Validation of CHIMERE simulations against ADRIMED measurements, and application to the study of the aerosol direct effect on photochemistry in Lampedusa.

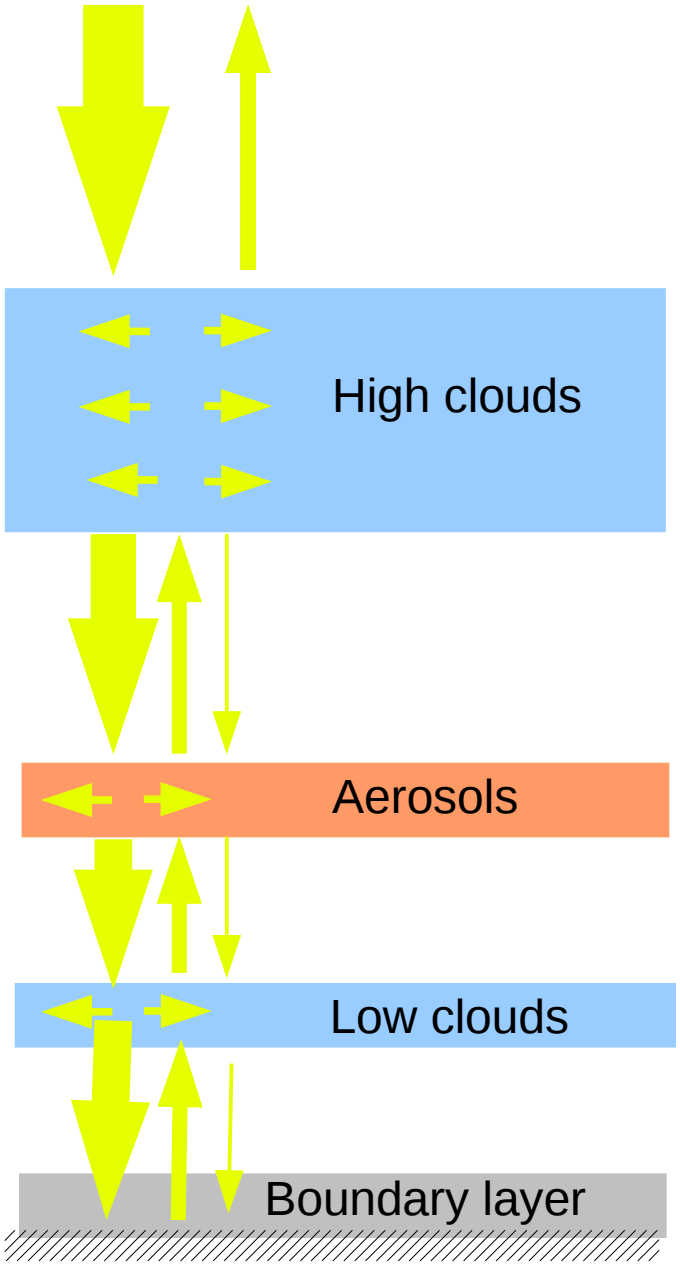
S. Mailler, 4th international workshop CHArMEx – ICTP, Trieste, 20-24 oct. 2014

Results from 2 studies :

1. Analysis of the atmospheric composition during the summer 2013 over the Mediterranean area using the CHARMEX measurements and the CHIMERE model. ACP, 2014 (submitted)
L. Menut, S. Mailler, G. Siour, B. Bessagnet,S. Turquety, G. Rea, R. Briant, M. Mallet, J. Sciare, P. Formenti

2. On the radiative impact of aerosols on photolysis rates: comparison of simulations and observations in the Lampedusa island during the ADRIMED campaign. To be submitted to ACP.
Sylvain MAILLER, Laurent MENUT, Alcide G. DI SARRA, Jose Luis Gomez Amo, Silvia Becagli, Roberto Udisti, Tatiana Di Iorio, Rita Traversi, Damiano Sferlazzo, Guillaume SIOUR, Bertrand BESSAGNET, Regis BRIANT, Solene TURQUETY, Geraldine REA

Radiative transfers in CHIMERE : what needs to be accounted for ?



Significant effects :

- **Rayleigh diffusion** by tropospheric and stratospheric ozone

- **Mie diffusion** by ice and water droplets within the clouds

- **Mie diffusion** by natural aerosols (volcanic ash, mineral dust, biomass burning, SOA ...)

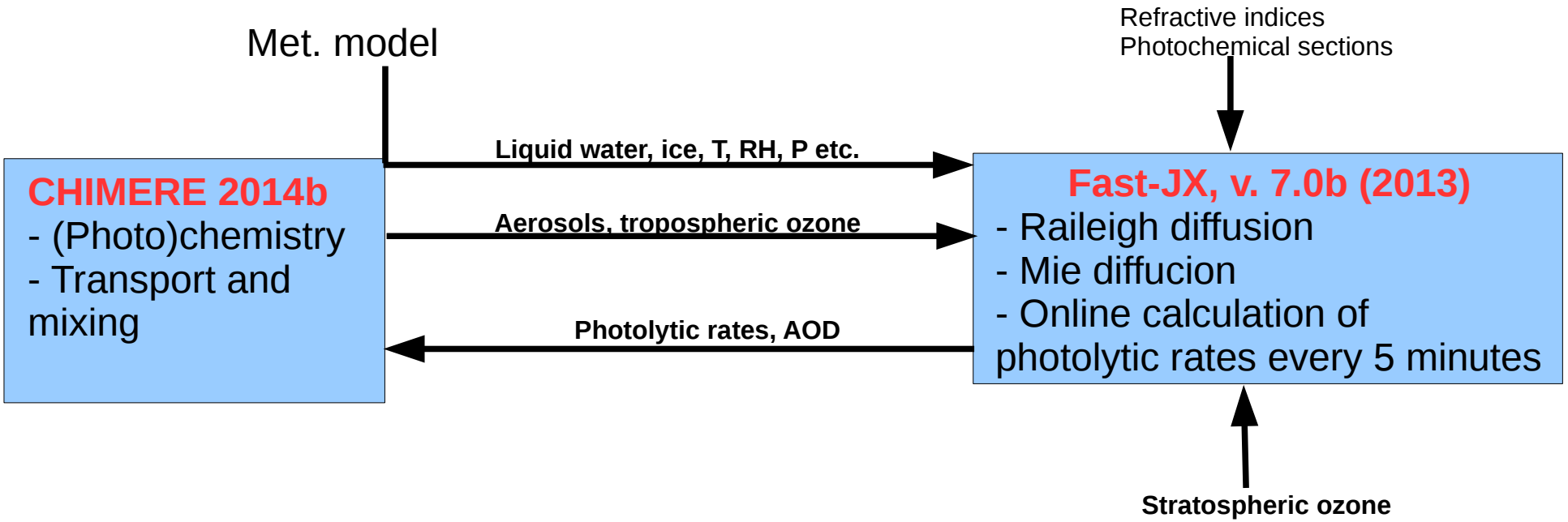
- **Mie diffusion** by anthropogenic aerosols

- **Reflexion and diffusion by the ground**

Relevant wavelengths :

- Visible + near-UV (200-1000 nm)

Radiative transfers in CHIMERE : How are they treated ?



- Fast-JX (*Wild, et al., 2000, J. Atmos. Chem.*) is a module specialized in the online calculation of photolytic rates, already in use in models such as Geos-chem, UKCA, Polair3d ...

- Takes into account all the effects we need to take into account : Mie diffusion, Raileigh diffusion etc.

- CHIMERE takes into account all the relevant particle sources : anthropogenic (trafic, industry, shipping, heating ...), natural (fires, dust, biogenic)

- Typically 15 species of aerosols, each one defined with 9 bins → detailed representation

→ All the ingredients there for a good representation of the radiative effects of aerosols !

Configuration of the simulation

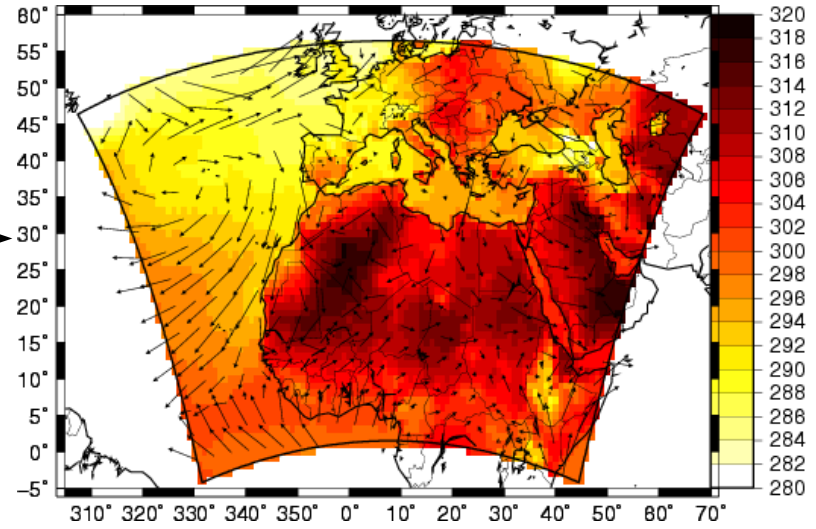
Met. model

Meteorology :
WRF 3.3
Forced by NCEP/GFS analysis, with spectral nudging inside the domain

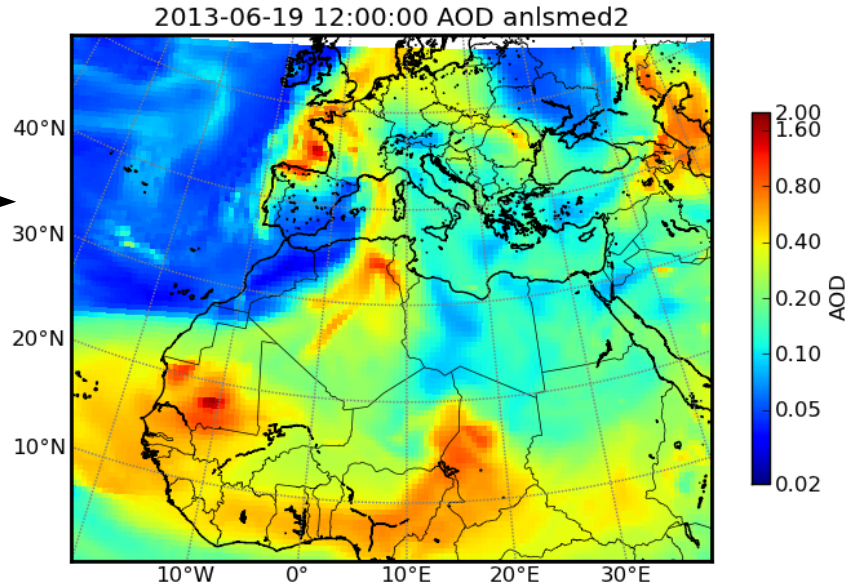
Chemistry-transport :
CHIMERE (development version)

