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Atmospheric
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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 #3

Received and published: 5 January 2007

1.1 Referee #3 - general comments

This paper presents a comparison of a mesoscale model with measurements conducted in the west Mediterranean region, with focus on the meteorology of the area and its influence on the chemical composition of aerosols measured by both surface,

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airborne and space measurements. The authors set two targets for their study: the analysis of the dynamic processes of the region, and the aerosol sources and composition over the whole western Mediterranean. In my opinion, the first goal is not original, since it is already discussed in detail by Flamant (2003), and the second is not complete, since (a) they do not discuss the whole western Mediterranean but only the area between France and Italy, and (b) they do not have sea-salt in their model, which is a very important (if not the most important) aerosol constituent above sea; further,sea-salt is not even included in the future work of the authors. For these reasons, as detailed below, I recommend that this paper should be rejected in its present form.

We agree with the reviewer about the two key points. For the study of the dynamics over this region, this is right that the analysis was previously published by Flamant (2003). In our study, the goal was not to publish a second time this analysis but to relate the dynamical processes with the transport of particules and pollutants. According to the reviewer comments, the description of the evolution of the flow pattern was shortened, the originality of the study was made clearer in the introduction, which was reformulated (see replies to the detailed comments).

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 are included in the revised version and the thoroughly discussed (in order to present the relative amount of AOD due to sea-salt, we present complementary maps: without sea-salt, with sea-salt, the two being compared to satellite data) (see replies to the detailed comments). The reason behind focusing on the region of southern France and western Italy is the availability of measurements from the FETCH campaign and

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the SeaWIFS data in this region. The aerosols distribution is described in the regions to the south west of the Tyrrhenian Sea and to the north of the African north coasts. But the lack of measurements over these regions stopped further discussions.

1.2 Referee #3 - detailed comments

Introduction: it includes nice ideas, but not well connected to each other. A thorough restructuring is required.

Yes, the introduction was completely rewritten in order to be more concise and more accurate. And the new introduction is:

“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 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.

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 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."

Chemistry-transport model description: For a study above the Mediterranean Sea, and with wind speeds that exceed 10 m s^{-1} *(Figure 6a) most of the time, it is necessary to have sea-salt in the model calculations. It is not a surprise that the authors underestimate the AOD by a factor of two (page 11928, lines 20-25). It is wrong to make a comparison above sea, without including sea-salt.*

We agree with the reviewer that 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.

- 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.

Section 3.1 is almost identical with Flamant (2003) and should be removed. It also gives a lot of meteorological details that are not needed in the manuscript. It is just repetition of previously published results. The meteorological conditions have been *analysed in detail elsewhere, in a meteorological journal as expected, and only an outline should be given here with focus on the conditions of interest for the present study.*

Section 3.1 was shortened in order to give the key elements that give some light on the aerosol distribution over the western Mediterranean Sea. It is now:

“The March 24, 1998 Mistral event was featured by the existence of an upper level trough associated with a cold front progressing toward the Alps and a shallow vortex (1014 hPa) over the Tyrrhenian Sea between Sardinia and continental Italy, at 0600 UTC. As the day progressed, the low over the Tyrrhenian deepened (from 1014 to 1008 hPa between 0600 and 1500 UTC) while remaining relatively still. From 1500 UTC on, the low continued to deepen (from 1008 to 1002 hPa) while moving to the southeast. It was located over Sicily on March 25, 1998 at 0600 UTC.

The multistage evolution of the Alpine lee cyclone over the Tyrrhenian Sea induced a very nonstationary wind regime over the Gulf of Lion (also see Flamant, 2003). The diurnal evolution of the Mistral and the Tramontane on March 24, 1998 are evidenced on the wind field simulated in the ABL (at 950 hPa) by the MM5 model at 0600, 0900, 1200, 1500, 1800 and 2100 UTC (Fig. 2). In the early stage (low at 1014 hPa, 0600 UTC), the Tramontane flow prevailed over the Gulf of Lion due to the high position (in terms of latitude) of the depression. The Mistral extended all the way to Southern Corsica, wrapping around the depression. To the north, a weak easterly outflow was observed over the Gulf of Genoa. As the low deepened (1010 hPa), the prevailing wind regime shifted to a well-established Mistral peaking around 1200 UTC. The Mistral was observed to reach Southern Sardinia where it wrapped around the depression. At this time, the outflow from the Ligurian Sea (i.e., Gulf of Genoa) had become stronger. In the afternoon, the Mistral was progressively disrupted by the strengthening outflow coming from the Ligurian Sea in response to the deepening low over the Tyrrhenian Sea (1008 hPa, 1500 UTC) and the channelling induced by the presence of the Apennine range (Italy) and the Alps. In the evening, the Mistral was again well established over the Gulf of Lion as the depression continued to deepen (1002 hPa, 2100 UTC), but moved to the southeast reducing the influence of outflow from the Ligurian Sea on the flow over the Gulf of Lion. During this period, the Tramontane flow appeared to be much steadier than the Mistral and less disrupted by the return flow associated with the depression.

