# SPECIAL PROJECT FINAL REPORT

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| **Project Title:** | Monitoring Atmospheric Composition and Climate - Phase 3 (MACC-III) |
| **Computer Project Account:** | sp DEFRIU |
| **Start Year - End Year :** | 2014 - 2017 |
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## Summary of project objectives

The overall objective of MACC-III (Monitoring Atmospheric Composition and Climate – Phase 3) was to function as the bridge between the project MACC-II and the Atmosphere Service envisaged to form part of Copernicus Operations. MACC-III has sustained the pre-operational atmosphere activities developed in MACC-II in order to avoid any interruption in the critical handover phase between the pre-operational and fully operational service. In October 2015 MACC-III went over to the fully operational Copernicus Atmosphere Monitoring Service (CAMS). CAMS provides continuous data and information on atmospheric composition. The service describes the current situation, forecasts the situation a few days ahead, and analyses consistently retrospective data records for recent years. CAMS has been developed to support policymakers, business and citizens with enhanced atmospheric environmental information. The Rhenish Institute for Environmental Research at the University of Cologne (RIUUK) plays an active role in subproject CAMS_50.

## Summary of problems encountered

End of 2014 we run out of computing time due to the use of a computational demanding module for the computation of biogenic emissions for the prototype operational air quality forecast.

## Experience with the Special Project framework

We are satisfied with the application procedure and with the requirements on project reporting.

## Summary of results

The scientific plan of this special project is fully based on the concepts of MACC-III and CAMS_50, as it is detailed in the corresponding Descriptions of Work in more comprehensive terms.
In MACC-III RIUUK was leading subproject EDA (*Data assimilation for European air quality*) and took an active part in sub-projects ENS (*Ensemble air quality forecasts for Europe*), and EVA (*Validated Assessments of air quality for Europe*). In CAMS_50 RIUUK is engaged in sub-projects 50.2 (*Continuous Model Development)* and 50.3 *(Distributed Regional Production)*.

The delivery of the European-scale air quality data is based upon a geographically distributed ensemble of 7 individual air quality models under the lead of Meteo France. RIUUK provides a member of this ensemble with its comprehensive chemistry transport model EURAD-IM (Elbern et al., 2007).
Three data streams are provided in CAMS_50:

- on a daily basis, hourly analyses for the previous day and forecasts up to + 96 h;
- with a delay of a few weeks (in order to maximize the number of observations) interim re-analyses are produced daily with systems frozen in their configuration of January 1st every year;
- with a delay of up to 2 years (due to the delay in getting fully validated data), re-analyses are processed with frozen systems, which are only updated every few years.

The provision of interim re-analyses is carried out in addition to the service established in MACC.
An important component of MACC-III and CAMS_50 is the further development of the individual air quality forecast models and of the data assimilation systems. Especially in MACC-III the principal objective of sub-project EDA was to assure increasing and skilful use of new trace gas and aerosol measurements or retrievals, within a scenario of changing earth observation data compositions, retrieval versions and model configurations. Furthermore EDA aimed to provide for validity of the data assimilation in case of extraordinary geophysical or atmospheric events.

## 1. Summary of development activities in MACC-III and CAMS_50

### 1.1 IASI $O_3$ and CO data assimilation

IASI is an infrared Fourier transform spectrometer developed jointly by CNES (the French spatial agency), and by EUMETSAT. IASI is mounted on-board the European polar-orbiting MetOp satellite. It contributes to atmospheric composition measurements for climate and chemistry applications with high horizontal resolution and sampling, and with 1 km vertical resolution (Clerbaux et al., 2009). To reach this objective, IASI measures the infrared radiation of the Earths surface and of the atmosphere between 645 and 2760 $cm^{-1}$ at nadir and along a 2200 km swath perpendicular to the satellite track. A total of 120 views are collected over the swath, divided as 30 arrays of 4 individual Field-of-views varying in size from $36 \times \pi$ $km^2$ at nadir (circular 12 km diameter pixel) to $10 \times 20 \times \pi$ $km^2$ at the larger viewing angle. IASI offers in this standard observing mode global coverage twice daily, with overpass times at around 9:30 and 21:30 mean local solar time.

An observation operator and its adjoint for FORLI (Fast Optimal Retrievals on Layers for IASI) (Hurtmans et al., 2012) $O_3$ columns have been developed and integrated in the EURAD-IM assimilation system. FORLI provides partial $O_3$ columns for 19 atmospheric layers. Levels 1 to 18 correspond to 18 atmospheric layers with 1 km thickness, level 19 to the layer from 18 to 60 km. The observation operator computes model equivalents of $O_3$ partial columns for these layers between the surface and the model top at 100 hPa.

