**Atmospheric
Chemistry
and Physics
Discussions**

Interactive comment on "Transport and dispersion of atmospheric sulphur dioxide from an industrial coastal area during a sea-breeze event" *by* C. Talbot et al.

C. Talbot et al.

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The author and co-authors thank the referee for his remarks. We have chosen here to respond point by point on the S7055-S7058 comments.

Answer to specific comments:

1) Why was September 15th, 2003 chosen as the case study? Is there any motivation for this choice? Is this a typical sea-breeze event?

September 15th was chosen between several sea-breeze days of the experimental campaign where meteorological conditions were favourable to the development of the sea breeze, with a synoptic wind direction opposed to the direction of sea breeze prop-

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agation. These conditions help to distinguish the sea-breeze onset from the synoptic wind. These general meteorological conditions represent moreover a typical sea-breeze event with a 31 % occurrence of sea-breeze cases.

2) When giving an overview of the case study it would be helpful to provide a synoptic chart showing the synoptic flow pattern for this specific case study.

This will be updated for clearer reading of the figure.

3) How is sea breeze identified? Are other variables such as temperatures, humidity and gustiness used or only wind speed and wind direction?

The sea breeze is identified by a sudden switch of the wind direction turning from a southerly to a northerly direction, and is measured by the ground meteorological stations and the sodar. Moreover, a strong sodar echo is measured during the passage of the sea breeze front. For this day, there has also been a slight decrease of the air temperature and an increase of the relative humidity. At 13:00 UTC, the sea breeze front is observed on the vertical cross section of the lidar signal variation. These features were shown in detail in the previous publication Talbot et al. 2007.

4) In the introduction extreme values for rain PH and SO2 concentrations are given. It would be useful to have some average rain PH and SO2 values to compare to.

The usual given value in the literature for natural rain PH is 5.6 (Seinfeld and Pandis 1998). Whelpdale and Miller (1989) displayed a map of the worldwide rain PH, which indicated an average PH of 4 of the rain precipitation over northern Europe, during the background air pollution monitoring program of WMO (the map may be found also in Seinfeld and Pandis 1998). There is unfortunately no monitoring of the rain PH in the Nord-Pas de Calais region. The extreme PH value of 2.8 is extracted from a series of measurements during a field campaign led by the ULCO university, in the industrial area in 2001 (Ledoux 2003). Averaged daily value during September 2003 of SO2 concentrations, at the DK7 station, were 11 μg/m3 while the maximal hourly value for

this month was 169 μg/m3. This maximal SO2 concentration is typical of the industrial plumes in the region. The 920 μg/m3 value of SO2 concentration is an illustration of the extreme values recorded in the area.

5) I'm unsure what is meant by "the sea-breeze front brought up air masses from the gravity current in the above layers". Could this be explained better?

The model simulated an acceleration of the sea-breeze flow during its first hours, causing the marine air inside the gravity current going faster than the propagation of the sea-breeze front above land. In other words, the wind speed intensity of the gravity current is greater than the propagation of the sea-breeze front inland. As a result, during this short period of acceleration of the sea-breeze flow, air masses inside the gravity current reach the sea-breeze front and are uplifted in altitude by the ascents of the front.

6) How is the upper highly reflective layer measured? What does it represent? Is it a proxy for the top of the gravity current?

The High Reflectivity Layer (HRL) is detected by the lidar from 15:30 UTC to 22:00 UTC on September 15th, 2003 (Talbot et al., 2007). The lidar signals provide information about the atmospheric structure because of the different aerosol and gas loading of the diverse layers. When the laser beam crosses the HRL, the lidar signals increases, mainly due to the local high reflectivity of the aerosols. The HRL transitions heights (black diamonds and white rectangles in figure 3) are computed by using the inflexion point method (Menut et al., 1999). The top of the HRL decreases continuously from 400 m at 15:00 down to 250 m ASL at 22:00 exactly like does the top of the gravity current deduced from sodar measurements and modelling results. The HRL top is featured by high pollutants concentration as established in the paper (figure 8.b).

