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Atmospheric Chemistry and Physics Discussions



# *Interactive comment on* "Aerosol distribution over the western Mediterranean basin during a Tramontane/Mistral event" *by* T. Salameh et al.

**T. Salameh et al.**

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## 1 Anonymous Referee #1

Received and published: 4 January 2007

*This paper uses two models and a variety of observation data to analyze the aerosol distribution during a Tramontane/Mistral event. While the event is interesting, the analysis has very little physical explanation for the discrepancies between model and measurement. Some paragraphs are one-page long, without focusing on any central questions. Furthermore, the paper lacks the scientific importance needed for publication in ACP. The paper should be rejected in the current form. My major concerns are listed*

*in below.*

### 1.1 Referee #1 - comment 1

*The objective of this paper is unclear. In the introduction part, the authors stated that the article is designed to (a) analyze dynamical processes driving the Mistral flow and its relationship with aerosol distribution observed by lidar and satellite, and (b) aerosol source, composition and distribution over the whole Mediterranean basin. However, their findings as stated in their abstract and conclusion part don't echo these two objectives, in particular, the second objective. Under the Tramontanel/Mistral wind and their interaction with topography, would that be expected that the aerosol distribution is less stable? I don't see anything new in here. It would be interesting to quantitatively show the amount of aerosol mass transported by the Mistral and how that enhances the background aerosols?*

We agree with the reviewer that confusion can be made between the scientific context and motivation and the actual objectives of the paper (we can not address quantitatively the aerosol radiative impact since there is no feedback between the dynamical model MM5 and the chemistry transport model CHIMERE). We thus reformulated the introduction to state more clearly the original points of our manuscript, following the reviewer suggestions. The introduction is now:

"The Mediterranean basin is featured by an almost-closed ocean basin surrounded by mountain ranges in which numerous rivers rise, a contrasted climate and vegetation from south to north, numerous and rapidly growing built-up areas along the coast with several major cities having industrial activities emitting a large number of gas substances and aerosols. Highly aerosol loaded air masses are found from the surface up



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to the upper troposphere and contribute to decrease air quality on a large scale and reduce precipitation in the region (Lelieveld et al. 2002). The aerosols found below 4 km height originate from regional sources especially from western and eastern Europe (e.g. Sciare et al. 2003; Traub et al. 2003; Schneider et al. 2004), and from the Saharan desert (e.g. Bergametti et al. 1992; Moulin et al. 1998; Guieu et al. 2002). Industrial activity, traffic, forest fires, agricultural and domestic burning are the main source of pollution in Europe whereas the close vicinity of the Saharan desert provides a source for considerable amounts of dust. Above 4 km height they are usually linked to transport due to global-scale motions and teleconnections, for instance with the Indian monsoon and the North Atlantic Oscillation (Moulin et al. 1997).

Aerosols are harmful for ecosystems and human health, and they affect the Mediterranean climate and water cycle. Indeed, they affect the atmospheric energy budget by scattering and absorbing solar radiation (reducing solar radiation absorption by the sea and altering the heating profile of the lower troposphere), and contribute to the suppression of evaporation and moisture transport, in particular to North Africa and the Middle East (Lelieveld et al. 2002). Aerosols also affect the Mediterranean biogeochemistry by deposition of dissolved inorganic phosphorus (Bergametti et al. 1992; Bartoli et al. 2005), silicon (Bartoli et al. 2005) and iron (Guieu et al. 2002) from soil-derived dust from desert areas of North Africa and anthropogenic emissions from European countries. These are atmospheric inputs internal sources of nutrients and constitute an important pathway for nutrients to the photic zone of the Mediterranean Sea (Migon et al. 1989; Béthoux 1989; Bergametti et al. 1992; Prospero et al. 1996; Guerzoni et al. 1999; Benitez-Nelson 2000; Béthoux et al. 2002). In the context of climate change in which the Mediterranean basin appears quite vulnerable due to hydric stress and ever increasing pollution levels, it is thus crucial that knowledge be improved concerning the mechanisms linking the dynamics of the main flow regimes and the existing pollution sources scattered around the basin in order to provide insight into future trends at the regional scale.



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The western Mediterranean climate is frequently affected by the Mistral and its companion wind, the Tramontane (Georgelin and Richard 1996; Drobinski et al. 2001a). The Mistral is a severe northerly wind that develops along the Rhône valley (while the Tramontane blows in the Aude valley) (Fig. 1), and occurs between 5 and 15 days per month. The development of the Mistral is preconditioned by cyclogenesis over the Gulf of Genoa and the passage of a trough through France. The Mistral occurs all year long but exhibits a seasonal variability either in terms of its strength and direction, or in terms of its spatial distribution (Mayençon 1982; Orieux and Pouget 1984). At the regional scale, the Mistral is frequently observed to extend as far as a few hundreds of kilometres from the coast (Jansà 1987) and is thus associated with low continental pollution levels near the coastline as it advects the pollutants away from their sources of emission over the Mediterranean Sea (Bastin et al. 2006; Drobinski et al. 2006).