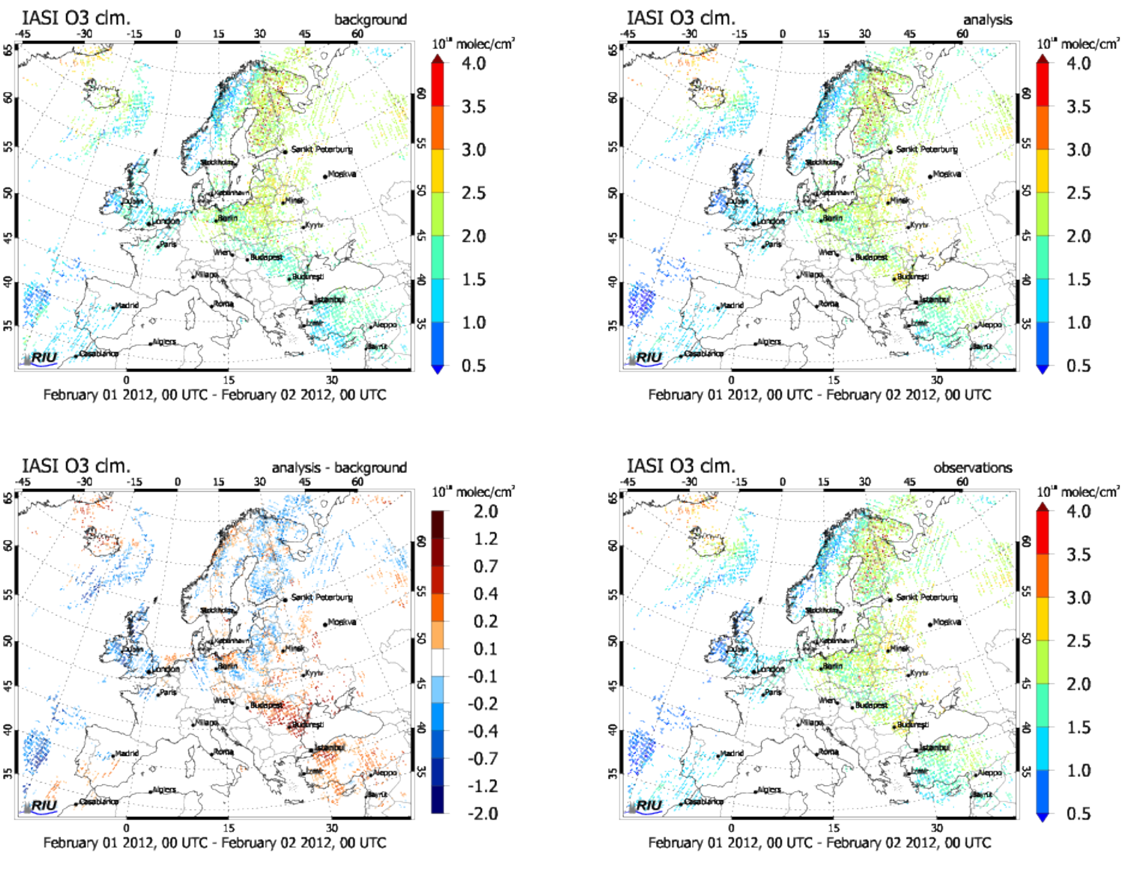



*Figure 1: FORLI partial O3 columns integrated between the surface and 100 hPa for February 1, 2012. Upper left: EURAD-IM forecast without data assimilation, upper right: EURAD-IM analysis, lower left: differences between analysis and forecast, lower right: observations.*

The 3d-var assimilation of FORLI $O_3$ data was tested for two episode types, an episode with high Ozone concentrations from July 7 - 16, 2010 and a case with high particulate matter concentrations from January 16 to February 3, 2012. Figure 1 shows FORLI partial $O_3$ columns for 1. February 2012. The assimilation of IASI $O_3$ data was independently validated with Ozonesonde measurements. Figure 2 shows Ozone profiles at Uccle for 27. January 2012. The assimilation of

FORLI $O_3$ data has clearly improved the skill of EURAD-IM to simulate upper tropospheric Ozone concentrations. The improvement of near-surface concentrations originates from the assimilation of surface in situ measurements.

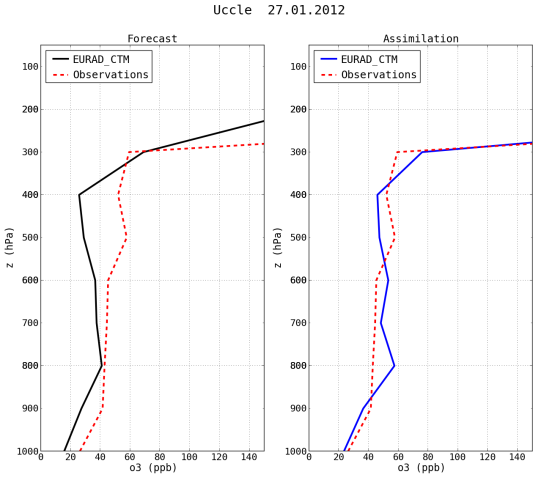



*Figure 2: Ozone profiles at Uccle for January 27, 2012. Black: EURAD-IM forecast, blue: EURAD-IM analysis, red: observations.*

An observation operator for FORLI CO data and its adjoint has been developed and was integrated in the EURAD-IM assimilation system. In a first step, the observation operator computes CO partial columns for all FORLI layers between the surface and the model top at 100 hPa. Finally, the FORLI averaging kernel vector for the partial columns is applied to compute the model equivalent. Figure 3 shows results from a 3d-var assimilation experiment of IASI FORLI-CO data for March 25, 2013. The EURAD-IM background CO columns do not have a general bias: at March 25 the CO column content was rather over estimated in north-east Europe and under estimated in south-east Europe. The bias was significantly reduced in the analysis run.

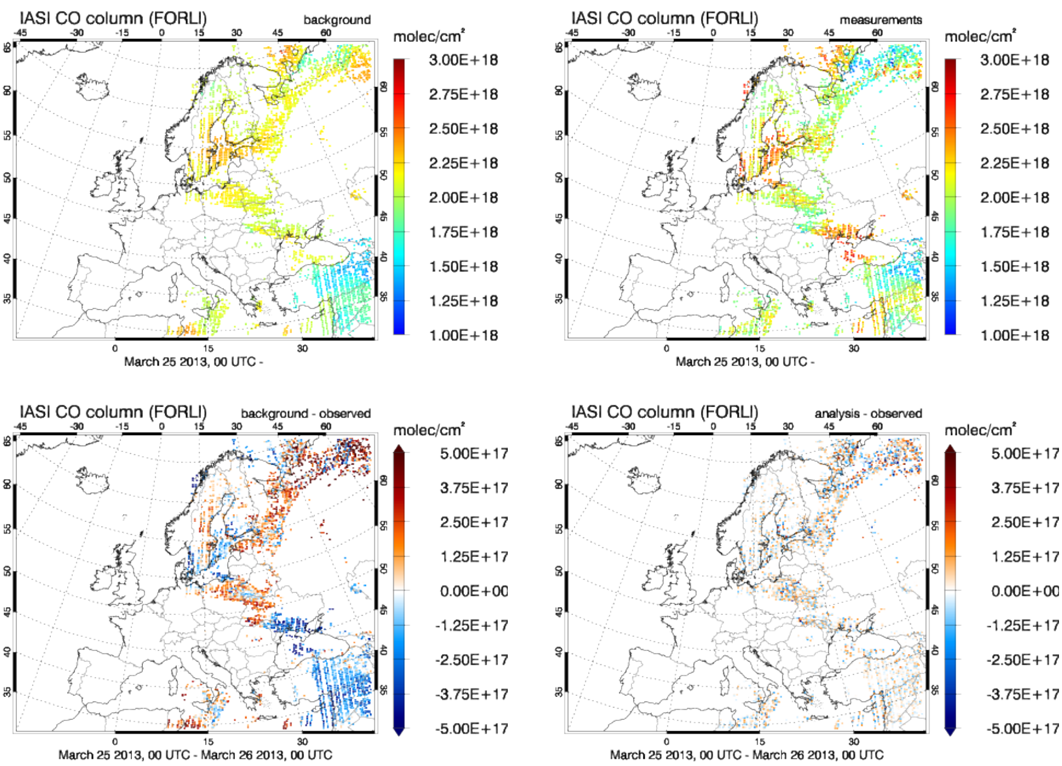



*Figure 3: Atmospheric CO columns (below 100 hPa) for March 25, 2013. Upper left: background model equivalents, upper right: IASI FORLI-CO data, lower left: background minus measurements, lower right: analysis minus measurements.*

The assimilation has been independently validated with surface in situ data from the eReporting database of the European Environmental Agency. Results are depicted in Figure 4 for the Period May 1 to 14, 2013. The large negative EURAD-IM CO bias in the near surface layer has been slightly decreased due to the assimilation of IASI CO column retrievals. In May 2013 most of the retrieval data was located over Eastern Europe. Because surface in situ CO measurements for this area are very sparse, the impact of IASI CO column assimilation on the near surface CO concentration is potentially under estimated in the time-series shown.