- 107x157 grid points (about 60km resolution)
- MELCHIOR2 chem. Mechanism
- 20 vertical levels from 2 hPa a.g.l. To 300 hPa
- Includes online calculation of photolysis rates and of AOD using Fast-JX (Wild et al. 2000)

Emissions :
- Anthropogenic (HTAP)
- Biogenic (MEGAN model)
- Dust emissions : Menut et al. (2013)
- Fire emissions : Turquety et al. (2014)

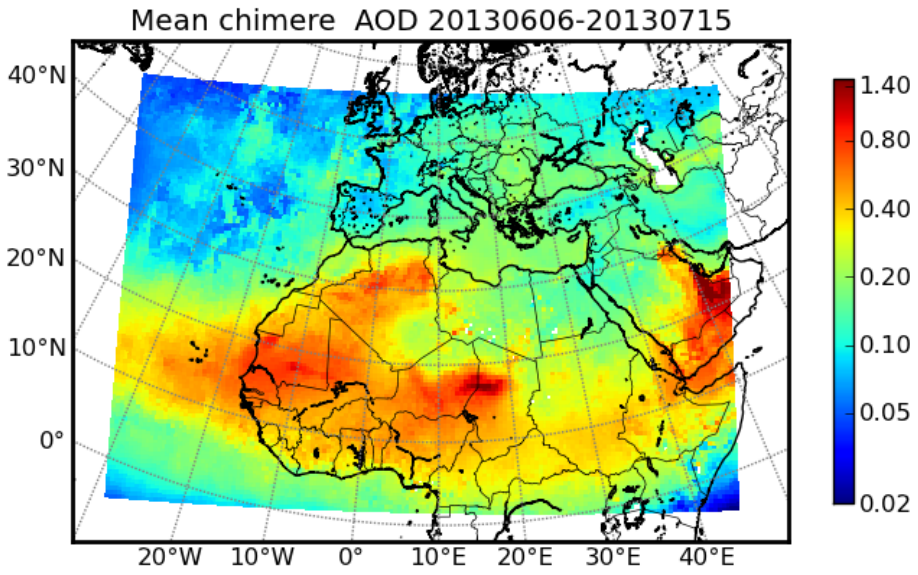


Simulation domain. Horizontal resolution is 60 x 60 km. 2M-temperature (K) and 10-m wind speed for June 21st, 12 UTC

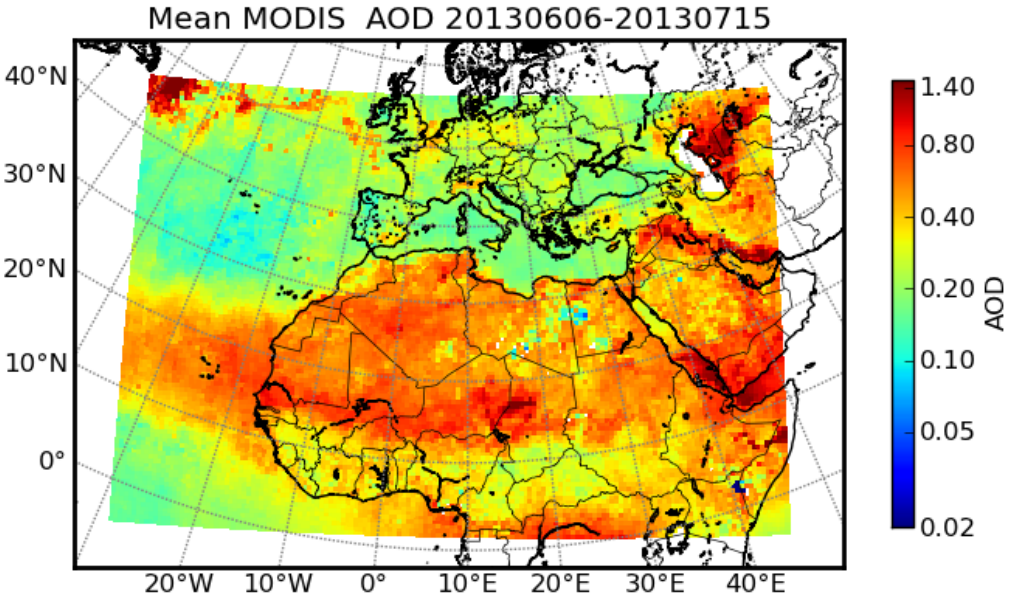


Simulated AOD (600 nm) for Jun. 19th,12UTC

Validation of the simulation : 1. average AOD



Average CHIMERE AOD, 06-06-2013-15-07-2015



Average MODIS AOD, 06-06-2013-15-07-2015

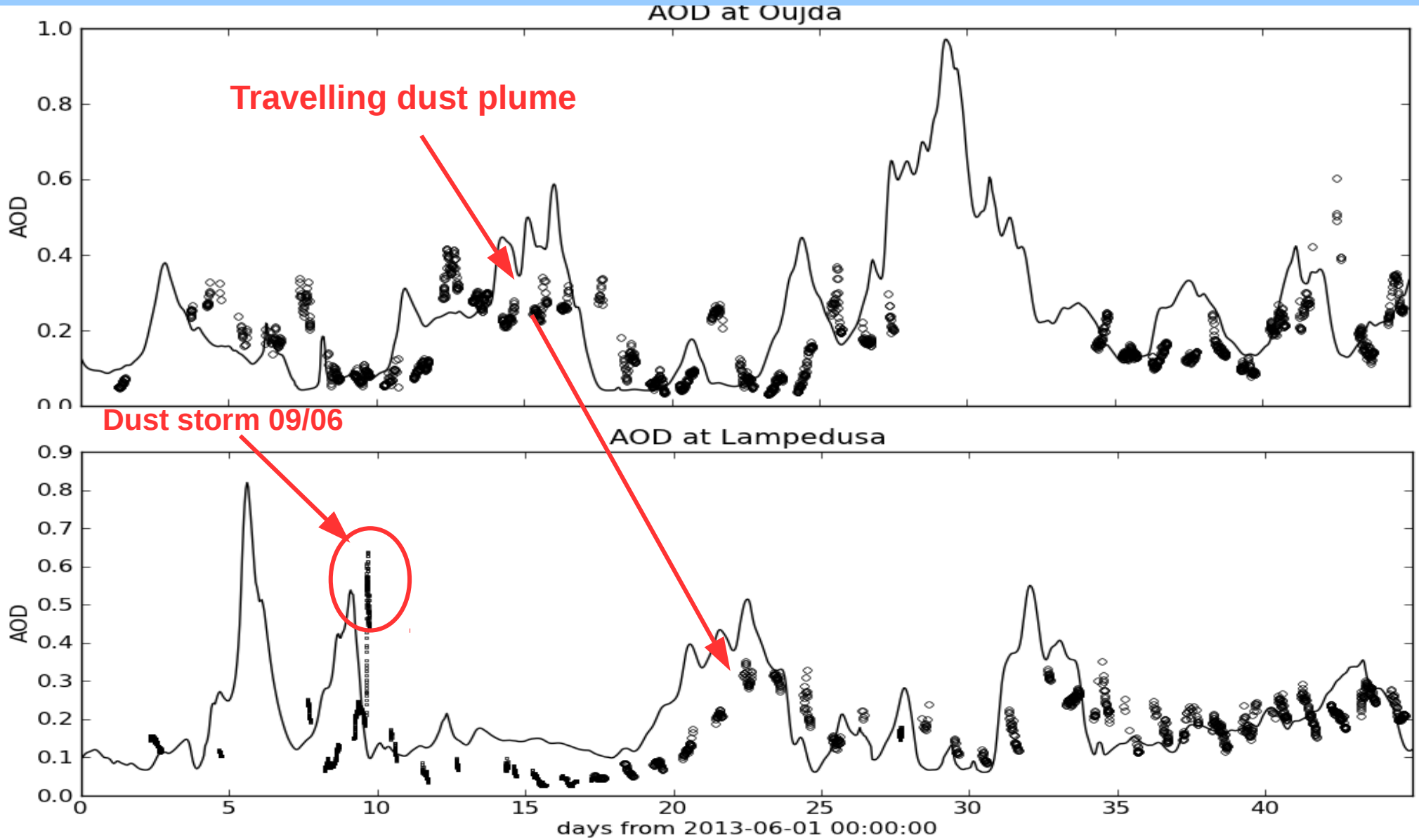
- Good agreement over the source areas particularly in NW Africa, the Atlantic dust plume and the Med Sea

- CHIMERE underestimation over industrial Europe

- Discrepancies in the eastern part of the domain, also in north-Atlantic (advection from ouside the domain?)