An important feature of the cold air outbreak over the Gulf of Lion is also observed in the form of banners of weaker winds (sheltered region) separating (i) the Mistral and the Tramontane (in the lee of the Massif Central) and (ii) the Mistral and the Ligurian Sea outflow (in the lee of the western Alps, see Fig. 2). The unsteady nature of the western Alps wake (as opposed to the steadier Massif Central wake) is caused by the complex topography of the Alps which amplifies any variation of the wind speed and direction, or ABL depth upstream of the Alps (Guénard et al. 2006). This complex evolution of the flow affects directly the aerosol distribution over the western Mediterranean."

Page 11925, lines 27-28: figure 6a shows an overestimation of the wind speed and not an underestimation.

The reviewer is right, it is now corrected in the revised manuscript.

Section 4.2: How do you define cloud-free? Are the satellite cloud-free conditions consistent with the model's? What optical properties do you use? How do you treat hygroscopic growth? Sea-salt will alter your results significantly, when you include it, especially in regions with strong winds. Do the model and sampling times agree? The authors should discuss the meteorological conditions change with respect to their importance on transport and reflectivity.

In the original manuscript, the cloud-free regions used as a mask for comparison between the observations (1132 UTC) and simulations (1200 UTC) were those identified from the SeaWIFS measurements. The SeaWIFS passage is at 1132 UTC and the model output is at 1200 UTC. The times corresponding to the SeaWIFS observations and the MM5 model are now specified in Fig. 8 caption. The simulated field of surface downward shortwave radiation which indicates the cloud-free (large shortwave radiation > 800 W m 2) and cloudy regions (low shortwave radiation) shows that the SeaWIFS cloud-mask matches well the MM5 cloudy region over the Gulf of Lion. In order to illustrate the time variability, the MM5 surface downward shortwave radiation was calculated at 1100 and 1200 UTC (model outputs just before and after the Sea-WIFS passage) and evidences no significantly different cloud cover between the 1100 and 1200 UTC. In terms of aerosol transport, there is no significant difference between 1100 and 1200 UTC outputs.

I disagree with the author's statement from page 11929 line 28 to page 11930 line 2: In figure 10a and 10c, a local maximum appears to the west, same as in reflectivity, while 10b appears to be uniform. What is the effect of sea-salt? Changes from 40 % to 60 % in relative humidity will strongly affect the sea-salt aerosol size due to its high hygroscopicity.

We agree with the reviewer that when the relative humidity increases from 40 to 60 % aerosol uptakes more water (deliquescence region) and therefore the aerosol distribution changes and also its total mass (mass of water increases). So, this can cause an error on the lidar signal especially from the sea salts. In order to check this point we computed the lidar reflectivity with constant relative humidity along the leg (60 % and 40 %). The results show that the overall structure of the lidar reflectivity is not affected by the relative humidity. The variability along the legs still stands with homogeneous relative humidity. The gradient of relative humidity along the leg thus plays a minor role (less than 1 % due to the hygroscopicity of the sea salts). Despite the effect of relative humidity on the lidar reflectivity, the original conclusions of the manuscript are still verified. In the revised version, we included this sensitivity analysis to make our

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conclusions more robust.

Page 11933, lines 8-9: Once more, how can the authors make such a statement, and do not include sea-salt in their model?

In order to address this issue, the sea-salt module was implemented in a new CHIMERE run, the results of which are thoroughly discussed in the revised manuscript. In addition, the absence of a careful validation of this sea-salt module up to now, is now a side objective of the revised manuscript

Figures 11-13: In the back trajectory analysis, the time of the back-trajectories should be presented (for example by 6-h points), together with its height. If the air parcels move above a city, but in very high altitude, their chemical composition will be completely different when compared to surface air masses.

The 24-hr trajectories of the particles ending at the time specified in the figure captions (Figs. 11 and 12) are represented with 1-hr resolution. The altitude of the particle is constant during the calculation. This was made clear in the revised manuscript.