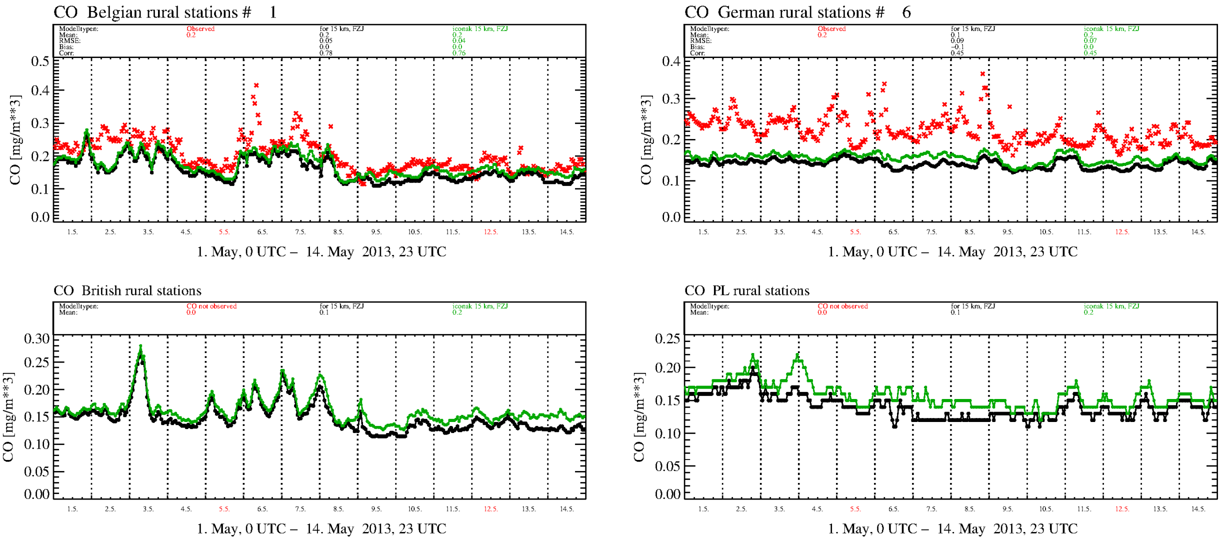



*Figure 4: CO time-series averaged over all available background stations from the eReporting database of the European Environmental Agency (EEA) for May 1 to May 14, 2013. Upper left: Belgian stations, upper right: German stations, lower left: UK Stations, lower right: Polish stations. Black: EURAD-IM forecast without any data assimilation, green: EURAD-IM analysis of LATMOS FORLI CO column retrievals, red: observations.*

**1.2 Assimilation of MODIS AOD measurements**

The EURAD-IM assimilation system has been extended by an observation operator for the aerosol optical depth at 550 nm. Aerosol particles are treated as internally mixed. Refractive indices of the individual aerosol species (Schroedter-Homscheidt, 2010) are averaged according to their weight fraction. Inorganic aqueous species are merged to one component whose refractive index is computed with the Biermann (2000) mixing rule. A fast approximation according to Evans and Fournier (1990) in combination with a 50 point Gauss-Legendre quadrature is used for the calculation and integration of the extinction efficiency with respect to particle size.

The observation operator was applied on MODIS AOD measurements in a 3d-var assimilation experiment. Figure 5 shows model equivalents of the aerosol optical depth at 550 nm for March 24, 2013. The assimilation has strongly improved high AOD values measured over the Mediterranean Sea south of Italy, which most probably originate from a Sahara dust outbreak. In some areas e.g. over the Baltic Sea AOD values are over estimated in the analysis. We suppose that this over estimation is an implication of the truncation of the aerosol water gradient. The consideration of aerosol water is necessary for the construction of an accurate adjoint AOD observation operator. However, in the assimilation procedure the aerosol water gradient is set to zero because aerosol water is currently not a prognostic variable of the EURAD-IM aerosol module.

