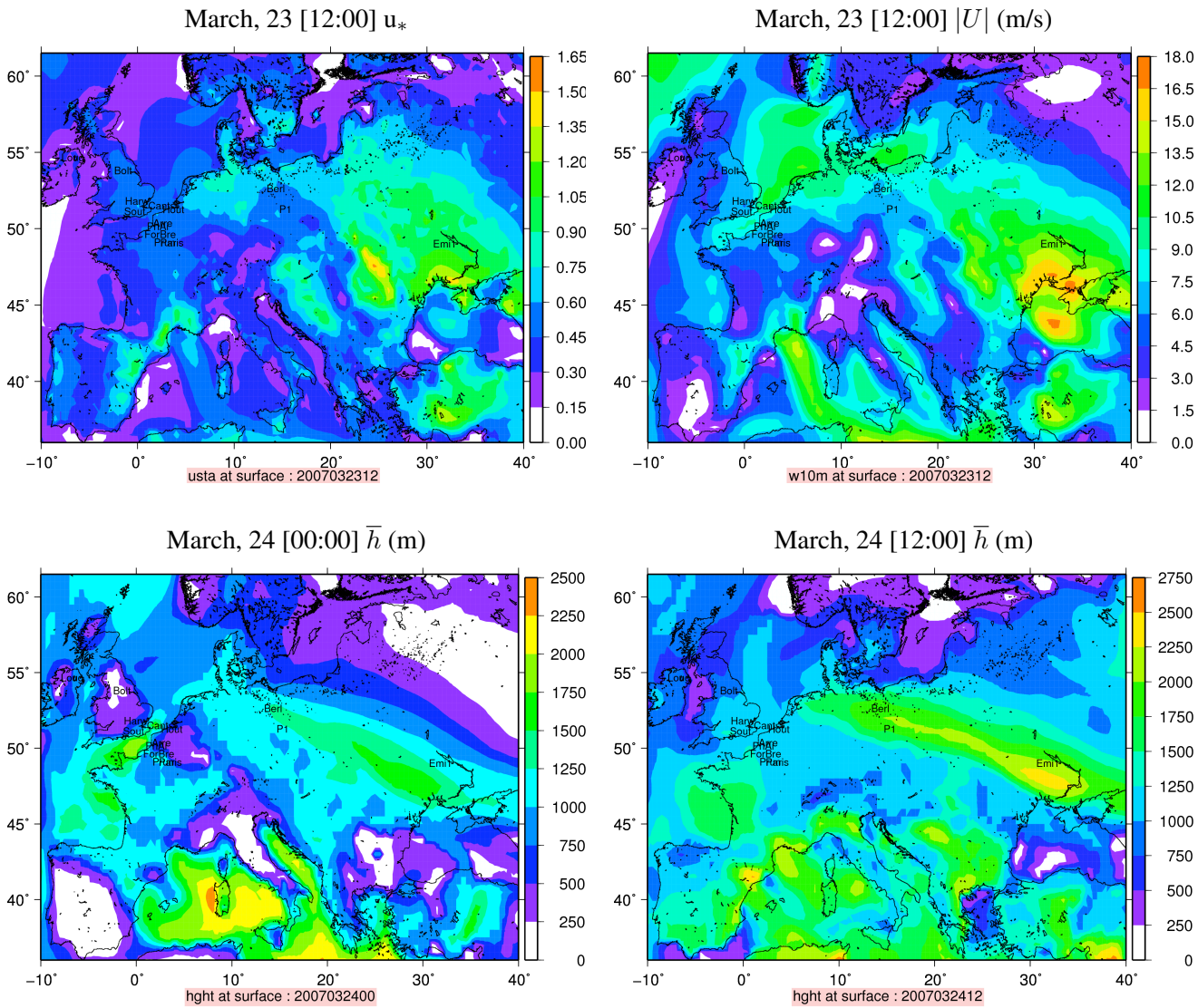

Figure 1. *[top] Friction velocity and horizontal 10-m velocity calculated on March, 23 2007, 12:00, [bottom] Wind speed on March, 23 2007, 12:00 and boundary layer height* $\overline{h}$ *on March, 24 [00:00].*

