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Modélisation d'épisodes de pollution particulaire en région parisienne (2003)

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TABLE DES MATIERES

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1. INTRODUCTION

Les particules atmosphériques ont une influence importante sur le climat, la visibilité et principalement sur la santé, comme cela a été démontré dans de nombreuses études épidémiologiques (Katsouyanni et al., 1997 ; WHO, 2003 ; Pope et al., 2002). Elles font l'objet d'une surveillance de plus en plus systématique, notamment pour la fraction fine (PM2.5). En période hivernale, il est fréquent d'observer en Europe des zones de haute pression de la Mer du Nord à la Scandinavie, dirigeant vers le Nord de la France des masses d'air froid souvent chargées en polluants d'origine continentale qui s'ajoutent ainsi à la charge de particules d'origine plus locale.

Les modèles de qualité de l'air sont à l'heure actuelle capables d'estimer les concentrations de fond de matière particulaire à une échelle allant de quelques kilomètres à plusieurs dizaine de kilomètres. Des incertitudes importantes demeurent cependant. Elles sont dues à la qualité parfois insuffisante des inventaires d'émission de particules primaires, et également à une connaissance limitée de certains processus physicochimique. Le modèle CHIMERE est utilisé par l'INERIS pour des études de scénarios de réduction de polluants et pour la prévision de la qualité de l'air en France et en Europe (PREV'AIR). Il permet aussi une meilleure compréhension de la phénoménologie des aérosols à travers des études d'épisode plus ciblés.

Ce rapport présente l'analyse de plusieurs épisodes de pollution particulaire en Ile-de-France qui ont eu lieu durant l'hiver 2003 (Février-Mars). Ce travail a fait l'objet d'une publication acceptée dans *Atmospheric Environment* (en annexe), et il fait suite au rapport pour la LCSQA 2003 référencé DRC-Bbe-03-45600-135 qui détaillait en partie les évènements de pollution particulaire observés. Il s'agit d'une étude de modélisation appuyée par de nombreuses mesures expérimentales de PM10, sulfates, nitrates, et ammonium réalisées sur deux sites à proximité de Paris.

2. LES EPISODES DE POLLUTION ETUDIES

2.1 SITUATION DU 21 FEVRIER 2003

Du 13 au 18 Février
A partir du 13 Février 2003, un anticyclone stationne sur le Danemark et dirige sur la France un flux d'Est à Nord-Est très froid, les températures sont très basses et ne dépassent 0° C dans l'après-midi que très localement près des côtes. La masse d'air est très sèche, le vent d'Est est faible mais non nul.

Du 19 au 20 Février
Le flux en surface s'oriente au Sud-Est avec un vent plus fort les 19 et 20 sur le Nord de la France. Une perturbation s'approche du Sud-Ouest de la France et sera rejetée vers le Nord-Ouest de la France avant de quitter la bordure Nord-Ouest du pays le soir du 20 Février. Cette légère dégradation aura eu pour conséquence un radoucissement des températures (de 3°C à 10°C à Paris du 19/02 au 20/02 à 12h GMT) et une augmentation de l'humidité dans le Nord-Ouest de la France.

Le 21 Février
Le matin du 21 Février, le Nord-Ouest de la France connaît quelques brouillards (aucun brouillard sur Paris). Les vents sont très faibles et l'humidité relative est importante sur le Nord-Ouest de la France. Le 22 Février, la situation se dégrade franchement par l'Ouest de la France.

2.2 SITUATION DU 21 MARS 2003

Du 13 au 20 Mars
Un anticyclone centré sur l'Ouest de l'Angleterre oriente un flux d'Est sur la France. A 12h GMT les températures sont comprises entre 10 et 15°C sur la France. Quelques gelées sont observées du Sud-Ouest au Nord-Est. Des brumes et brouillards persistants sont observés en Allemagne, aux Pays-Bas et en Grande-Bretagne. En France l'atmosphère est sèche en début de période, des brouillards sont observés les 18,19 et 20 sur le Nord, mais ils se dissipent dans la journée. Sur la région Parisienne, le vent devient très faible et s'oriente au Nord-Ouest le soir du 20 Mars. Des entrées maritimes envahissent alors le Nord-Ouest du pays.

Le 21 Mars
Des brouillards ont envahi le Nord-Ouest du pays jusqu'à la région Parisienne. Le vent est quasi nul. Les brouillards persistent une grande partie de la journée. La température à Paris est de 10°C à 12h GMT. Le 22 Mars, une dégradation intervient par le Sud-Ouest de la France et met fin provisoirement à cet épisode.

3. MODELISATION DES EPISODES

Le modèle de chimie transport CHIMERE a été appliqué sur l'Europe avec une résolution de 0.5°x0.5° avec un zoom sur la région Ile-de-France d'une résolution d'environ 5 km. Le modèle CHIMERE dans sa version aérosol est largement détaillé et évalué par Bessagnet et al. (2004) ainsi que dans le précédent rapport LCSQA référencé DRC-Bbe-03-45600-135. Les calculs ont été effectués du 10 Février 2003 au 31 Mars 2003. Pour ces épisodes, le module d'aérosol modélise 11 sections de 10 nm à 20 µm.

Plusieurs types d'observations sont disponibles sur les 2 sites de mesures: Gennevilliers (site urbain de fond) et Prunay (site rural). Pour les PM10, les mesures d'AIRPARIF (TEOM), ainsi qu'une mesure gravimétrique PARTISOL sont disponibles. De plus, des mesures de sulfate, nitrate et ammonium ont été réalisées.
Sur la figure 1 apparaissent les séries temporelles des observations et des résultats du modèle. Si globalement les résultats simulés pour les PM10, nitrate et ammonium semblent assez bien corrélés avec les observations, le modèle semble avoir plus de difficultés pour simuler les sulfates. En effet, si l'hiver la chimie aqueuse est une importante source de sulfates elle reste difficile à simuler car largement dépendante de paramètres délicats à déterminer : le pH des gouttelettes d'eau et la quantité d'eau liquide dans l'atmosphère notamment (faiblesse des modèles météorologiques).

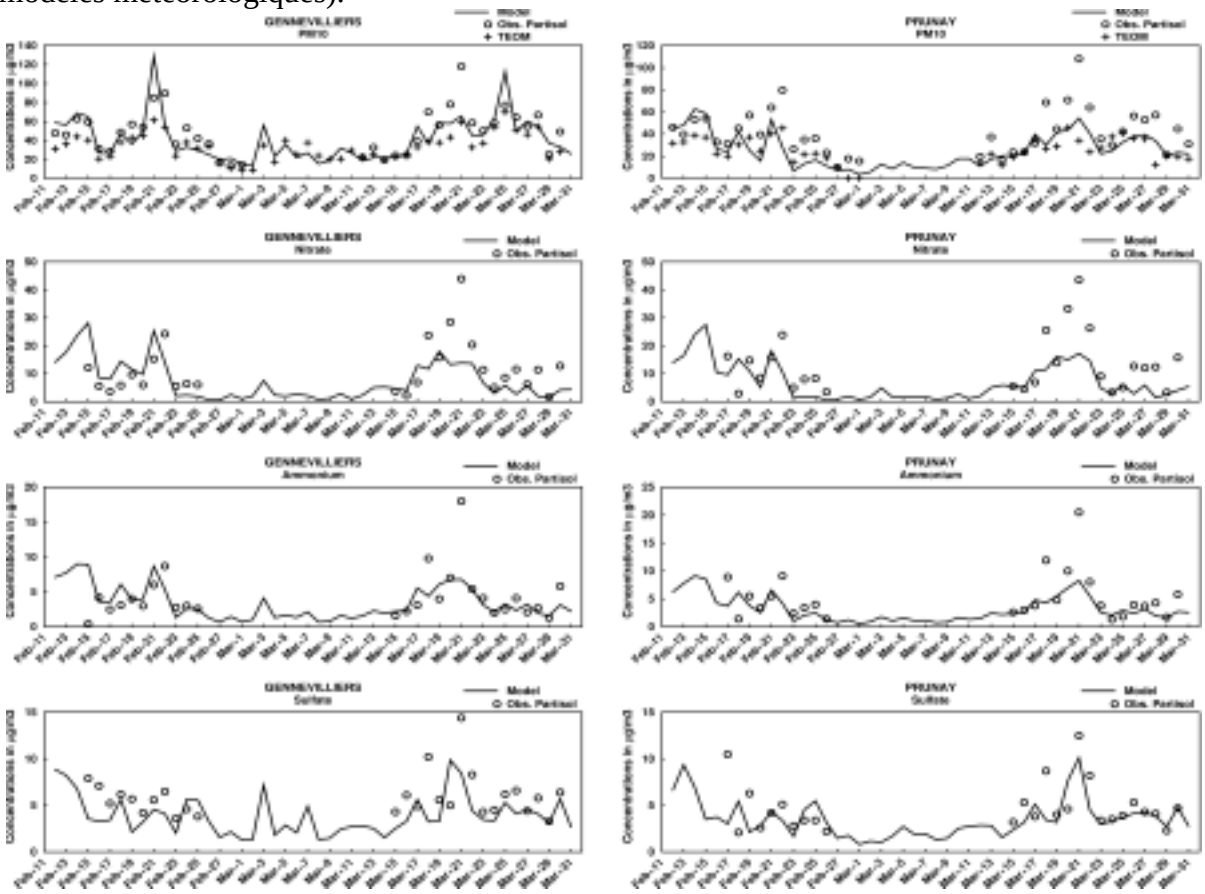

Figure 1 : Comparaison Mesure/Modèle pour les PM10, Sulfate, nitrate et ammonium

4. ORIGINE DES EPISODES DE POLLUTION

Une analyse académique est proposée pour déterminer la contribution de différentes zones géographiques. Elle concerne d'une part, l'influence des apports extérieurs au Bassin Parisien, et d'autre part l'influence de la zone Ile-de-France sur le reste du domaine. Pour ce faire, les émissions dans les zones géographiques dont on souhaite qualifier l'influence sont annulées.

4.1 INFLUENCE DE LA REGION PARISIENNE

La figure 2 représente la différence de concentrations entre deux simulations : l'une avec l'ensemble des émissions et l'autre obtenue en supprimant les émissions de la région Parisienne. On constate que l'influence des émissions de la région Parisienne s'étend sur plusieurs centaines de kilomètres. Le 21 Mars cette influence atteint la région Bordelaise avec une différence variant de 10 à 15 µg/m^3.