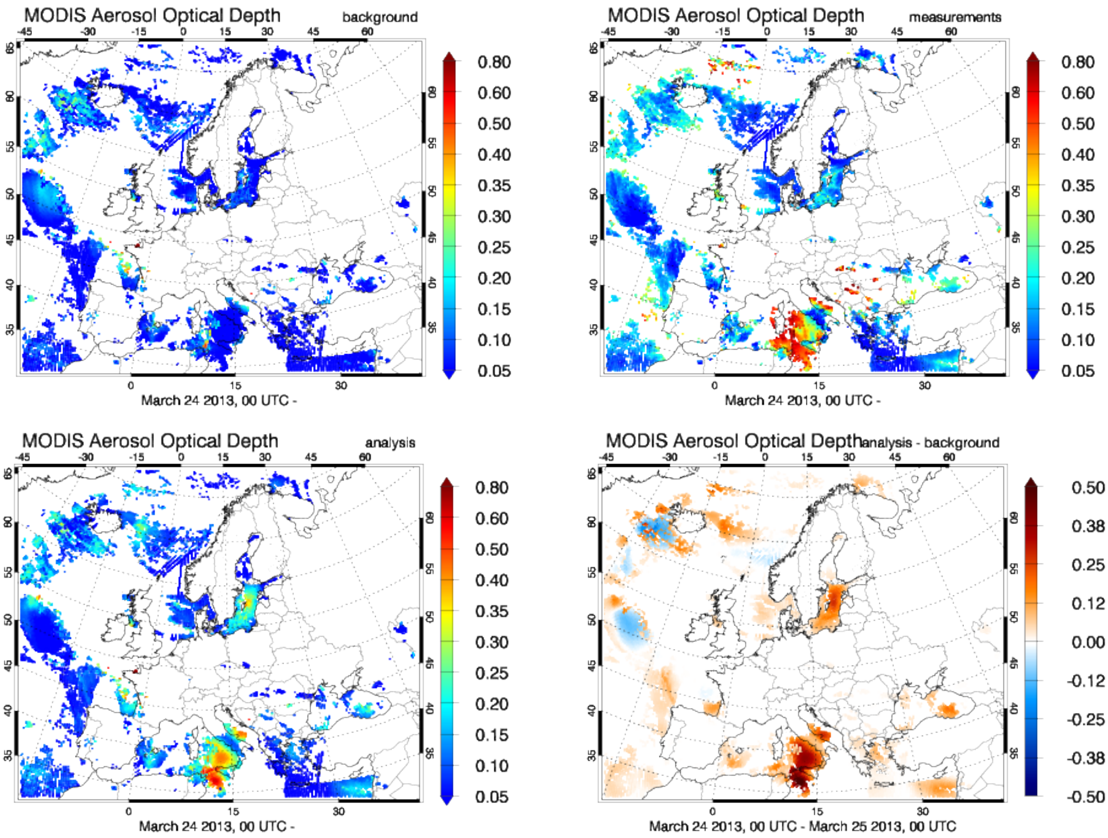


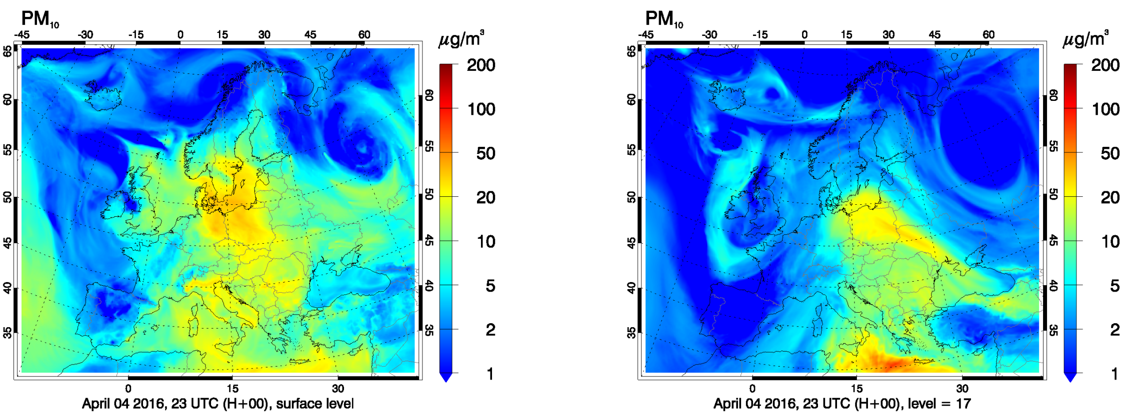

*Figure 5: AOD at 550 nm for March 24, 2013. Upper left: background model equivalent, upper right: MODIS measurements, lower left: analysis model equivalent, lower right: analysis minus background.*

**1.3 Further development of the EURAD-IM mineral dust forecast**

In case of Sahara dust outbreaks towards Europe, mineral dust concentrations are probably under estimated by the EURAD-IM forecasting system. The aim of this activity was an improvement of the EURAD-IM skills in such situations. The impact of two measures on the EURAD-IM Sahara dust forecast has been investigated.

*1.3.1 Activation of the EURAD-IM mineral dust emission module*

DREAM (Nickovic et al., 2001) is used in the EURAD-IM CTM for the computation of the mineral dust emission source strength. Figure 6 shows a comparison of $PM_{10}$ concentrations obtained from forecasts with EURAD-IM in the current operational configuration and with DREAM enabled.



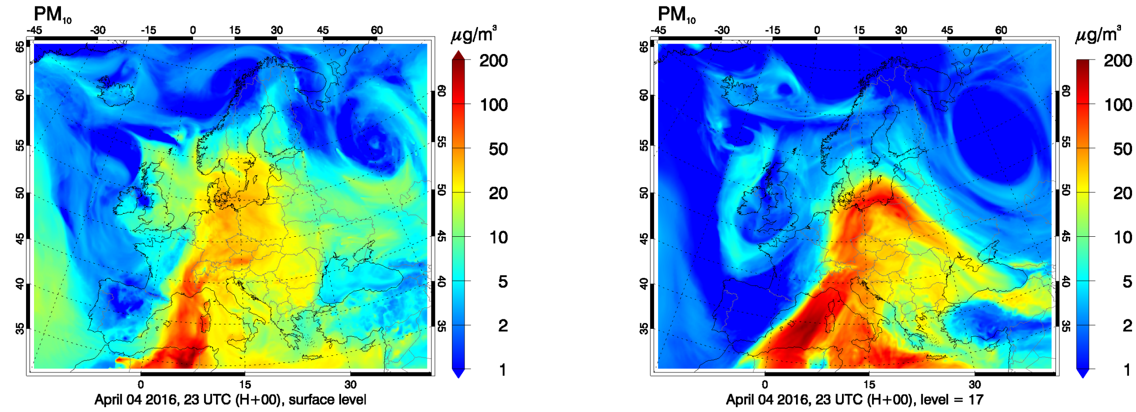



*Figure 6: $PM_{10}$ concentration at April 4, 2016. Upper panel: EURAD-IM forecast in the current operational configuration, lower panel: EURAD-IM forecast with the additional application of mineral dust emissions from DREAM. Left: surface level, right: level 17, approximately 3000 m height.*

The performance of DREAM in EURAD-IM forecasts has been evaluated for two mineral dust episodes: from April 25 to Mai 7, 2013 and from March 29 to April 7, 2016. The evaluation shows that the application of DREAM significantly reduces the negative bias of EURAD-IM $PM_{10}$ forecasts during mineral dust events, compared to the current operational configuration of the EURAD-IM forecast without mineral dust emissions from DREAM. This is depicted in terms of averaged $PM_{10}$ and $PM_{2.5}$ time-series in Figures 7 and 8. At least for the period in 2013, $PM_{2.5}$ is less influenced by mineral dust events than $PM_{10}$ in the model simulations. May be this is a result of the representation of mineral dust in the EURAD-IM CTM by coarse mode particles only. Due to the large-scale nature of mineral dust events, the application of DREAM could improve the aerosol forecast independent of station characteristics.