Due to favourable dispersion conditions, air quality studies generally do not focus on Mistral events. However the transport and resulting concentration distribution of chemical compounds and aerosols over the Mediterranean Sea has never been documented. The FETCH (Flux, Etat de mer et Teledetection en Condition de fetcH variable) experiment (Hauser et al. 2003) offers an ideal framework for such a study. The FETCH experiment took place from March 12 to April 15, 1998 and was dedicated to improve the knowledge of the interactions between the ocean and the atmosphere in a coastal environment under strong wind conditions (e.g. Drennan et al. 2003; Eymard et al. 2003; Flamant et al. 2003). The March 24, 1998 Mistral case is a strong episode, typical of intermediate season Mistral events compared to weaker summer Mistral events (e.g. Drobinski et al. 2005, Bastin et al. 2006). Flamant (2003) analyzed the complex structure of atmospheric boundary layer (ABL) observed using an airborne lidar over the Gulf of Lion at the exit of the Rhône valley during the March, 24 1998 Mistral event. Even though the study by Flamant (2003) suggested the existence of a marked east-west aerosol concentration gradient offshore to be related to larger concentrations of pollution aerosol from the city of Marseille and the industrial petrochemical complex of Fos/Berre, the highly spatially resolved aerosol measurements needed to (un)validate



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this hypothesis simply did not exist. By combining lidar observations and numerical simulations from the MM5 mesoscale model (Dudhia 1993; Grell et al. 1995) and the chemistry transport model CHIMERE (Schmidt et al. 2001; Vautard et al. 2001), we better address this question in the present article. Therefore this article is designed to analyze:

- the relation between the dynamic processes driving the small-scale structure of the Mistral flow and the aerosol distribution observed by the airborne lidar and the satellite imagery,
- the amount of aerosol mass transported by the Mistral and Tramontane in addition to the background aerosol mass,
- the aerosol sources, composition and distribution over the whole western Mediterranean basin, especially the sea-salts which can have a significant contribution under strong wind conditions.

The instrument set-up (i.e. FETCH-related sea-borne, airborne and space-borne observations) and the numerical models used in this study are described in Section 2. In Section 3, the meteorological environment leading to the Mistral episode is analyzed as well as the fine-scale structure of the Mistral flow. In Section 4, the aerosol distribution over the western Mediterranean basin is discussed, before conclusions are drawn in Section 5 and suggestions for future work are presented."

As also suggested by the reviewer, we also addressed the issue of the amount of aerosol mass transported by the Mistral and Tramontane in addition to the background aerosol mass. The background aerosol mass was computed from the new simulations that include sea-salts by taking the minimum value of the aerosol loading between March 23rd and March 25th at every point of the simulation grid. The amount of aerosol transported by the Mistral and Tramontane in addition to the background aerosol loading is thus the difference of the total aerosol loading minus the background



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aerosol loading. In the revised manuscript, we show that the aerosols transported by the strong winds (Mistral, Tramontane and associated Ligurian outflow) are from marine origin and from the main industrial sources (Marseille/Fos-Berre; Pô valley). The amount of aerosol loading solely due to the Mistral and Tramontane is as large as 3-4 times the background aerosol amount. All these points were made clear in the revised version (section 4.3. before the conclusion and conclusion).

### 1.2 Referee #1 - comment 2

*They mentioned this paper is motivated by the aerosol radiative effects. However, the authors didn't conduct any analysis of aerosol radiative effects in the paper, and seemly have no future plan to do so (their last sentence in the conclusion part says: future work will be dedicated to assesses the representativity of this case study). Hence, the paper lacks the scientific importance needed for publication in ACP. Here are some questions that should be addressed in this paper.*

In this paper, we do not explicitly address the radiative impact of the aerosols on the dynamics of the low level troposphere, which is not possible with our off-line chemistry transport model (no feedback of the aerosol radiative properties on the dynamics). Here, we just quantify as a first step, the aerosol distribution over the Mediterranean Sea in one very frequent flow regime and the aerosol optical depth as a preliminary proxy of the possible radiative impact of the aerosols during winter-time under strong wind conditions. Even if the radiative impact of the aerosols is a motivation for the article, its quantification is not an objective. In the introduction of the revised manuscript, we try to make clearer the objectives of the paper without interfering with the scientific context and motivation.



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*What are the aerosol radiative effects on surface temperature and atmospheric lapse rate? Are they important to explain the discrepancies between model and measurements in Figure 3, Figure 4, and Figure 6?*

The CHIMERE model is a chemistry transport model and there is no on-line coupling between the dynamical and chemical models but only an off-line interface. Consequently, it is not possible to answer this specific comment since there is no feedback of the aerosol radiative effect on the low-level dynamics. We made that point clear in the revised version (in the section describing the CHIMERE model).