7) Which upper part does the headwind refer to? Is this the upper branch of the circulation which is advected air offshore?

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The headwind is a descent of air masses located behind the sea-breeze head, whose horizontal wind direction is oriented by the synoptic southerly wind at the same altitude. It differs from the synoptic wind, since the wind accelerates locally due to the presence of the sea-breeze front, which also creates the descent of the air masses behind the front.

8) How are the SO2 concentrations normalised in figure 5?

Measured and simulated SO2 concentrations are normalized with the respective daily maximal value.

9) Why are comparisons between measured and modelled SO2 concentrations only shown for 2 sites? Does the model capture the peak in SO2 seen at the seashore site (DK3)?

Emissions in the model are located at ground level and comparisons were given for only 2 stations because most stations are too close from the sources, according to the model resolution, to compute correctly the dispersion of these emissions. One would need a finer model resolution to be able to include all industrial stations for a comparison. The other stations are not downwind of the industrial emissions at the sea-breeze onset. The model does not see any peak of concentration at DK3 because the wind direction veered from south-easterly to southerly direction before the sea-breeze onset, which means DK3 never was downwind of the industrial emissions during that day.

10) I don't understand how pollution can be "channelled up to the sea breeze front". From figure 7 there does not appear to be any low-level convergence.

Pollutants are channelled inside the gravity current from their sources up to sea-breeze front due to the acceleration of the sea-breeze flow (cf. answer to question 5). The convergence of winds between synoptic and gravity current was recorded by stations with different times of sudden change in the winds directions. Stations registered the

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sudden change in wind direction at different hours depending to the relative position of the stations on the coastline. The low-level convergence isn't obvious on the figure 7 because the representation of wind is an orthogonal projection in the vertical cross-section.

11) What causes the layering of gas and aerosol pollutants above the sea-breeze circulation?

The sea-breeze system enhances the process of atmospheric stratification by a disappearance of atmospheric convection above the TIBL. The income of fresh marine air induced a decrease of the mixing layer height from 1100 m (the top of the ABL) to about 200 m above the city (the top of the TIBL). The first consequence is an increase of the pollutants concentrations at ground level due to the loss of vertical dispersion in the new mixing layer. The second one is redistribution by the sea-breeze system of pollutants above the gravity current, in an area where there is no more vertical ascents. Such a phenomenon favours the layering of gas and pollutants at a relatively low altitude (above the sea-breeze gravity current) and several hours before night time.

12) What does the lidar measure? Are the positive and negative values in figure 8(c) aerosol layers? Could the coastline be marked on this figure?

During this experiment, the lidar was used to continuously deduce the structure of the lower troposphere and to measure the vertical distribution of ozone (not showed in this paper) up to 1400 m. The lidar was located at Petite-Synthe, 6 km from the coastline, and the maximal range of the lidar measurements (1400 m) is too short to mark the coastline on the figure 8(c). The variations of the lidar signals provide qualitative information on the structure and dynamics of the lower troposphere. The derivative of the lidar signals, here referred to as the Negative Lidar Signal Variation (NLSV), is used for deducing the main structure of the lower troposphere. The figure 8(c) represents the vertical section of the NLSV deduced from the lidar before the land-breeze onset (from 20:45 to 21:07) and shows a zone of optical heterogeneity

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consisting in an horizontal multilayered structure below 600 m.

13) Most of the SO2 appears to have been advected larges distances from the source regions in figure 9. If this is the case how can it contribute to the photochemical activity at the start of the next day? It would be interesting to calculate by how much the SO2 concentrations are enhanced the following day due to the sea-breeze circulation. More evidence is needed to support the conclusion that the SO2 is recycled the next day and thus adds to the pollution levels.