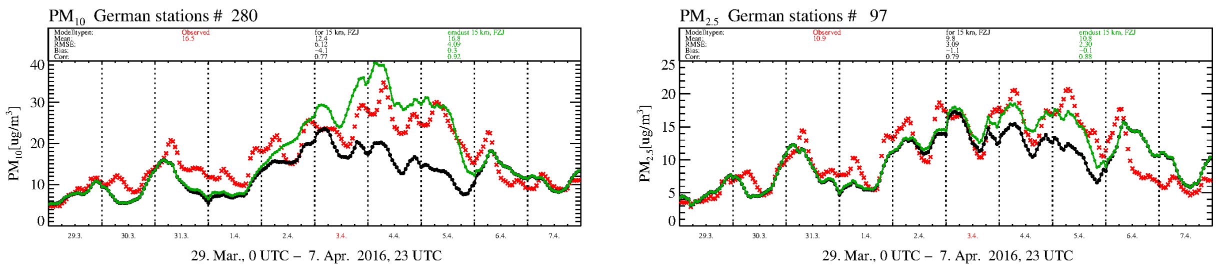



*Figure 7: $PM_{10}$ (left) and $PM_{2.5}$ (right) time-series averaged over stations from the German Umweltbundesamt for March 29 to April 7, 2016. Black: EURAD-IM forecast in the current operational configuration (aerosol boundary values from the global C-IFS forecast applied), green: EURAD-IM forecast with the additional application of mineral dust emissions from DREAM, red: observations.*

### *1.3.2 Assimilation of MODIS AOD retrievals to improve PM forecasts*

For the mineral dust episode from April 25 to Mai 7, 2013 the impact of an AOD analysis on the forecast performance has been investigated. For this episode the MODIS Aerosol Optical Depth Land and Ocean of MOD04 L2 retrieval from the Terra and Aqua satellites has been assimilated with EURAD-IM in the configuration, which is currently used for the operational analysis (hourly 3d-var). Results from this analysis were taken as initial values for a forecast with mineral dust emissions from DREAM enabled. Generally, the application of initial values from the AOD analysis can further improve the EURAD-IM performance during mineral dust events (see Figure 8).

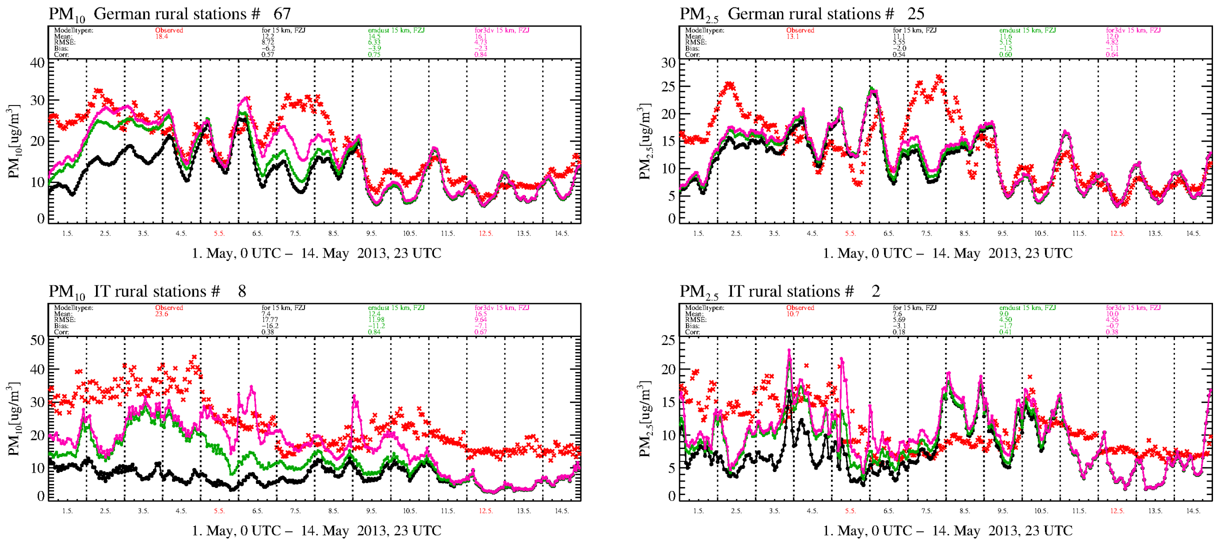



*Figure 8: $PM_{10}$ (left) and $PM_{2.5}$ time-series averaged over available stations from the eReporting database of the European Environmental Agency (EEA) for May 1 to May 14, 2013. Upper panel: German stations, lower panel: Italian Stations. Black: EURAD-IM forecast in the current operational configuration (aerosol boundary values from the global C-IFS forecast applied), green: EURAD-IM forecast with the additional application of mineral dust emissions from DREAM, magenta: EURAD-IM forecast initialised with an AOD analysis, mineral dust emissions from DREAM applied, red: observations.*

### *1.3.3 Investigation of the impact of accumulation mode mineral dust on the aerosol forecast*

In September 2016 the implementation of an additional mineral dust variable for the accumulation mode of the modal representation of the aerosol size distribution used in EURAD-IM has been completed. The investigation of the impact of the additional accumulation mode mineral dust variable on the EURAD-IM aerosol forecast has been introduced to the integrated action "Investigating aerosol episodes using quality modeling with data assimilation and data fusion techniques" between RIUUK and CESAM & Department of Environment and Planning at the University of Aveiro, Portugal.

The dust episode from April 20 to Mai 10, 2013 was simulated with EURAD-IM with climatological boundary values and without anthropogenic emissions. Two sensitivity runs were set up: the first with 90% of the emitted mineral dust mass assigned to the coarse mode and 10% assigned to the accumulation mode, the second run with 95% assigned to the coarse mode an 5% assigned to the accumulation mode. Figure 9 shows AOD time series for three AERONET measurement sites. As expected from Mie scattering theory, the time series show that the AOD is slightly larger, if the mineral dust load in the accumulation mode is larger.