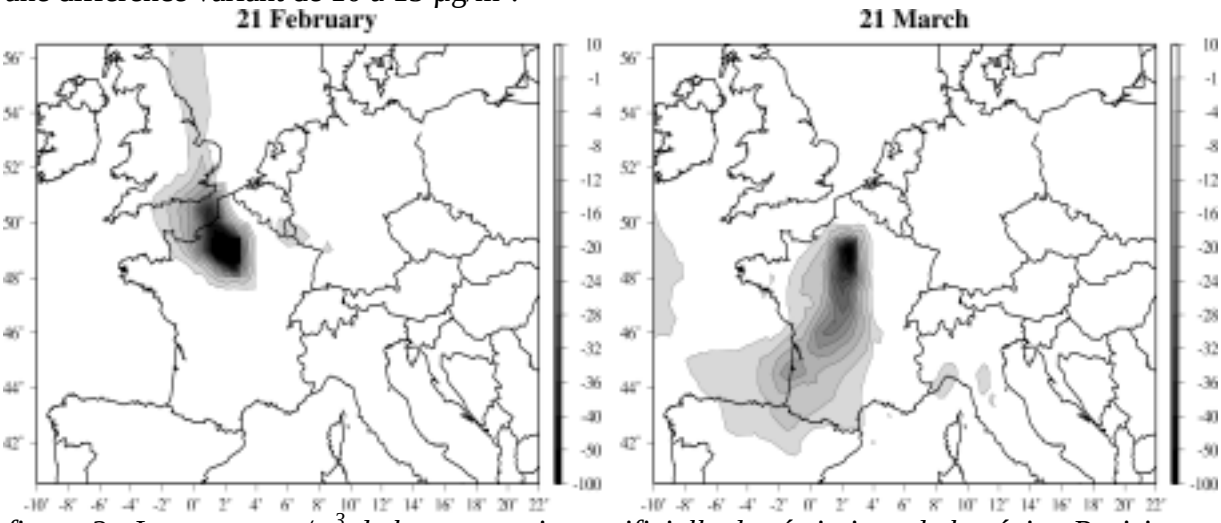

figure 2 : Impact en µg/m^3 de la suppression artificielle des émissions de la région Parisienne sur le reste du domaine

En région Parisienne, à Prunay et Gennevilliers, les séries temporelles des concentrations en espèces particulaires issues des contributions extérieures à Paris (figure 3), montrent que lors des épisodes, celles-ci peut atteindre 50% pour les PM10 à Gennevilliers, et 75% à Prunay. Cette contribution en pourcentage est donnée par le rapport de la concentration donnée par la simulation sans les émissions de la région Parisienne divisé par la concentration donnée par la simulation avec toutes les émissions. Cette contribution est largement due à la présence de nitrate d'ammonium transporté loin des sources de ses précurseurs. Notons que les sulfates en région Parisienne proviennent largement de l'extérieur de Paris.

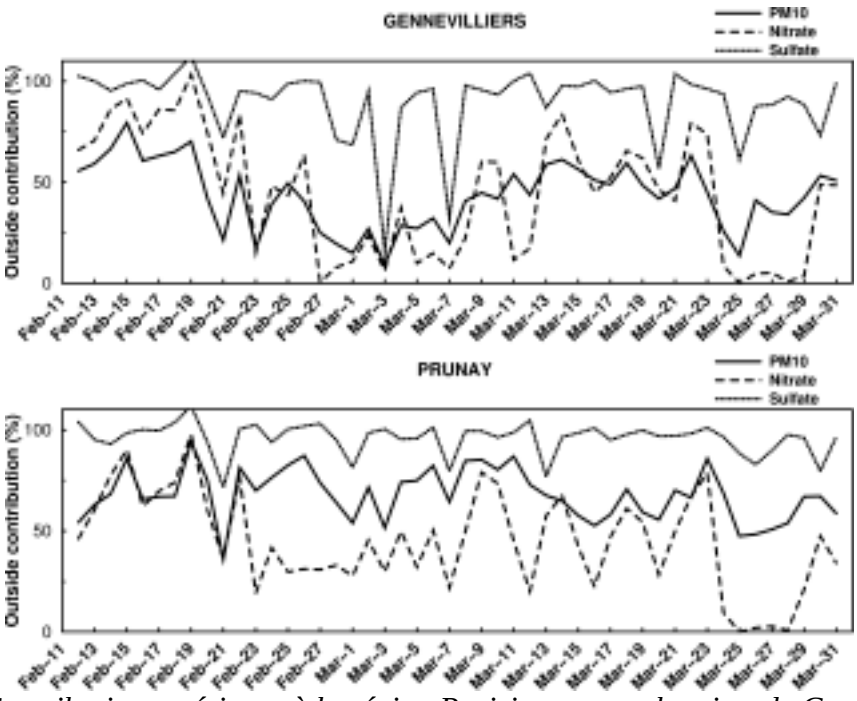

figure 3 : Contribution extérieure à la région Parisienne pour les sites de Gennevilliers et Prunay

Remarque importante

Lorsque l'on parle de contribution, il ne s'agit pas de directement d'évaluer la quantité de polluant imputable à une partie du domaine. En effet, les phénomènes modélisés sont parfois fortement non-linéaires et il possible d'obtenir une contribution légèrement supérieure à 100%, c'est le cas pour les concentrations de sulfate en figure 3. De même, une molécule d'acide nitrique formée dans une région A régissant avec une molécule d'ammoniac émise en zone B peut former du nitrate d'ammonium particulaire en zone C. Il est alors délicat d'imputer la formation du nitrate d'ammonium en zone C aux polluants émis en zones A ou B. Cela pourrait être possible en marquant toutes les molécules en fonction de leur zone d'émission, mais c'est pour l'instant irréaliste en terme de temps calcul. C'est pourquoi le terme de « contribution » doit être pris avec prudence et considéré comme une indication de l'influence d'une région sur une autre.

4.2 INFLUENCE DES PAYS VOISINS

Dans le paragraphe précédent, il a été clairement mis en évidence l'influence des émissions extérieures à Paris sur les concentrations de PM10 en région Parisienne. Par le même type d'étude académique, il est donc possible d'étudier l'influence des pays voisins sur les concentrations de PM10 en région Parisienne.

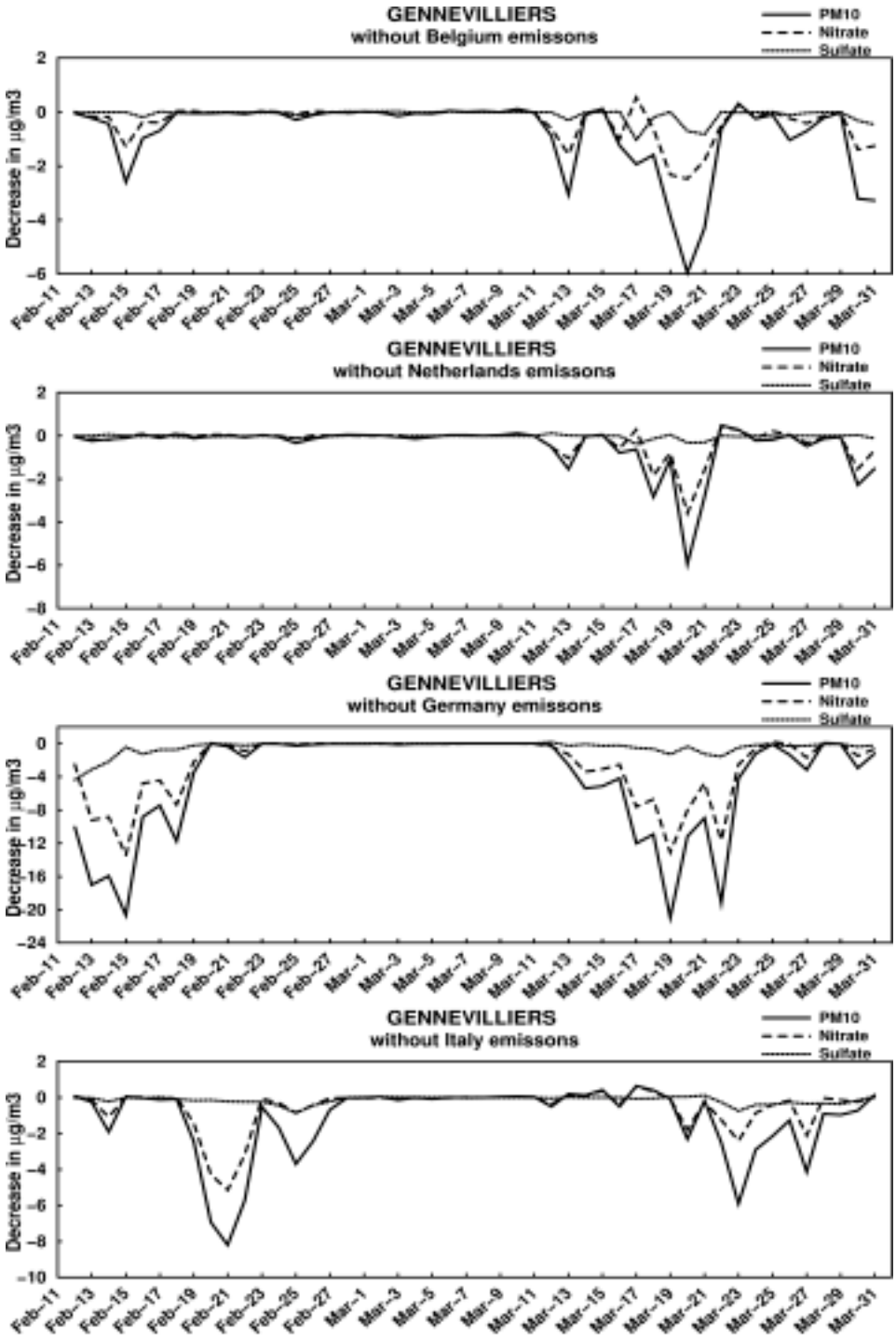

figure 4 : Evolution de l'influence de la suppression des émissions des pays voisins sur les concentrations de PM10 en région Parisienne

La figure 4 montre les séries temporelles des différences entre la nouvelle simulation sans les émissions du pays considéré et la simulation de référence comprenant toutes les émissions. Lors du premier épisode début Février 2003, seule l'Allemagne influence assez fortement les concentrations en région Parisienne avec toutefois une moindre contribution le 21 Février au plus fort de l'épisode. L'Italie a un effet assez important mais décalé dans le temps suite à la rotation du flux au Sud-Est. Lors de l'épisode de Mars 2003, l'Allemagne, les Pays-Bas et la Belgique ont une influence significative sur les PM10 en Ile-de-France. L'Italie présente toujours un effet décalé dans le temps.

5. INFLUENCE DE LA FRANCE

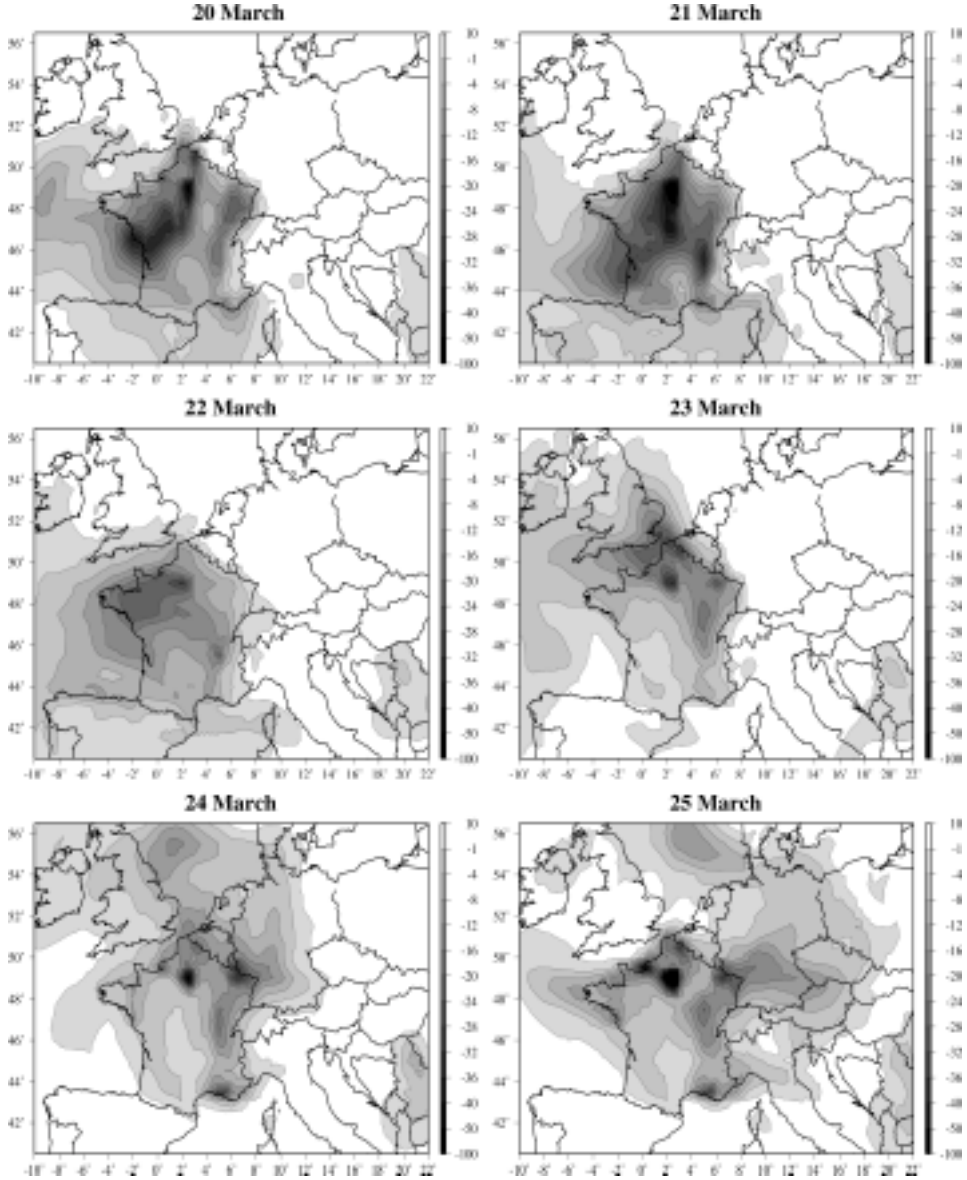

figure 5 : Influence de la France sur les pays voisins (écart de concentrations en µg/m3)

