ESA project

Impact of Spaceborne Observations on Tropospheric Composition Analysis and Forecast (ISOTROP)

Final report

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Date: 8 March 2017, version 2.0, final

ESA Contract No: 4000105743/11/NL/AF

<table>
<tr><td>ESA Contract No:
**4000105743/
11/NL/AF**</td><td colspan="2">SUBJECT:
Final report
TITLE**:**
Impact of Spaceborne Observations on Tropospheric Composition Analysis and Forecast (ISOTROP)</td><td>CONTRACTOR:
KNMI</td></tr>
<tr><td>ESA CR( )No:</td><td>No. of Volumes: **1**
This is Volume No: **1**</td><td colspan="2">CONTRACTOR'S REFERENCE:
ISOTROP_final_report_20170308.docx</td></tr>
<tr><td colspan="4">ABSTRACT:
This document is the final report of the ISOTROP project.</td></tr>
<tr><td colspan="4">The work described in this report was done under ESA Contract. Responsibility for the contents resides in the author or organisation that prepared it.</td></tr>
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Executive summary

The ISOTROP project proposal was a response to the ESA ITT "Impact of Spaceborne Observations on Tropospheric Composition Analysis and Forecast", AO/1-6845/11/NL/AF. The general aim of this activity is to assess the benefit of the LEO+GEO satellite system for the understanding of local to regional scale tropospheric composition with a focus on Europe. The ISOTROP consortium involved researchers from the European institutes CNRS-GAME, TNO, NILU, FMI, and was led by KNMI. The study focussed on Sentinel 5-P (S5P) and Sentinel 4 (S4) observations, with a sensitivity study for the Sentinel 5, which will have a different overpass time than Sentinel 5-P. The study focussed on four chemical species: NO_2, HCHO, CO and O_3.

Within ISOTROP a comprehensive series of Observing System Simulation Experiments (OSSEs) were performed. Two European air quality modelling and assimilation system, MOCAGE from France and LOTOS-EUROS from the Netherlands were used for this. A so-called cross-OSSE approach was followed, in which both models produced Nature Runs at the spatial and temporal resolution of the satellite instruments. The observations produced from the Nature Runs are used in the OSSE runs of the other model.

ISOTROP has followed the steps of a well constructed OSSE study, which are detailed below:

- Nature Runs for the four species NO_2, HCHO, CO and O_3 were produced for 3 summer months and 3 winter months with both modelling systems. The resolution is 7x7 km, and runs are available over a large European domain. These simulations were compared with surface observations to investigate the realism of these runs.
- Based on the Nature Runs, synthetic observations for NO_2, HCHO, CO and O_3 were produced for Sentinel 4 and Sentinel 5-P separately, in total for 6 months. The synthetic observation generation follows the DOAS approach for NO_2, HCHO and optimal estimation for CO and O_3. Detailed error estimates are produced, and averaging kernels are provided in the product for individual pixels. Synthetic retrievals of the effective cloud fraction and top pressure are produced from the ECMWF meteorological analyses. The synthetic observations account for the geometry of individual pixels, and the corresponding cloud properties and albedo. Synthetic surface observations were also produced.
- Reference runs are produced by both models. Where relevant, this is based on an assimilation of surface observations.
- S4 and S5P CO and ozone OSSEs were performed with the MOCAGE system based on synthetic observations computed from the LOTOS-EUROS Nature runs, extended into the stratosphere by simulations with the TM5 model.
- S4 and S5P NO_2 and HCHO OSSEs were performed with the LOTOS-EUROS system based on synthetic observations computed from the MOCAGE Nature runs. An Ensemble Kalman filter approach is used which adjusts emissions to optimise the trace gas concentrations.

All these steps are discussed in this report, and in more detail in the various report deliverables of the ISOTROP project (section 3). The four species were analysed individually, and the main results of the OSSE studies are summarised in section 4. The available datasets and spin-off from ISOTROP are listed in sections 5 and 6. We end this report with a set of recommendations and ideas for follow-up studies.

1 ISOTROP project aims: ESA ITT

The ISOTROP project proposal was a response to the ESA ITT "Impact of Spaceborne Observations on Tropospheric Composition Analysis and Forecast", AO/1-6845/11/NL/AF. The following text is a quote from the ITT:

> OBJECTIVES OF THE ACTIVITY
>
> The general aim of this activity is to assess the benefit of the LEO+GEO system for the understanding of local to regional scale tropospheric composition with a focus on Europe.
>
> The first objective of this activity is to assess the value of a LEO+GEO satellite observation system measuring in the UV, visible, near infrared, and short wave infrared at nadir for tropospheric composition monitoring and forecast using a data assimilation scheme, using the Sentinel-4/-5 observation system as an example.
>
> The following questions shall be addressed in this context:
>
> 1. What is the gain in model and forecast skill by assimilating observations from LEO and GEO?
> 2. What is the improvement for boundary layer concentrations?
> 3. What is the improvement for long range transport of trace gases and its impact on boundary layer concentrations?
> 4. What is the improvement for components from episodal sources and from temporally constant sources?
> 5. What is the improvement regarding optimization of surface emission rates?
>
> The second objective of this activity is to study the impact of cloudiness and of uncertainties in the dynamic fields, especially the vertical transport in the lower troposphere on model and forecast skill. A number of critical issues shall be addressed regarding e.g. the way to treat uncertainties in cloudiness, aerosol load, and surface albedo and emission rates to optimize this assimilation scheme.

Additional requirements from the ITT text:

> The contractor shall adapt/develop an assimilation scheme capable of combining LEO and GEO satellite and in-situ observations covering the following tropospheric key species: O3, NO2, CO, HCHO. Reservoir species must be taken into account for these key species within the model and the choice of the assimilation technique must be appropriate for short lived species. The following key features for the model must be taken into account:
>
> 1. Spatial resolution better than 50 km for Europe, better than 10 km x 10 km for selected target regions in Europe according to Sentinel-4/-5 observational capabilities; The target regions are to be agreed by the agency.
> 2. Adequate treatment of the vertical sensitivity (averaging kernels) of trace gas observations.
> 3. Temporal resolution hourly according to Sentinel-4 (diurnal cycle in target regions).
> 4. Accurate representation of the PBL.

The targets for this study are therefore very ambitious and both the observation simulations and model runs required are very extended and diverse. These requirements can be summarised as follows:

- Full-complexity Observing System Simulation Experiments (OSSEs) are required
- Such OSSEs are based on two independent models, one producing the Nature Run, the other producing both reference and future observations assimilation runs.
- The call includes both GEO and LEO platforms: two sets of synthetic data need to be generated for very different platforms.
- Four target species are mentioned: O3, CO, NO2, and HCHO.
- The new Sentinel 4 & 5 satellite instruments are high-resolution sounders, and the amount of data produced by these sensors is many times larger than for current satellite instruments like OMI.
- Model simulations should be performed at high resolution, representing the resolution of the satellites.
- The set of 5 questions to be addressed (see above) is very extended, and sometimes requires different OSSE solutions to obtain optimized answers. For instance, the long-range transport question could have a very different model set-up, domain, time period, resolution and data assimilation approach than the emission rate question, which basically requires e.g. inverse modelling techniques to be applied.

The ISOTROP response to the diversity of the questions was the introduction of two modelling systems. The MOCAGE system was performing OSSE assimilation runs over a larger European domain, with a focus on long-range transport and the species ozone and CO. The LOTOS-EUROS system was using an assimilation approach in which the surface emissions were adjusted, focussing on NO2 and HCHO and on the question of optimising surface emission rates. In this way all five science questions have been addressed to some degree. The main focus was on questions 1 and 2, on the gain in model skill and impact on boundary layer concentrations. Episodal sources were studied by focussing on the fires in Purtugal in Summer 2003. Long-range transport aspects were treated in the CO OSSE with MOCAGE. Surface emission improvements were briefly discussed in the LOTOS-EUROS NO2 OSSE study.

2 The ISOTROP approach

In response to the ITT, the consortium of researchers from KNMI, TNO, CNRS-GAME, NILU and FMI prepared the **ISOTROP** proposal in the form of a technical and a financial part:

> *Impact of Spaceborne Observations on Tropospheric Composition Analysis and Forecast (ISOTROP); Volume I, Technical Proposal; A proposal in response to ESA Invitation to Tender AO/1-6845/11/NL/AF; date: 3/10/2011.*
>
> *Impact of Spaceborne Observations on Tropospheric Composition Analysis and Forecast (ISOTROP); Volume II, Financial, Management and Administrative Proposal; a proposal in response to ESA Invitation to Tender AO/1-6845/11/NL/AF; date: 3/10/2011.*

Below we will give a short summary of the ISOTROP proposal and work plan. More details can be found in the proposal text. Details of the modelling and assimilation systems, and approach to generate the synthetic observations can be found in the proposal and the project deliverables which are the topic of the next chapter.

The ISOTROP project approach to provide answers to the study objectives presented in the previous section is by performing Observing System Simulation Experiments (OSSEs), see Fig.2.1. These OSSEs were performed with two state-of-the-art chemistry transport models, or air quality models, equipped with data assimilation capabilities, namely MOCAGE and LOTOS-EUROS. In particular:

- Synthetic observations of ozone, CO, NO2 and formaldehyde (HCHO) were generated to reflect as closely as possible the performance of the future S5-P, S4 and S5 missions. Full detail (averaging kernels, covariance matrices, a-priori) is passed from the optimal estimation synthetic observations to the model OSSEs. This detail is essential for a realistic OSSE.
- The “Nature Run” (used to make the synthetic observations) was performed with great care: it should reflect state-of-the-art uncertainties in present-day air quality modelling, and should be independent from the model performing the OSSE. Therefore it was proposed to involve two models, which provide each other’s Nature Run. Note that the LOTOS-EUROS high-resolution simulations have been extended to include the free troposphere and stratosphere by means of TM5 simulations at 1x1 degree. This in order to provide meaningful profiles for the ozone OSSE in particular.
- An OSSE has been performed with the MOCAGE-PALM modelling system, which has focussed on ozone and CO, which are determined strongly by long-range transport.
- An OSSE has ben performed with LOTOS-EUROS, which has focused on NO_2, HCHO, and the optimization of surface emission rates, with an evaluation of O_3.
- The S5P, S4 and S5 missions will have enhanced observational capabilities to characterise the cloud cover and determine surface albedo and aerosols. This aspect has been included and detailed error budgets were provided with the synthetic observations.

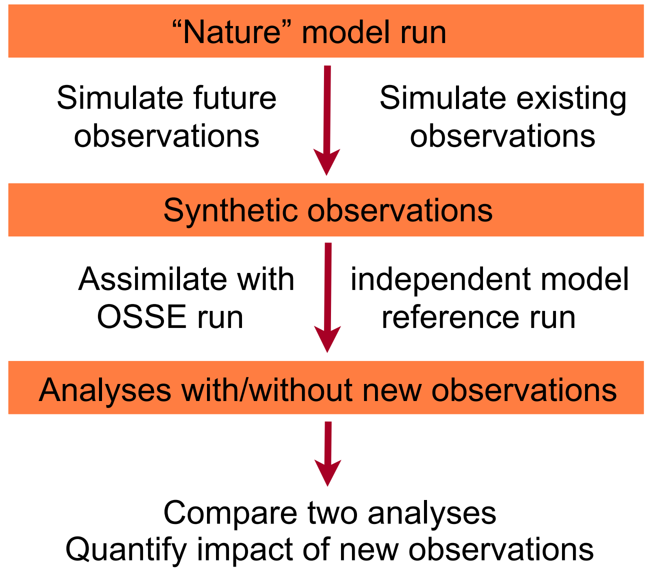


Fig. 2.1. Schematic diagram for an Observing System Simulation Experiment. Based on an independent, high-resolution realistic Nature Run synthetic observations are simulated for both existing observations and the future observations for which the impact on the system is estimated. These observations are assimilated and the run with future observations added is compared with a reference run based on existing observations. In this way the expected impact of the new observations is quantified.