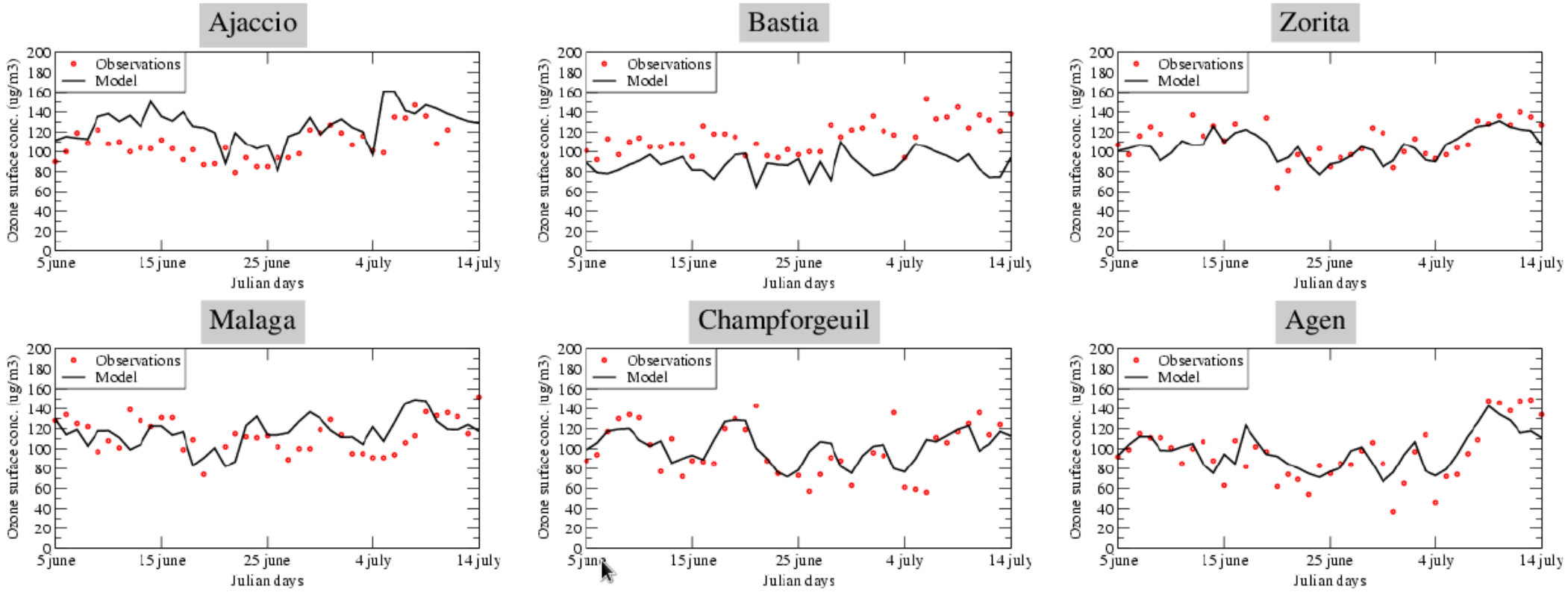
→ **Globally good agreement of average AOD**

Validation of the simulation : 2. Variations of the AOD



Comparison of CHIMERE outputs to measured AOD at 532 nm, for Lampedusa (AERONET data + MFRSR thanks to **D. Meloni**), AOD at Oujda (AERONET, thanks to PIs Diouri Mohammed and Djamaleddine Chabane)

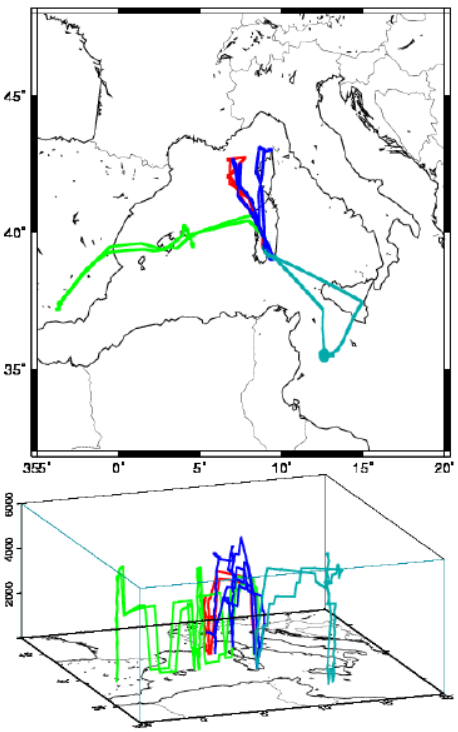
Validation of the simulation :
3. Variations of the ozone concentrations



Comparison of daily maximum O_3 values to station data (EMEP background station data)

Model biases are rather small
The model reproduces realistic day-to-day variations

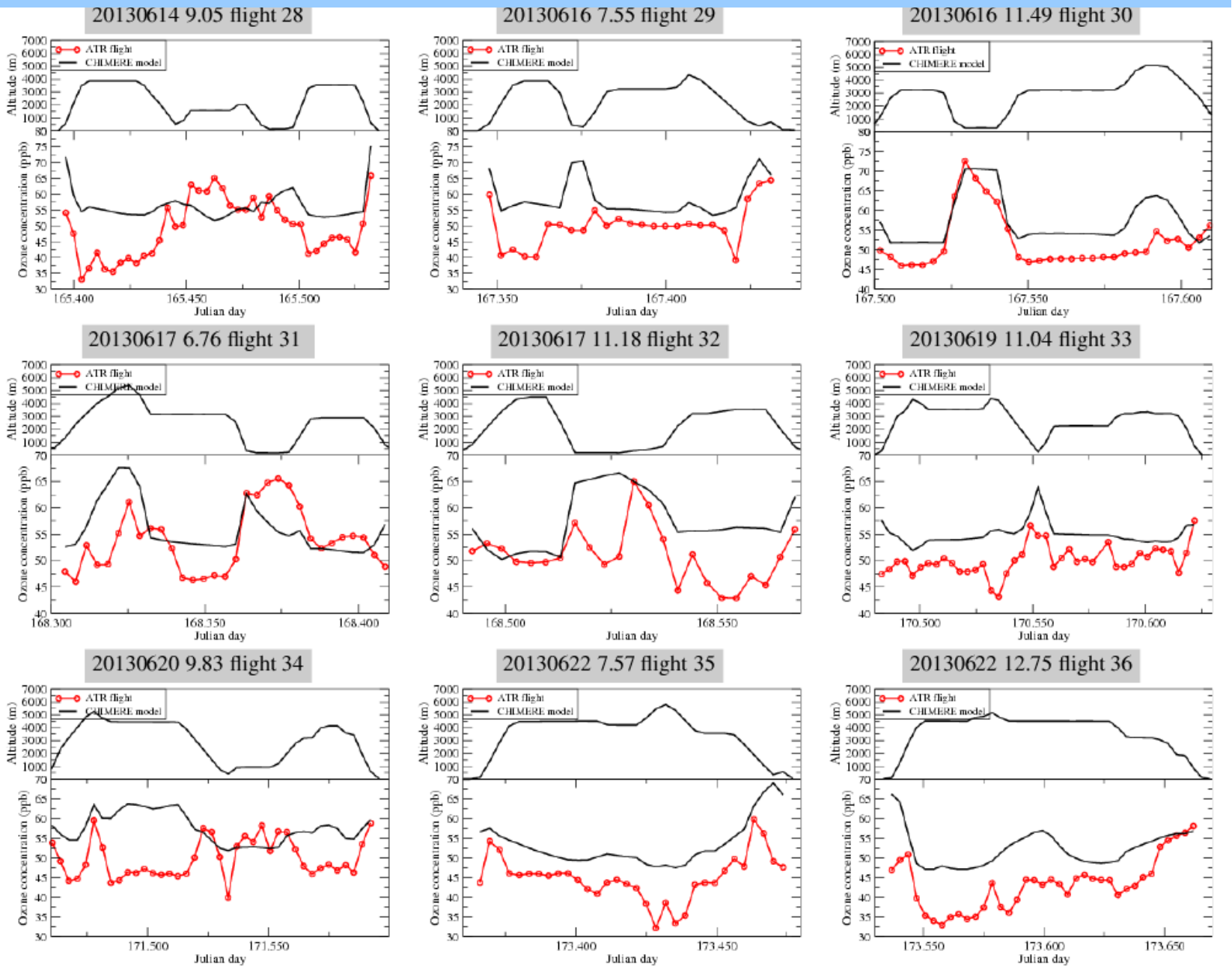
Validation of the simulation :
4. Comparison of ozone concentrations to airborne measurements



Flight trajectories

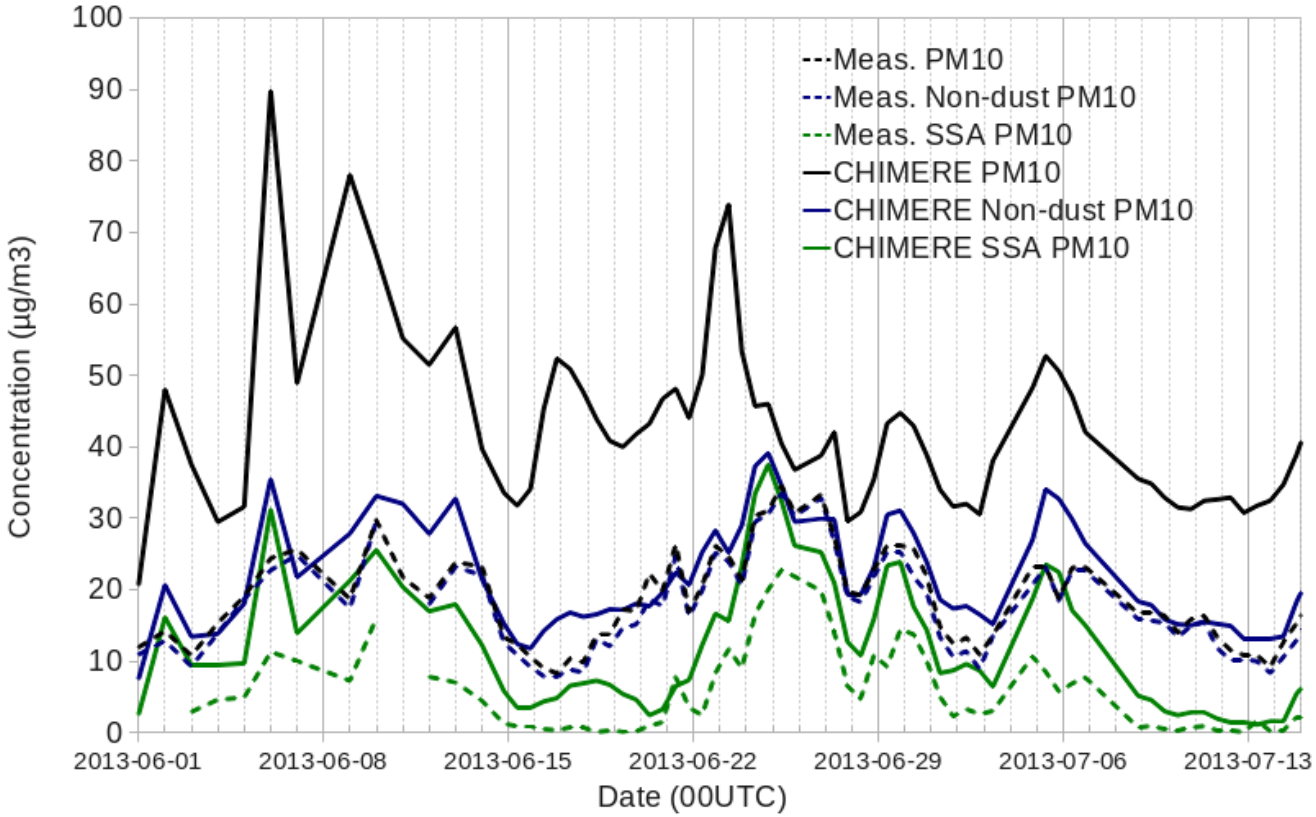
- Model bias about +5 ppb

- The model captures many variations seen by observations



Comparison of measured (red) and modelled (black) O_3 concentrations over flight paths