Une simulation a été effectuée en supprimant les émissions de la France afin de déterminer l'influence des émissions Françaises sur les concentrations en PM10 en Europe. La figure 5 représente l'écart avec la simulation de référence comprenant l'ensemble des émissions pour les journées du 20 au 25 Mars 2003. Au plus fort de l'épisode sur Paris, les 20 et 21 Mars, les émissions Françaises ont une influence uniquement sur le Nord de l'Espagne. En revanche, lorsque l'épisode se termine, le vent tournant au Sud-Est puis Sud-Ouest, la France rejette de nombreux polluants, d'abord sur les Iles Britaniques puis successivement sur la Belgique, les Pays-Bas et l'Allemagne, jusque vers les pays de l'Est.

6. CONCLUSION

Le modèle CHIMERE dans sa version aérosol a été utilisé sur l'Europe avec un zoom sur la région Parisienne pour analyser des épisodes de pollution particulaire. Deux épisodes de pollution hivernale (Février-Mars 2003) ont été étudiés. Dans un premier temps, une analyse a été menée afin d'évaluer le modèle sur deux sites instrumentés proches de Paris. Il reproduit correctement les concentrations en nitrate, ammonium et PM10. Le modèle semble avoir plus de difficultés pour les sulfates notamment à cause de la chimie aqueuse difficile à modéliser en période hivernale. Les différences obtenues entre les deux systèmes de mesure TEOM et PARTISOL largement dues à l'évaporation du nitrate d'ammonium, ont de nouveau été mis en évidence.

Les épisodes étudiés ont une origine largement continentale et pourraient être notamment dus aux polluant primaires (NOx et PM essentiellement) émis par les pays voisins tels que l'Allemagne, la Belgique et les Pays-Bas mais aussi l'Italie en fin d'épisodes lorsque le flux bascule au Sud-Est. Une quantification de ces contributions est proposée et montre une forte influence de l'Allemagne sur les concentrations de particules en région Parisienne (environ 20 $\mu g/m^3$) lors des épisodes de pollution. L'Italie, les Pays-Bas, et la Belgique ont une contribution moindre (autour de 10 $\mu g/m^3$). Une autre analyse a permis de montrer qu'après un épisode de pollution, la France pouvait à son tour avoir un impact significatif sur les concentrations de PM dans les autres pays au fur et à mesure que le flux de masse d'air s'oriente à l'Ouest (environ 10 $\mu g/m^3$).

ANNEXE

Origin of particulate matter pollution episodes in wintertime over the Paris Basin.

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Abstract

Several wintertime pollution events due to particulate matter on the Paris Basin in 2003 are investigated in this paper. High pressure systems close to Scandinavia or the North Sea involve highly stable conditions with slight Northeasterly flux on France leading to high airborne pollutant concentrations. An evaluation of the CHIMERE model results against observations over the Paris area is proposed. While PM_{10}, nitrate and ammonium seem fairly well reproduced, sulfate concentrations remain difficult to predict. A specific study, by removing Ile-de-France emissions, displays on February 21 and March 21 episodes an important ammonium nitrate contribution, mainly originating from outside Paris area. According to the model results, the Paris Basin has also a large influence up to the South West of France. In a similar way, an investigation of the possible sources outside the Paris basin, displays a strong influence of emissions from Germany, the Netherlands and Belgium during these episodes. To a lesser extent, Italy has an influence on the Paris area at the end of the episodes. It is also demonstrated that in some situations, the contribution of locally produced or emitted particles is prevalent at the ground level. The influence of French emissions is also studied from March 20 to 25, displaying an influence on Spain and a strong impact at the end of the episode successively on Great Britain, Belgium, the Netherlands when winds veer Southeast and West. This influence is also significant up to Eastern Europe.

Keywords : modeling study, aerosols, episodes, model evaluation, emission inventory, ammonium nitrate

Introduction

At the global scale, particles act on climate by affecting the Earth's radiative balance ; altering the scattering properties of the atmosphere (direct impact), and changing cloud properties (indirect impact). Visibility impairment, as a result of scattering and absorption of light by particles, is another problem due to particulate matter (PM) pollution. Particles in the atmosphere have recently received much interest because of increasing epidemiological and experimental evidence of their impact on human health (Katsouyanni et al., 1997 ; WHO, 2003 ; Pope et al., 2002 ; EPA, 2004).
Since most people live in large city areas, focussing at the urban scale allows for a better evaluation of the exposure of their inhabitants. Although particulate matter measurements have been deployed in most cities, the modeling approach is essential at local scales in order to investigate processes leading to PM pollution episodes and to predict their concentration levels. Modeling tools are also used to study emission control scenarios. Seigneur (2001) reviews the current status of the mathematical modeling of atmospheric particulate matter and the ability of such tools to simulate

pollution episodes. This review suggests that several models (Jacobson, 1997 ; Pai et al., 2000 ; Ackermann et al., 1998 ; Meng et al., 1998) provide a fairly comprehensive treatment of the major processes, but the author stated that some uncertainties remain. For the CHIMERE model, Bessagnet et al. (2004) reported a complete panel of error statistics for rural stations in Europe, and Hodzic et al. (2005) proposed a similar evaluation over the Paris Basin. These analyses were performed over an entire year to get representative results. They showed the ability of the CHIMERE model to deal with the simulation of atmospheric aerosol components.
The topic of this paper is to study several wintertime PM pollution episodes over the Paris Basin at the continental and local scales. In France, high pressure systems in the North of Europe give highly stable and windless atmospheric conditions, sometimes favoring the advection of very cold air masses from the Central Europe. These situations lead to strong PM pollution episodes over the North of France. Hereafter, three wintertime episodes occurring in February and March 2003 over the Paris basin are studied with the CHIMERE model. Two PM measurement systems, TEOM and PARTISOL, are available to get data to assess model performances. The TEOM instrument is routinely used to get continuous measurements. However, artifacts may occur during the conditioning procedure (sample heated at 50°C) due to the volatilization of PM components such as ammonium nitrate (Allen et al., 1997) or semi-volatile organic species. To avoid and quantify the TEOM measurement uncertainties, the gravimetric method PARTISOL is used. PARTISOL filters are analyzed to determine sulfate, nitrate and ammonium concentrations. Due to instrumental constraints, only daily measurements are available with the PARTISOL system. These data are used to evaluate the model at two sites near Paris. The question of the origin of these high PM concentrations during the episodes is addressed. The respective contribution of PM production by local sources and long range transport is investigated. A sensitivity analysis on the emission inventories is proposed to understand the impact of Paris area and neighboring country emissions on PM concentrations observed during these episodes. The influence of French emissions on the rest of Europe is also discussed.

Model setup

In this study, the regional version (V200501G) of the Chemistry Transport Model, CHIMERE, applied over Europe (Schmidt et al., 2001; Bessagnet et al., 2004 ; Vautard et al., 2005) is used to constrain a local version applied over the Ile-de-France area (Hodzic et al., 2005). The model grid at the European scale ranges from 10.5°W to 22.5°E and from 35°N to 57.5°N with a ½ degree resolution both in latitude and longitude. The domain covers most of the Western Europe and the Western Mediterranean basin. The Ile-de-France subdomain grid covers the Paris Basin with a 6 km resolution. 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) with recent improvements reported in Vautard et al. (2003). The aerosol module is presented in Bessagnet et al. (2004) and Bessagnet and Rosset (2001). The full model documentation for the updated version can be found via internet at : http://euler.lmd.polytechnique.fr/chimere.
In this version, particles are assumed to be composed of 7 chemical species : PPM (Primary Particle Material), Dust (desert dust), SOA (anthropogenic and biogenic Secondary Organic Aerosol), Sulfate, Nitrate, Ammonium and Water. Sea salts are not considered in this exercise, this species has only an influence on coastal regions. PPM includes not only anthropogenic primary particle materials but also biogenic particles like vegetative debris, insects, plant waxes and particles produced by soil erosion (detailed in Vautard et al., 2005). These latter species are expected to be negligible in winter (due to high soil humidity). In this version, particle diameters are described by 11 bins ranging from 10 nm to 20 µm. The model accounts for the coagulation process as described in Gelbard and Seinfeld (1980), and Warren (1986). The dynamic of the absorption process of organic and inorganic semivolatile species is parameterized with a first order equation. For the ternary system, Sulfate/Nitrate/Ammonium, the thermodynamic equilibrium is computed with the ISORROPIA model (Nenes *et al.*, 1999). Heterogeneous chemical processes on particles and fog droplets (nitrate production) and a simplified sulphur aqueous chemistry (sulphate production) have

been implemented. Moreover, a preliminary chemical module describing the formation of secondary organic aerosols was introduced.
Meterological are provided by the NCEP (National Center for Environmental Prediction) analyses, refined by the 5th generation Pennsylvania State University model : MM5 (Dudhia, 1993), version 2.3.6. In order to have low computational cost, MM5 is used at the continental scale with a relatively low resolution (36 km) over a domain encompassing the CHIMERE domain, with 25 vertical levels. At the local scale, a 5 km horizontal resolution is chosen.
In order to account for pollutants transported through the model domain boundaries, the continental runs are driven by GOCART model climatologies (Ginoux et al., 2001; Ginoux et al., 2004) for aerosol species, while the MOZART climatologies (Horowitz et al., 2003) are used for the gas phase species. PM components at the boundaries include desert dust, organic and elemental carbon, and sulfates.
All simulations listed in Table 1 are run on the February 10 – March 31 period. Results from February 12 are presented to leave a two days spin-up.