- A separate OSSE has been performed by both models to study the impact of including cloudy observations, as compared to assimilating cloud-filtered scenes only.

Special about the ISOTROP project is the so-called "cross-OSSE" approach. This is shown in Figure 2.2. Nature Runs are produced for the four target species by both models, and the Nature Run results have been compared with observations. Synthetic observations for Sentinel 4 and 5P for NO2 and HCHO are generated from the MOCAGE Nature Run. Synthetic observations for Sentinel 4 and 5P for O3 and CO are generated from the LOTOS-EUROS / TM5 Nature Run.

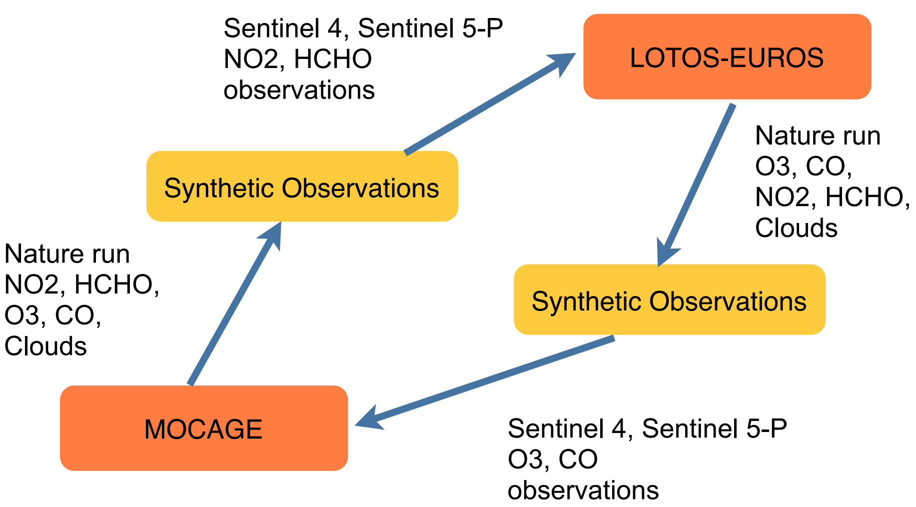


Fig 2.2. The cross-OSSE approach used in ISOTROP.

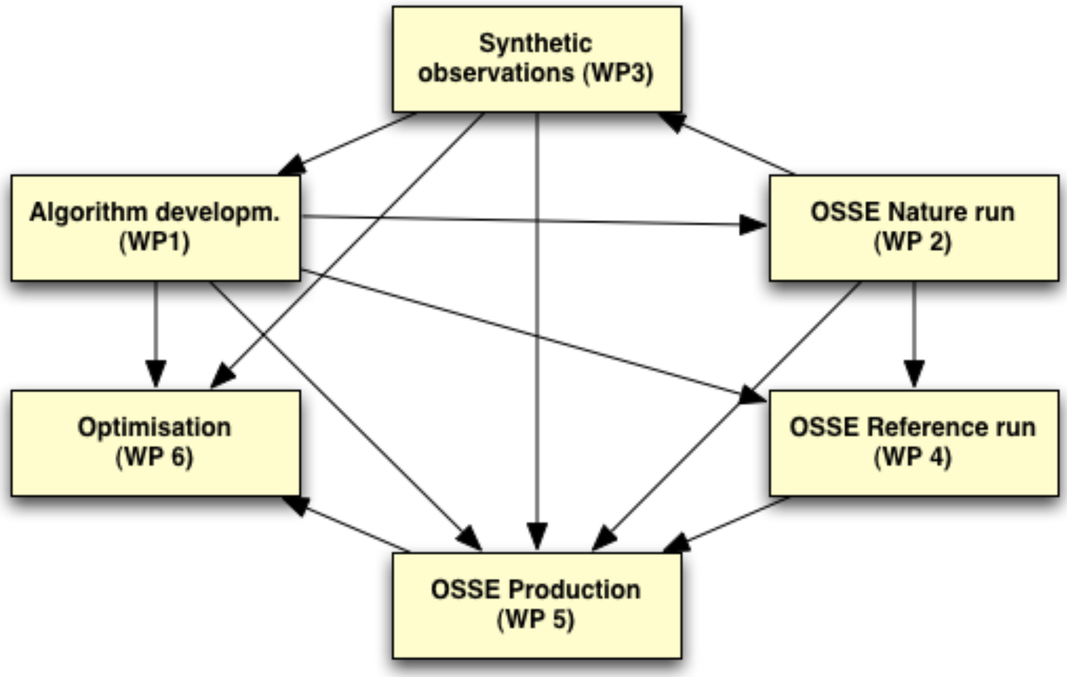


Fig. 2.3. Work breakdown diagram of ISOTROP. The arrows indicate the output-input dependencies of the tasks.

A schematic overview of the work performed in ISOTROP is provided in figure 2.3, which also introduces the tasks or work packages (WPs). The corresponding work package tables can be found in the financial proposal. Each of these tasks is further detailed below. The corresponding deliverables are discussed one-by-one in the next section. Study domains and time periods are shown in Fig. 2.4.

The work started with the setup of the models (WP1), definition of study domains, resolution and period. A specification of the synthetic observation format (from WP3) was used to code the observation operators (WP1). When the setup was complete, the Nature Runs with MOCAGE and LOTOS-EUROS/TM5 were produced (WP2). Based on the Nature Run, the synthetic observations were produced (WP3). In parallel the reference run was produced (WP4). Based on the inputs from WP1, WP2, WP3 and WP4, the OSSEs were performed and analysed (WP5). The impact of cloud-covered observations was studied in WP6.

The consortium has proposed to focus on S5P and S4. One major reason for this is that the instrument details are much better known, and ISOTROP could make use of the (draft) ATBDs. ESA indicated that they also wanted results for S5. The main difference between S5 and S5P is the overpass time, morning (S5) vs. afternoon (S5P). For simplicity, within ISOTROP we have assumed similar instrument specifications for all three instruments. The effect of the difference in overpass time was studied with the use of the S4 dataset, which is available hourly. Assimilation runs were done with the S4 9 UTC and 13 UTC dataset only. It was checked as well if the 13 UTC S4 dataset leads to comparable results as the S5P dataset.

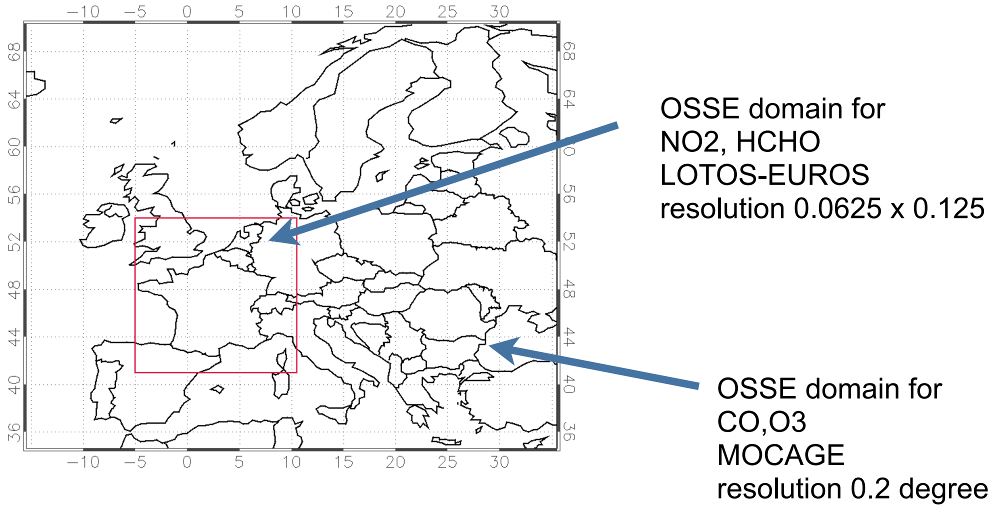


Fig. 2.4. The domains defined for the OSSE studies are the MACC European domain (entire figure) and the Prev'Air extended domain (France and surroundings, red square). Study periods are JJA 2003 and NDJ 2003-2004, 6 months in total. The LOTOS-EUROS model is extended in the vertical with TM5 simulations of free troposphere and stratosphere. TM5 is run globally at 3x2 degree, with a zoom domain at 1x1 over Europe covering the full MACC domain. TM5 at 1x1 degree provides the boundary conditions for the LOTOS-EUROS model.

The project duration and available funding were limited, as compared to the goals of the project. In ISOTROP ten OSSE studies were performed (for four species, for S5P and S4, and for S4 and S5P combined). The analysis of these OSSE runs has focused on showing the impact of the observational datasets: how much are we able to reconstruct the Nature Run concentrations? Based on these runs we briefly discussed aspects like long-range transport (especially for ozone and CO) and emission fluxes (NO_2 and HCHO). The Portugal fires (episodal fluxes) in 2003 were studied in more detail. However, a more thorough discussion of emissions improvements (science question 5) and long-range transport (science question 3) was not feasible within the limited number of person months. Emission improvements will depend strongly on the ability of the model to accurately describe model processes (chemistry, transport) that link concentrations to emissions. This is best studied in a system that assimilates multiple species datasets and may require sensitivity studies, to study the dependence of the emission estimates on individual model uncertainties, which are beyond the scope of ISOTROP.

3 ISOTROP project results

An overview of the ISOTROP activities and results obtained will be given by reviewing the individual project document deliverables one by one. For more details we refer to the content of these reports.

3.1 Effective cloud parameters for the synthetic retrievals

> *"Comparison of cloud parameters calculated using ECMWF and ARPEGE/MOCAGE input variables for use in retrievals,"*
> *prepared by Jason Williams, Henk Eskes and Albert Oude Nijhuis,*
> *8 January 2013.*

The retrieval approaches which are applied for the ISOTROP multiple OSSEs are using effective cloud parameters - cloud fraction and cloud top height - which are derived from the satellite spectra. In the project we decided to construct the satellite derived effective parameters from the weather model simulated cloud properties. In the first months of the project we decided to have an extra short study - not explicitly defined in the work plan - to study the realism of the cloud properties derived by comparing the two sets of cloud distributions and by comparing with observations of OMI.

The derivation of effective cloud parameters involves several steps:

- Extracting liquid water and ice water paths, effective humidity, and cloud cover from the meteorological archives
- Conversion of these cloud fields into cloud optical depths for each model layer
- Relating the cloud optical depths to radiance levels at the top of the atmosphere
- Computing the effective cloud fraction from this radiance level, and the effective cloud height from the distribution of cloud optical depth values.

When comparing the results for ARPEGE (0.2 degree resolution) and ECMWF (0.25 degree resolution), we found major differences in the cloud distributions, as can be seen in Fig 3.1. For the cloud effective altitude better agreements were found between the two meteorological inputs. In conclusion, the best agreement with the OMI monthly-averaged distribution is obtained for the ECMWF meteorological data. Comparisons with OMI have to be considered with some care, because of differences in effective resolution between the model and observations. Nevertheless, it seems that also for ECMWF (2003 operational data) there is a tendency to produce more cloud-free synthetic observations, and less observations with partially clouded scenes than what is observed by OMI.