Validation of the simulation :
5. Focus on Lampedusa : Speciation data



Comparison of CHIMERE outputs towards measurements of **S. Becagli**, for total PM10 (black), non-dust PM20 (blue), and SSA (green)

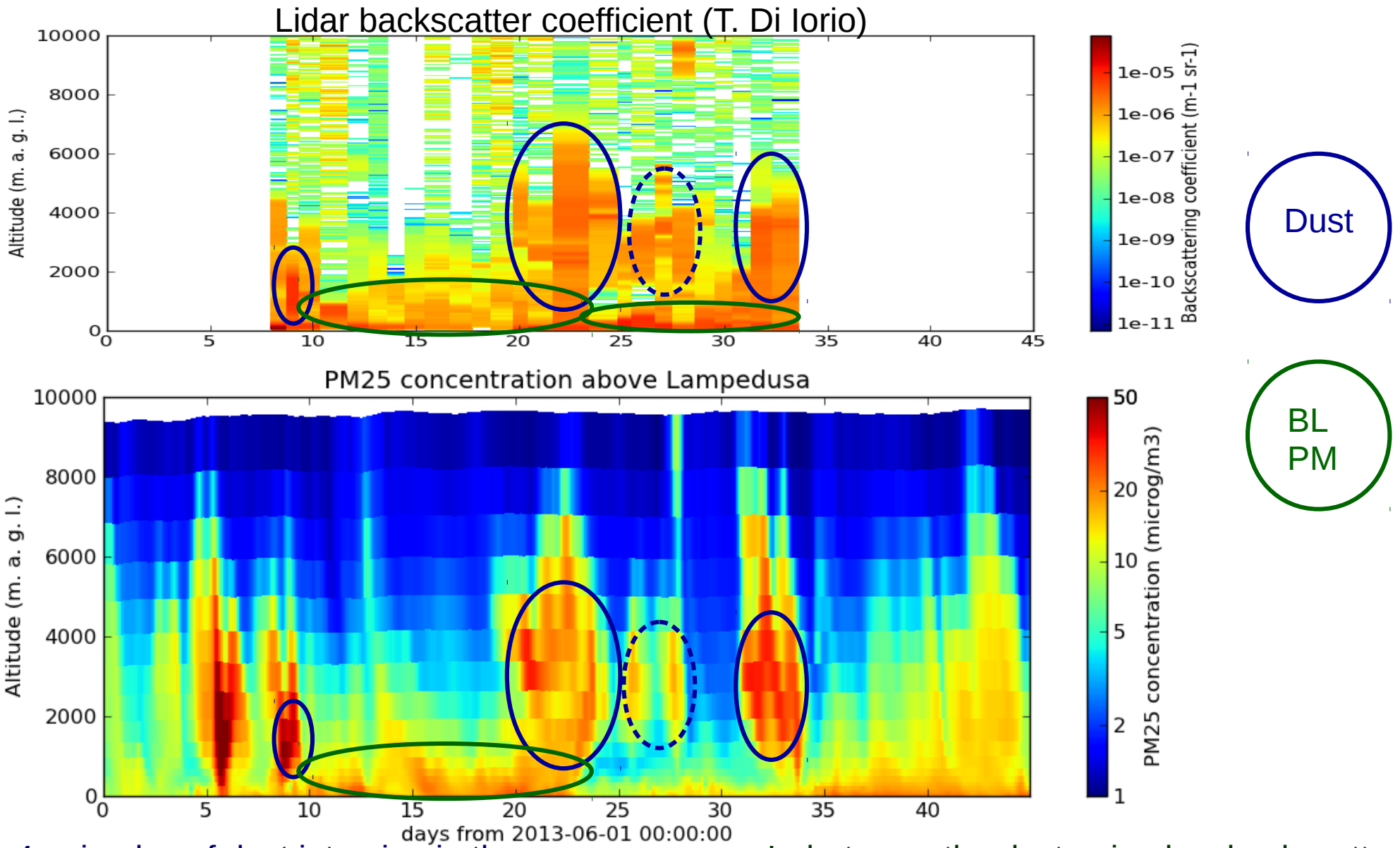
- At ground level, CHIMERE overestimates dust : excessive sedimentation or vertical diffusion ?

- Measurements show that **PM10 at ground-level is dominated by Sea-salt**

- **Non-dust aerosols (blue) correspond very well between CHIMERE and the measurements**

- Sea-salt aerosols overestimated in CHIMERE : **at the model grid scale (60x60km), Lampedusa is not representative of its entire grid cell**. But time correlation is excellent (0.86)

Validation of the simulation :
6. Focus on Lampedusa : LIDAR data

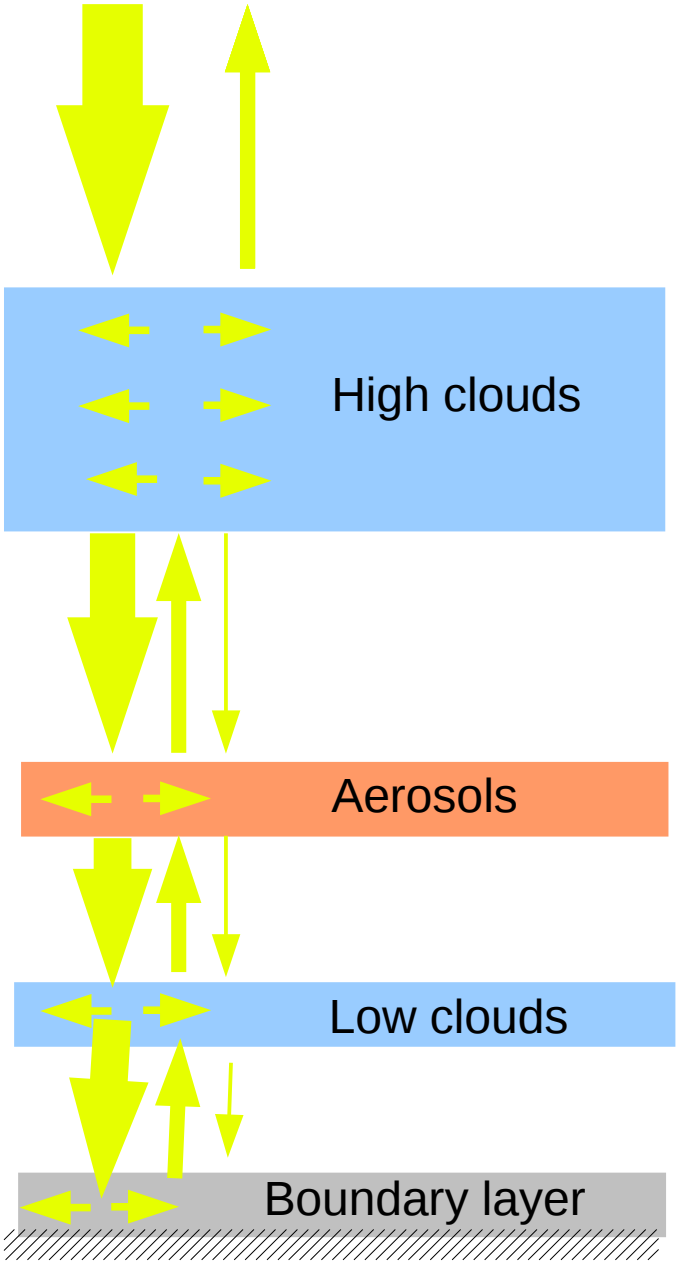


- 4 episodes of dust intrusion in the common period between LIDAR measurements and simulation. **Good synchronisation and vertical structure**

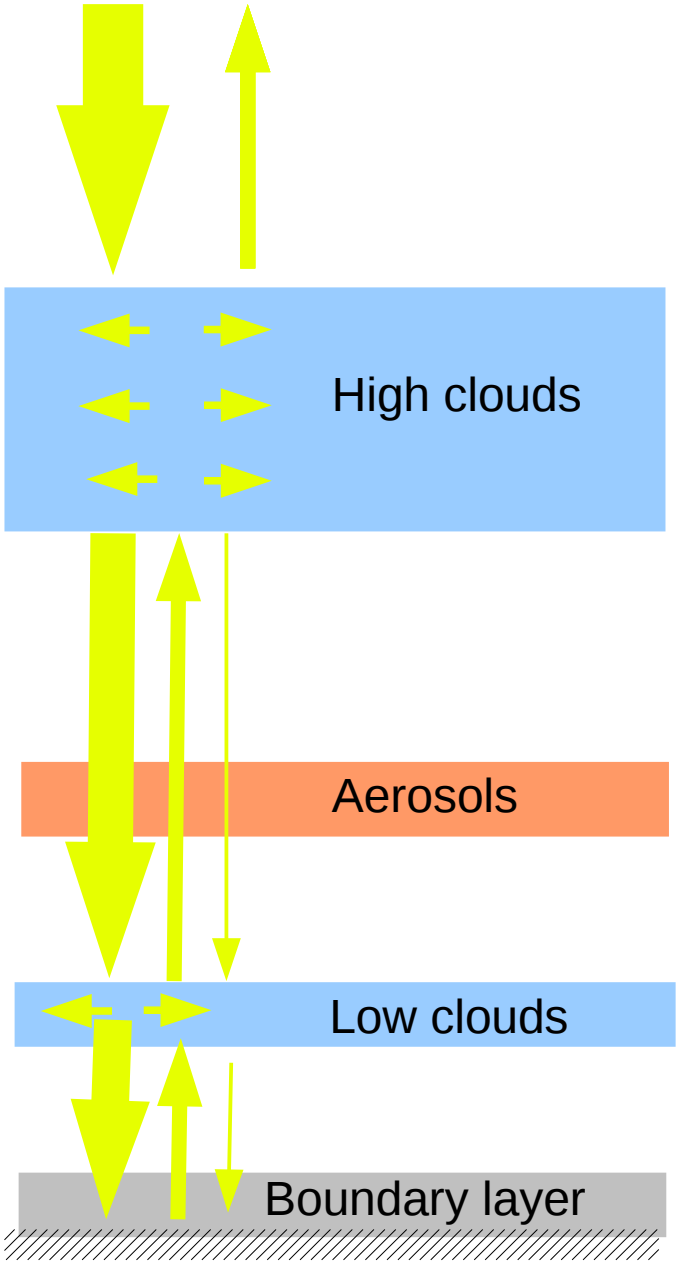
- In-between the dust episodes, backscatter is due mostly to **boundary-layer aerosols (mostly, Sea-salt)**.