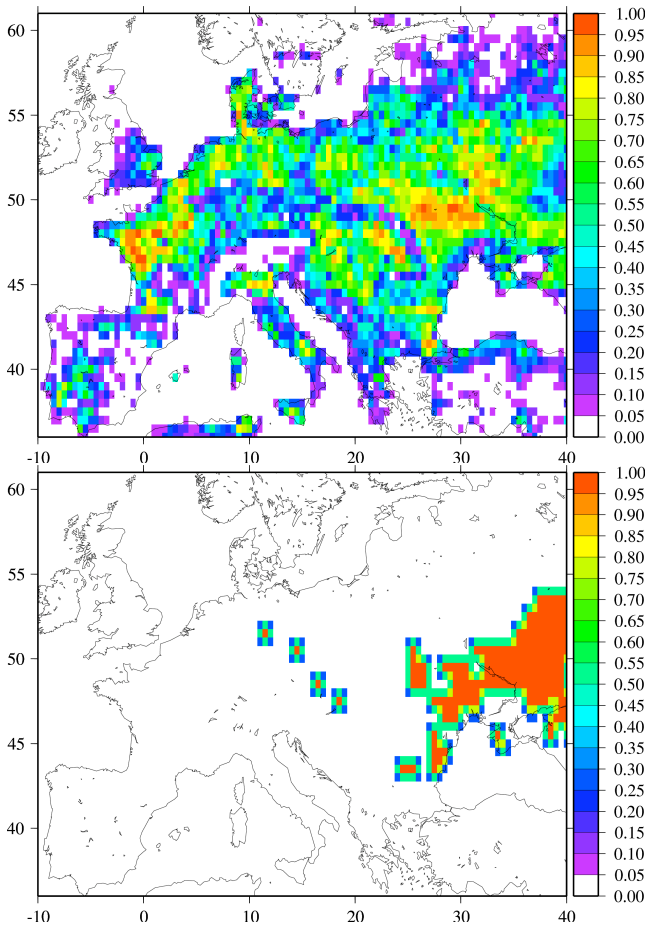

Figure 2. *Fraction of [top] cropland and [bottom] Chernozemic soil in the simulation domain*

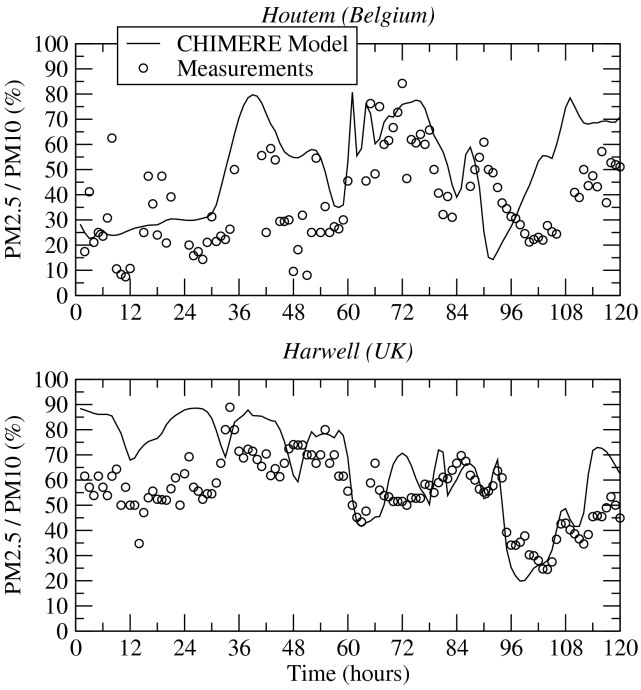

Figure 3. *Modelled and measured PM2.5/PM10 ratio in Houtem (Belgium) and Harwell (United Kingdom) for the period from 20 to 25 March 2007.*

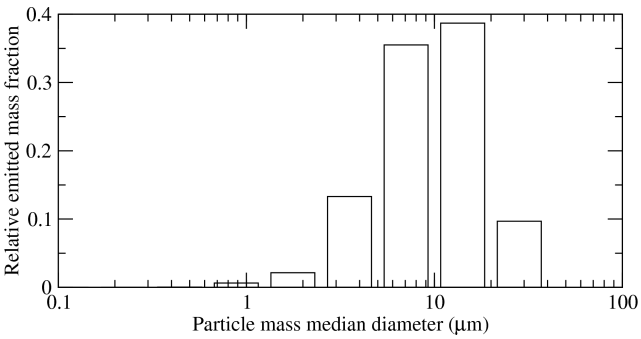

Figure 4. *Histogram of the relative mass fraction of dust emissions in each model bins.*