These results were discussed explicitly during one of the early ISOTROP meetings. Because of the deviating distribution of ARPEGE, and the lack of fully clouded pixels, we decided to base the ISOTROP synthetic observations on the ECMWF cloud analyses only. In the case of the NO2 and HCHO OSSE with LOTOS-EUROS, this means that both the model simulations and the synthetic observations are based on similar cloud information from ECMWF. The MOCAGE runs for CO and O3 are based on the ARPEGE meteorology and cloud distributions, which therefore differ from the cloud informations used in both the Nature runs and synthetic observations.

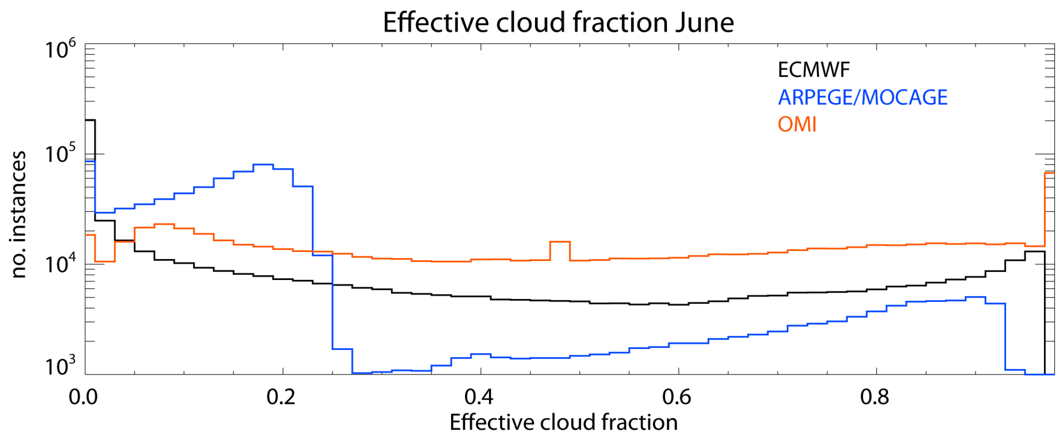


Fig 3.1. Comparison of the synthetic observation effective cloud fractions derived from the ECMWF (black) and ARPEGE (blue) meteorological cloud fields, with the effective cloud fraction retrieved from actual OMI observations using the O2-O2 algorithm (orange), for June 2003. The model computations produce on average smaller cloud fractions than what is observed by OMI.

3.2 Description of the assimilation schemes

"Assimilation Scheme Description",
prepared by Lyana Curier, Renske Timmermans, Arjo Segers, Laaziz El Amraoui, Rachid Abida, Jean-Luc Attié, Henk Eskes
2 October 2013

The Assimilation Scheme Description Document (ASDD) is containing a description and motivation of the model and assimilation scheme choices, and a description and motivation of the target regions and periods.

The document provides detailed descriptions and a list of references of the two model-assimilation systems:

- *The MOCAGE model and the MOCAGE-PALM assimilation system:* the OSSE assimilation runs are based on the 3D-FGAT technique, and are applied to the ozone and CO observations.
- *The LOTOS-EUROS model and EnKF assimilation system:* the Ensemble Kalman Filter approach is used to optimize model parameters, and emissions in particular, which has been used to study the impact of sentinel data on emission fluxes.

The suitability of these models and data assimilation schemes to answer the ISOTROP project goals were also discussed.

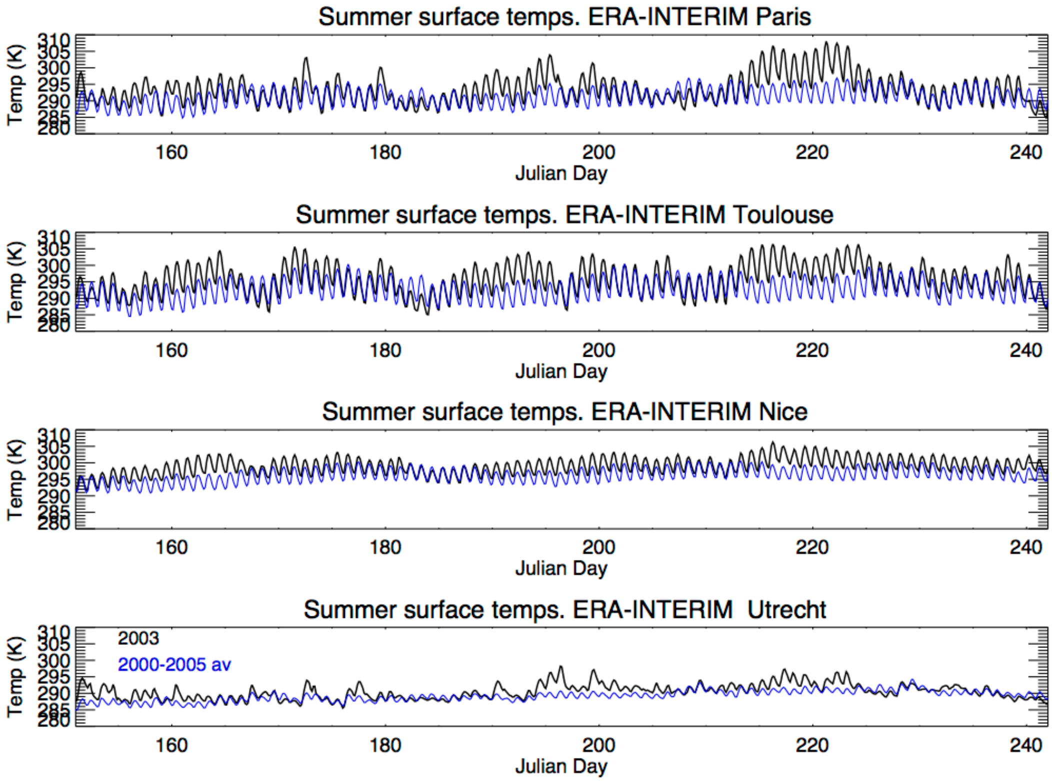


Figure 3.2: The temperature series at Paris, Toulouse, Nice and Utrecht for the summer of 2003 (June, July, August). The actual temperature (black) is compared with 6-year mean temperature records.

For the ISOTROP study we chose the summer and winter of 2003 for the simulation. The motivation for this is the following: During summer 2003 there were extensive heat wave periods with very high ozone peak values in western Europe. Because ISOTROP covers a large period - three full months in summer and three months in winter - there are also more normal periods. Large fires occurred during the heat waves, especially in Portugal. Therefore this period allows us to study major events within the same time window.

In the study we have focused on different domains (see Fig. 2.4):

1. **Europe** (resolution of about 15x15 km)
2. **Zoom** or Prev'Air domain (resolution of about 7x7 km)
3. **Paris** domain (resolution of about 7x 7km; this is just a selection of grid cells from the 'Zoom' domain around the city of Paris)
4. **Fire** domain (detection of fire plumes, Iberian peninsula, resolution of about 7x 7km)

3.3 Nature Runs

"Description of Nature Runs used in ISOTROP OSSEs",
prepared by Jean-Luc Attié, Rachid Abida, Laaziz El Amraoui, Arjo Segers, Philippe Ricaud, Lyana Curier and Henk Eskes
October 2013

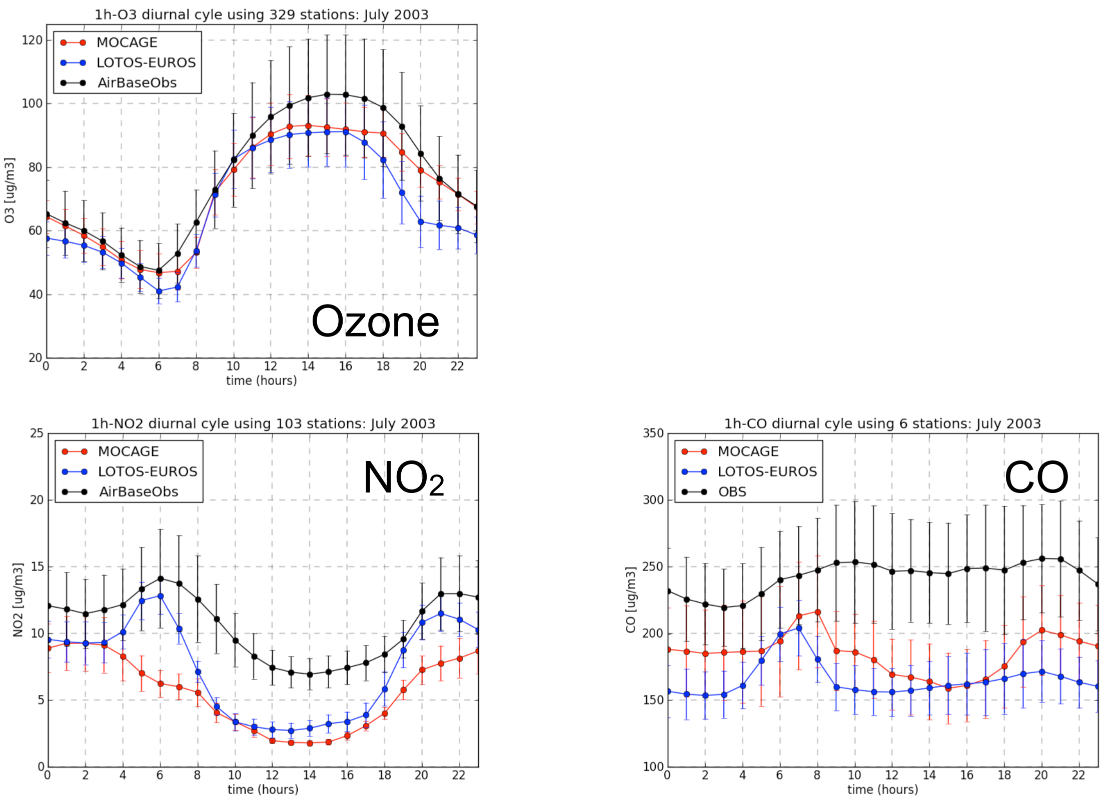


Fig 3.3. Comparison of the LOTOS-EUROS and MOCAGE Nature Run simulations with surface observations for July 2003.

Nature Runs represent the true atmosphere that will be used to build the synthetic observations. In this Nature Runs Description (NRD) document, the LOTOS-EUROS and MOCAGE model configurations used and the Nature Runs obtained are described. In addition, a comparison between the different NRs is performed to evaluate the differences (variability, bias, ...) between the resulting Nature Run simulations of the two modelling systems and available surface observations in Europe. Both modelling systems generated Nature Run output for all four target species: O_3, CO, NO_2, HCHO.

Strictly speaking, for the OSSE studies only output for two model species is needed. However, the availability of the four fields allows us to explore the differences between the models and observations. This study provides an indication if the model-model differences are a realistic estimate of the uncertainties in the individual models. The document provides an extensive set of evaluation statistics for the two models for the species NO2, O3 and CO, compared to observations at the surface in Europe.

Comparisons between Nature Runs from MOCAGE and LOTOS-EUROS show quite similar biases (see Fig. 3.3). These results suggest that a cross assimilation (OSSE) may be conducted without debiasing the model outputs. This bias will be taken into account in the discussion when assessing the OSSEs. The comparisons between the two models show roughly a similar behaviour concerning all evaluated species. The variability between models is quite similar and concentrations obtained are in the same order of magnitude.