Effect of the aerosols on photolytic rates : Two companion simulations

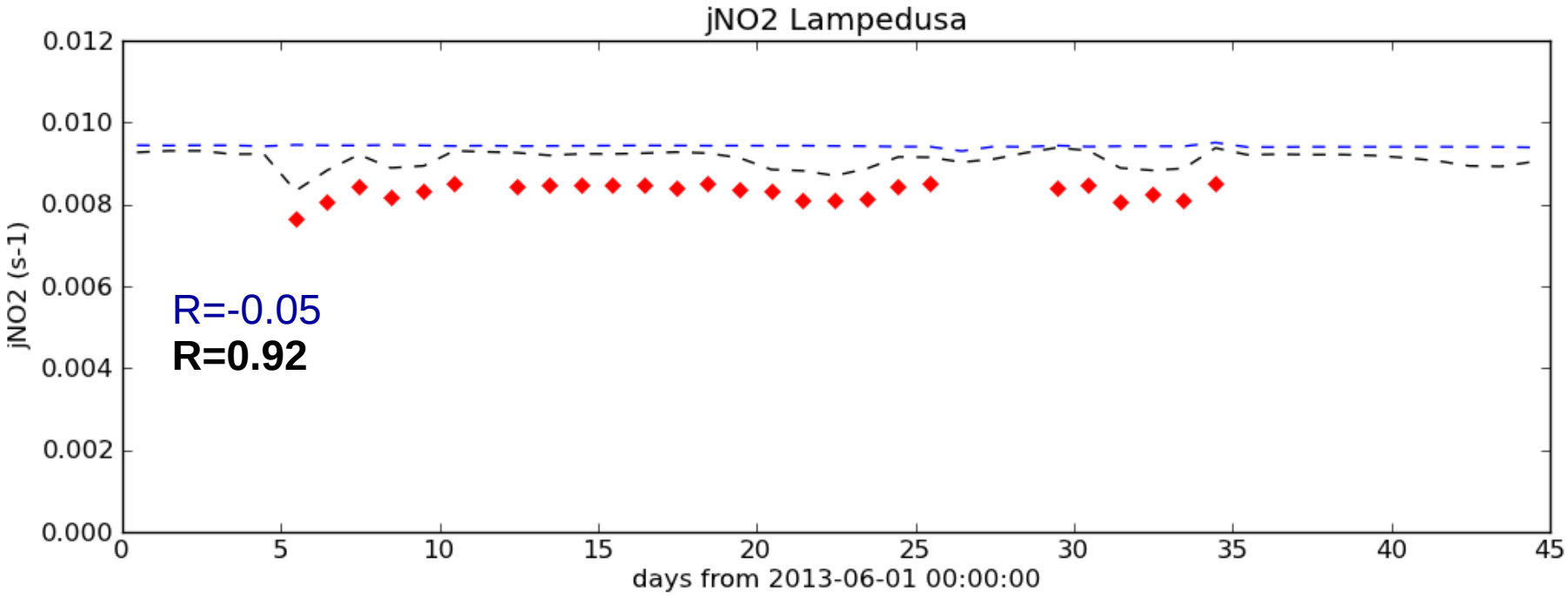
REF simulation : realistic aerosols



NA simulation : all aerosols are PERFECTLY TRANSPARENT



Effect of the aerosols on photolytic rates : The case of jNO2

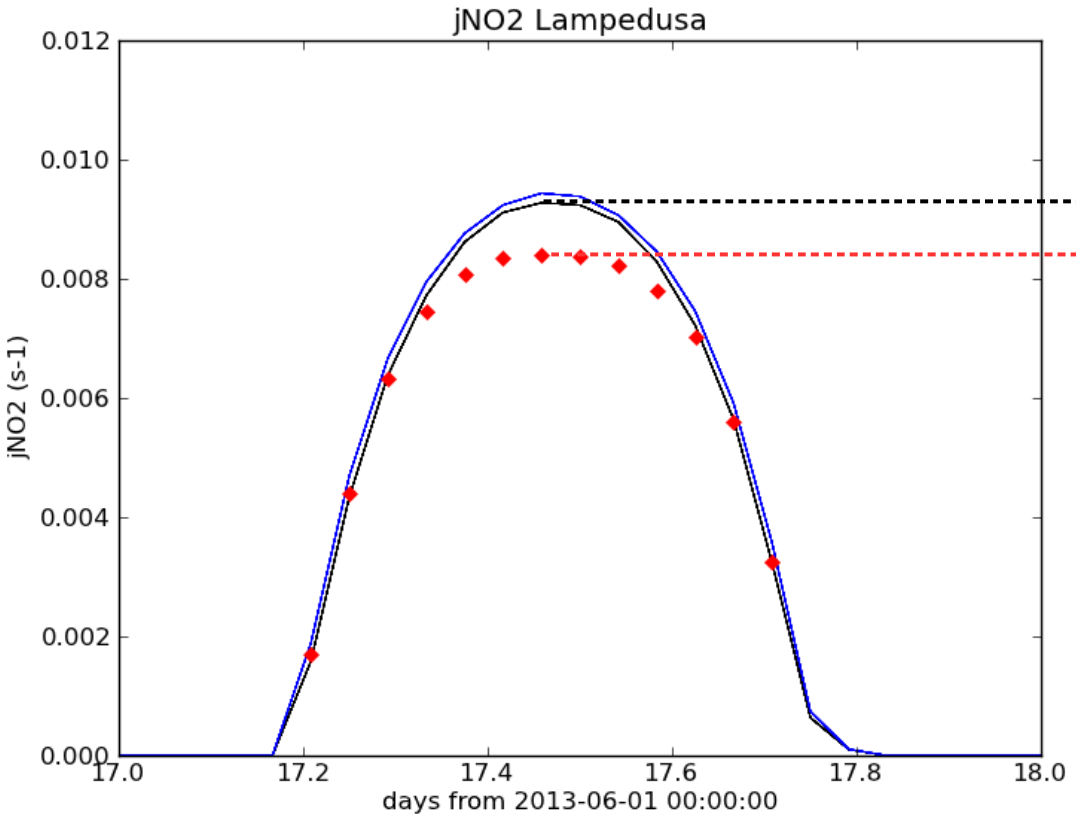


Red : Measured values from **A. G. Di Sarra** (daily max) ; black : REF simulation ; blue : NA simulation

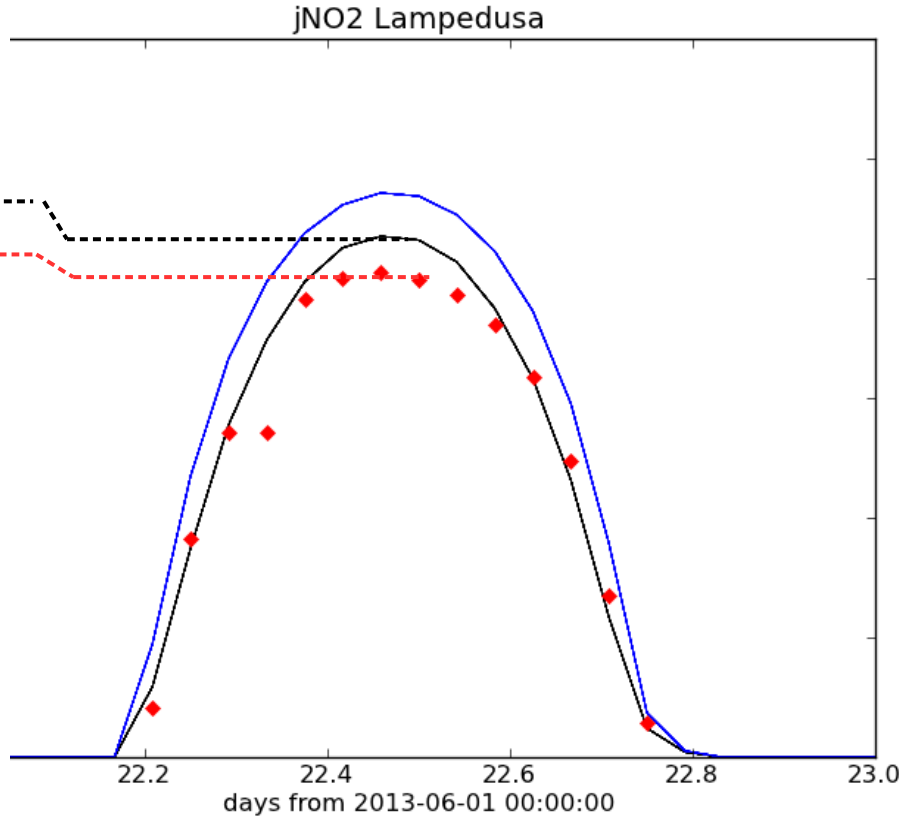
- In the absence of clouds, the REF simulation has an **excellent ability to reproduce daily variations of jNO2**. This ability is entirely **due to the inclusion of the effects of the aerosols**

- Slight bias of model compared to observation. Possible causes include the simplifications in the model, as well as measurement uncertainties.