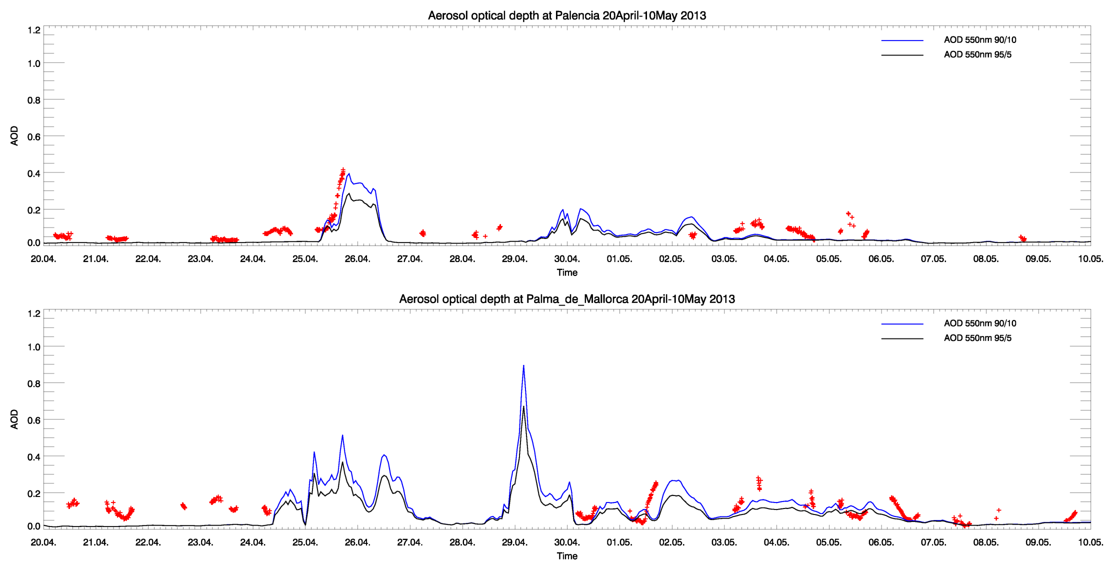


*Figure 9: AOD time series at two measurement sites of the Aerosol Robotic Network (AERONET) for April 20 to Mai 10, 2013. Blue: EURAD-IM forecast with 90% of emitted mineral dust mass assigned to the coarse mode of the aerosol size distribution and 10% of emitted mass assigned to the accumulation mode, black: EURAD-IM forecast with 95% of emitted mineral dust assigned to the coarse mode and 5% assigned to the accumulation mode, red: AOD measurements.*

Because of the longer atmospheric life time of aerosol particles in the accumulation mode, one would expect that the total mineral dust concentration is higher in the model run with a larger fraction of emitted mineral dust assigned to the accumulation mode. This is surprisingly not the case. The differences in total mineral dust between the two model runs is very low and does not show a specific sign (see figure 10).

We do not intend to change the current practice in EURAD-IM to assign only 5% of the emitted mineral dust to the accumulation mode.

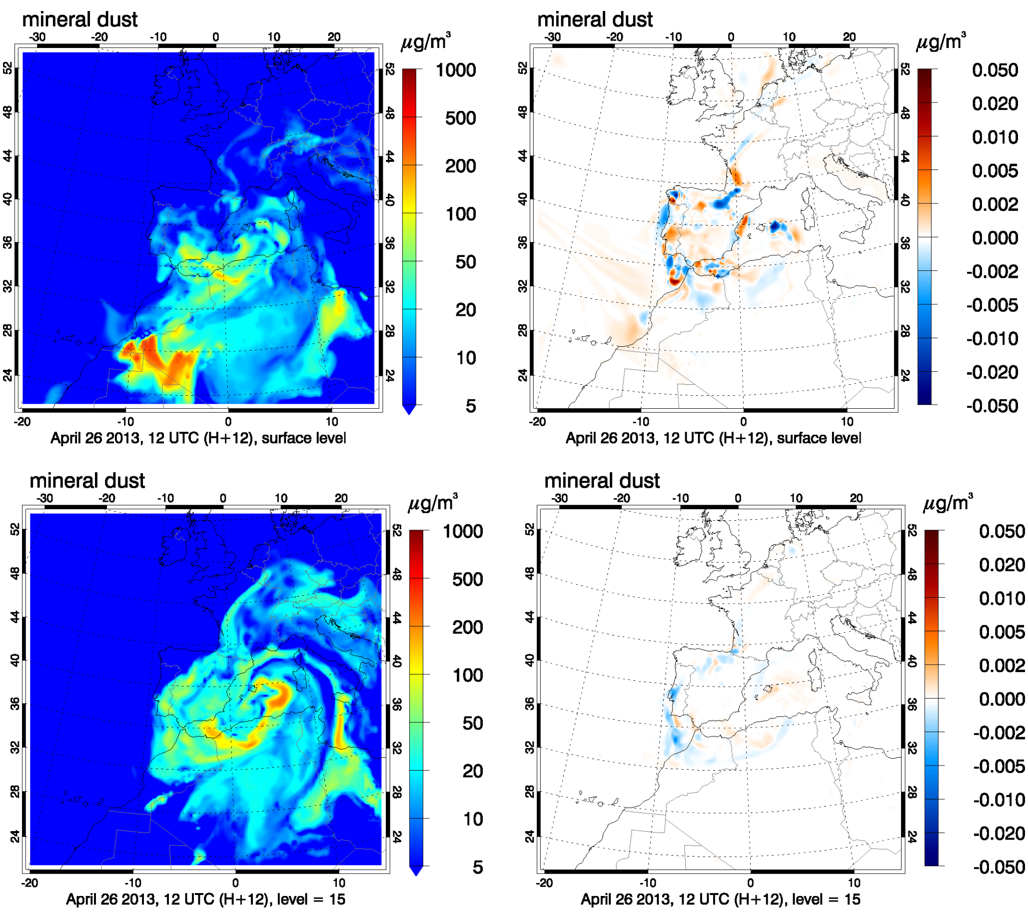



*Figure 10: Left: EURAD-IM mineral dust forecast for April 26, 2014 at 12:00 UTC with 90% of emitted mineral dust mass assigned to the coarse mode and 10% assigned to the accumulation mode at the near surface layer (above) and at model layer 15 (about 2000m) (below). Right: Difference between this forecast and a sensitivity run with 95% of emitted mineral dust assigned to the coarse mode and 5% assigned to the accumulation mode.*

**1.4 Volcanic emission data assimilation**

To improve volcanic ash and gas emission dispersion forecasts, we performed a 3d variational data assimilation study, using vertically highly resolved lidar (light detection and ranging) observations from NASA's CALIPSO satellite. Therefore, we developed a corresponding observation operator that maps the modeled quantity into the observation space. This includes in detail a look-up-table approach whereas the radiative transfer of the lidar signal is simulated based on Mie-theory (Fast et al., 2006; Wiscombe, 1980) and the lidar equation according to Huneeus u. Boucher (2007). A case study regarding the ash cloud of the Eyjafjallajökull eruption in April 2010 over Western Europe is accomplished, discussed, and published in Wilkins et al. 2016 (see Fig. 6).

