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1 Introduction

This report presents the validation of LOTOS-EUROS v2.0. The new version is compared with v1.10.005 (the previous official release) and with EMEP observations.

Previous model versions were run and validated for 2006. For the new version, a new year (2012) has been chosen for validation. Firstly, the quality of input (meteorology, but also boundary conditions from MACC/Copernicus) has improved. In addition the observations network has changed for more recent year, with more stations with PM2.5 observations and monitoring of additional species (e.g. Na). We have also applied more strict criteria regarding quality and availability of observations (>75% coverage) in the validation.

In the following Chapters the model developments will be mentioned, then the simulation setup will be presented. For detailed process descriptions we refer to the Reference Guide (Manders et al 2016). Incremental changes due to various model developments have been tested and their impact is discussed.

First the surface concentrations of various species will be presented: annual mean surface concentration values of v2.0 and 1.10.005 are compared and time series at EMEP station locations are compared with observations. Appendix A presents statistics for a selection of stations. Also dry and wet deposition of SOx, NOy and NHx are presented, and wet deposition values are compared with monthly wet deposition observations. Finally an overall conclusion is given.
2 Changes

Several improvements have been made with respect to v1.10.005. Important changes with respect to this version are:

Functional changes
- Implementation of Land-sea mask based on GIS map.
- Division of sea spray aerosol (Na) and dust over 5 size classes to get a better differentiation of deposition velocities for the particles in different size bins. The artificial discrimination of deposition velocity for sodium over land and over sea that compensated for this was therefore removed.
- Use of snow cover from meteorological fields instead of being computed using snowfall and surface temperature (ECMWF, RACMO).
- A new dust emission parameterization was included based on a combination of new and existing parameterization. Dust emissions from arable land are excluded since variations in agricultural practices across Europe are too large, no uniformly applicable parameterization was developed yet.
- Roughness length for “desert” was changed to 0.013 (was 0.01). Local roughness length for dust emission parameterization was set to 0.0008.
- For arable land, the roughness length is made explicitly dependent on leaf area index. Outside growing season \( z_0 \) equals that of “desert”, when plants start to grow (May) it immediately increases to 0.10 (its previous default value). In fall the roughness length is set back to 0.013.
- Climatological boundary conditions from Logan and EMEP have been changed to include observed trends and monthly mean desert dust concentrations.
- A modification of the growing seasons was made to include equatorial region (LAI and SAI always maximum) and southern hemisphere (183 days shift w.r.t. northern hemisphere).
- A simple parameterization of NOx emissions from soils following Novak and Pierce (1993) was added as extra option.
- VBS scheme (optional) is modified to exclude terpene contribution since it is too uncertain.
- The structure of the meteorological input has changed from grib to netcdf format. Due to differences in resolution, in particular for clouds, this leads to small differences with respect to the use of the original grib files. In addition, calculation of relative humidity and soil moisture has been changed.
- Soil moisture is set to 1 in the code for grid cells with sea fraction >0.1, since in the original set of ECMWF meteorology that was used it was set to 0 (completely dry). This has led to spurious dust emissions in these cells. In the new ECMWF set the soil moisture for sea was correctly set to 1.
- Grib support is removed (for ECMWF meteo and MACC_r_ens boundary conditions).
- Exchange of concentrations between vertical layers has been updated.
- An optional extra reservoir layer has been added (5 layers).
Technical changes

- Rearrangement of structure for land-use dependencies. All land use dependent parameters were removed from the DEPAC routine and included in le_landuse_data.F90.
- Option to pack output data during simulations (less decimals stored).
- Checks on budget output consistency (check whether all necessary N and S tracers, accumulated tracers).
- Extra checks in rc-file for erroneous combinations of emissions, in particular outside Europe (glc+ trees).
- Add possibility to use single point source at user-defined location in rc-file for model testing.

Bug fixes

- Sedimentation velocity in lowest layer was not taken properly into account in mix2ground routine, now consistent with Seinfeld & Pandis.
- Dry deposition fluxes: sign of sedimentation flux for deposition budget.
- Cloud chemistry: redundant multiplication with cloud cover removed.
- Definitions of time and latitude swap in CIFS boundary conditions.
3 Test runs

The following simulation was performed for the validation:

V1.10.5  1-1-2012 to 1-1-2013
V2.0 1-1-2012 to 1-1-2013

Both simulations use MACC-III emissions, emissions of agricultural dust and road resuspension dust, MACC forest fires, biogenic VOC emissions, sea spray and wind-blown dust. V2.0 includes soil NOx emissions, which are not present in v1.10.5. Both simulations use climatological boundary conditions. The simulations use a different set of meteorological data. For v1.10.5, the GRIB data are used, in v2.0 the netCDF data are used. Both data sets stem from the same IFS cycle but are interpolated differently.