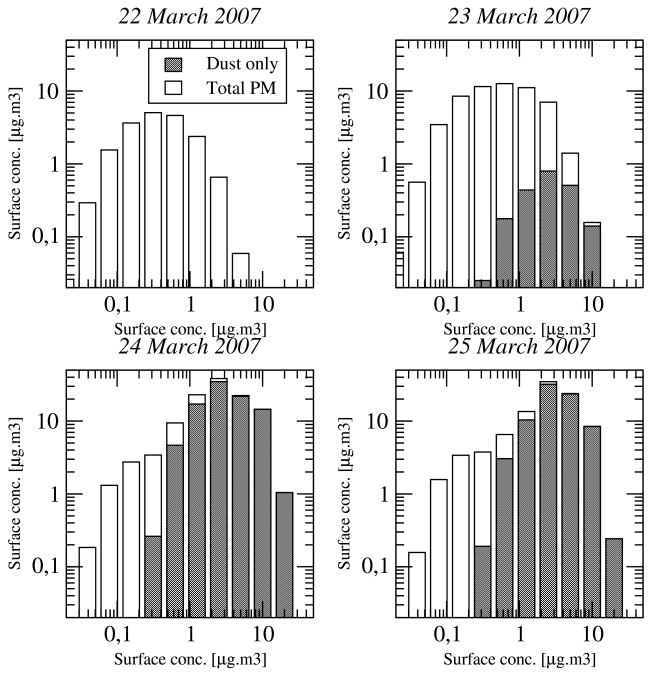

Figure 5. *Particle size segregated concentrations in the twelve model bins, corresponding to the Melpitz surface stations and for four consecutive days, 22, 23, 24 and 25 March 2007 (daily mean values).*

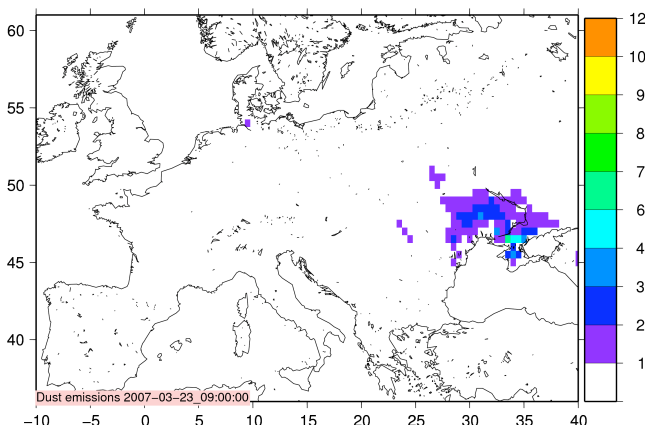

Figure 6. *Dust emissions ($g.cm^2.s^{-1}$) for the March 23, 2007 at [09:00] with improvements in the cropland landuse fraction, associated to Chernozemic soils characteristics.*

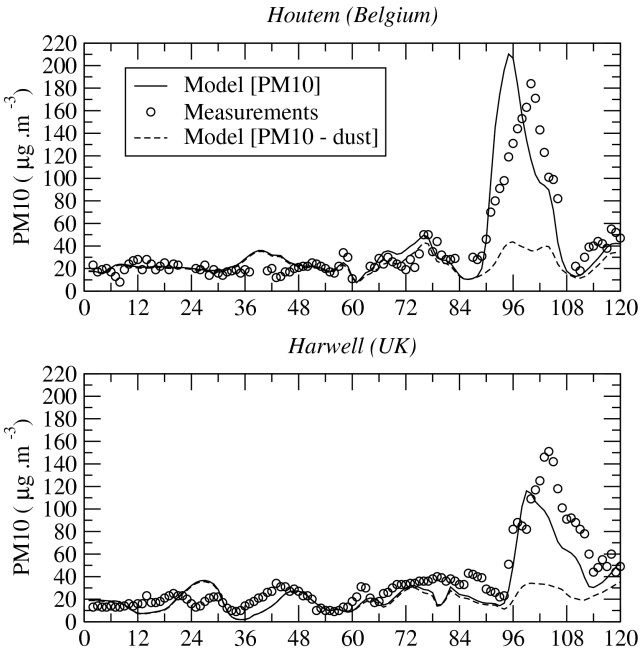

Figure 7. *Concentrations time series of PM10 modelled and measured in Houtem (Belgium) and Harwell (United Kingdom) for the period from 20 to 25 March 2007.*

