Atmos. Chem. Phys. Discuss., 6, S6930–S6944, 2007
www.atmos-chem-phys-discuss.net/6/S6930/2007/

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

T. Salameh et al.

Received and published: 23 February 2007

The two referees of our paper state that our paper can not be accepted in the current form and suggest major revisions. They both suggest that:

- the objectives should be better highlight and their originality made clearer
- the absence of sea-salts on our simulation is a drawback that prevents relevant discussion of our results

We agree with the two reviewers about the two major points and all the other minor points. We agree that confusion could be made between the scientific context and motivation and the actual objectives of the paper (e.g. 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 two reviewer suggestions.

Also, 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 with the sea-salts are included in the revised version and the thoroughly discussed (in terms of AOD, lidar reflectivity). Finally, as suggested by the reviewers, we also addressed the issue of the amount of aerosol mass transported by the Mistral and Tramontane in addition to the background aerosol mass.

We thus found the remarks of the referees valuable and we used all of them to improve the quality of the manuscript in the manner described on the pages enclosed.

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. Therefore this article is ow 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.

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 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

ACPD

6, S6930–S6944, 2007

Interactive Comment

EGU

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.

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

ACPD

6, S6930–S6944, 2007

Interactive Comment

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.

Table 1. The difference of simulated and measured sulfates and nitrates is quite large,

ACPD

6, S6930–S6944, 2007

Interactive Comment

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

2 Anonymous Referee #3

Received and published: 5 January 2007

2.1 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, 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 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.

2.2 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. We also state more clearly the original points of our manuscript, following

ACPD

6, S6930–S6944, 2007

the reviewer suggestions. Therefore this article is ow 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.

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

ACPD
6, S6930–S6944, 2007

Interactive Comment

EGU

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

ACPD

6, S6930–S6944, 2007

Interactive Comment

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

ACPD

6, S6930–S6944, 2007

Interactive Comment

(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 SeaWIFS 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 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.

ACPD

6, S6930–S6944, 2007