The figures show the annual mean surface level concentration of v2.0. for 2012 and a validation with the available EMEP stations and scatter plots of annual averages. For ozone, also the summer average daily maximum and daily maximum 8-hour average were calculated, as well as the indicators aot40 and somo35. Appendix A contains tables with temporal correlations (mean, bias, root mean square error, correlation) for a selection of EMEP stations. These selected stations were chosen such that they cover both ozone and PM10 and such that they represent different regions in Europe. For the component-wise scatter plots, all available EMEP stations for that component were included, with the restriction that the station should be below 700 m above sea level.
4 Results and discussion: concentrations

Main impacts of the changes are

- The change in roughness length for bare arable land during winter leads to lower deposition rates and higher concentrations for all species.
- The new land sea mask with separation of sea and inland water results in difference in sea salt emissions and in lower deposition velocities and higher concentrations of species over inland water.
- The differentiation in 5 size classes for sea spray and dust leads to overall larger transport distances due to a better differentiation of deposition velocities.
- The new meteorological data have a small but noticeable impact on all variables. In particular SO2 conversion to SO4 is affected due to slightly different cloud patterns, and slightly higher wind speeds over sea result in higher sea spray emissions.
- NOx emissions from soil lead to local small increases in NO (0.35ppb), NO2 (0.9ppb), HNO3 (0.07ppb), NO3a (0.4 μg/m^3), NH4 (0.1 μg/m^3) and SO2 (0.001ppb) concentrations, but a decrease in NH3 (0.1ppb), SO4 (up to 0.05 μg/m^3). For ozone a reduction is observed over northwestern Europe (0.5ppb), but an increase in southern Europe (order 1 ppb). Total PM2.5 and PM10 increase locally with 0.5 μg/m^3.
- The change in vertical mass exchange resulted in a small changes of surface concentration for several species.
- The two bug fixes in the deposition lead to an increase in concentration of coarse mode particles in surface level concentrations (not on model levels) and an increase of coarse mode mass in the deposition fields.

Now the results per component are discussed.

4.1 Ozone

Ozone concentrations have increased due to several factors. Soil NOx emissions have increased NOx concentrations and thus O3 concentrations. The largest impact comes from the use of the new meteorological data. The new set of meteorology has slightly different temperature and radiation values, leading to an increase of around 1.5 ppb over large parts of the domain, in particular when 5 layers are used. Slight differences in temperature have also affected the isoprene emissions and ozone formation rates. Also the improvement of exchange between the vertical layers has contributed to the general increase in concentration. The change in roughness length of arable land has changed deposition velocities and residence times of O3 and its precursors in winter. The net effect of all these small changes is however notable, with increases of up to 7 ppb. The spatial correlations have diminished slightly and for time correlations the pattern is mixed with some improvements and some reductions.
4.2 NO2, NO, NO3 aerosol

NOx and NO3 are affected by many processes. Including soil NOx emissions has increased the NO2 and NO3 concentrations whereas the increased sea spray concentrations (due to the new meteorology and changes in deposition velocity) and a bug fix in the mix2ground routine have contributed to the change in coarse mode NO3.
Furthermore, NO3_f is affected by the change in SO4_f concentrations due to differences in clouds, and in general concentrations are changed by the change in deposition velocity over arable land in winter. The net effect is a general small increase in concentration of NOx and a clear increase in NO3 aerosol concentrations, bringing them closer to the observed levels on most locations. There is one station where NO2 and NO concentrations are largely overestimated, this is the coastal location De Zilk in the Netherlands. This mainly due to an overestimation of the low concentrations. This could be related to the resolution of the model, the emission hotspot around Schiphol airport is in the same grid cell as this rural station. This feature indeed disappears when running the model at higher resolution.
4.3 NH3 and NH4

Ammonia concentrations have mainly increased due to the change in roughness length over arable land. Agricultural lands are often also the hotspots of NH3 emissions, leading to larger increases in these areas, and due to increased residence time also to slightly larger concentrations in remote areas. The changes lead to clearly higher concentrations, which bring most of the measurements closer to the observed levels. The increased NH3 concentrations are reflected in increased NH4 concentrations.
4.4 CO

For CO the results show a mixed pattern with small increases over large parts over domain, but decreases in Northern Italy and parts of Romania.