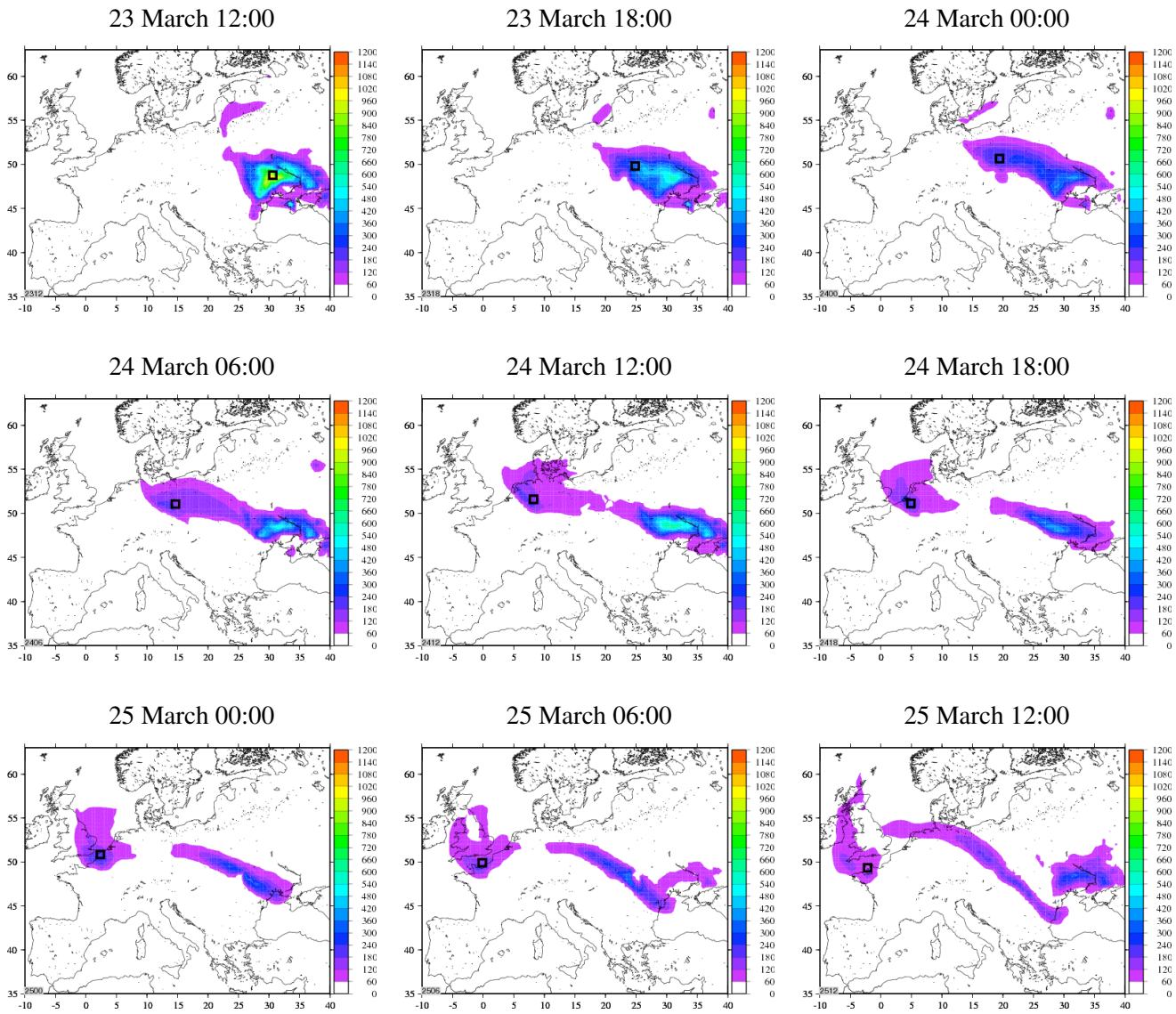

Figure 8. *Surface dust concentrations ($\mu g.m^{-3}$) modelled from the 23 March 2007 [12:00] to the 25 March 2007 [12:00], every 6h.*