3.4 Synthetic observations

"Synthetic Observation Product Specification (SOPS) Deliverable D4 / WP3", prepared by Jukka Kujanpää, Albert Oude Nijhuis, Henk Eskes, Johan de Haan, Pepijn Veefkind, Johanna Tamminen, 12 August 2015

"Synthetic Observation Product Specification (SOPS-Val), validation results", prepared by Jukka Kujanpää, 13 December 2013

"ISOTROP synthetic observation generation by using look-up tables", prepared by Albert Oude Nijhuis, Johan de Haan, Henk Eskes, Pepijn Veefkind, May 2013

"Synthetic Observations Product Specification Level-2 user manual", prepared by Albert Oude Nijhuis, Henk Eskes, Johan de Haan, 19 February 2013

The generation of the synthetic satellite observations is non-trivial and involves a series of steps. The satellite synthetic data were stored in HDF-5 format files, one file per orbit / hour, inspired by the OMI data format. The individual species are stored in separate files. A series of scripts were developed to implement the various steps. The implementation was done by KNMI, and all steps were independently checked by FMI. The individual steps are:

- The generation of the orbit tracks for the sentinel 4 and sentinel 5P missions.
- The construction of a full set of orbits for the three summer months and three winter months, for both S4 and S5P, by a repetition of the initial orbit tracks in space and time.
- The construction of the individual observations (7km footprints) and the correct geometries (latitude, longitude, time, solar/viewing zenith/azimuth angles).
- The interpolation of the meteorological cloud fields to the satellite footprints, and the computation of the effective cloud fractions and cloud top heights.
- The interpolation of the Nature Run results to the satellite footprints. Nature Run profiles are stored for the four target species O3, NO2, CO, HCHO into the orbit files for S5P and the hourly observation files for S4, limiting the data to the European domain. Addition of model grid information (pressure levels).
- Addition of ancillary data: surface albedo values for the four species.
- Generation of lookup tables for the satellite retrieval uncertainties (covariances) and the corresponding averaging kernels, for the four target species O3, NO2, CO, HCHO.
- Generation of the individual synthetic observations (retrieval state, averaging kernel, retrieval uncertainty estimate and a-priori (if relevant)) by using the available geometry information, Nature Run profiles, albedo and cloud information, and the lookup tables. Noise is added to the observations based on the estimated retrieval uncertainty.

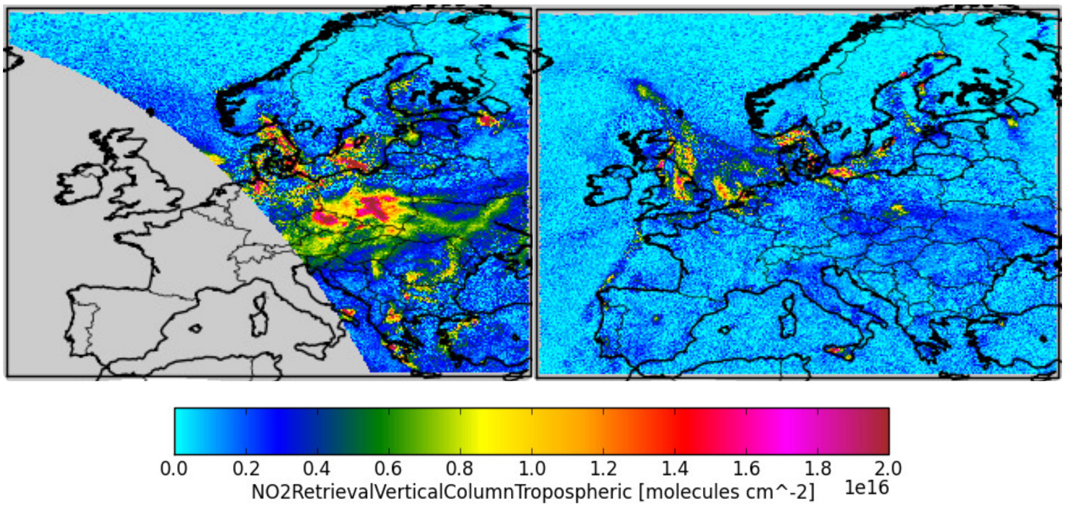


Figure 3.4.a: Example of simulated noisy tropospheric NO2 observations for two S4 orbits, early morning (left), 4:00, and mid-day (right), 12:00, on 1 June 2003. The plots show the large diurnal cycle present in the MOCAGE Nature Run. Data is plotted for SZA < 85 degree. All observations are shown, independent of their cloud fraction.

Lookup tables have been generated for the four species separately. Entries, resulting kernels and uncertainties are available for a representative sample of geometries (viewing/solar angles), vertical layers (20 layers), surface pressure and albedo values. The lookup tables have been constructed by repeated calls to the DISAMAR radiative transfer and retrieval software developed by KNMI (de Haan, 2010). Based on these lookup tables, averaging kernels and retrieval uncertainties (covariances) are computed for each of the observations individually.

A version-1 set of scripts and lookup tables were delivered in 2013. Based on these, observational datasets were produced for S4 and S5P, for all four species. The product description and a short user manual were produced (the two documents with Albert Oude Nijhuis as first author). A verification of the product was done by FMI and reported at the end of 2013 (SOPS-Val document). The resulting synthetic observation results were more or less as could be expected. The datasets were shared with the teams performing the OSSE studies.

During 2014 feedback was received on these datasets. Apart from that, the synthetic observation software was inspected in detail. Several issues came up:

- The brute-force kernels/covariances produced for ozone were leading to extremely large observational datasets, in particular for ozone. The MOCAGE group indicated that the datasets were problematic.
- Inspection of the v1 code revealed several bugs, interpolation issues and sub-optimal assumptions. In the meantime, advanced S5P ATBDs had become available to fine-tune some of the settings/assumptions in the code.

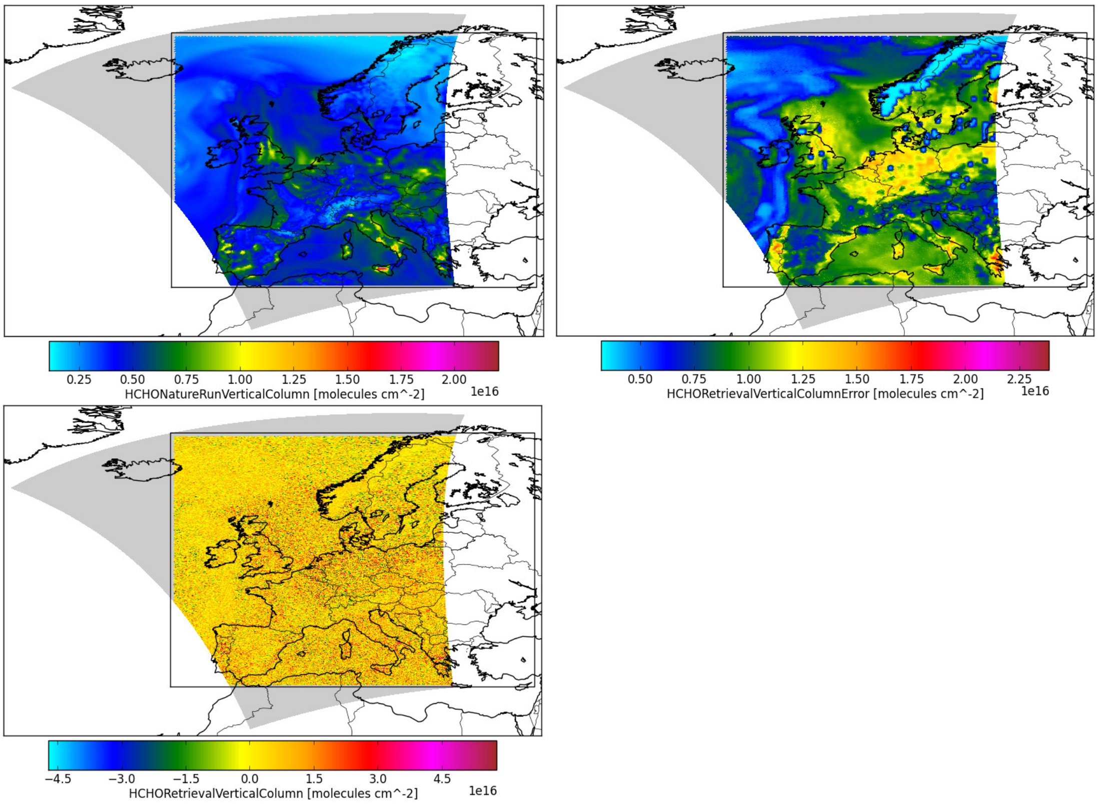


Figure 3.4.b: Nature Run (top-left), retrieval slant column error (top-right), retrieval vertical column (bottom) for HCHO. S5P, orbit 21, 1 June 2003, time 12:34. Grey: no-data in synthetic observations (mainly outside nature model domain).

Based on this, it was decided that a version 2 of the datasets would be generated before continuing with the OSSE runs. This caused a major delay in the project. All parts of the code to generate the synthetic observations were reviewed, and large parts were re-written. All steps in the observations generation were explicitly tested. A completely new approach for ozone was developed. New datasets became available in spring/summer 2014. The results are described in the SOPS document (final version 12 August 2015), which is briefly summarised below.

For NO_2, the synthetic observations retrieval and error propagation is based on the DOAS approach, and the steps are very similar to the baseline L2 processors for S5P and, more recently, S4. An example of two hourly datasets generated for S4 is shown in Fig. 3.4.a.

The formaldehyde (HCHO) simulations are also based on the DOAS approach and have a similar error propagation approach, and the data product also contains scene-dependent averaging kernels. The uncertainty of the product is dominated by the noise in the slant column fitting. Because HCHO concentrations over Europe are relatively low (see Fig. 3.4.b, top-left) compared to e.g. the tropical regions or the South of the USA, and because noise is added according to the simulated uncertainty (top-right panel), the final result for individual overpasses are noise dominated, as is evident from the lower panel in Fig. 3.4.b which contains a large number of negative column values. However, when these results are averaged over a week or a month, one will see the formaldehyde features emerge as simulated by the Nature Run.