4.5 Isoprene

Isoprene emissions have changed slightly due to the different meteorology. In addition, changes in ozone due to the soil NOx contribution and changes in temperature-dependent conversion velocities due to the new meteorology contribute to changes. This leads to changes in isoprene concentrations.
4.6 Elemental carbon

Differences in ec distributions are mainly due to the change in roughness length and for ec also the fix in the mix2ground routine leads to an additional increase.

4.7 SO2 and SO4

SO2 and SO4 are affected more directly by the new meteorology than most other species since the different interpolation/higher resolution has resulted in slightly different cloud cover, to which SO2 to SO4 conversions are very sensitive. For SO2 there is an overall decrease in concentrations, for SO4_f concentrations increase in areas with high concentrations in Central Europe, but diminish slightly over the Atlantic. Coarse mode SO4_f has decreased nearly everywhere, but differences are very small. The spatial correlations have increased slightly.
4.8 Sodium

The use of 5 size classes leads to a slight increase in concentrations of total coarse sodium (<1 μg/m³ on annual basis) and a very small decrease in concentrations of total fine sodium (<0.01 μg/m³).

Sodium concentrations decrease everywhere, since inland lakes do no longer emit sodium (for example the large Swedish lakes).

The new meteorology leads to a small decrease in concentrations of the fine mode aerosol in all sea areas and a small increase in concentrations of the coarse mode aerosol over the Atlantic and a decrease over the Mediterranean. Sodium concentrations are slightly overestimated (were slightly underestimated in v1.10.5) and have a good spatial correlation.

Note that the changes in sodium concentrations also have an impact on the formation of coarse nitrate, mainly in the area close to the Strait of Gibraltar with intensive shipping.
4.9 Dust

Note that dust concentrations are artificially low in this simulation, since the inflow of Saharan dust through the southern boundary was not taken into account. Differences between v2.0 and v1.10.005 are locally quite large, mainly due to the reduction of spurious emissions due to the fixes in soil moisture for emissions at beaches. The impact of using 5 size classes is minor in this case, since dust concentrations are here dominated by anthropogenic emissions (road resuspension) which are still attributed to the original two size classes. Only for natural emissions these size classes apply.

4.10 PM2.5

The overall effect of all changes is an increase in PM2.5 concentrations over land and a slight reduction over sea. This reduces the overall bias without reducing the good spatial correlations. Time correlations at a selection of stations has improved slightly.
4.11 PM10

PM10 surface concentrations have increased substantially as a result of the model developments, bringing them closer to observed values. PM10 concentrations are still underestimated. In particular the higher concentrations have increased more, improving the slope of the linear fit. Time correlations at several stations show a mixed picture with in terms of improvement.
5 Results and discussion: deposition

In the figures for difference between dry deposition fields, care should be taken in the interpretation of the sign. LOTOS-EUROS has the convention in LOTOS-EUROS that dry deposition is negative so that NH3 re-emissions have a positive sign. For the difference plots of dry deposition positive values indicate that v1.10.5 has higher deposition values than v2.0. For wet deposition positive values indicate that v2.0 has higher deposition values.

5.1 Dry and wet SOx

Dry deposition of SOx has increased, wet deposition has increased with about the same amount. Differences at observation locations are relatively small. Inspection of time series shows that in particular high concentrations in spring were underestimated, as well as coincident peaks in NHx and NOy wet deposition (see corresponding sections). Spatial correlation of wet deposition with observations is very good, but with a severe underestimation.
5.2 Dry and wet NOy

NOy dry deposition has decreased nearly everywhere, but wet deposition has increased. The large increase in the Alps is probably related to differences in resolution of meteorology, combined with high NOy concentrations around the Po valley. Note the difference in scale between wet and dry deposition differences. Differences at observation locations are small, with a slight increase in values that were underestimated.
5.3 Dry and wet NHx

For NHx both wet and dry deposition have increased rather substantially in hot spot areas. This has brought modelled values closer to observed values. The spatial correlation of wet deposition values with observed values is good, the time correlation again shows the largest deficit in spring. This is probably due to seasonal variation in ammonia emission.
6 Conclusions

The net effect of functional changes and bug fixes has led to an overall the increase in concentrations of most species. For most species the new version has led to a reduction in bias, since model values were and are lower than the observed values. Only for ozone the values were in the right order of magnitude in the previous version (1.10.005) and tend to be slightly too high in the new version (v2.0). Spurious dust emissions from beaches have disappeared. Correlations in space and time have changed only little.