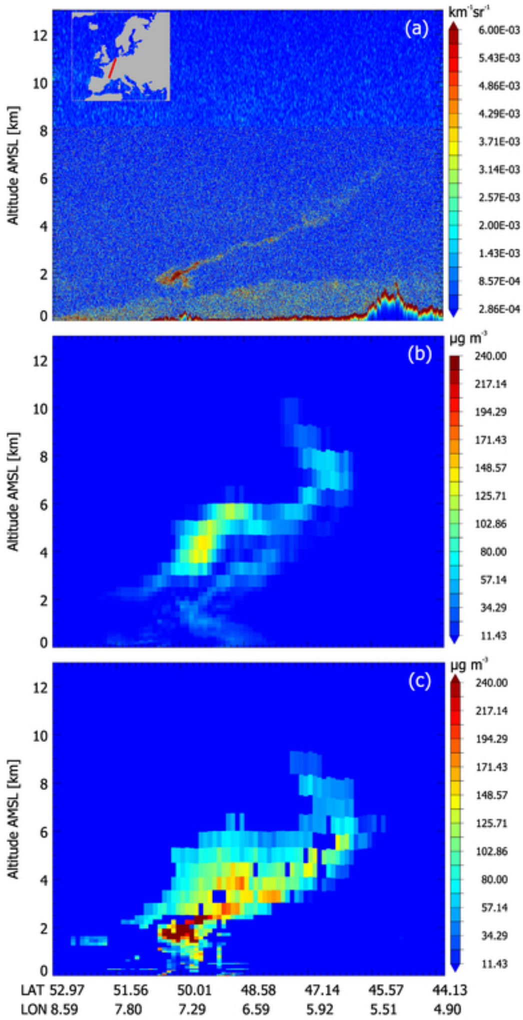



*Figure 6: (a) Vertical cross section of the CALIOP total attenuated backscatter signal (532 nm) on 17th April 2010, on its flight path indicated in the European map in the upper left corner. (b) Vertical cross section of the a priori volcanic ash concentration by EURAD-IM for the same time and location as in (a). (c) Analysis of the volcanic ash concentration corresponding to the observations (a) and the a priori concentrations.*

Further research activities are related to the last eruption of the Bardarbunga/Iceland (Aug. 2014 to Feb. 2015), which was characterized by a strong release of $SO_2$ into the lower troposphere while ash emissions were negligible. Hence, we aim to assimilate $SO_2$ concentrations in terms of vertical column densities observed by polar orbiting satellites such as GOME-2 and OMI. As a first case study a particular episode (Sep. 2014) with extremely high $SO_2$ concentrations recorded at several European measurement sides at ground level is considered. EURAD-IM is able to predict these high concentrations quite accurately looking upon forward simulations with roughly estimated emission parameters (see Figure 7). Furthermore, the forecast results indicate that the unusual high $SO_2$ values, which were recorded at several European stations, are due to the transport of $SO_2$ rich air from Holuhraun towards continental Europe. However simulated concentrations stay in many cases below measurements of ground stations. The application of the EURAD-IM 4D-var system based on satellite observed $SO_2$ concentrations may improve these results.

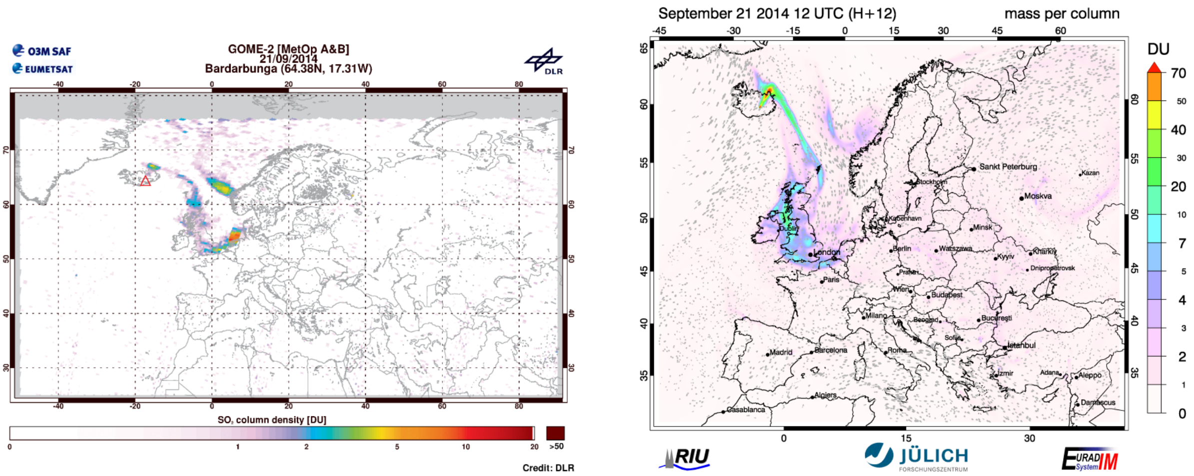



*Figure 57 $SO_2$ vertical column density, 21.09.2014. Left: GOME-2 measurements (source: DLR), right: EURAD-IM forecast at 12:00 UTC.*

## 2. Ensemble Air-Quality Forecasts for Europe

This activity focuses on the delivery and verification of the operational European-scale regional NRT air-quality services. In direct continuation from MACC and MACC-II, this service is based upon an ensemble of forecasts performed at seven centres in Europe, including RIUUK.

### 2.1 Air-quality forecasts

This task corresponds to the operational provision of forecasts for key air-quality compounds with the EURAD-IM system up to 96h for a range of molecules and vertical levels in GRIB2 format. Core products of MACC-II were forecasts of $O_3$, NO, $NO_2$, $SO_2$, CO, $PM_{2.5}$, and $PM_{10}$ at the surface level and at 500, 1000, and 3000 m height. These core products are verified at the surface on a daily NRT basis. Following a request of MACC users, the forecast products have been amended by concentrations of $NH_3$, NMVOC, and total PAN. The vertical resolution of the forecast products has been increased by additional provision of data at 50, 250, 2000, and 5000 m height. Each year starting at the beginning of March Birch pollen forecasts are provided.

### 2.2 Air-quality analyses

This task corresponds to the operational provision of analyses for key air-quality compounds ($O_3$, NO, $NO_2$, $SO_2$, CO, $PM_{2.5}$, and $PM_{10}$) with EURAD-IM. Hourly analyses for the previous day are delivered daily. At first, this is done every day in the early morning with a reduced set of surface in situ observations to provide improved initial values for the subsequent forecast. A second analysis of the previous day has been set up at 08:00 UTC, which includes as many as possible NRT observations. Currently these are surface in situ observations from the EEA and OMI and GOME-2 $NO_2$ column retrievals.