Emission inventory

The 2001 anthropogenic emission data from the EMEP data base have been used for the continental runs (Vestreng et al., 2004). Annual emitted amounts of NO_x, CO, SO_x, NMVOC, NH_3, PPM_{10} and $PPM_{2.5}$ are available for 11 SNAP activity sectors. These data, initially provided by each country, are given on the EMEP grid (50 km resolution), following the methodology described in Vestreng (2003).
Calculation of model species emissions is made in several steps. First, the spatial emission distribution from the EMEP grid to the CHIMERE grid is performed using an intermediate fine grid at 1km resolution. The knowledge of soil types on the fine grid allows for a better distribution of the emissions according to urban, rural, maritime and continental areas. This high resolution land use inventory comes from the GLCF (Global Land Cover Facility) data set. Time profiles of NO_x, CO, SO_x and NMVOC are considered depending on SNAP activity sectors and are provided by the IER (University of Stuttgart). For NH_3, no time variability is considered. According to Aumont et al. (2003) HONO emission is set to 0.8% of NO_x while NO_2 emission is set to 9.2% of NO_x emissions, the remaining NO_x emissions being NO. Afterwards, for each SNAP activity sector, the total NMVOC emission is split into emissions of 227 real individual NMVOC according to the AEAT speciation (AEAT, 2002). Finally, real species emissions are aggregated into model species emissions. For instance, the MELCHIOR chemical mechanism, used in CHIMERE, accounts for 10 NMVOC. Mass-reactivity weighting of real emission data is done following the methodology of Middleton et al. (1990), so that the overall ozone production capability of the emission mixes is kept constant through the emission processing procedure.
For the local scale simulations over Ile-de-France area, the official emission inventory developed by the Paris air quality monitoring network AIRPARIF is used. This inventory is based on 2001 data, a detailed description is given in Hodzic et al. (2005).

Available observations

Two sites of the AIRPARIF air quality network have been equipped with monitoring instruments : a urban site near Paris, Gennevilliers (48°56'N, 2°18'E) and a rural site, Prunay (48°52'N, 1°40'E) located 50 km in the West side of Paris. Two kinds of PM10 measurements are available during these episodes. A gravimetric method PARTISOL-Plus (daily data) and a routine method TEOM (hourly data). The PARTISOL-Plus has been assessed as an equivalent method compared to the EU reference method (EN 12341 standard). The TEOM method is a well known method largely used in France ; however, it is not considered as a standard method.
The PARTISOL filters were weighed using a microbalance (Mettler Toledo MT5) with a sensitivity of ± 1 µg. The used filters were stored in a room controlled in temperature (20 ± 1°C) and relative humidity (50 ± 3%) and equilibrated with these conditions for at least 24-h prior to weighing. The balance was first calibrated and zeroed and the electrostatic charges on the filters were eliminated using an ionization system (HAUG Discharge system, Multistat). Each filter was weighed three

times and the mean of the three values was recorded with a standard deviation less than 4 µg. All filters were weighed again after 24-h to control possible bias. Two control filters were weighed during each weighing session and used to correct for weight changes in the sampled filters cause by variations in the balance room atmosphere. Sampling semi-volatile compounds in airborne particles is complicated by the fact that positive or negative artifacts (Solomon et al., 1999) may occur, due to volatilization during the sampling process or to adsorption of gaseous substances on deposited particles or on the filter material itself. However, Chow et al. (1994) reported that losses of nitrate from the Teflon filter are lower in winter (period of the study). Moreover, losses of semi-volatile compounds are minimized during the sampling process, by maintaining the sampler temperature near ambient conditions and conditioning the filters in a protective container during the transit to the laboratory at a temperature below 20°C and storing the filters in a freezer after weighting.
A TEOM series 1400 was used in its standard configuration (Patashnick and Rupprecht., 1991). The TEOM filter was heated to 50°C and correction factors were applied to the TEOM data (b_0 = 3 μgm^{-3} and b_1 = 1.03). The same PM_{10} inlet type used on the Partisol-Plus was used on the TEOM.
An other rural station, Fontainebleau (48°24'N, 2°42'E), is also used to evaluate PM_{10} (TEOM) and NO_2 model concentrations because measurements are not available at Prunay.
Sulfate, nitrate and ammonium daily measurements are available. Moreover, some size-segregated measurements have been carried out using a low pressure LPI-30 Berner impactor (60 nm – 16 micrometer) and Tedlar foils. Observations are presented for a small period in February 2003 in the form of raw data without inversion calculation of artifact corrections *e.g.* the volatilization due to the low pressure.

Description of the pollution episodes

February 2003 - episode EP1

Meteorological conditions are characterized by slight Northeasterly winds over Paris due to a high pressure zone in Scandinavia from the 13th to the 18th. The air mass is dry and cold in the North of France with highest temperatures close to 0°C. Figure 1 shows a back trajectory of an air parcel on Paris the 17th. From the 19th to the 20th winds veer Southeast, a low pressure area appears on the Western coast of France. Thus, a cooler oceanic air mass comes in the North West of France, increasing temperature and humidity. The 21st in the morning, the situation is very stable with fogs in Normandy, high PM concentration levels are observed over the Paris area and in Belgium (confirmed by measurements and model results in Figure 2). Winds turn Southeast at the end of the episode. In this paper, the episode is called EP1, it is followed by non polluted conditions until March 17.

March 2003 - episodes EP2 and EP3

The second episode is similar to the previous one. From March 13 to 20, an anticyclone over the Great Britain drives Northeasterly winds over the North of France. Hazes and fogs are observed in Germany, Netherlands and Great Britain. In France, the atmosphere is dry at the beginning of the period and turns moist on the 18th. On the 21st, high humidity levels are observed on the Northwestern part of France up to the Paris region, winds are very weak, and high PM concentrations are observed. This episode is called EP2 in the following. Afterwards, Southerly winds generated by a low pressures system on the Atlantic ocean blow over the Western part of France.
A very stable situation occurs from March 24 to 29 (EP3) due to high pressure over Eastern Europe. During this stagnant episode, on March 26, an air mass coming from the Sahara towards the North of France at 2500 m in altitude (Figure 1).

Simulation results against observations

Spatial representation of model results

In Figure 2, daily mean PM_{10} concentrations simulated over Europe on February 21 and March 21 are displayed together with some observations located at rural and near city sites. Measurements show background daily mean values exceeding 40 µg m^{-3} in a large part of Western Europe. The most striking feature in Figure 2 is the apparent overestimation of the model in Prunay. In France, one has to keep in mind that PM_{10} routine data are issued from TEOM 50°C measurement system known to evaporate a large part of ammonium nitrate. Indeed, according to the modeling results at the continental scale, these episodes seem correlated with high nitrate levels.
A zoom on Ile-de-France with the local scale version of CHIMERE is presented in Figure 3, showing a PM pollution plume coming from Paris. These too high predicted concentrations are mainly attributable to primary particles because the local inventory for PM is largely overestimated over the Paris city as demonstrated by Hodzic et al., 2005.

Transport and mixing processes

The pollutant NO_y-NO is used to evaluate the transport and the mixing processes in the model. Actually, AIRPARIF measures NO_y-NO concentrations as an equivalent observation for NO_2. In Table 2, predicted NO_y-NO concentrations using the local simulation against observed values indicate satisfactory error statistics for the whole period at Gennevilliers : a small absolute bias, a low normalized error with a correlation coefficient of 0.75. These good results are in agreement with the time series of NO_2 presented in Figure 4. These results confirms that transport processes are well parameterized in the model for urban areas. At Fontainebleau, a significant negative bias is observed in Table 2. An overestimation of inversion layer heights and an underestimate of NO_x emissions for the local inventory in rural areas can explain this statement. In table 2, the differences obtained on error statistics regarding the simulation scales show that the continental inventory is slightly overestimated in rural areas (Fontainebleau) with low scores for the continental model.

Particulate components

Due to the expected evaporation of ammonium nitrate, the PM_{10} measured with TEOM instrument is expected to be comparable to the PM_{10}-AN predicted concentrations (as PM_{10} - ammonium - nitrate) assuming that ammonium and nitrate are mainly under the ammonium nitrate form. With this assumption, error statistics are calculated, and the same scores as for NO_y-NO are observed for PM10-AN, they can be explained by following the same interpretation. The very large negative bias at Fontainebleau could also be due to missing biogenic emitted species. When PM_{10} are directly compared to the TEOM values, the evaporation of ammonium nitrate is exhibited in Table 2 by a large positive bias. Regarding the influence of the resolution in Table 2, the continental model gives better results for PM10-AN at Fontainebleau (low normalized error and bias) compared to the local simulation. The opposite is observed for the urban site Gennevilliers.
Time series at Prunay and Gennevilliers from February 11 to March 31 are presented in Figure 5. The temporal evolution of ammonium and nitrate is quite fairly reproduced at Gennevilliers and Prunay. However, observed concentrations are globally underestimated by the model. During the episodes, the mass ratio Nitrate/Ammonium ≈ 3 (close to the theoretical ratio of the molar masses : 3.4) and the weak values of sulfate concentrations confirm the presence of ammonium nitrate in particles. For sulfate, observations are not very well predicted. Indeed, the aqueous chemistry depends on pH (estimated by the model), on the liquid water content (a parameter difficult to compute in meteorological models) and H_2O_2 concentrations. The H_2O_2 concentration levels showed in Figure 4 are of the same order of magnitude as the ones reported by Sakugawa et al. (1990) for urban areas in winter.
In Figure 5, a good temporal correlation is observed for PM_{10} at Gennevilliers, particularly with PARTISOL data. Simulation results show a peak on February 21, due to the overestimate of concentrations coming from the Paris city as described in section 0. The underestimate of predicted PM_{10} the 18 and 21 March is correlated with a large underestimate of nitrate and ammonium,

primary particle concentrations could also be underestimated. The global underestimation of PM concentrations is due to a lack of ammonium nitrate production or advection at the continental scale. Compared to other PM pollution events, EP3 measurements display weak nitrate and ammonium concentrations particularly at Gennevilliers.

Size distribution

Evaluating an aerosol model requires information on the size distributions of the main components. The mean diameter for sulfate, nitrate and ammonium distribution for a three days period in February 2003 is in the range 0.7-0.9 µm (Figure 6), all these species absorb onto fine particles. Model results are in good agreement with observations but model spectral distributions are wider than the observed ones. The numerical diffusion generally due to the sectional approach could explain such a behavior (Zhang et al., 1999).

Modeling results confirm that differences between TEOM and PARTISOL measurements during these events are largely explained by ammonium nitrate volatilization induced by the TEOM measurement procedure. Such model studies give information on the spatialization of PM pollution events and the chemical characteristic of particles. As an example, a simulated map of ammonium nitrate concentrations is useful to identify the regions concerned by the differences expected by the two PM measurement methods. In the present study, model results show that most of the episodes over Ile-de-France also affect a large part of Western Europe. The question of the origin of such wintertime episodes is treated in the next section.

Analyses on emissions

Methodology

In this section, sensitivity analyses are carried out on anthropogenic emissions. The principle is the following : emissions of a specific area A are completely removed and the resulting influence on PM concentrations elsewhere is observed. The idea is to compare a base case scenario with all emissions (BC) to a particular test case (WA) without emissions in area A (a country or a subdomain). Modeling results have to be carefully interpreted ; the difference Δ of the concentrations between two simulations (WA *minus* BC) gives an information about the impact of area A on concentrations at point P, but not the real quantity directly coming from A. For instance, local nitric acid produced at the point *P* can be neutralized by ammonia emitted in zone A and transported at point P. Therefore, the resulting ammonium nitrate in particles can have two distinct origins.
This way of performing such analyses must be improved by marking all component emissions of each source, but this methodology implies non realistic computation times. In the next subsections, influences of the Paris Basin, Germany, Netherlands, Belgium and France are discussed.