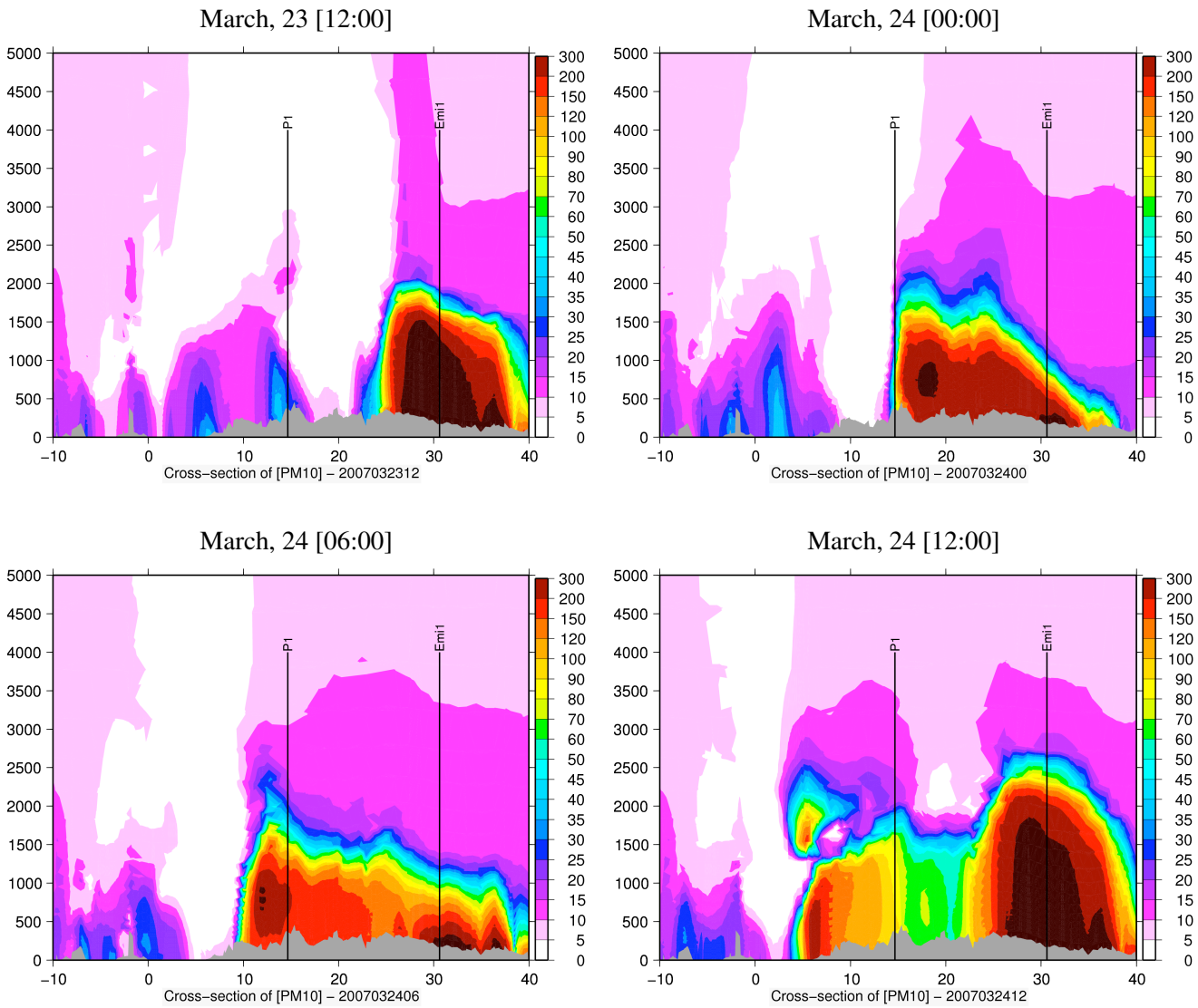

Figure 9. *Time series of dust concentrations ($\mu g.m^{-3}$) along the plume. Color scale stops at 300 $\mu g.m^{-3}$, even if some very important values are modelled.*

