Reply to Reviewer #1

This paper presents a through, comprehensive comparison of three tropospheric NO2 VCD data sets from car-MAX-DOAS measurements, OMI satellite products (DOMINO v2.0), and a regional chemical transport model (CHIMERE) for a megacity. The authors report a unique and valuable data set derived by car-MAX-DOAS measurements in and around Paris, which covers two relative long-term periods with 25 days in the summer of 2009 and 29 days in the winter of 2010. They perform a detailed analysis of the advantages and limitations of the three data sets, and then improve each data set by making use of the advantages of others in a synergistic way. A new and interesting result of this study is that CHIMERE model results can be improved by rotating around the center of Paris based on the plumes measured by car-MAX-DOAS.

The manuscript is well written and structured in general, although the text seems to be lengthy and some parts, e.g. in description and presentation of regression results, can be more concise. I would recommend the manuscript to be published after the following comments have been addressed.

Author reply:
We thank the reviewer for the positive assessment and the useful suggestions. We addressed almost all of ths suggestions as described in detail below.

Specific comments:

While the rotation of model results around the center of Paris based on the car-MAXDOAS measurements appears to be effective for improving the model simulation of urban plumes, the physical picture behind this operation is not clearly described.

First, it is stated that disagreements in the locations of the NO2 maximums between the car-MAX-DOAS measurement and CHIMERE simulation are caused by biases in surface wind direction used by CHIMERE, but the reason for this poor model behavior is not sufficiently explained in the paper. The paper merely refers a previous study at the SIRTA site, which is at 20 km SW from the center of Paris, for surface wind observations (Page 2454, Line 1-3). Are there any concurrent wind observational data to validate the wind fields simulated by MM5 for the experimental periods in this study?

Author reply:
The referee asks for reasons of wind direction errors in MM5 simulations driving CHIMERE chemistry-transport model simulations. First, we would like to stress that the optimal rotation angles up to ±25° found in our study do not necessarily reflect a poor model behavior. Indeed, surface (10m) wind field errors in mesoscale meteorological models of about ±30° (Heinke Schluenzen, personal communication, Pielke et al., 2013) are considered as typical. In our paper, we made reference to a comparison with wind observations (at 10 m height) at the SIRTA site 20 km in the SW of Paris center. This site is chosen, because it delivers observations of meteorological and dynamical parameters of high quality and with a known spatial representativity (Haeffelin et al., 2005). The measurement site is located on a plateau and in mostly free terrain. The

comparison referred to in Zhang et al. (2013) does cover a large part of the summer measuremement period, that is all July 2009. Typical wind direction errors during this period are generally below ±20° (Zhang et al., 2013, except for days with very low windspeed below 2m/s). These errors are compatible with the typical mesoscale model errors on one hand, and with the optimal rotation angles found in our study, on the other hand.

In order to make these points clear, we added the following text at the top of page 2454 instead of the simple reference to SIRTA measurements:
"Such differences between simulated and observed surface wind direction of this order are frequently observed for surface winds (at 10m height) at the SIRTA site at Ecole Polytechnique, Palaiseau, at 20 km SW from the town center. This site is chosen, because it delivers observations of meteorological and dynamical parameters of high quality and with a known spatial representativity (Haeffelin et al., 2005). The measurement site is located on a plateau and in mostly free terrain. For July 2009 (covering most of the summer measurement period in this paper), wind direction errors were generally below ±20° (except for days with very low windspeed below 2 m/s) (Zhang et al., 2013). These errors are compatible with the typical mesoscale model errors on one hand (Pielke et al., 2013), and with the optimal rotation angles found in our study, on the other hand.

At what time intervals did MM5 provide meteorological data for CHIMERE (Page 2446, Line 3-4)? It is stated in the paper that CHIMERE data are available in hourly time steps (Page 2448, Line 1). Is it possible that the wind direction change dramatically (by up to 25 degree) within this time interval?

Author reply:
MM5 provides meteorological data for CHIMERE in an hourly time step. This is now explicitly stated in the paper. Observed wind speeds used for the comparisons are available at an hourly time step. In general, observed (and simulated) wind directions smoothly change form one hour to the next, at a rate of typically 10 to 20 degrees (see figure 2 in Zhang et al., 2013). However, especially during low wind speed periods, much larger jumps in observations are possible, which are then often not simulated. In these cases however, the average transport time between NOx emission sources and the measurements is of several hours, which averages out sudden jumps in wind direction.
We added this information at the end of section 2.3.

Second, it is acceptable if the locations of several model grid cells with peak NO2 from CHIMERE are moved (rotated) to match car-MAX-DOAS observations. However, if all the model grid cells for NO2 VCDs in a larger area are rotated around the center of a city as performed in this study, there might be a problem, e.g. in the case that there are strong emission sources in the upwind area of the city. In that case, the wind fields instead of NO2 VCD results should be corrected.