Evaluation of the PM continental transport over the Paris Basin

A first step consists in evaluating the impact of PM continental transport over the Paris basin during the pollution events described previously. Anthropogenic PM and gas emission sources are removed in the Paris Basin. Therefore, the predicted concentrations within the domain are representative of concentrations coming from outside. Of course, this approach also allows for the quantification of the impact of Ile-de-France emissions on the rest of France.
With the notations defined in Table 1 for the simulation WP and BC, Figure 7 displays the spatial patterns of the differences Δ as previously defined by :

(PM_{10} concentrations from WP simulation) – (PM_{10} concentrations from BC simulation)

During EP1 the 21 February, Paris emissions seem to have an impact on the PM concentrations in the South East of The United Kingdom (up to 10 µg m^{-3}). That corresponds to a traditional situation following a PM accumulation over the North of France during stable pollution episodes. A South East flux in front of a low pressure system over the Atlantic Ocean drives pollutant towards United Kingdom. On March 21, it is noteworthy that Paris emissions have a strong impact on PM_{10}

concentrations in the South West of France (10-12 µg m^{-3}), hundreds kilometers further, essentially due to ammonium nitrate formation.
To analyse the Paris Basin concentrations, the outside Paris Basin contribution is expressed by $100 \times \frac{C_{WP}}{C_{BC}}$ (in %) with C_{WP} and C_{BC} respectively the concentrations for the WP and BC simulations.
This ratio can theoretically exceed 100% because of non linearities in the model. The temporal variability of the ratio for sulfate, nitrate and PM_{10} is presented in Figure 8. Sulfate observed at Gennevilliers and Prunay generally comes from outside the Ile-de-France domain, but concentrations remain low (usually less than 5 µg m^{-3}). Nitrate and ammonium come from outside during the first two episodes with an average contribution often exceeding 50%. For PM_{10}, the outside contribution largely exceeds most of the time 50% at Prunay and sometimes 80%. The contribution is less important in Gennevilliers (urban site) due to important local PPM emissions and is usually in the range [20-50%] with the highest values during pollution events.
For the last episode, EP3, PM_{10} concentrations over the Paris Basin are locally produced (ammonium nitrate) or emitted (primary particle material). The outside contribution of PM_{10} is close to 30 %.
The relative contribution of local and continental sources to the pollution episodes have been quantified. The origin of such PM events is investigated hereafter.

Possible origin of wintertime PM pollution episodes over Paris in February and March 2003

European origin?

As previously done, the influence of neighboring countries over the Paris Basin concentrations is assessed. Influences of Belgium, Netherlands, Germany and Italy are studied. Because of their location, these countries are likely to have an influence over the PM concentrations on the North part of France during such episodes. Thus, emissions of these countries are removed by turns (Table 1). Only results for March 21 are presented, this episode covering a very large part of Western Europe. The PM_{10} concentration difference Δ for the case "without Italy emissions" is defined by :

(PM_{10} concentrations from WI simulation) – (PM_{10} concentrations from BC simulation)

and so on for the other cases presented in Table 1. In Figure 9, results are presented for each country. The influence of Belgium emissions spreads out from the North up to the South West of France. A decrease of 10 to 15 µg m^{-3} in PM concentrations is observed close to Belgium and about 5 µg m^{-3} up to the South West of France. For the Dutch contribution, it is quite different : the influence seems most important in the South part of France near Lyon. Ammonia from The Netherlands is transported in these regions and can react with nitric acid locally produced, to form ammonium nitrate. Germany seems to have the most significant impact over the North East and South West of France, while Italy emissions have solely an influence in the very South Eastern part of France.
In Figure 10, the temporal variation of Δ for each country at Gennevilliers is presented. For EP1, only Germany contributes significantly to PM_{10} concentrations near Paris (up to 20 µgm^{-3}) until the 20 February. Belgium and The Netherlands have a weak influence (less than 2 µgm^{-3}). The influence of Italy is important on February 21 (up to 10 µg m^{-3}). For EP2, Germany, Netherlands and Belgium impact significantly (between 6 and 20 µgm^{-3}), and as for EP1, Italy brings a contribution the days after. Generally, Italy has a specific effect compare to other countries. Its geographic location involves a time-shifted effect when the North East flux turns South East over Paris. As discussed in the previous section, Figure 10 confirms the local origin of the EP3 event, with only few µg m^{-3} of PM concentrations due to surrounding countries on March 24-29.

Extra European origin?

On March 24-29, Southerly winds blow over the Atlantic ocean with an air mass directly coming from Sahara. Hodzic et al. (2004) already observed with Lidar measurements near Paris a thin PM layer at 2500 m above the ground level suspected to be desert dusts. In Figure 11, the simulated dust concentrations (µg m^{-3}) at 2500m in altitude on March 26 at 12:00 GMT over the domain is presented. A dust layer is observed from Ireland to the North of France. This particular episode has been simulated with a 18 levels model version. The ability of the model to simulate the correct altitude of this dust layer (Figure 11) can be noticed. As described in Rodriguez et al. (2001), wintertime dust outbreaks in the South of Europe (Spain and Portugal) are classically due to the cyclonic activity over the Atlantic as shown with EP3. Unfortunately, no specific measurements highlighting the desert dust contribution were available. According to model results at the ground level, desert dusts should contribute to a minor extent to the PM load (about 5 µg m^{-3}). However the dust concentrations are certainly underestimated. Actually, 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 uneasy to process, the average of monthly mean values taken from runs over years 2000 and 2001 was used. Since dust events are very sporadic during the year, boundary conditions for dusts are tuned (reduced by a factor of 3) to be representative of the dust background level (Vautard et al., 2005). In doing so, dust outbreaks in Europe are expected to be underestimated, then, it is difficult to conclude on the influence of this event on the EP3 episode.

Influence of France

A simulation without French emissions was run to assess their influence during episodes. Modeling results in Figure 12 display the spatial pattern of the results from March 20 to 25. At the height of the episode, on March 20-22, French emissions have only an influence on Spain (up to 10 µgm^{-3} on the North of Spain), the main plume extending over the Atlantic Ocean. When the Northeasterly flux veers Southeast at the end of the event, as it was the case for Italy, the plume extends Northwest up to Ireland through Great Britain during on March 23. Afterwards, the plume structure disappears. Nevertheless, the influence of France emissions remain significant up to Eastern Europe, close to 10-15 µg m^{-3} in Germany, Netherlands and Belgium.

Conclusions

The CHIMERE model has been used over Europe with a zoom on the Paris Basin. Wintertime PM pollution episodes during February and March 2003 were studied. Firstly, a model evaluation was carried out to assess model performances. TEOM and PARTISOL measurements display large differences due to ammonium nitrate evaporation in the TEOM system processes. Although model results are quite good for PARTISOL PM_{10}, ammonium and nitrates concentrations, model deficiencies are observed for sulfate. Indeed, sulfate in winter is produced in clouds, involving modeling parameters which are sometimes difficult to obtain from meteorological outputs. Error statistics show large differences between the continental and local simulations due to the resolution and the two different inventories used.

Most of these episodes seems to have a continental origin, and a qualitative approach attempts to estimate the advected part from outside Paris Basin to the total PM concentrations. On February 21 and March 21 episodes, the outside contribution reaches 50%, while during non episode situations the contribution drops down to 20%. When the episode reaches maximum concentrations, the “PM outside contribution” is mainly composed of ammonium nitrate. On March 24-29, although a dust layer was observed and simulated by the model at 2500 m above ground level, the modeling study confirms the local character of the pollution episode, essentially due to primary particle material emitted in the Paris area.

In the last section, the origin of the outside Paris contribution is investigated. A very preliminary sensitivity study is performed by removing the emissions of the neighboring countries supposed to influence PM concentrations over Paris area. Germany, The Netherlands, and Belgium seem to have a strong impact during EP1 and EP2. To a minor extent, Italy has an influence at the end of the episodes when the North East flux veers South East.

Removing all French anthropogenic emissions allows to assess the influence of French emissions on PM concentrations observed in the rest of Europe. During the studied episodes, France has only an influence on Spain. But at the end of the episodes, when the North East flux veers South East and West, France has successively a strong influence on PM concentrations in Great Britain, Belgium, Netherlands, Germany and up to Eastern Europe to a lesser extent.

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Tables

Simulation name	Description
BC	Base simulation with all emissions
WP	Simulation without Paris basin emissions
WB	Simulation without Belgium emissions
WN	Simulation without The Netherlands emissions
WI	Simulation without Italy emissions
WF	Simulation without the whole France emissions

Table 1 : Description of the simulations

Simulation scale (resolution)	Pollutants	Fontainebleau					Gennevilliers				
		Obs. [c]	Mod.[d]	Corr.[e]	Nerr.[f]	Rms.[g]	Obs.[c]	Mod.[d]	Corr.[e]	Nerr.[f]	Rms.[g]
Local (~5km) (AIRPARIF inventory)	PM_{10}-AN [a]	23.6	13.4	0.64	43.2	13.8	32.8	30.2	0.65	41.4	18.9
	PM_{10}	23.6	23.0	0.63	39.4	10.8	32.8	40.8	0.67	56.1	23.6
	NO_y-NO [b]	18.4	12.2	0.61	41.0	11.6	57.6	56.4	0.75	29.2	19.7
Continental (~50km) (EMEP inventory)	PM_{10}-AN [a]	23.6	19.5	0.65	32.6	11.5	32.8	35.1	0.64	48.3	18.7
	PM_{10}	23.6	28.8	0.64	48.3	13.7	32.8	45.5	0.63	69.8	26.4
	NO_y-NO [b]	18.4	19.7	0.56	60.1	11.5	57.6	49.9	0.76	34.4	20.2

[a] PM_{10} (TEOM) without ammonium and nitrate, [b] NO_y-NO equivalent NO_2
[c] Observed mean concentration (µg m^{-3}) : TEOM data for PM_{10}, [d] Mean predicted concentration (µg m^{-3}), [e] Correlation,
[f] Normalized Error (%), [g] Root mean square error (µg m^{-3})

Table 2 : Error statistics based on hourly data for NO_y-NO, PM_{10}, and PM_{10} without ammonium nitrate at Fontainebleau and Gennevilliers for the 12 February – 31 March period. Statistics are reported for model results at the continental and local scales. Observed values for PM_{10} and PM_{10}-AN are issued from the TEOM system.