Effect of the aerosols on photolytic rates : The case of jNO2



June 18 (clear day)

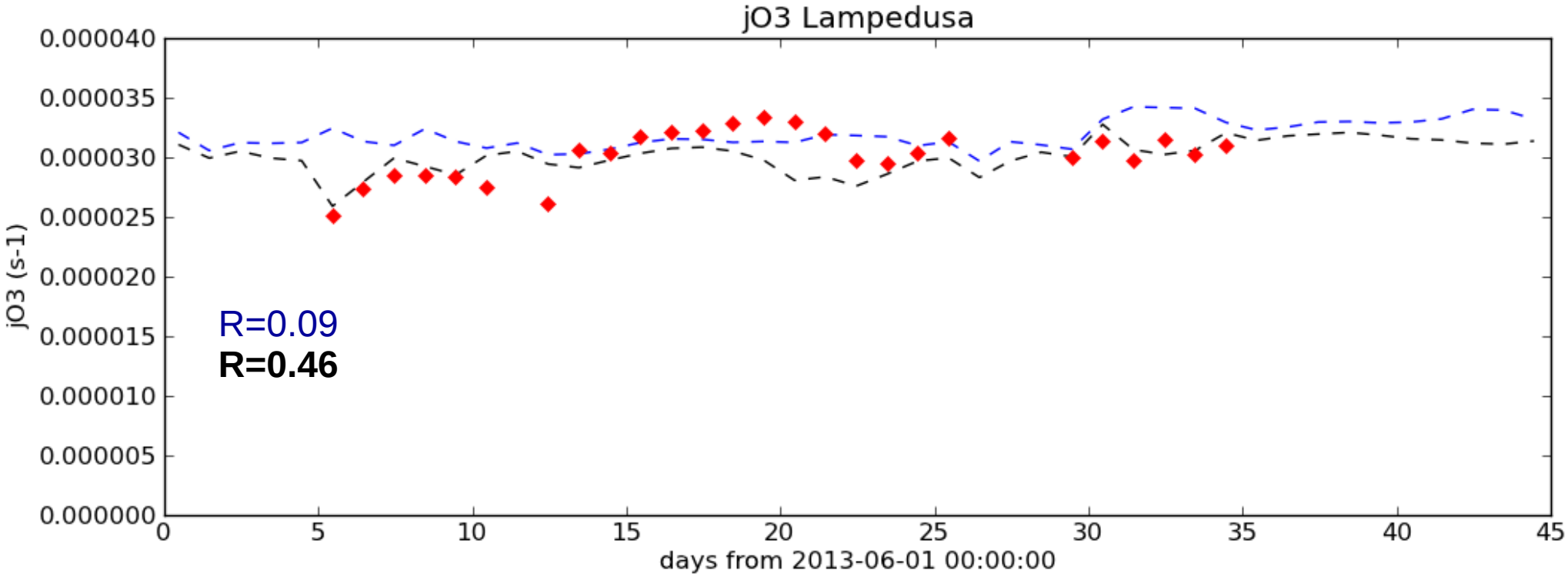


June 23 (Dusty day)

- Diurnal cycle is good in the model, except overestimation around noon

- Effect of the aerosols is very similar between model and observations

Effect of the aerosols on photolytic rates : The case of j(O1d)

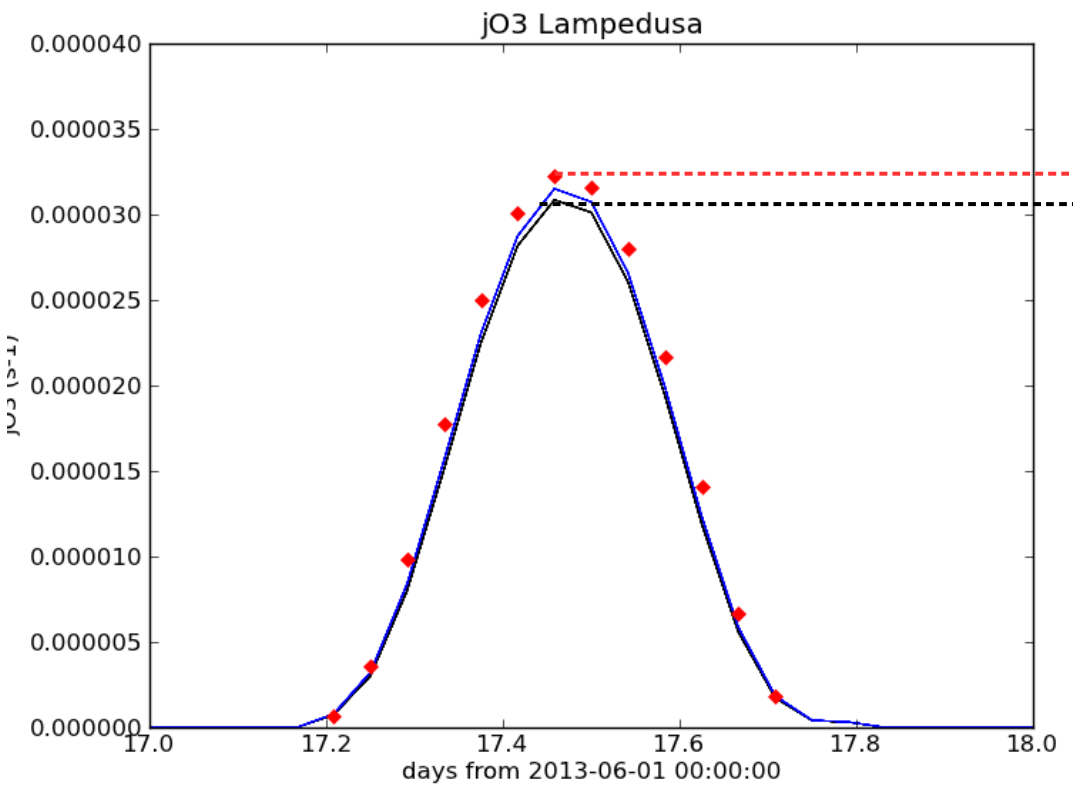


Red : Measured values (daily max) ; black : REF simulation ; blue : NA simulation

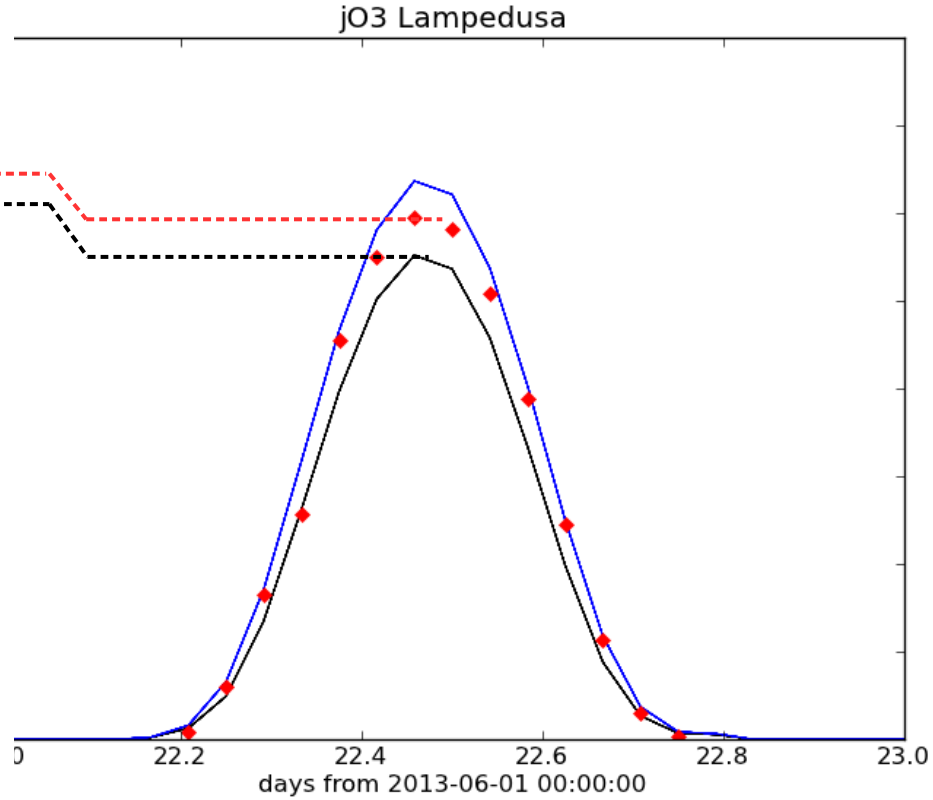
- No systematic bias of the model compared to the observations

- Including the aerosols permits the model to capture some variations. Other variations, due to, e.g., stratospheric ozone are missed.

Effect of the aerosols on photolytic rates : The case of j(O1d)



June 18 (clear day)



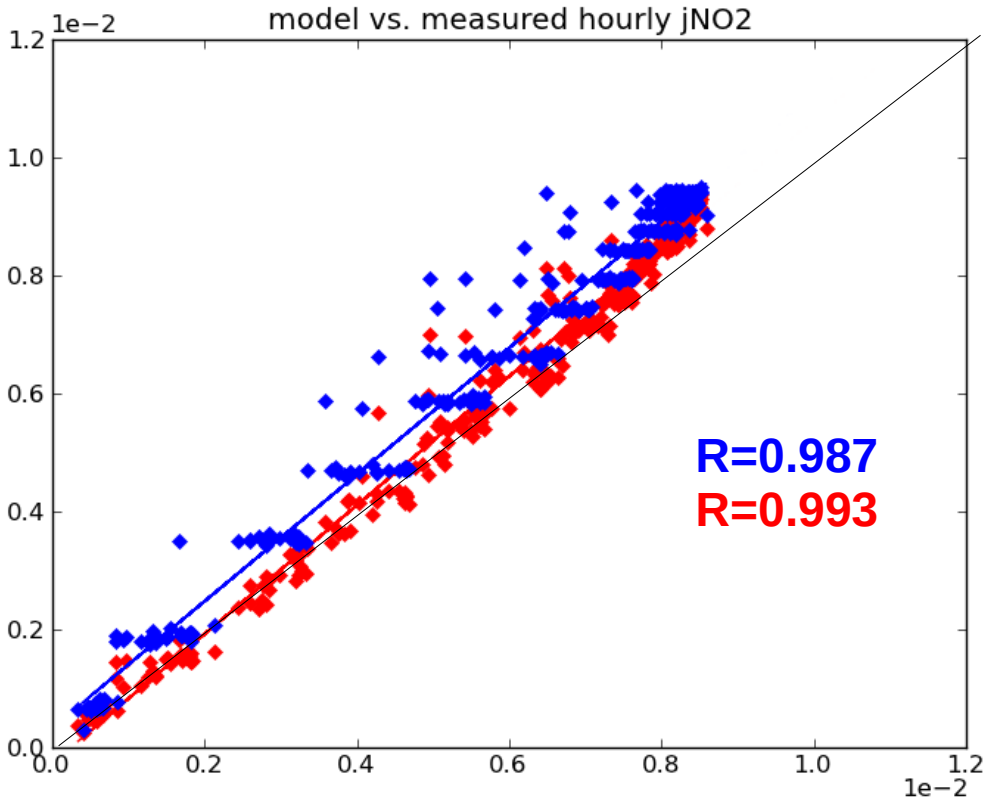
June 23 (Dusty day)