Author reply:

Here it should be noted that instead of rotating the model results, it would have been more correct to rotate the wind fields before using them in the model simulations (and leaving the emissions sources unchanged). However, this procedure would be very time-consuming, since complete model simulations would have to be performed for each rotation angle of the wind fields. Fortunately, for the model results the errors caused by rotating the whole wind fields are small, because the NOx emissions within the Paris agglomeration (within about 20 -30 km distance from the Paris center) are on the average about 2 orders of magnitude larger than emissions outside the agglomeration (see Fig. 1c, Petetin et al., 2014). The next larger cities (Orléans, Reims, Rouen) are at more than 100 km distance from the agglomerations, but their population is much smaller (100.000 to 200.000 inhabitants) than that of the Paris agglomeration (nearly 12 million inhabitants). Outside the Paris agglomeration, emissions are concentrated along several highways and also along the Seine and Marne rivers. But again, these emissions are much smaller than those within the agglomeration.
We added this information to section 4.1.

Again, CHIMERE data were available in hourly time steps while car-MAX-DOAS observations persisted 1-2 hours with 1 min for an individual observation. Therefore, it might not be necessary to match simulated NO2 VCDs in all the model grid cells outside the plume to car-MAX-DOAS measurements due to the time differences.

Author reply:
In principle it could be a valid option to remove some measurements from the comparison, which are dominated by sources far away from the center and with a large time difference. However, this would require a) the definition of clear selection criteria, b) exact knowledge about these sources. While this procedure might be a good option for other cities, for Paris it seems to be not so important.

Overall, while the rotation method proposed in this study appears to be applicable to Paris, some assumption and limitations should be given and discussed.

Author reply:
We added the following information to section 4.1:
'Here it should be noted that for many other cities, probably less ideal conditions exist. In such cases, modified rotation methods might be applied, by e.g. excluding areas with strong interfering sources outside the city center.'

Technical issues:

P2440, L10-11: This phrase needs to be polished. It can be "with the European annual limit value of 40 _g m-3 being exceeded not only at the urban traffic sites, but also frequently at urban background sites".

Author reply:
corrected.

P2440, L22: Add a comma between "(2012)" and "can".

Author reply:
corrected.

P2443, L9: should be “5_22_”?

Author reply:
Many thanks for this hint! Actually, our submitted manuscript was correct. The error was introduced during typesetting, and we did not recognise that. We will take care after typesetting of the revised version.

P2443, L27: Delete “(“ in front of “depending” or add “)” somewhere.

Author reply:
corrected.

P2445, L15-: Are the diurnal variations in emission rates (e.g. from traffic) considered in the inventory?

Author reply:
Yes, activity and country specific diurnal (hourly) variations in emission rates are considered. They are applied during the emission preprocessing procedure (Menut et al. 2013).
This information is added in section 2.3.

P2462, L15: Delete “)“.

Author reply:
corrected.

P2464, L3: It seems not so informative to use “Sects. 4 and 5” in a section title.

Author reply:
we changed the title to’ Comparison with the results of the bilateral comparisons OMI versus CHIMERE and car-MAX-DOAS versus CHIMERE’

P2485 and P2496: There are some inconsistencies between the rotation angle values shown in Fig. 6 and in Fig. 17. On 18 July 2009, for example, it is lower than –10 degree in Fig.6, but higher than 10 degree in Fig. 17.

Author reply:
In section 4.1. we already discussed the probable reasons for the disagreement between rotations determined with either car-MAX-DOAS and OMI:

‘The rather low correlation is probably caused by the fact that the comparisons of the model data with both observational data sets are made for different times and locations. In particular the comparisons versus OMI observations are performed for a much larger area (see Figs. 3 and 4).’

In the revised version we changed the text (in section 5.2):

‘Like for the comparison with the car-MAX-DOAS measurements, for most days the rotation of the CHIMERE data leads to an improvement of the correlation coefficients. However, for the slopes and y-axis intercepts only small changes are found.’

into:

‘Like for the comparison with the car-MAX-DOAS measurements, for most days the rotation of the CHIMERE data leads to an improvement of the correlation coefficients (as has to be expected). However, the improvement of the correlation coefficient is smaller than for the comparison with the car-MAX-DOAS measurements. Also, for the slopes and y-axis intercepts only small changes are found. Both findings indicate that the determination of the rotation is less well constrained by the OMI observations compared to the car-MAX-DOAS measurements. This can be explained both by the much coarser resolution of the OMI data and the frequent gaps due to clouds. Since the spatial resolution of the CHIMERE data is much finer than the OMI resolution, the shape of the OMI ground pixels has no significant effect on the determination of the rotation angles, as CHIMERE data is re-sampled to the OMI pixel extent.’