## 3. Validated assessment of air quality in Europe

An important aim of MACC-III and CAMS_50 is the yearly production of air quality assessment reports for Europe. The state and the evolution of background concentrations of air pollutants in Europe are described in these reports. Validated observation and modeling data are combined in re-analysed maps and numerical fields, to propose the best available representation of air pollutant concentration fields for a spatial resolution of 0.1 deg. The validated assessment reports are based upon an ensemble of models hosted at seven institutions in Europe including RIUUK. During the accounting period the air quality re-analyses for the years 2012 to 2014 and the 2016 interim re-analysis have been completed. The observation data assimilated in the re-analysis consists of surface in situ data for the pollutants $O_3$, $NO_2$, $PM_{10}$, and $PM_{2.5}$, the troposhperic $NO_2$ column content retrieved from the OMI and GOME-2 instruments on the Aura and MetOp satellites provided by KNMI. For the 2014 re-analysis CO profile data retrieved from the MOPITT instrument onboard of the Terra satellite provided by UCAR/NASA has been added. Intermittent 3d-var data assimilation has been applied. 30% of surface in situ background stations were held back from assimilation to allow for an independent validation of the assimilation results. Figure 9 exemplary shows bias and root mean square error of daily averaged $O_3$, $NO_2$, and $PM_{10}$ concentrations averaged over all Airbase background measurement sites, which were held back from assimilation for the year 2013.

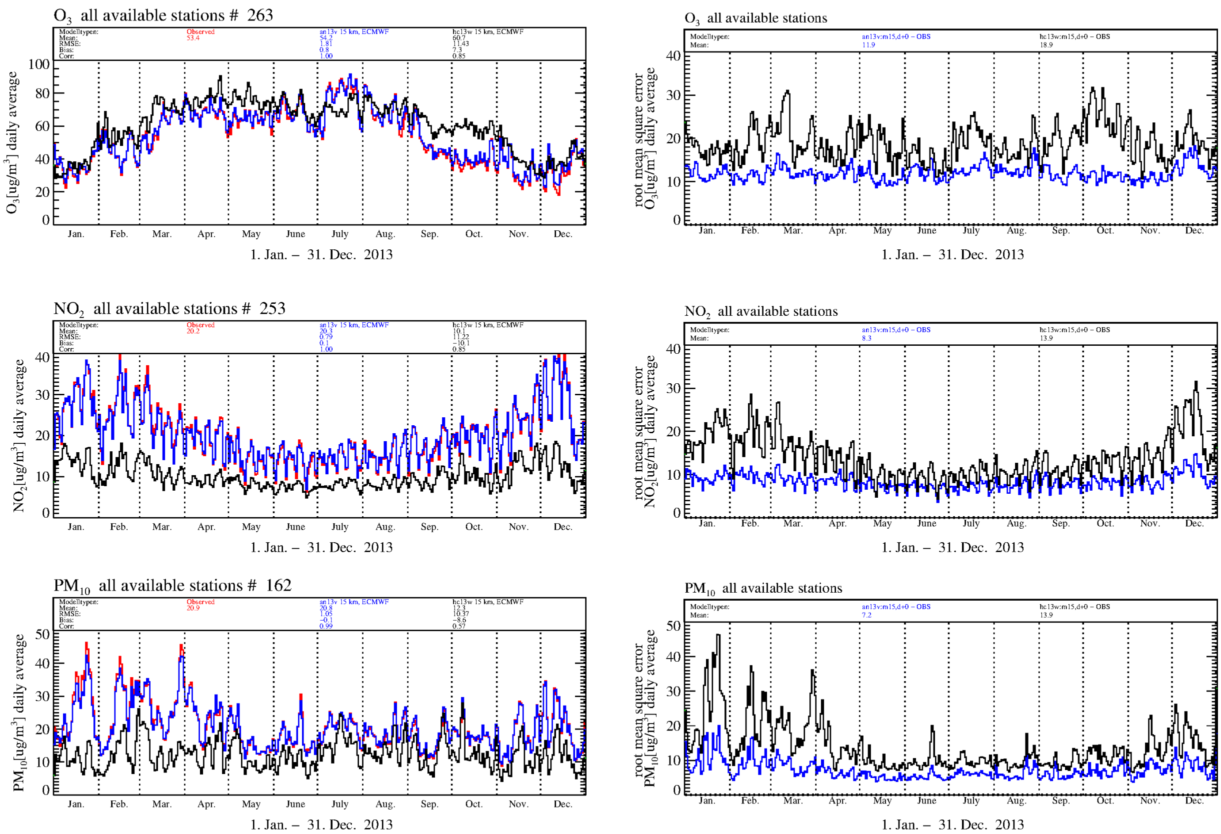



*Figure 9: Daily averaged concentration (left) and its root mean square error (right) of $O_3$ (first row), $NO_2$ (second row), and $PM_{10}$ (last row) averaged over all surface in situ measurement sites, which were held back from assimilation for the year 2013. Red: observations, blue: EURAD-IM 3d-var re-analysis, 30% of stations held back from assimilation, black: control run (no data assimilation at all).*

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## Future plans

Several actions are planned in CAMS_50 to improve the performance of EURAD-IM analyses above surface. EURAD-IM has established ability to assimilate in situ aircraft measurements. This ability will be applied to IAGOS (In-service Aircraft for a Global Observing System) measurements of $O_3$, CO, and $NO_2$. Furthermore, the EURAD-IM system will be augmented by 3d-var assimilation of $SO_2$ column retrievals. NRT $SO_2$ column retrievals are provided by the AURA/OMI and MetOp/GOME-2 instruments. In 2017 MOPITT CO data will be deployed in near-real time via the NASA LANCE system. This opportunity will be used to develop and assess the assimilation MOPITT CO retrievals also in EURAD-IM. The ability of EURAD-IM to assimilate MODIS AOD data will be further developed, with the aim to assimilate the MODIS AOD products over land.

In October 2018 CAMS will enter the second phase. The development activities in CAMS_50.II aim at the introduction of relevant evolutions into the regional air quality models in order to keep the regional production at the state of the art level of quality. New developments will be mostly driven by requirements of the ITT, user requirements, evolutions of the input data stream (IFS, boundary values, emission inventories), developments coming from own research and collaborations, and outcomes from the research and development component of CAMS (CAMS_61 ITT).