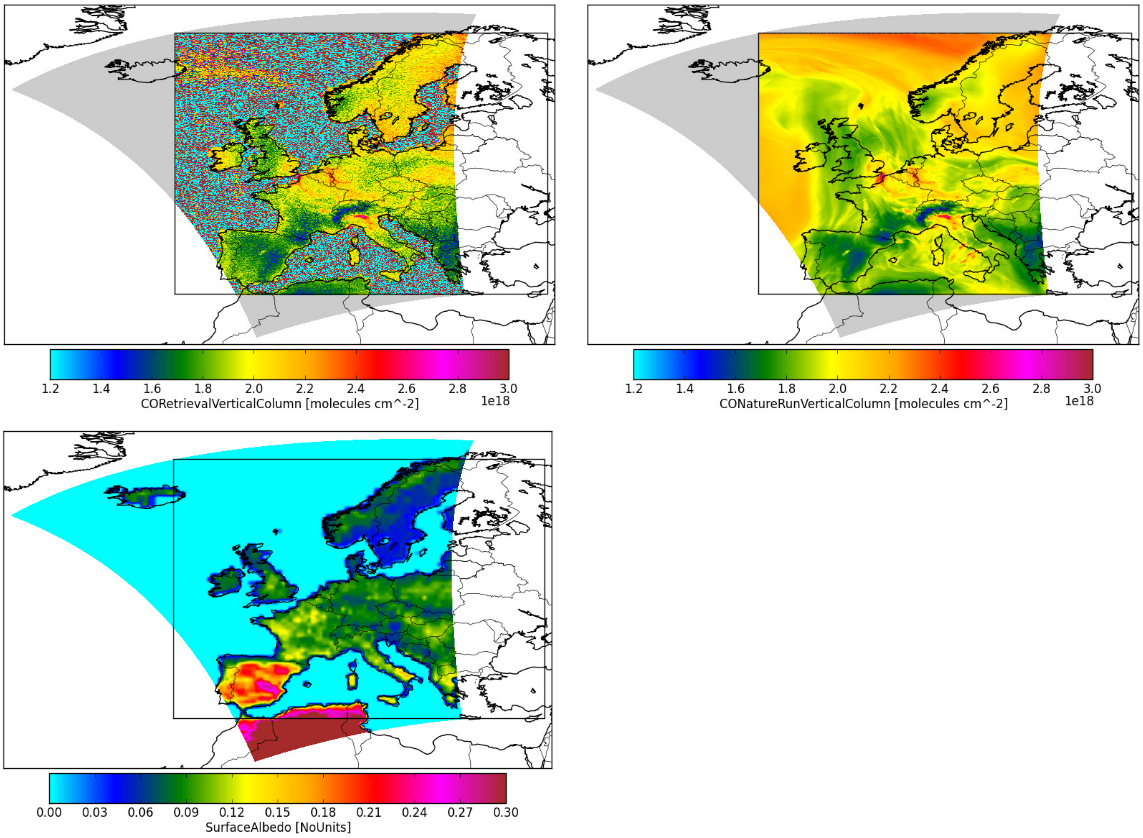


Fig. 3.4.c. Retrieval vertical column, Nature Run vertical column, and surface albedo for CO. S5P, orbit 21, 1 June 2003, time 12:34. Grey: fill values.

An example of CO observation simulations for S5P is shown in Fig. 3.4.c, top-left. Note the large signal-to-noise ratio over land, and the large noise-to-signal ratio over the ocean. The dominating factor for CO is the value of the surface albedo, which is very small over water (lower panel). A patch of clouds is present East of Iceland, and the figure indicates that meaningful results can be obtained in case of clouds over the oceans and seas. The simulated noise levels (range 2-10% over land) are quite consistent with the S5P ATBD on CO. Note that S4 does not have a SWIR channel, and therefore no CO observations were generated for the geostationary case.

Finally, as mentioned above the approach for v2 ozone profiles was completely revised. Here we closely follow the approach suggested by Migliorini (2008) in a publication called "Use of the Information Content in Satellite Measurements for an Efficient Interface to Data Assimilation". Instead of providing full covariance matrices and square averaging kernels, the retrieval results are provided in the eigenspace of the radiative model. The DISAMAR code was extended to support this kind of output. The mathematical equations are presented in the SOPS document. The approach is a major innovation pioneered by ISOTROP, and has major benefits:

- The storage for the retrieval product is significantly reduced. In our case, where the retrievals are specified on 20 vertical levels, this reduction is about a factor of 8.
- The number of independent observations presented to the assimilation system is reduced from 20 to 6. With this reduction, larger number of profiles can be analysed simultaneously in the assimilation system.

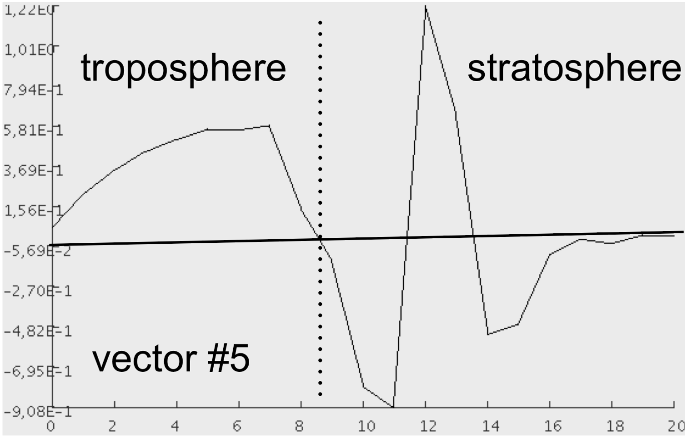


Figure 3.4.d. One example of one of the kernel vectors (presented in log(ppm) space) for the fifth eigenvector. The first three vectors are mainly sensitive to stratospheric ozone, but vectors 4, 5, 6 demonstrate the sensitivity of the satellite UV observations to tropospheric ozone.

- The observation covariance is diagonal by construction, which is consistent with the assumption of independent observations often made in assimilation.

One more choice was made, which is a restriction of the wavelength range to 300-320 nm. Going to smaller wavelengths rapidly decreases the signal-to-noise ratio, and in practice spatial co-adding of observations is needed. For this relatively narrow window retrievals can be performed at the full spatial resolution of the instruments. Removing the shorter wavelengths will reduce the amount of profile information in the middle-upper stratosphere. For ISOTROP the focus is on the (lower) troposphere, and therefore 300-320 nm was perceived as the most logical choice. Datasets were produced for S4 and S5P.

3.5 Reference runs

"Description of ground-based data used in the Reference Runs",
prepared by Arjo Segers, Lyana Curier, Jean-Luc Attié, Rachid Abida, Laaziz El Amraoui, Philippe Ricaud
16 October 2013

"Reference Run Description Document",
prepared by Arjo Segers, Renske Timmermans, Lyana Curier, Jean-Luc Attié, Rachid Abida, Laaziz El Amraoui, Philippe Ricaud,
3 April 2015

These two documents describe the reference runs: the reference runs are runs that have the same model configuration as the OSSE runs, except that the S4 and S5P datasets are not assimilated. This is a reference to investigate how much is gained by introducing the sentinel datasets.

The first report only describes the ground-based datasets, which are used in the reference runs. European regulatory AIRBASE observations are used according to their classification, e.g. Fig. 3.5.a shows only the background rural stations, which measure ozone and NO_2. Furthermore, part of the observations (validation stations) can be removed from the assimilation to test the effectiveness of the analysis to improve concentrations at sites not included in the analysis. The uncertainties of the observations are also discussed.

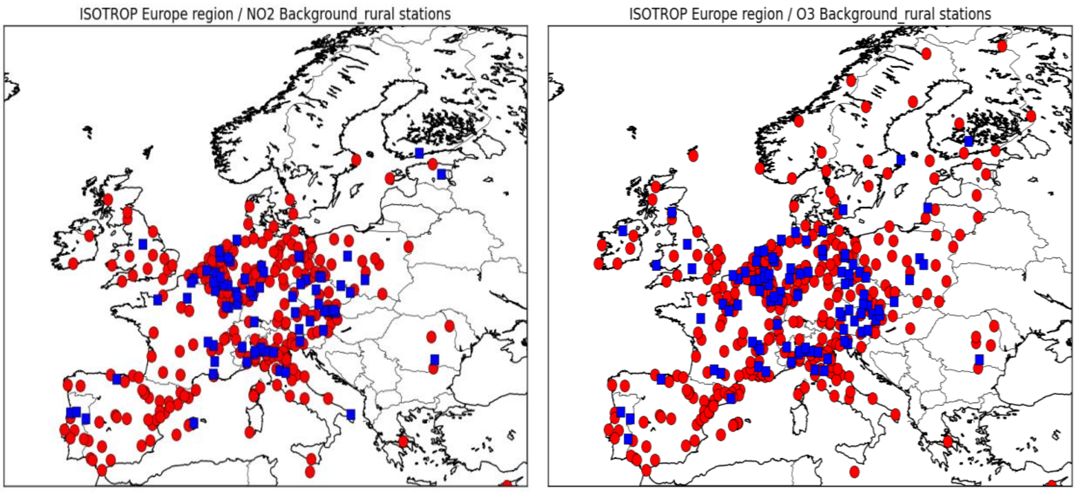


Fig. 3.5.a. Composite map of the sampling locations for the ozone and NO_2 stations for the European domain; red circles denote assimilation stations, blue squares validation stations.

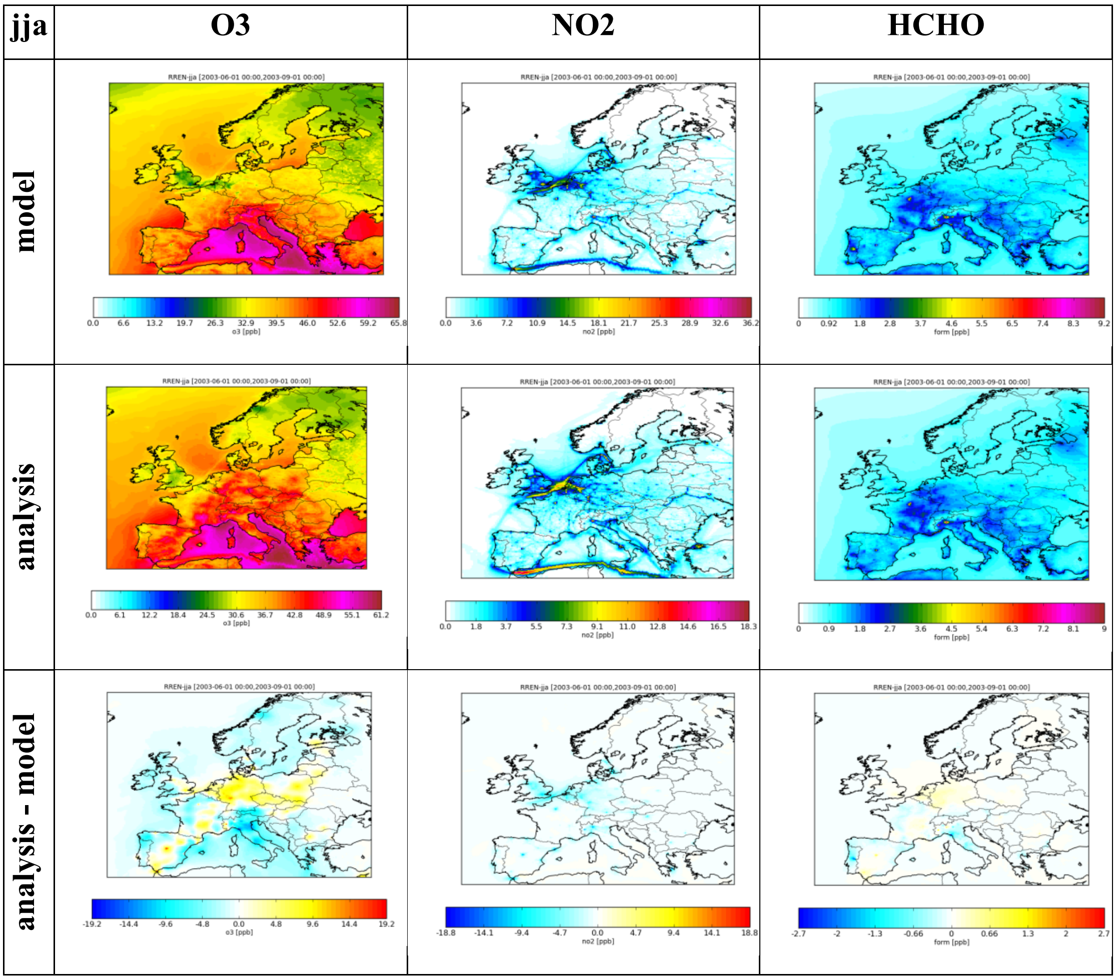


Fig. 3.5.b. Average surface concentrations in LOTOS-EUROS in the free model run (top row) and when synthetic ozone surface observations, derived from the MOCAGE Nature Run, are assimilated.