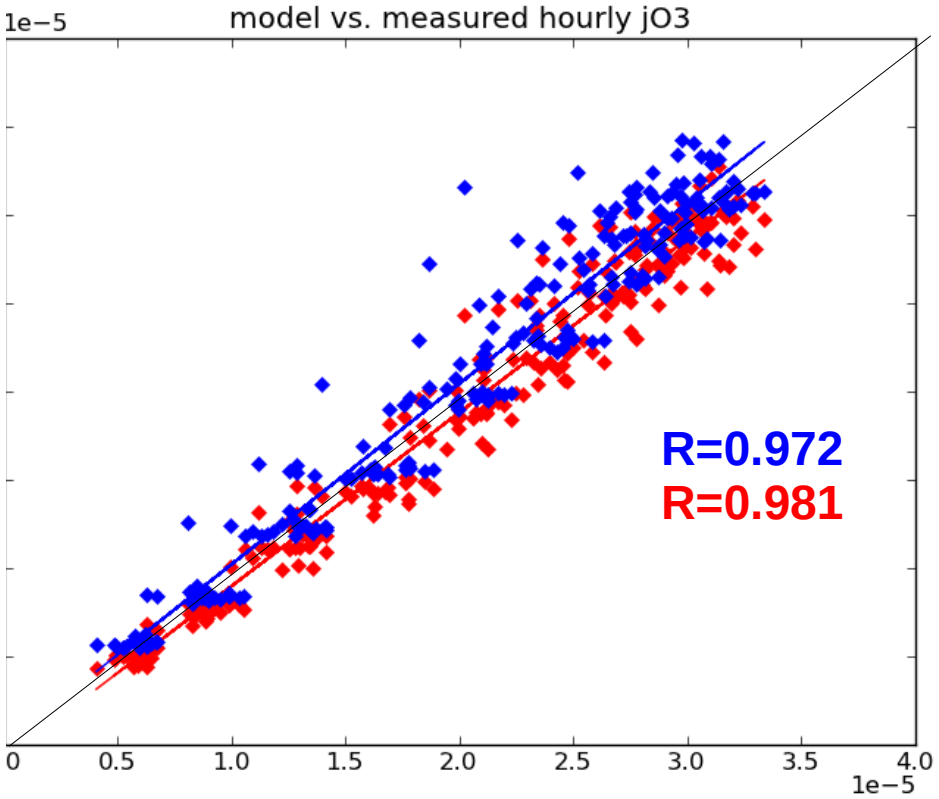
- Diurnal cycle is good represented well in the model, even though biases persist

- Possible causes of bias include the stratospheric ozone column

- Effect of the aerosols is very similar between model and observations

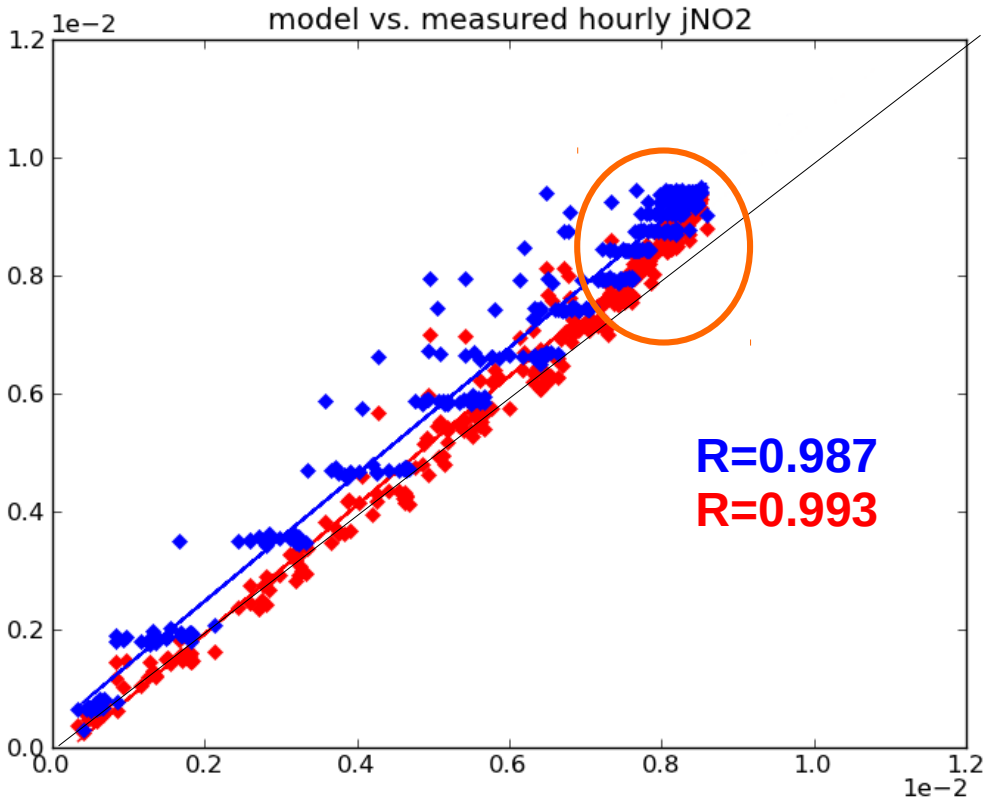
Effect of the aerosols on photolytic rates : The case of j(O1d)

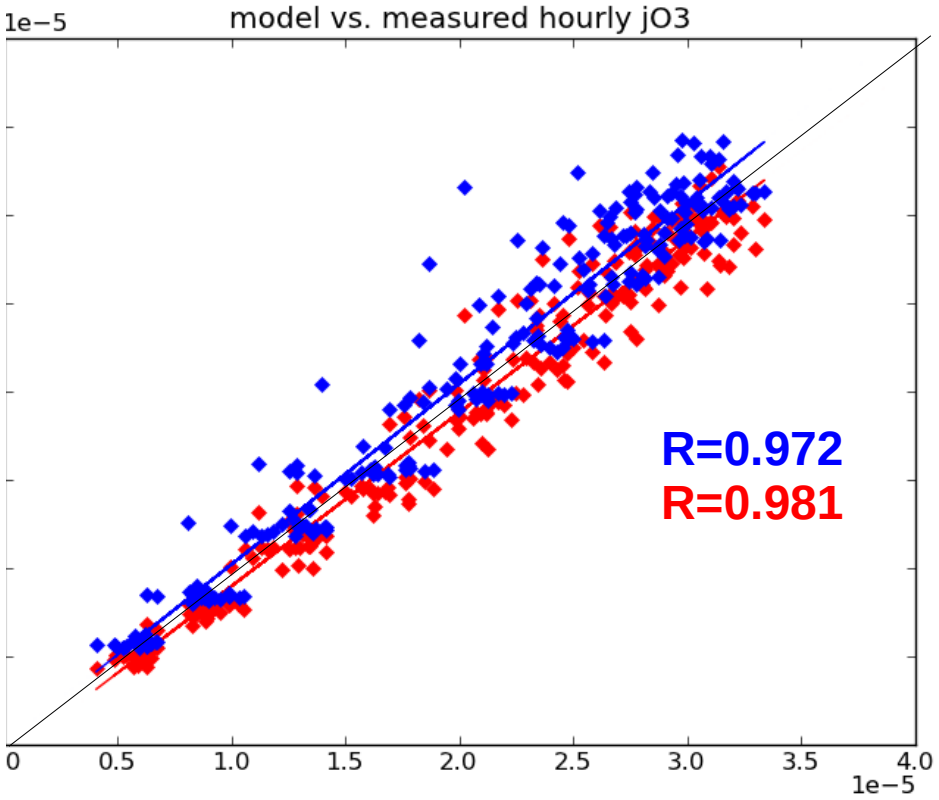




Excellent correlation rates and small biases reflect the excellent ability of the model to reproduce observed photolytic rates. Uff !

Effect of the aerosols on photolytic rates : The case of j(O1d)

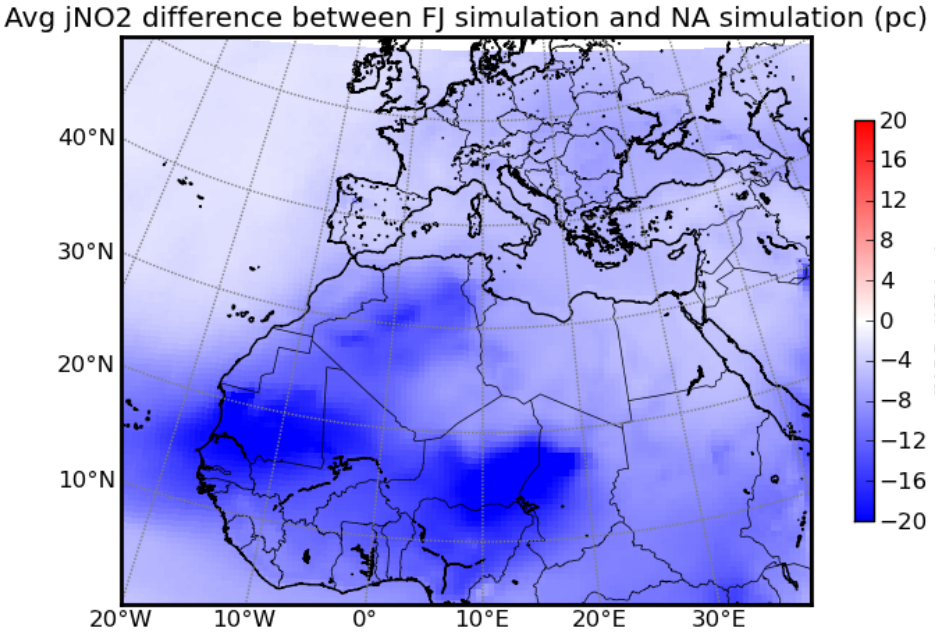


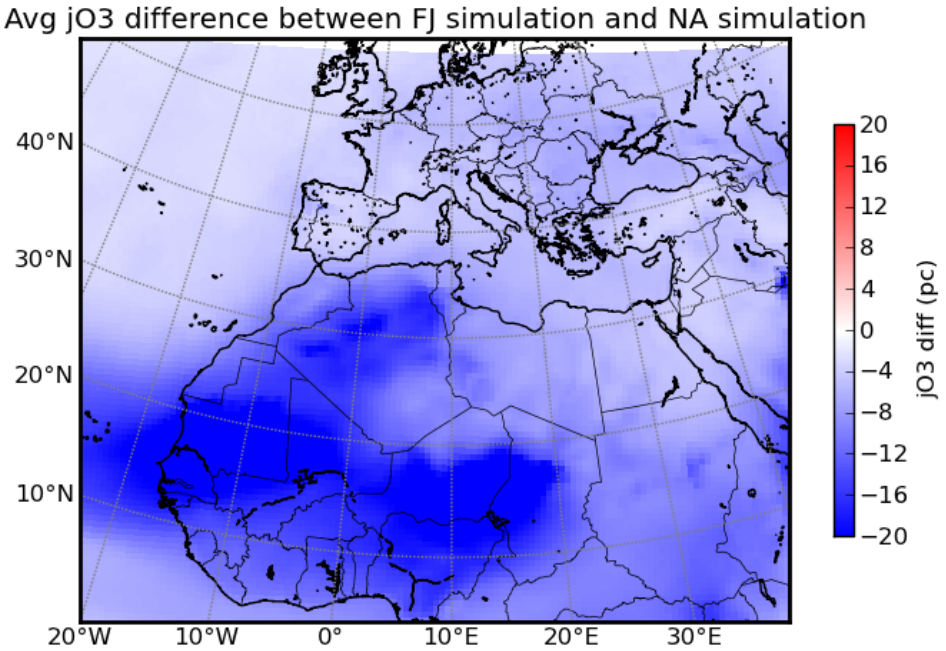


Excellent correlation rates and small biases reflect the excellent ability of the model to reproduce observed photolytic rates. Ouf !