Illustrations

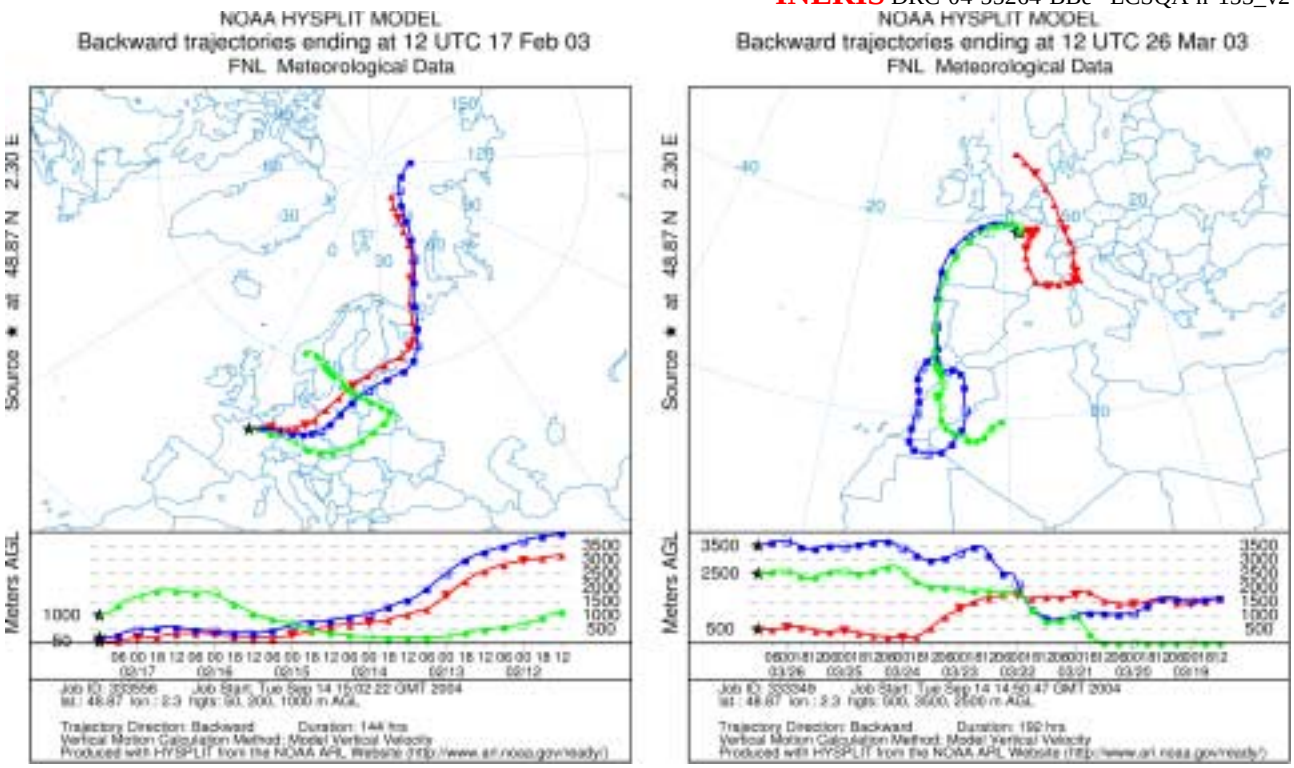

Figure 1

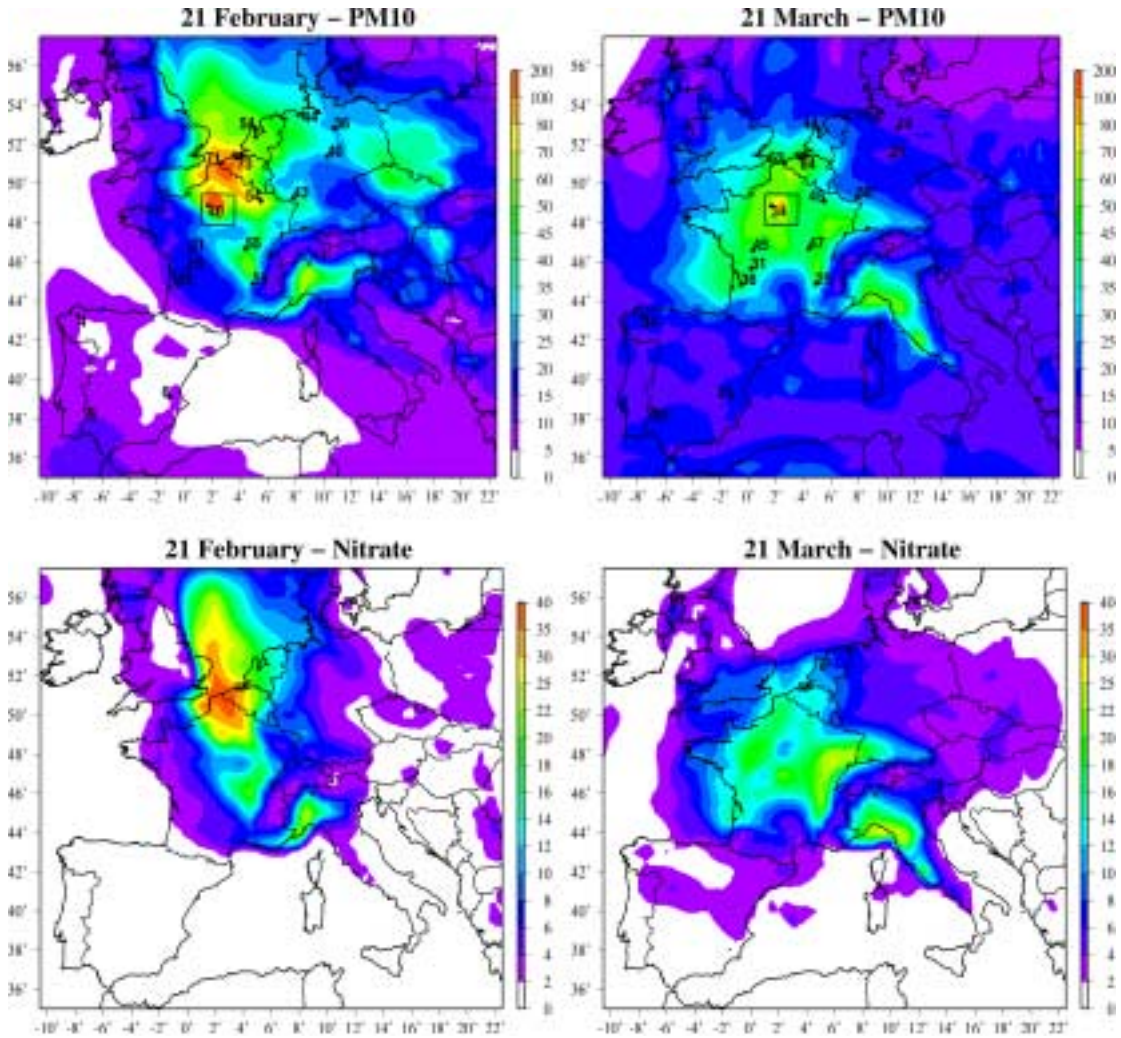

Figure 2

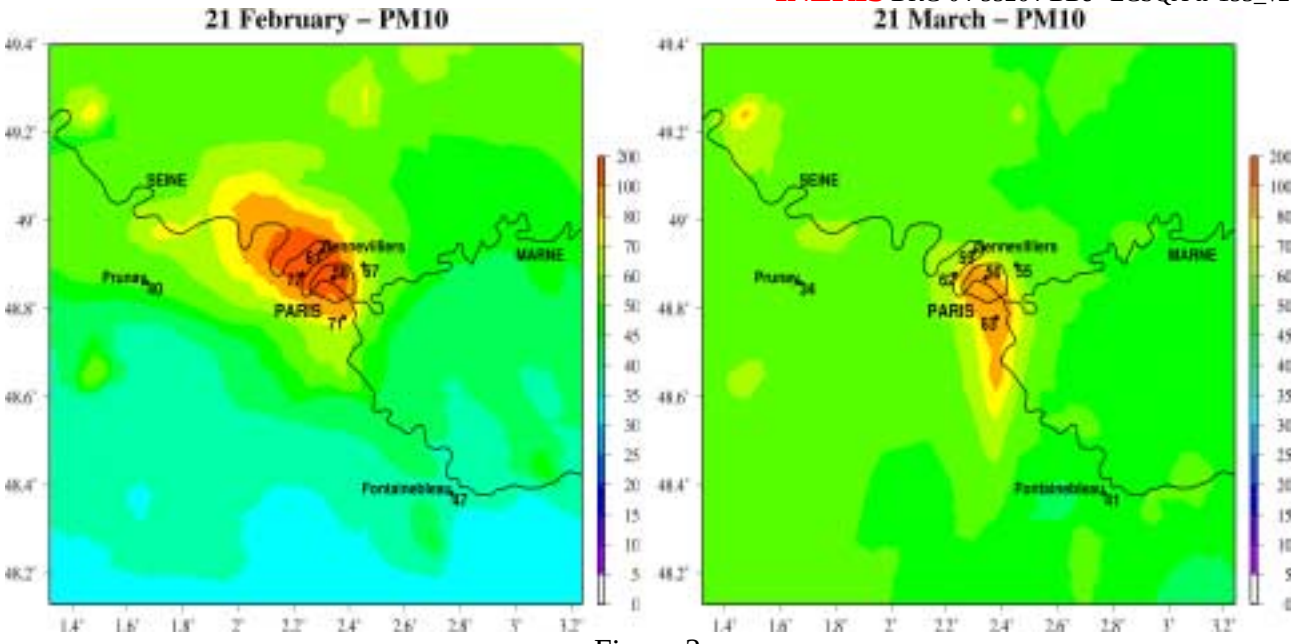

Figure 3

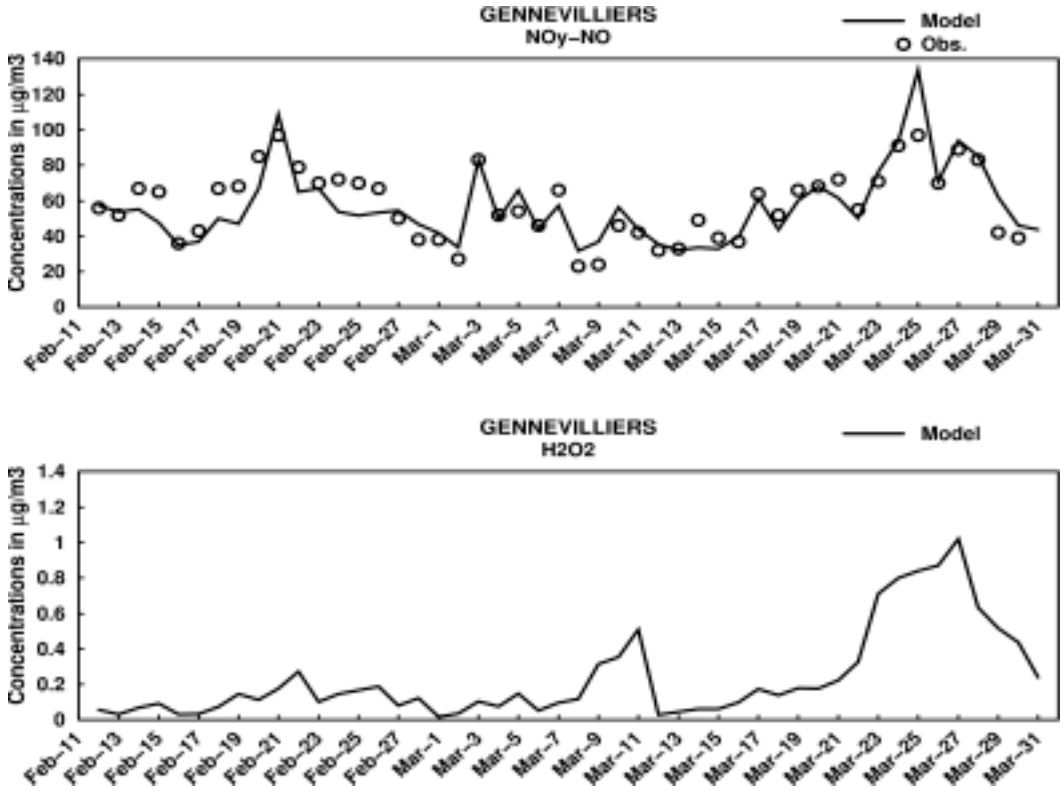

Figure 4

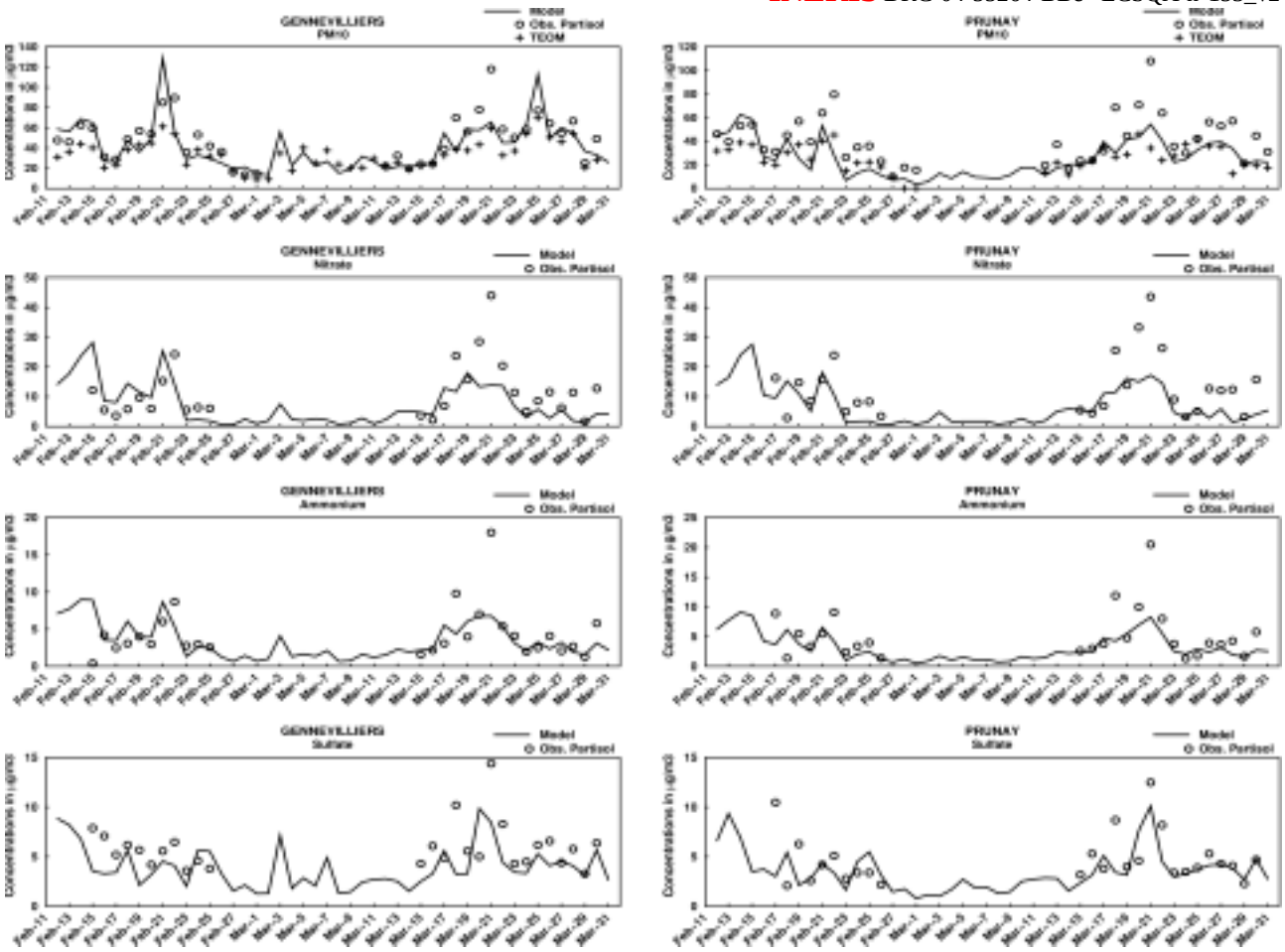

Figure 5