The second document describes the reference runs (RR) produced in the ISOTROP project. Reference runs are made with each of the two considered models (around MOCAGE and around LOTOS-EUROS), and might assimilate synthetic ground based observations of ozone. Multiple reference runs are produced for each of the models, for the large European domain, the Prev'Air domain and a fire domain over Portugal.

Examples are shown in Figure 3.5.b, which demonstrates the impact of assimilating the synthetic surface observations in the LOTOS-EUROS system during the summer months. This not only impacts the ozone concentrations, but also NO_2 and formaldehyde.

3.6 The CO OSSEs

"OSSE production and analysis (S5 CO OSSE)",
prepared by Jean-Luc Attié, William Lahoz, Rachid Abida, Laaziz El Amraoui, and Philippe Ricaud,
November 2015

Abida et al., accepted for publication in ACP, 2016

Several regional-scale Observing System Simulation Experiments (OSSEs) were conducted over Europe to explore the impact of the LEO satellite mission S5P carbon monoxide (CO) total column measurements on lowermost tropospheric air pollution analyses, with a focus on CO concentrations at the surface and in the Planetary Boundary Layer (PBL). The OSSE runs were performed with the MOCAGE-PALM assimilation system and with the synthetic CO column observations derived from the LOTOS-EUROS Nature Run.

The OSSE results for summer 2003 (June, July and August) indicate that simulated S5P CO total column measurements benefit efforts to monitor surface CO. The largest benefit occurs over land in remote regions (Eastern Europe, including Russia) where CO sources are sparse. Over these land areas, and for the case when we remove the systematic error, we obtain a lower RMSE value (by ~10 ppbv) for the AR than for the CR, in both cases vs the NR. Over sea and Scandinavia, we also obtain a lower RMSE (by ~10%) for the AR than for the CR, in both cases vs. the NR. Consistent with this behaviour, we find the AR is generally closer to the NR than the CR to the NR, with a correlation coefficient R, reaching 0.9 over land (NR vs. AR), see Fig. 3.6. By contrast, the correlation coefficient between the CR and the NR is typically less than 0.5, with very low values over Eastern Europe, where CO sources are sparse. In general, for all the metrics calculated, there is an overall benefit over land from the S5P CO total column measurements. Significance tests on the CR and AR results indicate that, generally, the differences in their performance are significant at the 99% confidence level. This indicates that the S-5P CO total column measurements provide a significant benefit to monitor surface CO during northern hemisphere summer.

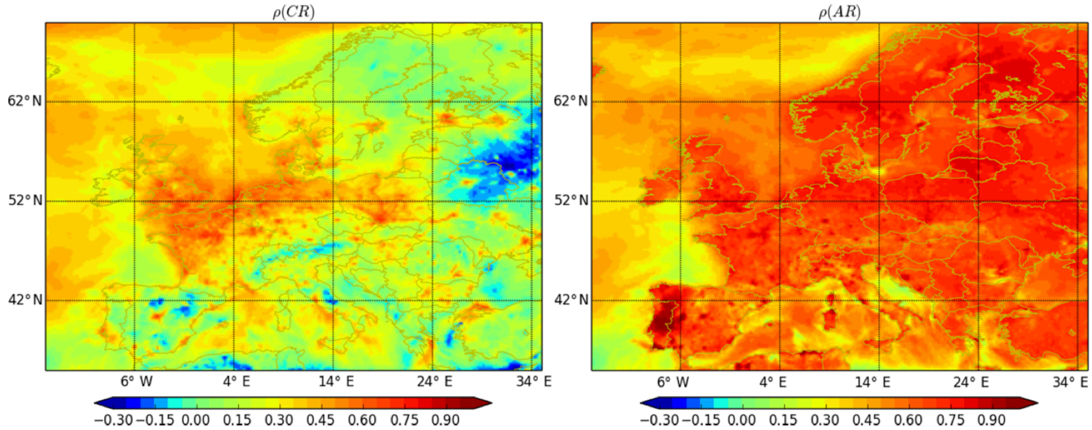


Fig. 3.6. CO OSSE: Correlation coefficient (R) between the CR and the NR (left panel) and the AR and the NR (right panel) at the surface and for the summer period (1 June – 31 August). The labels are longitude, degrees (x-axis) and latitude, degrees (y-axis). Red/blue colours indicate positive/negative values of the correlation coefficient.

We further show that, locally, the AR is capable of reproducing the peak in the CO distribution at the surface due to forest fires (albeit, weaker than the NR signal), even if the CR does not have the signature of the fires in its emission inventory. A second OSSE shows that this relatively weak signal of the forest fires in the AR arises from the use of a criterion to discard CO total column observations too far from model values, a criterion not appropriate to situations resulting in excessive values in the CO concentrations, as for forest fires. This second OSSE shows a much stronger signal in the AR, which is now much closer to the NR than the CR, confirming the benefit of S5P CO total column measurements.

The CO OSSE was also conducted for the three months during winter 2003- 2004 (November, December 2003 and January 2004). In contrast to the summer period, the winter experienced a generally cloudy situation. This period allowed us to study the impact of cloudy pixels in the OSSE. For this, we performed two OSSEs, one using all pixels with 100% cloud cover or less, and a second with pixels with 10% cloud cover or less.

3.7 The NO_2 and HCHO OSSEs

"NO_2 and HCHO OSSE results LOTOS-EUROS",
prepared by Renske Timmermans, Arjo Segers, Henk Eskes,
September 2015

This document contains the OSSE results for the Sentinel 5P and Sentinel 4 NO_2 and HCHO observations, using the LOTOS-EUROS model. It also includes the results from a delta study identifying the difference in impact of S5 versus S5P with a different overpass time.

This OSSE investigated the additional impact of the Sentinel 4 and Sentinel 5P observations of NO_2 and HCHO over the impact of the ground based ozone observations on the air quality analyses from the LOTOS-EUROS model. Surface NO_2 observations have not been used in the reference run, as the NO_2 observations are prone to contamination by other constituents. The assimilation of ground based ozone observations was the operational set-up of the model within the regional MACC (Monitoring Atmosphere and Composition Change) and CAMS (Copernicus Atmosphere Monitoring Service) services.

It is shown that both sentinel 4 and 5P NO_2 columns have a clear impact on modelled NO_2 values (Fig. 3.7.a and b). The additional assimilation of these observations on top of ground based ozone observations further decreases biases, RMSE and improves the temporal variability. The higher temporal resolution of the Sentinel 4 observations has a clear benefit resulting overall in a larger impact especially when the Sentinel 5P satellite has no observations. The added value of the satellite observations is visible in both modelled columns as well as in the surface concentrations of NO_2. For example, during the summer period over the zoom domain the RMSE in surface NO_2 is decreased by about 30% during daytime, while the temporal correlation is increased by the same amount. The impact on NO_2 columns is even larger.

In the winter period the positive impact of the surface ozone observations in some regions was counteracted through the additional assimilation of the satellite NO_2 observations. This was caused by a contradiction between the bias in satellite columns and bias in surface concentrations due to different NO_2 profiles in the MOCAGE nature run and LOTOS-EUROS. It is thus crucial to analyse the models performance for simulating NO_2 profiles.

The HCHO observations do show an added value in case of elevated HCHO values during a wildfire event (Fig.3.7.c). In other cases the noise in the product unfortunately is too large to provide a benefit to modelled HCHO fields.

When looking at surface ozone concentrations the satellite NO_2 and HCHO do not have a large influence, neither positive nor negative.

In a delta study it has been shown that the impact of S5P observations is very similar to the impact of S4 observations when only 13 UTC observations are used. Therefore we have used the sentinel 4 observations from 9 UTC as substitute for S5 observations to study the difference between the morning (S5) and afternoon (S5P) overpasses. The results show a similar benefit from both local times, except that the impact of sentinel 5P can be mainly seen in the afternoon while the impact of the S5 can be mainly seen in the morning (Figure 3.7a).

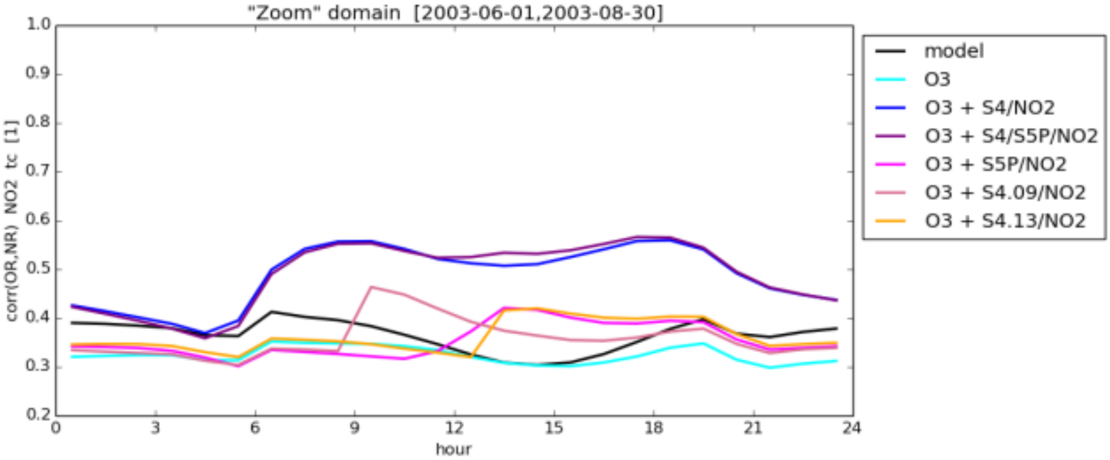


Fig. 3.7.a. Impact of the various satellite correlations (S4, S4+S5P, S5P, S4.09 and S4.13) on the NO_2 correlation between the nature run and assimilation runs, plotted as a function of the hour of the day. Sentinel 4 at 13h only has a very similar impact as S5P. The impact of S4 at 9utc is very similar to the impact of S4 at 13utc, which indicates that S5 and S5P will have very similar impacts. Note that in the LOTOS-EUROS implementation the improvements due to the S5(P) satellite observations remain for about 3 to 6 hours.