The SO2 map shows pollutants emitted in altitude by the sea breeze and thus represents the pollutants emitted before the end of the sea-breeze acceleration. During the hours following the sea-breeze event until the next day, pollutants emitted from the industrial area remained within a maximum range of 150 km from Dunkerque. The rotation of the wind keeps the pollutants during and after the sea-breeze event at a closer distance. SO2 was only used here as a passive tracer of the primary pollutants emitted from this industrial area, because this pollutant is emitted in great quantities. There is however no SO2 chemical reaction in the RELACS chemical scheme used in the model. Actually, the real SO2 concentrations would fairly drop with its transformation to H2SO4. Under such a consideration, the sea-breeze system may act as a sink for SO2 concentrations and a source for the creation of sulphate particles. In our discussion, the SO2 pollutant must be interpreted as the tracer of a wide variety of primary pollutants emitted from the industrial area and particularly VOC implicated in the formation of ozone. Let us note that half of the SO2 and VOC pollutants emitted in the Nord-Pas de Calais region comes from the only industrial area of Dunkerque. The chemical transformations of pollutants must be treated with a more complex chemical scheme in numerical simulations, which is out of the scope of this paper but will be subject of next studies.

14) The cumulus cloud in figure 10 appears to be 1 gridpoint only. Does this mean that it is < 1 km wide and < 60 m thick? Better vertical resolution is needed to identify this as a cloud. There is also another grid point further inland showing high condensed

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water, this is not referred to.

The coloured area represents a water condensation over a grid point in a zone with a 1-kilometer horizontal resolution of the grid mesh and 80-meter resolution for the vertical resolution at this altitude. However the fact that the model shows a cloud at the top of the sea-breeze front doesn't mean there really was a cloud formation at this instant and for this day. The water vapour mixing ratio is indeed hard to predict in numerical models since water vapour concentration is linked to the history of precipitations in nearby areas and the input numerical data are too coarse to give a correct representation of the water vapour distribution. On the other hand, the fact that the model shows a cloud formation in an area where marine air circulated above the industrial zone is interesting enough to be mentioned with respect to the occurrence of the sea-breeze phenomenon in 2003 (more than one day over three during summer period) and with respect to the uprising of pollutants in altitude by ascents in the sea-breeze front.

15) In the conclusions you state that you have made comparisons between the model simulations and the sodar observations. Are these comparisons discussed in the paper?

The sodar determined the height of the gravity current. Comparisons with the model allowed determining a discrepancy of about 150 m before 15:00 UTC from the simulations to the measurements. After 15:00 UTC, the height of the gravity current simulated by the model corresponded to the one measured by sodar. The comparisons were discussed in previous paper (Talbot et al. 2007).

16) Have you done any correlations between surface measurements and simulations of SO2 and or only qualitative comparisons? In the conclusions you state that correlations have been made.

Correlations were done but only from a qualitative point of view (cf. question 9). Simulations indicated a rather good restitution of the dynamics by the model by comparison with the measured concentrations.

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17) The modelled vertical distribution of SO2 (figure 8b) does not show the multilayer structure seen in the lidar measurements (figure 8c). Why is this? In the conclusions you state that the model does generate these layers.

Figure 8b shows a representation of a vertical profile of SO2 concentration for an ideal case at 18:00 UTC and figure 8c shows a lidar scan of the atmospheric stratification at 21:00 UTC. The lidar scan brings information about a heterogeneous optical atmosphere at low altitude which cannot be easily correlated with the only industrial primary SO2.

Answer to technical comments:

1) A south wind refers to a south-easterly wind, i.e. comes from the south-eastern direction.

2) The true meaning in line 22, section 2.2, is "coming onshore" instead of "coming offshore". Marine air masses are coming inland from the MBL. There has been a mistake in the manuscript.

3) Figures 6(a) and 6(b) will be updated for easier comparisons.

4) Figures 6,7,8,9 and 10 will be updated for easier reading.

5) In addition to the caption of figure 7: Dashed contour represents the sea-breeze front position; arrows represent coarsely the average wind direction for clearer reading (synoptic wind, wind in sea-breeze font, gravity current, the headwind is represented by the gray arrow and, finally, the wind above the gravity current).

6) Figure will be updated for easier reading.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 15989, 2007.

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