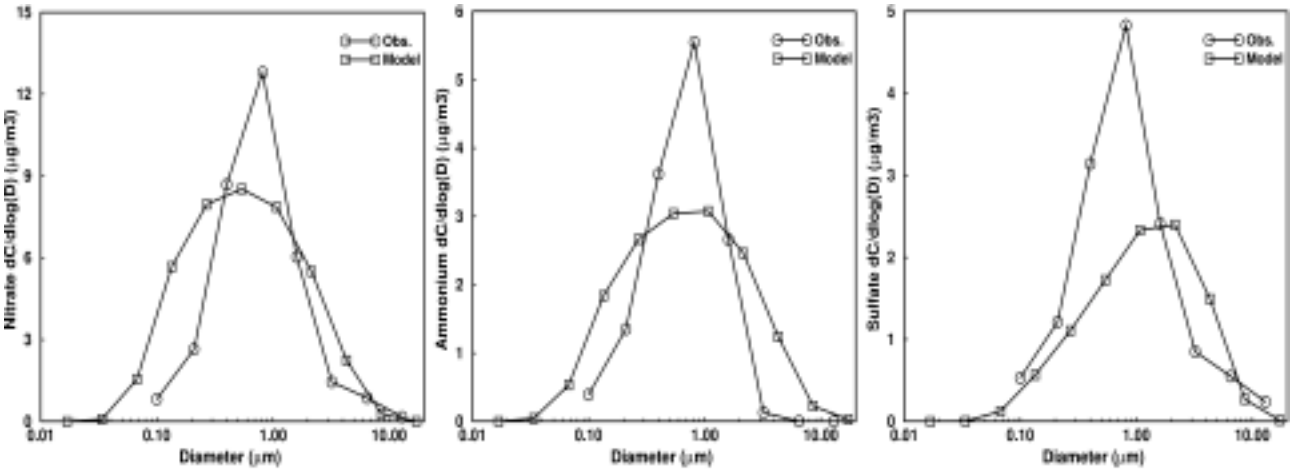

Figure 6

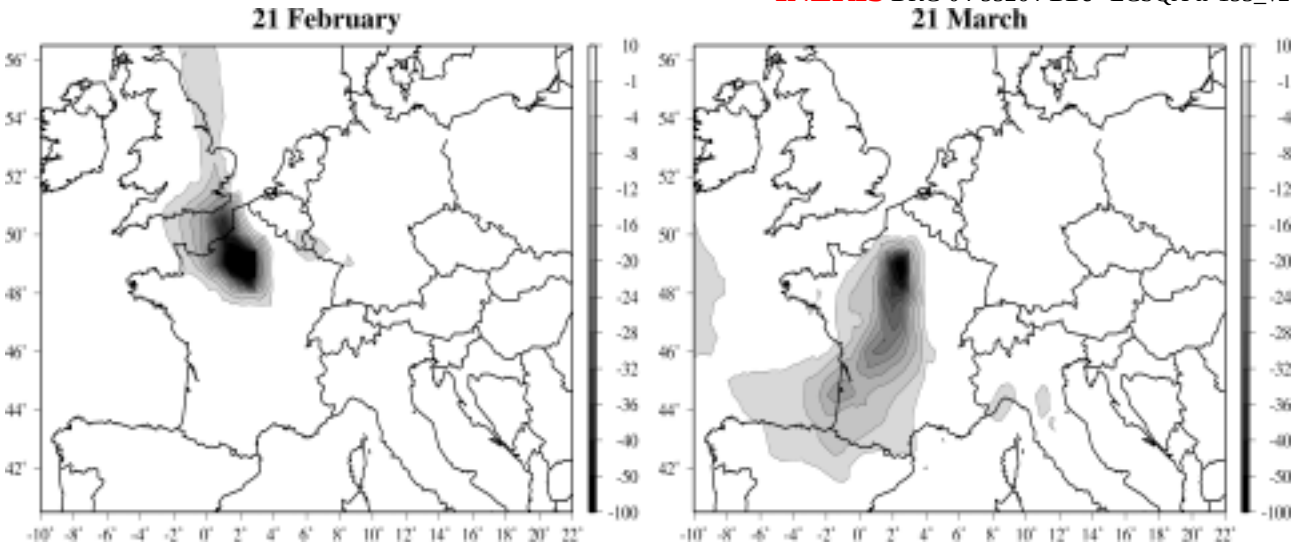

Figure 7

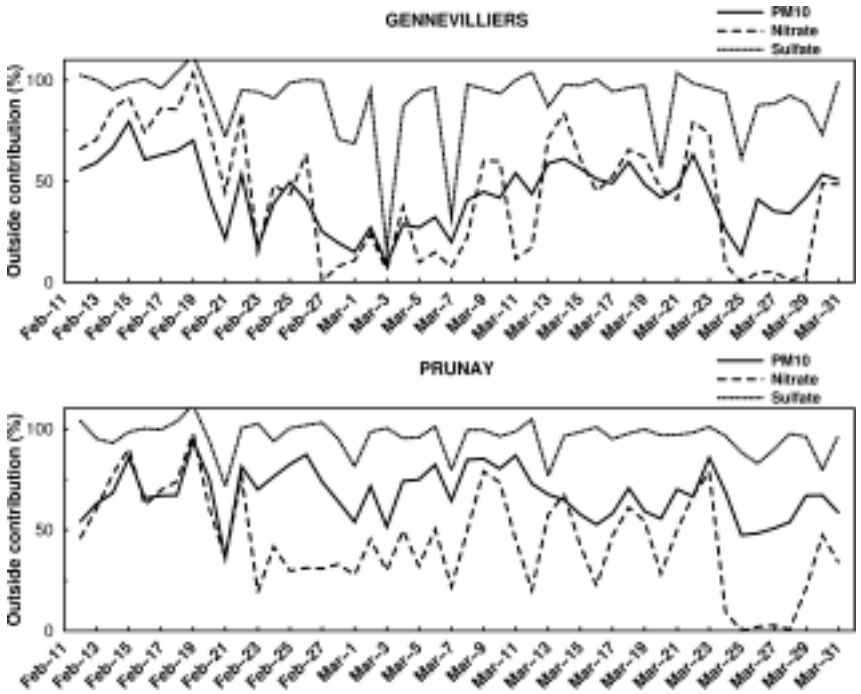

Figure 8

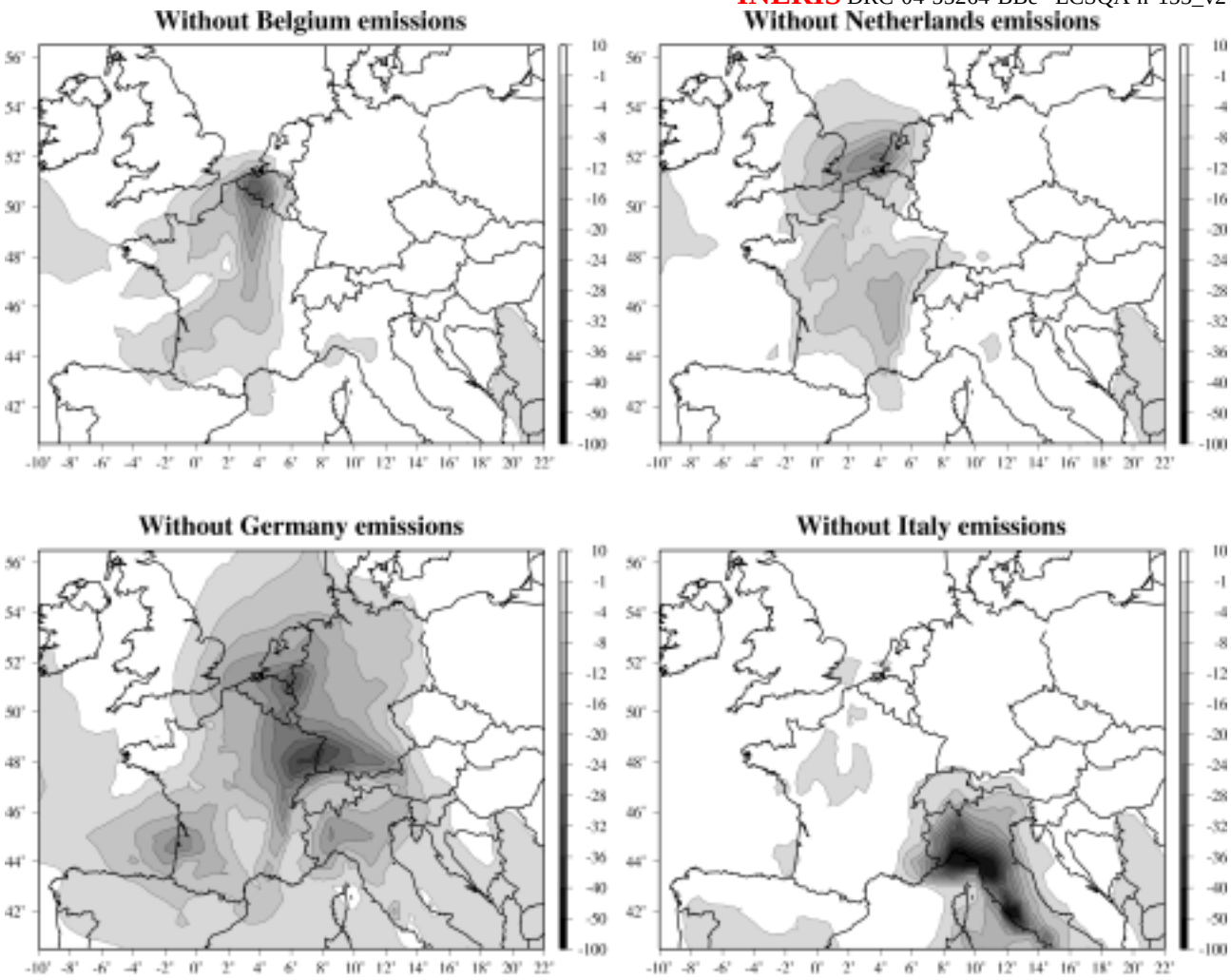
Without Belgium emissions
Without Netherlands emissions
Without Germany emissions
Without Italy emissions