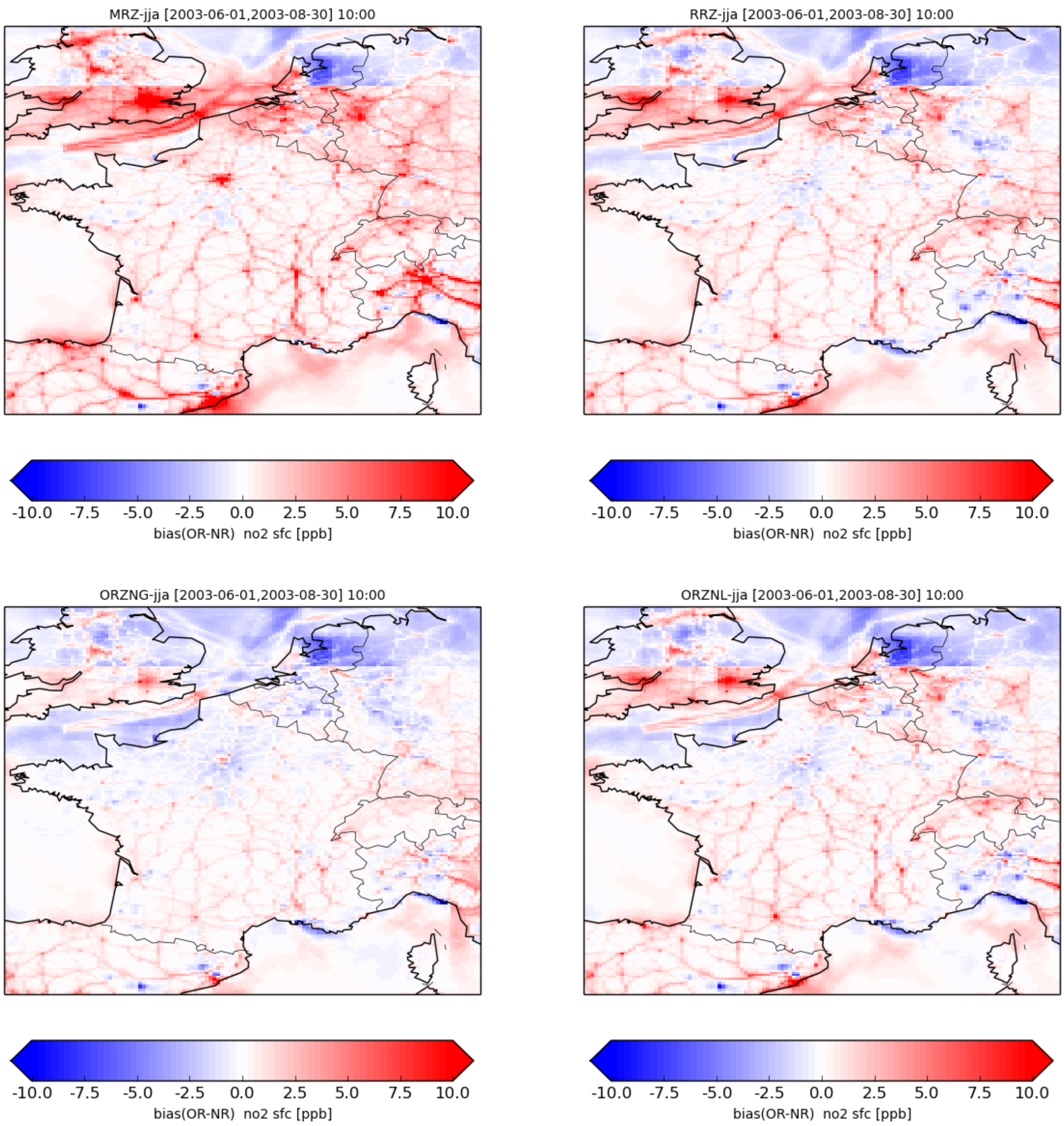


Fig. 3.7.b. NO_2 OSSE results with LOTOS-EUROS. Zoom domain, summer period, statistics over days at 10:00. Bias with nature run surface NO_2 before assimilation (top left), and after assimilation of ground-based O_3 derived from the MOCAGE nature run (top right), ground-based O_3 plus S4 NO_2 (bottom left), or ground-based O_3 plus S5P NO_2 (bottom right).

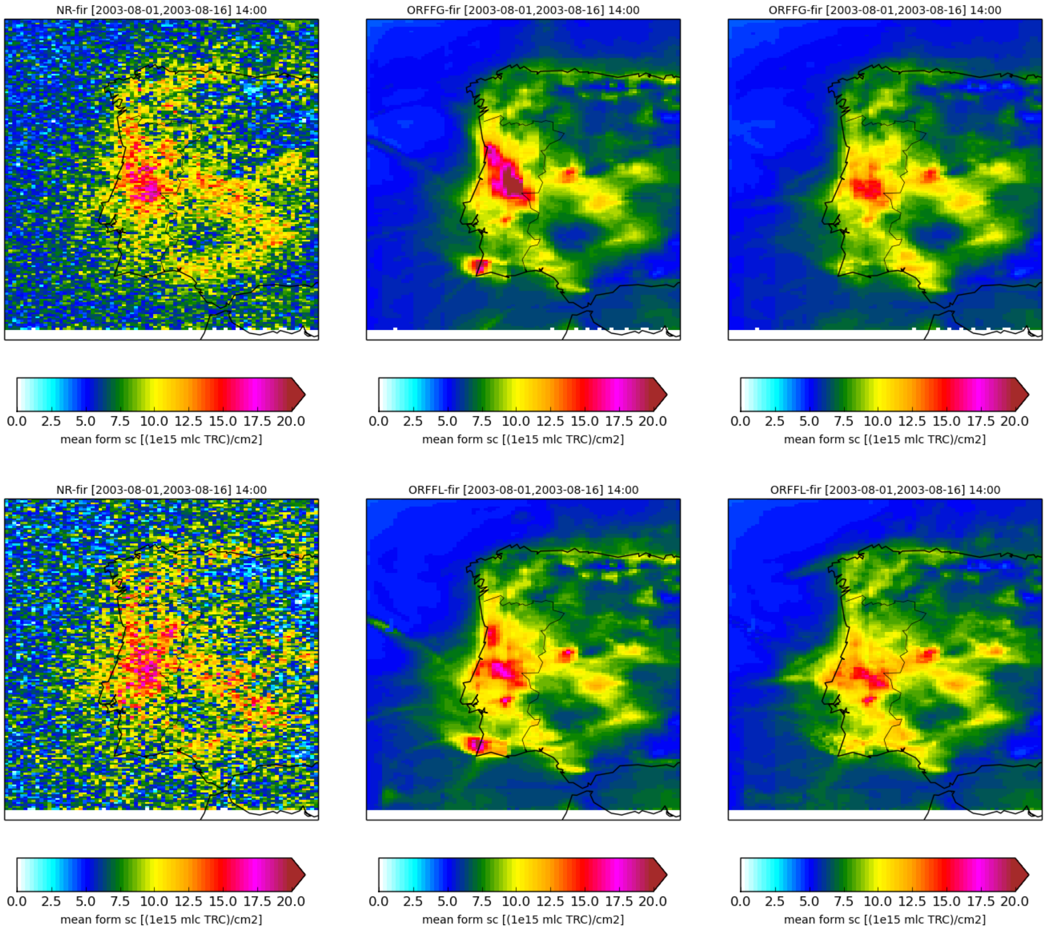


Fig. 3.7.c. Fire domain 1-16 August 2003 averaged synthetic HCHO columns at 14:00 (left) and collocated convolved HCHO columns from Model Run (middle) and Assimilation run (right) for O_3 ground-based plus S4 HCHO (top) and O_3 ground-based plus S5P HCHO (bottom).

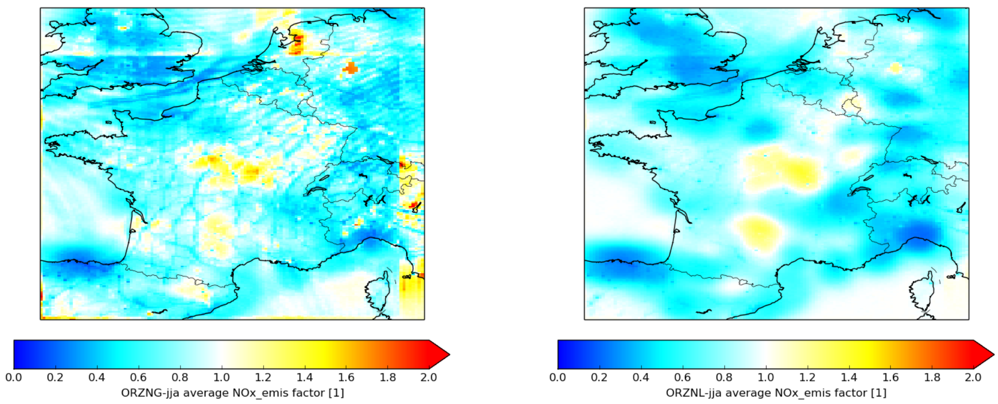


Fig. 3.7.d. Zoom domain, summer period: average NO_x emission increments for AR with S4 NO_2 observations (left) and AR with S5P NO_2 observations (right).

As mentioned earlier the data assimilation system updates emission factors in the model through the assimilation of the observations. Fig.3.7d shows that emission increments through the assimilation of S4 and S5P data are substantial.

3.8 The ozone OSSEs

"OSSE production and analysis (S4/S5P O3 OSSE)",
prepared by Jean-Luc Attié, Samuel Quesada-Ruiz, William Lahoz, Rachid Abida, Laaziz El Amraoui, Philippe Ricaud, Régina Zbinden and Henk Eskes,
November 2016

We performed several regional-scale Observing System Simulation Experiments (OSSEs) over Europe to explore the impact of GEO satellite mission S4 and LEO S5P ozone profile information on middle and lowermost tropospheric air pollution analyses. We focused on two periods during 2003-2004 separately: the summer of 2003 (JJA) and the winter 2003-2004 (NDJ). We also performed a 1-month OSSE considering cloudy pixels. In total we performed more than 3 years of assimilation to identify the added value of S4 and/or S5P ozone observations.

Special attention was given to the estimation of the representativity error, which is added to the retrieval error to obtain the total observation error used in the assimilation. In particular this leads to an increase of the error covariance for the leading retrieval modes with the highest signal-to-noise ratios. With the representativity error added the results and stability of the assimilation are improved.

These OSSE runs provide insight on the impact from LEO S5P and from GEO S4 O3 measurements on tropospheric O_3. We perform the standard steps of an OSSE for air quality, as identified in Timmermans et al. (2015), including a quantitative evaluation of the OSSE results, including statistical significance tests, and self-consistency tests. The useful statistical metrics for these OSSEs are mean absolute error (MAE), correlation coefficient and the skill score.

We focus on the surface and the troposphere by analysing the levels 200, 500 and 700 hPa. The reference run is the run with assimilation of the ground based station ozone data, as it is common in an operational system. The general conclusion of this study is that both S4 and S5P bring information from the middle troposphere to the upper troposphere. The maximum added value is above the height corresponding to 500 hPa. As expected, the assimilation of both S4 and S5P ozone shows better results than the control run and is closer to the Nature Run up to these altitudes (in terms of bias, skill score and correlation coefficient). The general conclusions for summer are:

- The behaviour of the assimilation runs with S4+S5P+GBS and S4+GBS is quite similar in terms of bias, variability (RMSE), reduction of RMSE (skill score) and in terms of correlation coefficient, and slightly better than the S5P ozone assimilation between 200 hPa and 700 hPa.
- There is a clear impact of S4 above the height corresponding to 500 hPa in terms of concentration (reduction of bias), and increase of correlation.
- Impact of S4 on the correlation coefficient for heights above 500 hPa.
- At 200hPa there is a reduction of bias from 60% to a more stable bias of about 30% (for S4+S5P+GBS). There is also a reduction in the variability (skill score) and a much better correlation with the NR.