Note the overestimation of jNO2 for small zenithal angles, systematic and very visible on the scatter plot. This problem does not exist for ozone.

Effect of the aerosols on photolytic rates : Where is it most important ?



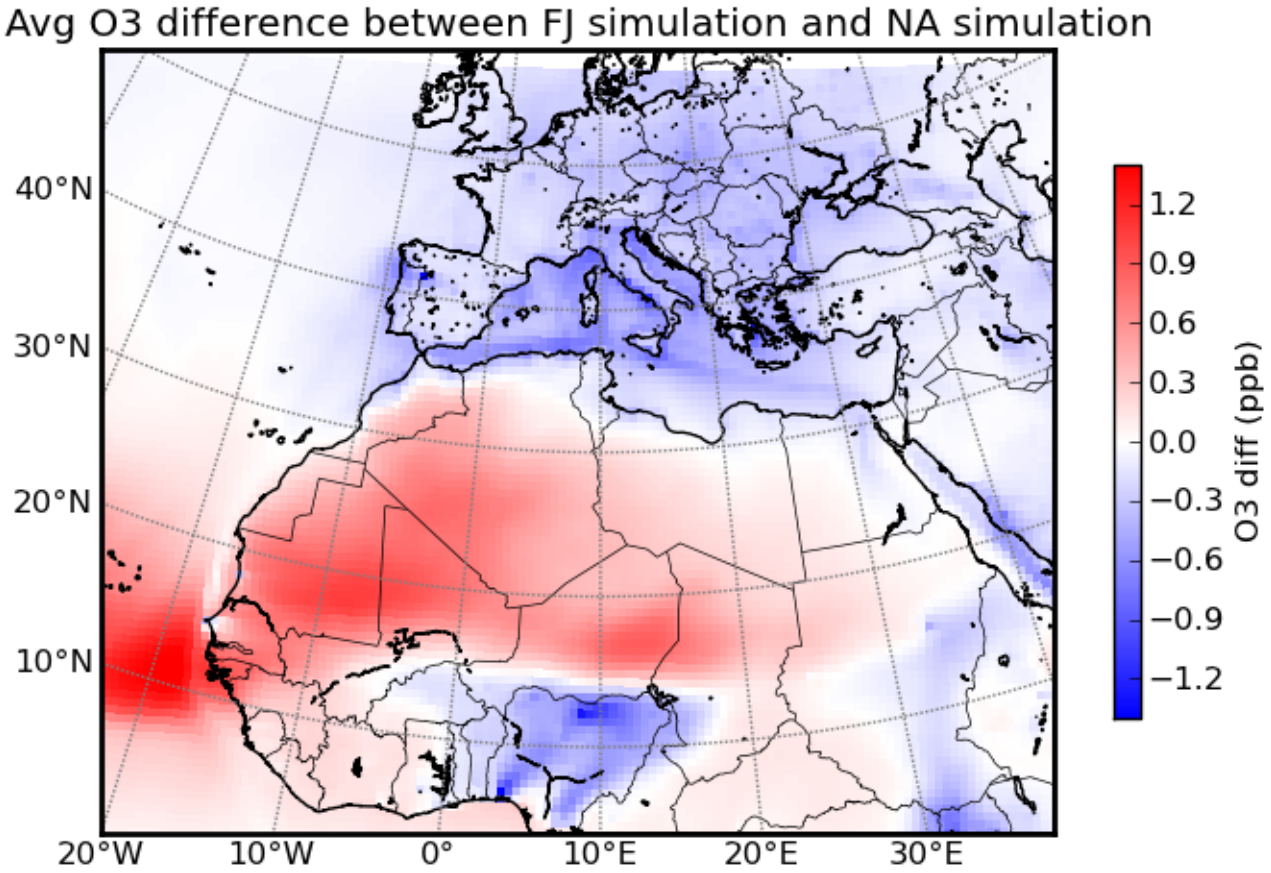


% difference in jNO2 (average)

% difference in j(O1d) (average)

Photolytic rates at ground level are systematically reduced by the effect of aerosols. This reduction is of more than 20 % close to source areas, and is stronger for j(O1d) than for j(NO2)

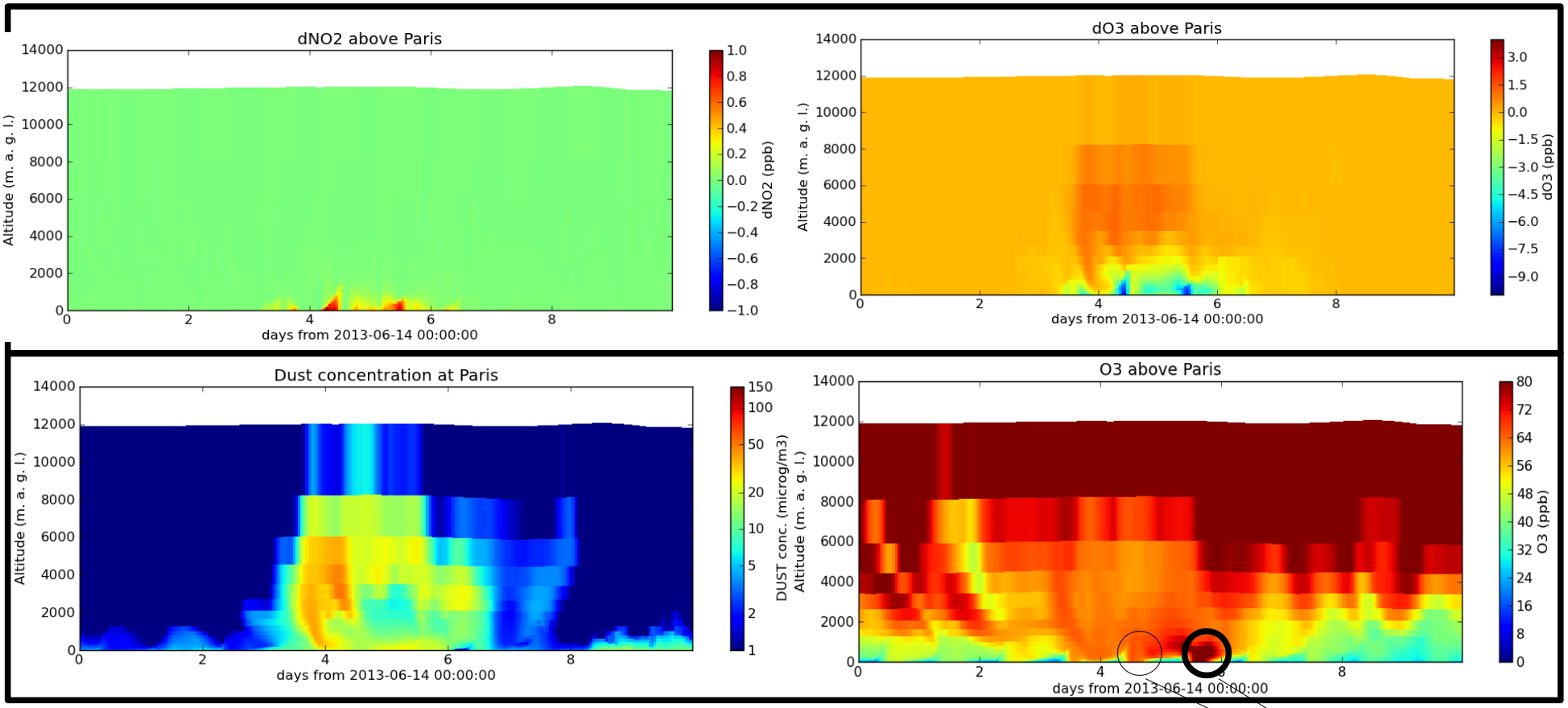
Possible effect of aerosols on ozone concentrations...
1. In average, weak effect ...



- Average effect is moderate, between -1.5 and 1.5 ppb

- The effect is either positive or negative depending on the area : Is it real ? Due to competition between the reduction in different photolytic rates ? Still to be investigated.

Possible effect of aerosols on ozone concentrations...
2. … but locally, possible strong effect



Increase or ozone concentration inside the dust layer
Reduction of ozone concentration in the boundary layer

Reduction of two strong ozone peaks by the dust radiative effect.

Conclusions

1. Model abilities :

CHIMERE is able to :

- Simulate atmospheric composition in the Mediterranean region (with some imperfections).

- Produce (relatively) good time series for the AOD

- Reproduce very well the photolytic rates at least for ozone and NO2

- Reproduce very well the day-to-day variations of jNO2 due to AOD

- Partly reproduce the day-to-day variations of j(O1D) due to AOD

2. Physical processes :

- The direct radiative impact of aerosols strongly reduces the photolytic rates.

- In the absence of clouds, it is almost the only driving factor of photolytic rates (except j(O1D))

3. Needs we indentified from this study, in terms of model development :

- Find a more realistic climatology for stratospheric ozone

- Find why dust concentrations at surface tend to be overestimated : vertical diffusion ? Sedimentation ?

Perspectives

1. Test and validate the photolytic rates in more complex situations, with multiple layers of clouds and aerosols.

2. Investigate more thoroughly the modulation of tropospheric chemistry : perform case study with intense photochemistry and strong aerosol load : summertime in a mediterranean megacity ? Need for a better horrizontal resolution.

3. Why is the effect of aerosols leading either to reduction or to increase of ozone concentrations ? By the way, does it happen in reality or just in the model?

Does it depend on the chemical regime ?

4. Use our model outputs to help treating the question that YOU are having in mind.

Validation of CHIMERE simulations against ADRIMED measurements, and application to the study of the aerosol direct effect on photochemistry in Lampedusa.

S. Mailler, 4th international workshop CHArMEx – ICTP, Trieste, 20-24 oct. 2014

Thank you for your attention !