Figure 9

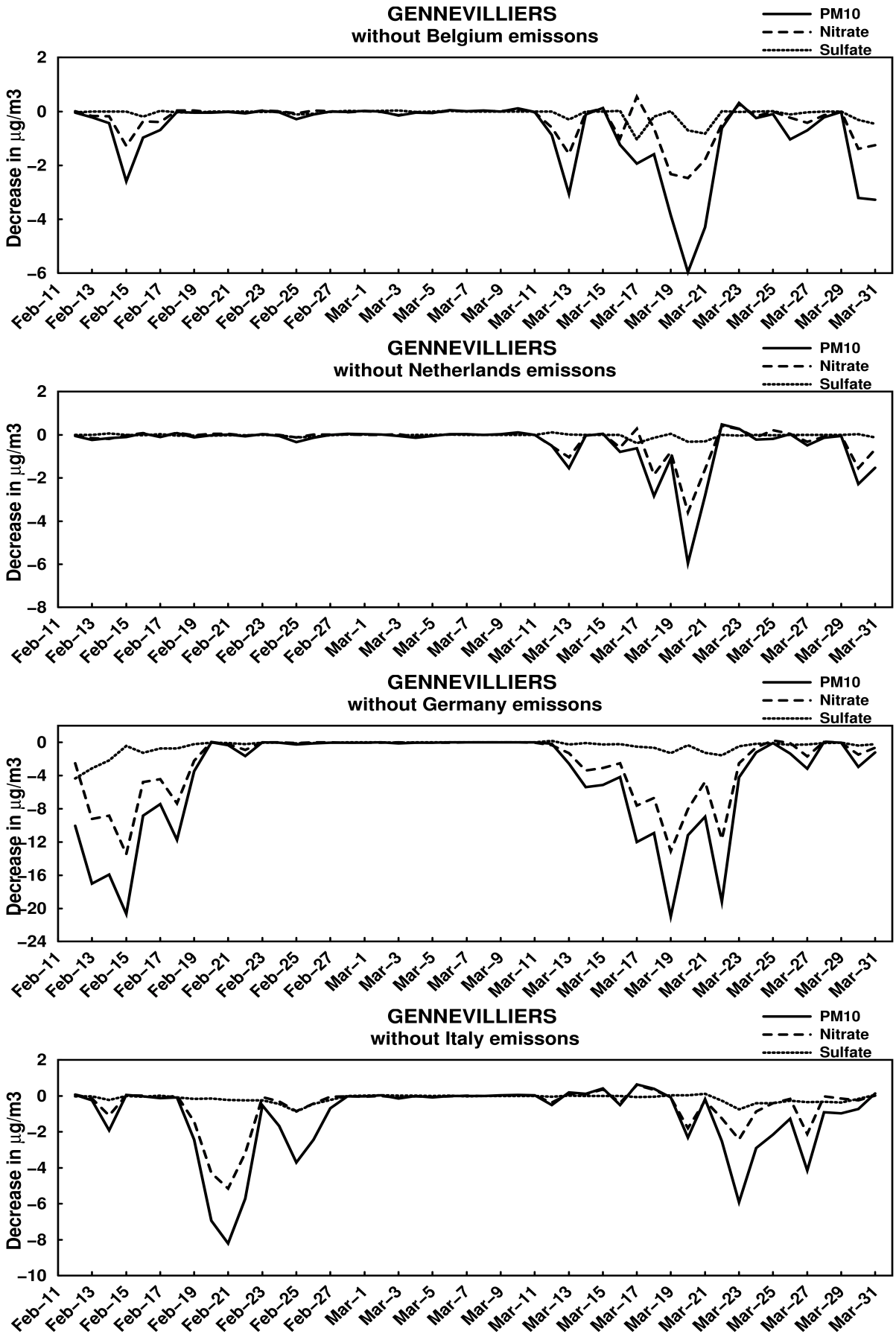
GENNEVILLIERS
without Belgium emissons
PM10
Nitrate
Sulfate
Decrease in µg/m3
2
0
−2
−4
−6
GENNEVILLIERS
without Netherlands emissons
PM10
Nitrate
Sulfate
Decrease in µg/m3
2
0
−2
−4
−6
−8
GENNEVILLIERS
without Germany emissons
PM10
Nitrate
Sulfate
Decrease in µg/m3
0
−4
−8
−12
−16
−20
−24
GENNEVILLIERS
without Italy emissons
PM10
Nitrate
Sulfate
Decrease in µg/m3
2
0
−2
−4
−6
−8
−10
Feb−11 Feb−13 Feb−15 Feb−17 Feb−19 Feb−21 Feb−23 Feb−25 Feb−27 Mar−1 Mar−3 Mar−5 Mar−7 Mar−9 Mar−11 Mar−13 Mar−15 Mar−17 Mar−19 Mar−21 Mar−23 Mar−25 Mar−27 Mar−29 Mar−31

Figure 10

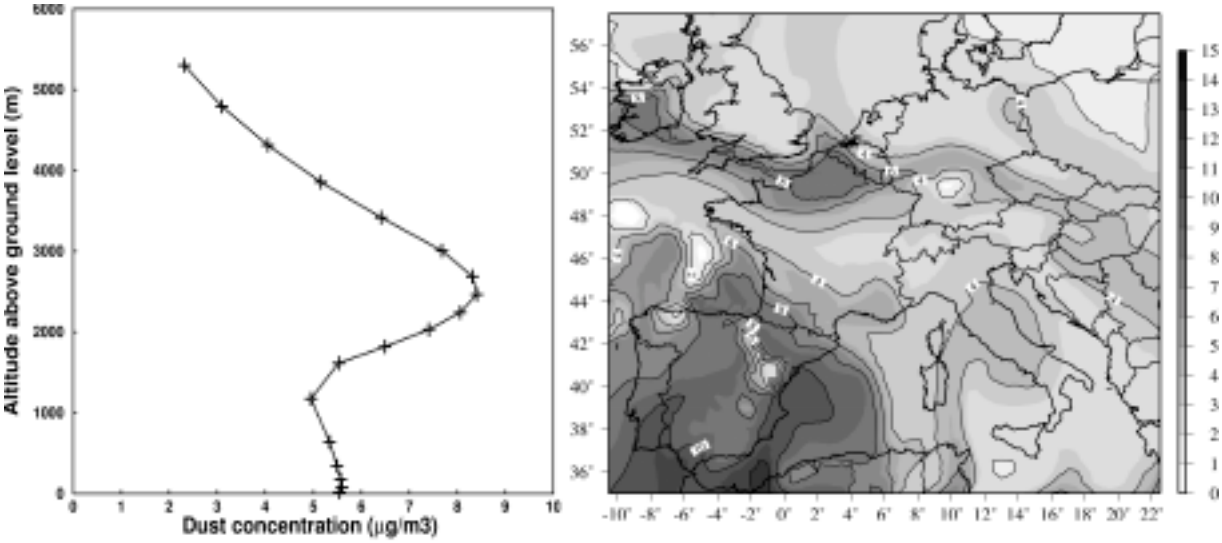

Figure 11

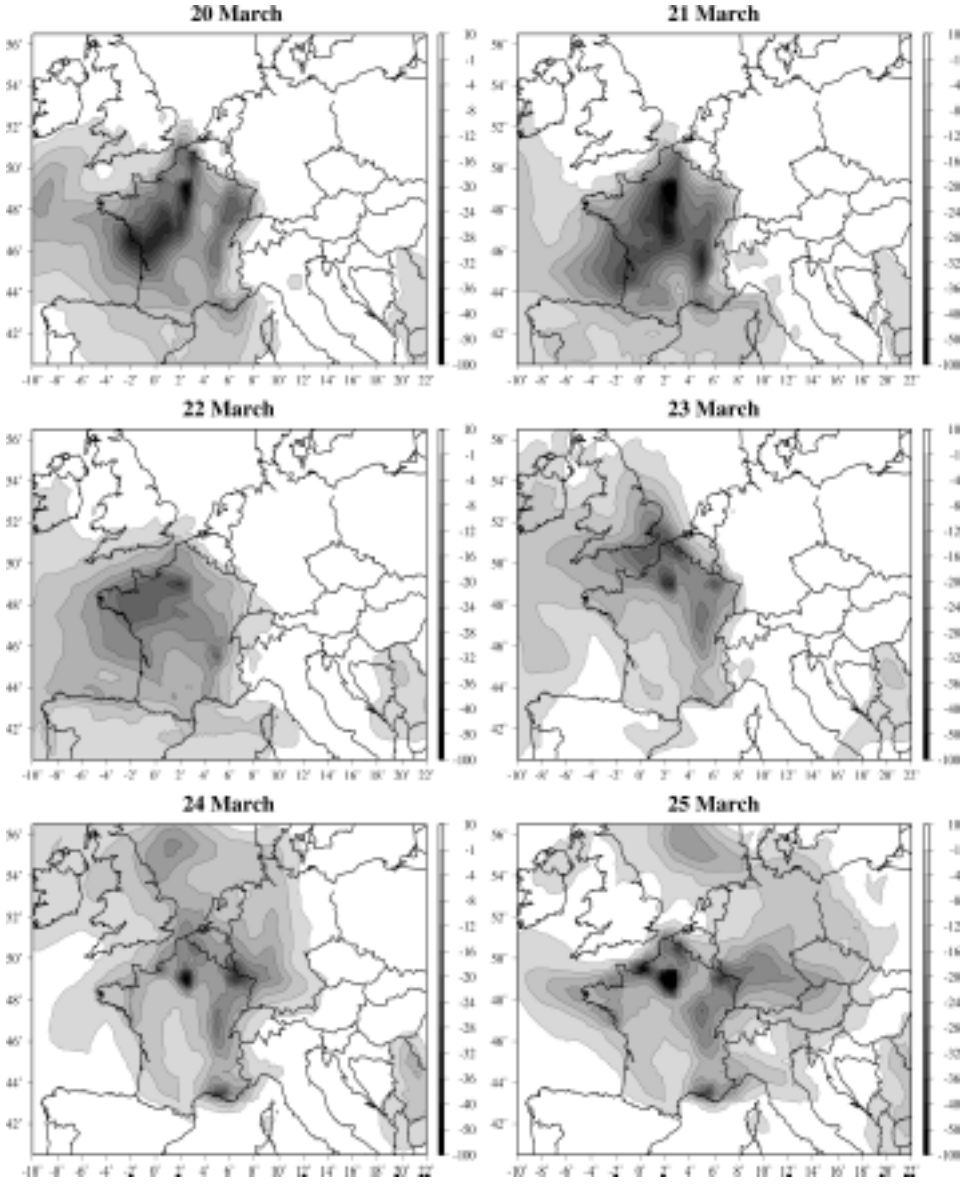

Figure 12