For rain water concentrations it was observed that all components show a peak in spring in the observations, that is not represented by the model. Modelled values during the rest of the year are in better agreement with the observations. When looking at annual averages, this leads to an overall underestimation by the model. This persistent feature is not specific for this version and needs further investigation. It may be related to growing seasons of vegetation and timing of emissions, in particular NH3.
7 Signature

Name and address of the principal

Names of the co-operators
R. Kranenburg MSc
Drs. C. Hendriks

Date upon which, or period in which the research took place
January–June 2016

Name and signature reviewer

Prof. Dr. M. Schaap

Signature:  Release:

Dr. A.M.M. Manders-Groot
Project leader

Ir. R.A.W. Albers MPA
Research Manager
Appendix A: Statistics for a selection of EMEP stations

Table A1. O₃ 8h (maximum all)

<table>
<thead>
<tr>
<th>station</th>
<th>Obs mean</th>
<th>Bias 2.0</th>
<th>Rmse 2.0</th>
<th>Corr 2.0</th>
<th>Bias 1.10.5</th>
<th>Rmse 1.10.5</th>
<th>Corr 1.10.5</th>
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<tr>
<td>Neuglobsow</td>
<td>66.545</td>
<td>9.067</td>
<td>20.068</td>
<td>0.724</td>
<td>1.004</td>
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<td>Vredepeel</td>
<td>59.051</td>
<td>3.512</td>
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<td>0.834</td>
<td>-2.044</td>
<td>14.822</td>
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<tr>
<td>Ispra</td>
<td>80.024</td>
<td>6.589</td>
<td>21.809</td>
<td>0.878</td>
<td>0.264</td>
<td>19.329</td>
<td>0.884</td>
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<tr>
<td>K-Puszta</td>
<td>90.251</td>
<td>-0.823</td>
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<td>Els Torms</td>
<td>87.291</td>
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<td>64.911</td>
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Table 2 Ozone 8h maximum summer

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<th>Rmse 1.10.5</th>
<th>Corr 1.10.5</th>
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<td>Neuglobsow</td>
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<td>Ispra</td>
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<td>K-Puszta</td>
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<td>76.667</td>
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Table A3. Daily correlations PM₁₀

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<th>Rmse 2.0</th>
<th>Corr 2.0</th>
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<th>Rmse 1.10.5</th>
<th>Corr 1.10.5</th>
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<td>Neuglobsow</td>
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<td>Vredepeel</td>
<td>23.863</td>
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Table A4. Daily correlations PM₂₅

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<th>Obs mean</th>
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<th>Rmse2.0</th>
<th>Corr2.0</th>
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<th>Rmse1.10.5</th>
<th>Corr1.10.5</th>
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<tr>
<td>Neuglobsow</td>
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<td>-4.281</td>
<td>8.239</td>
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<tr>
<td>Ispra</td>
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<td>Vredepeel</td>
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<td>Els Torms</td>
<td>7.901</td>
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<td>7.857</td>
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### Table A5. Daily correlations NO$_3^-$

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<th>Rmse 2.0</th>
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<th>Rmse 1.10.5</th>
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<td>Ispra(PM2.5)</td>
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<td>1.000</td>
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<td>0.292</td>
<td>0.083</td>
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<td>1.011</td>
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### Table A6. Daily correlations NH$_4^+$

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<th>Corr 2.0</th>
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<th>Rmse 1.10.5</th>
<th>Corr 1.10.5</th>
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<td>Ispra(PM2.5)</td>
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<td>-0.529</td>
<td>2.922</td>
<td>0.320</td>
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<td>Vredepeel</td>
<td>2.432</td>
<td>-0.642</td>
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<td>-0.927</td>
<td>1.981</td>
<td>0.798</td>
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<td>K-Puszta</td>
<td>1.316</td>
<td>0.043</td>
<td>1.164</td>
<td>0.581</td>
<td>-0.259</td>
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<td>0.611</td>
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<td>Els Torms</td>
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### Table A7. Daily correlations SO$_4^{2-}$

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