*Why the model-simulated AOD is only about 50 % of satellite-retrieved AOD? Which one should reader trust? If the model error is quite large, then further analysis of model results lack the credibility.*

One major drawback of the simulations we discussed in the original version of the submitted manuscript is the absence of the sea-salts. Under strong wind conditions, the sea-salts may no longer be negligible in the aerosol loading over the Mediterranean Sea. In order to address the reviewer comment, we included the sea-salt module in the CHIMERE model (Monahan et al. 1986). This module had never been validated up to now and this constitutes one of the objectives of the revised manuscript. The main results, now included in the revised version, are:

- Suspension of sea-salts occurs in very localized area which evolve during the day with respect to the evolving dynamics. The largest suspension of sea-salts occur in the most intense wind regions (over the Ligurian Sea). Between 1200 and 1500 UTC, large amount of sea-salt is found in the sheltered area (very weak wind). This can be explain by the stagnation of sea-salts previously emitted and transported by the Ligurian outflow over this region.



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- On average, in the regions where suspension of sea-salts is found, the contribution of the sea-salts to the overall aerosol loading ranges between 1 % (over the Ligurian Sea) and 10 % (to the west of Sardinia)

- At 1200 UTC, the north-south oriented band of sea-salts (starting at Toulon) explains the underestimation of the AOD by CHIMERE when compared to SeaWIFS in the original submitted manuscript. The AOD was recalculated and a better agreement is found between the simulations and observations. The contribution of sea-salt particles to the total AOT ranges from 1 to 10 %. These results are consistent with values reported by AEROCOM (http://dataipsl.ipsl.jussieu.fr/cgi-bin/AEROCOM/aerocom/) and in some published articles (Collins et al. 2002; Halthore and Caffrey 2006) for various open-sea locations. A detailed discussion has been included in the revised version.

Collins, W.D., P.J. Rasch, B.E. Eaton, D.W. Fillmore, J.T. Kiehl, C.T. Beck, and C.S. Zender, 2002: Simulation of aerosol distributions and radiative forcing for INDOEX: Regional climate impacts, J. Geophys. Res., 107 (D19), 8028, doi:10.1029/2000JD000032.

Halthore, R.N., and P.F. Caffrey, 2006: Measurement and modeling of background aerosols in remote marine atmospheres: Implications for sea salt flux, Geophys. Res. Lett., 33, L14819, doi:10.1029/2006GL026302.

Monahan, E.C., Spiel, D.E., Davidson, K.L., 1986. In: Monahan, E.C., Mac Niocaill, G. (Eds.), Oceanic Whitecaps. Riedel, Norwell, MA, pp. 167-174.

*Table 1. The difference of simulated and measured sulfates and nitrates is quite large, and in some cases, the difference is a factor of 3-4. The authors attributed these differences to the distance between the observation site and the location of simulated sulfate plume in the model. I don't think this explanation has any scientific value. What*



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*are the possibly physical reasons for this difference? Is it related to any non-ideality in the emission strength, boundary layer scheme, or other causes? If there is a 50km difference, can authors show the two model values, one in the pollution plume and another just over the station? Without a detailed analysis to explain the large difference, I doubt the fidelity of their other analysis.*

In the footnote of Table 1, we specified the value of the sulfate concentration (8-9 $\mu$g m$^{-3}$) in the plume located 50 km away from the measurement point. The simulated value over the station was shown in the last column of Table 1. For the nitrates, the relative error between the observed and simulated concentrations is about 20 % on average with two peaks of 75 % at NL09 on March 25th and IT04 on March 23rd. The 20 % uncertainty is due both to representativeness and instrumental accuracy. For the sulfates, the relative error between the observed and simulated concentrations is also about 20-25 % on average with one major peak of 430 % at FR03 on March 24th which corresponds to the mismatch between the simulated and observed sulfate plumes and is indicated by a footnote in Table 1. We detailed the discussion on the comparison between the EMEP measurements and the corresponding simulated values.

*More quantitative analysis is needed. What the correlation is between modeled and satellite AOD? What is their correlation with AERONET AOD? What are the correlation between modeled and measured sulfate and nitrate? A time-series of modeled and measured aerosol mass will be helpful as well.*

It is difficult to have a more quantitative comparison between the modeled and measured sulfate and nitrate concentrations since the EMEP data are daily averaged (all available data from the stations fitting within our simulation domains were used for comparison), the SeaWIFS passage and lidar legs are the only ones of the investigated day and the all available data from the AERONET stations located within our simulation do-



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mains were used. We thus do not have a more resolved time-series to compare our simulations with.



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