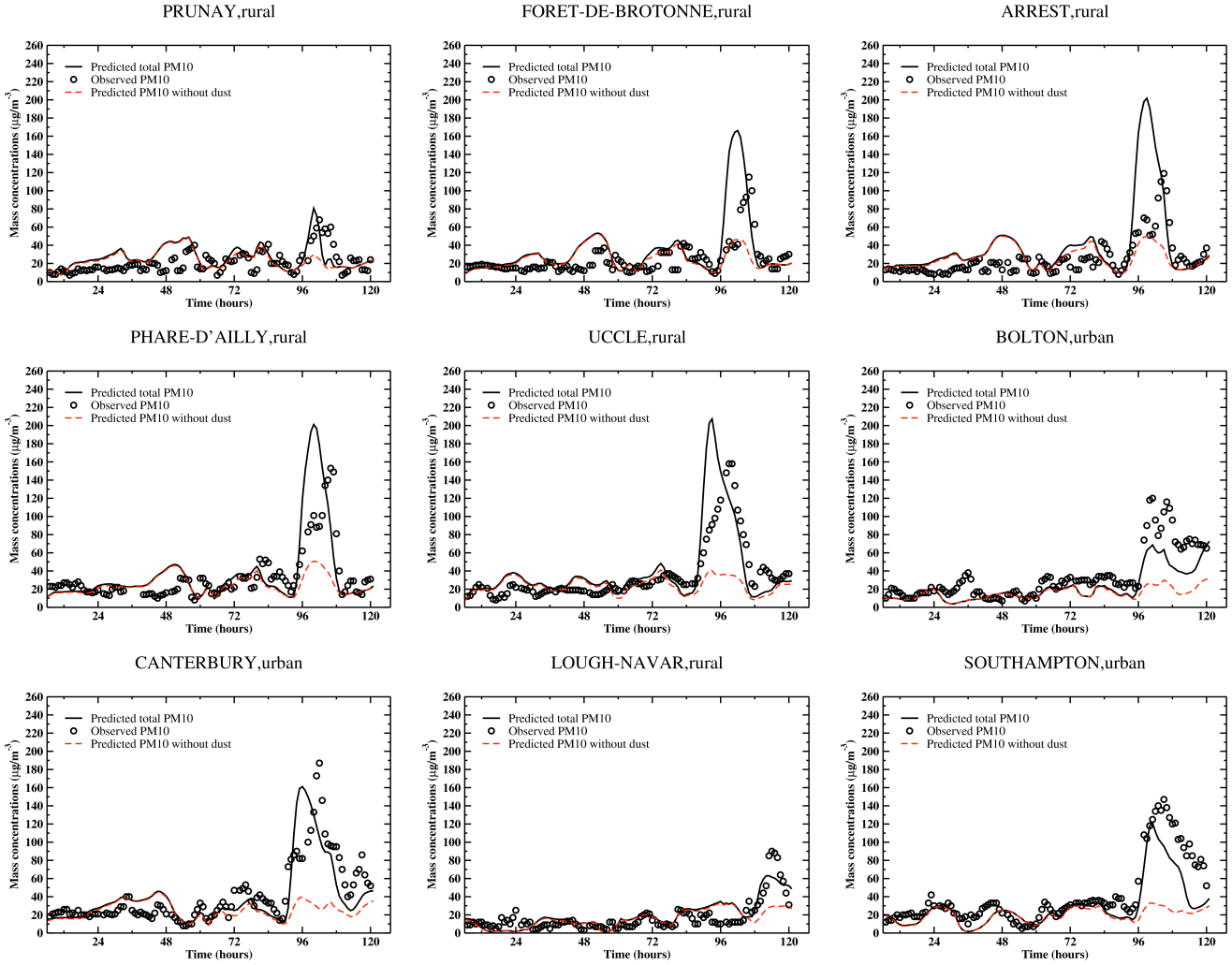

Figure 10. *Time series of dust concentrations along the plume.*

Tables

Station	Country	Latitude	Longitude
Uccle	Belgium	50.80	4.36
Houtem	Belgium	51.02	2.58
Prunay	France	48.86	1.68
Foret de Brotonne	France	49.41	0.69
Arrest	France	50.13	1.61
Phare d'Ailly	France	49.92	0.96
Canterbury	U.K.	51.28	1.08
Lough Navar	U.K.	54.43	-7.90
Southampton	U.K.	50.90	-1.40
Bolton	U.K.	53.58	-2.43
Harwell	U.K.	51.57	-1.32

Table 1. *Names and coordinates of stations presented for the time series results in the Figure 10.*