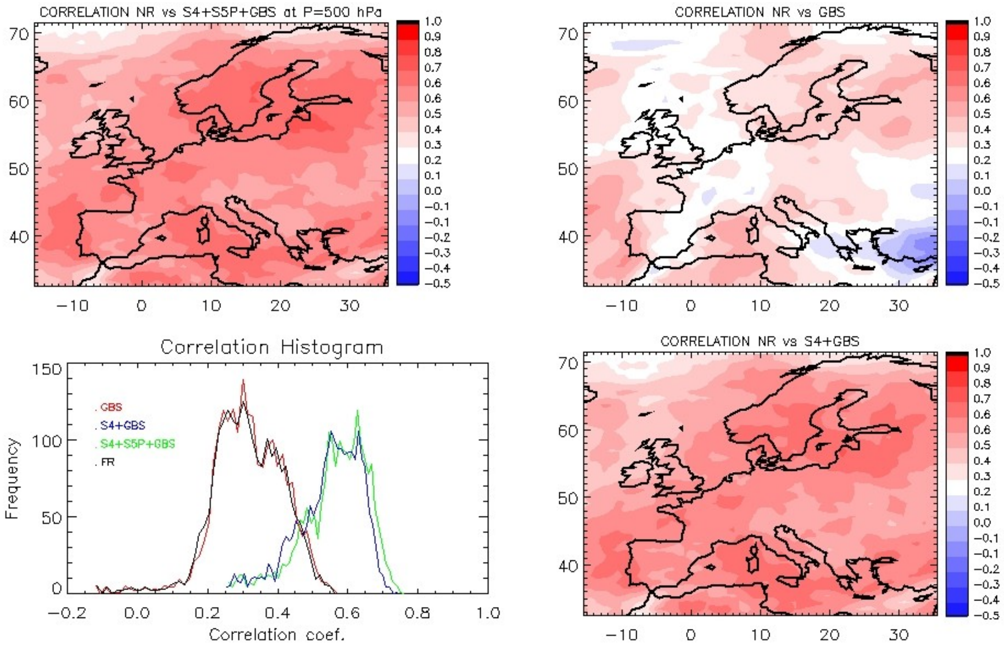


Fig. 3.8. Correlation scores at 500 hPa for S4, S5-P and the ground-based ozone observations (top left); for the ground-based data alone (top right); for S4+ground (bottom right). The histogram of the correlation coefficient during summer 2003 for all the assimilation runs and the free run (bottom-left) clearly demonstrates the improvement brought by the S4 data.

We did not find any significant impact at the surface from all the experiments. The system is strongly forced by the observations from the surface network, and the UV observations do not bring significant additional information. However, at 200 hPa the S4 increment values obtained are larger than at the surface, showing the added value obtained at this level from S4 UV ozone.

Headline message: Significant improvements from S5P and from S4 UV ozone retrievals were demonstrated during summer for O_3 information in the upper and middle troposphere for levels down to 500 hPa (i.e., heights above 500 hPa). Significant improvement for S5-P and S4 at 200 hPa was demonstrated for winter. No significant improvement was found at the surface. There is some improvement at 200 hPa when using only the cloudy pixels (cloudiness fraction greater than 0.4).

3.9 The impact of observations over clouds

The impact of observations over cloud-covered regions, and uncertainties were discussed in the following document deliverable,

> *Optimization and impact of observations made under cloudy conditions*
> *WP6, deliverables D8, D9,*
> *prepared by Henk Eskes and Arjo Segers,*
> *13 January 2017.*

The effect of using cloud-covered observations may be summarised as follows:

- For CO, adding cloudy pixels improves the analysis somewhat, by about 4% on average.
- For free tropospheric O_3, both clear and cloudy observations improve the analysis, but the results are somewhat better for the clear observations.
- For the analysis of surface NO_2, adding cloud-covered observations does not bring much added value. For columns the impact is somewhat higher.

There are two aspects of importance to understand these results: first, the cloud-cover prohibits the light to reach the surface, and the below cloud sensitivity is very small, leading to limited added value for surface concentrations, and second, the relative number of cloudy observations in the summer 2003 runs was rather small.

4 Summary of main project results: what have we learned

The ISOTROP project has produced a large series of carefully constructed OSSE experiments. An overview of the ISOTROP achievements was provided in the previous section. We performed the OSSE work following the recommended recipe for an OSSE for air quality, see Timmermans et al. (2015). These steps are:

- ✓ Production of high-resolution Nature Runs.
- ✓ Test of the realism of the Nature Runs by comparing with real observations.
- ✓ Generation of a set of synthetic observations based on state-of-the-art retrieval algorithms and realistic instrument characteristics, including detailed scene and geometry dependent averaging kernels and error estimates (covariances). Synthetic observations are produced for the future satellite observations as well as for the reference observations assimilated in the reference runs, which are surface observations from AIRBASE in our case.
- ✓ Use of models - independent from the model producing the Nature Runs - to produce the OSSE experiments, to create the control run or reference run, and the assimilation runs. The use of independent models is a requirement not to get overly optimistic results.
- ✓ Quantitative evaluation of the OSSE results, including performing statistical significance tests, and self-consistency and chi-squared tests.

There are more aspects which make the ISOTROP OSSEs state-of-the-art:

- High-resolution model runs have been performed, with a typical resolution close to the resolution of the satellites, which is about 7 km. With this we could study the full information content of the satellite instruments.
- Long-period datasets have been generated: three Summer months and three winter months, which make the results robust and highly significant, and which covers a range of weather regimes.
- A large domain was studied, namely the MACC domain which covers the largest part of Europe. This allowed us to investigate regional differences.
- A new approach was pioneered for the ozone profiles, which optimises the information content of the data sets for data assimilation applications.

The main conclusions can be summarized on a species-by-species basis:

Carbon monoxide:
The CO observations from S5P are expected to be of very high quality. The observations in the SWIR band show nearly constant averaging kernel profiles with high sensitivity at the surface over land. Typical estimated uncertainties are of the order 2-10%. Based on these synthetic observations the OSSE runs conducted demonstrate a very good skill to reproduce the Nature Run results over mainland Europe, Fig.3.6. Furthermore, the S5P CO total column observations are able to capture phenomena such as the forest fires that occurred in Portugal during summer 2003. Closer to the coast the results are more influenced by air masses coming from the ocean, less constrained by the observations which are much more uncertain over water. Cloud-covered observations over the ocean can be used, and the sensitive range is specified by the kernels in this case.

Nitrogen dioxide:
Compared to present-day capabilities (OMI) the nitrogen dioxide observations from S4, S5P and S5 bring considerable advances, namely 1) much improved resolution, from about 20 km to 7 km; 2) the hourly observations in the case of S4 providing full diurnal sampling; and 3) foreseen improved uncertainties (to about 15-30% for individual observations) due to advances in the characterisation of aspects like clouds, albedo, aerosol effects. The OSSEs were performed with an ensemble Kalman filter approach, which adjusts the NOx emissions, based on the NO_2 column observations. Despite the decrease of the NO_2 sensitivity towards the surface, the EnKF system is strongly adjusting also concentrations close to the surface. Note that this system is less flexible at remote locations with small emission fluxes. In the case of S4, many of the nature run features could be reconstructed, and the benefit is present throughout the entire day. For S5P or S5 a good impact was observed up to 3-6 hours after the overpass. With the increased observations resolution of 7 km we are able to provide constraints on source sectors such as road traffic, see Fig. 3.7.c.

Formaldehyde:
In Europe, mean concentrations of HCHO are relatively low compared to other parts of the World such as the tropics. For individual observations, the noise-dominated uncertainty is expected to be more than 100%, which results in very noisy images for individual orbits. When the data is averaged over a period of a week or longer, the distribution of concentrations starts to emerge from the noise, see Fig. 3.7.c. The LOTOS-EUROS assimilation system has not been specifically optimised to deal with such noisy daily data. Nevertheless, a positive impact from the data could be observed for the fire plumes over Portugal with elevated HCHO columns.

Ozone:
For the synthetic observations several choices were made. First, the spectral range was limited to 300-320 nm, discarding the shorter wavelengths, which allows us to retrieve ozone profiles at the full resolution of S4 and S5P (without co-adding). Secondly, we used the ideas of Migliorini to present the information content of the observations in a very efficient way to the (MOCAGE) assimilation system. At the proposal phase it was already clear that we could not expect major impacts in ozone at the surface based on UV ozone profile observations only. From these observations we obtain about one piece of information in the troposphere, with a larger sensitivity in the free troposphere compared to the boundary layer. On the other hand, ozone concentrations are constrained heavily in the boundary layer due to availability of a large number of hourly surface observations. Nevertheless, we could show that good impact from the S4 and S5P observations is found in the middle troposphere (Fig. 3.8). Cloud covered pixels contain similar or maybe even better information than the cloud free scenes.

5 Overview of available datasets produced during ISOTROP

Due to the high resolution of both the satellite instruments and the models, ISOTROP has produced large datasets. The ISOTROP database contains 1) the Nature Runs by MOCAGE, LOTOS-EUROS and TM5; 2) the synthetic observations for the four species; 3) a large number of OSSE runs.

5.1 Nature Run data sets

The Nature Run datasets are tabulated below. For more information we refer to the document "Description of Nature Runs used in ISOTROP OSSEs".

Model	**Domain**	**Resolution**	**Species**	**assimilation**
MOCAGE	MACC	0.2 x 0.2	NO2, HCHO	-
MOCAGE	MACC	0.2 x 0.2	O3, CO	Airbase O3
MOCAGE	Prev'Air Ext	0.1 x 0.1	NO2, HCHO	-
MOCAGE	Prev'Air Ext	0.1 x 0.1	O3, CO	Airbase O3

Table 5.1.a. MOCAGE Nature Runs. MACC domain: 15W- 35E, 35N-70N; PREVAIR extended domain: 5W-10E, 41N-53N. Period: 1-6-2003 to 1-9-2003, and 1-11-2003 to 1-2-2004.

Model	**Domain**	**Resolution**	**Species**	**assimilation**
LOTOS-EUROS	MACC	0.0625 x 0.125	NO2, HCHO, CO, O3	-
TM5	MACC	1 x 1	NO2, HCHO, O3, CO	-

Table 5.1.b. LOTOS-EUROS Nature Runs. MACC domain: 15W- 35E, 35N-70N; Period: 1-6-2003 to 1-9-2003, and 1-11-2003 to 1-2-2004. The LOTOS-EUROS runs extend to 3.5 km altitude. To construct the full vertical profiles, these lower tropospheric profiles have been extended into the stratosphere by adding the TM5 layers above 3.5 km.

5.2 Synthetic observation datasets

The available synthetic data are provided in orbit files for S5P and in hourly observation files for S4. The datasets are tabulated below.

Satellite	Species	Nature Run	Period
S4	NO2	MOCAGE	JJA 2003, NDJ 2003/4
S4	HCHO	MOCAGE	JJA 2003, NDJ 2003/4
S4	O3	LOTOS-EUROS TM5	JJA 2003, NDJ 2003/4
S5P	NO2	MOCAGE	JJA 2003, NDJ 2003/4
S5P	HCHO	MOCAGE	JJA 2003, NDJ 2003/4
S5P	O3	LOTOS-EUROS TM5	JJA 2003, NDJ 2003/4
S5P	CO	LOTOS-EUROS TM5	JJA 2003, NDJ 2003/4

Table 5.2. Available synthetic observations on the MACC domain: 15W- 35E, 35N-70N; Period: 1-6-2003 to 1-9-2003, and 1-11-2003 to 1-2-2004. To construct full Nature Run vertical profiles, the LOTOS-EUROS lower tropospheric profiles have been extended into the stratosphere by adding the TM5 layers above 3.5 km.

5.3 The OSSE runs

Run ID	Run	Domain	Resolution	Assimilation	
				Ground	Satellite
RRZ	Reference	PREVAIR ext	0.0625x0.125	Surface ozone	No
RRE		MACC	0.125x0.25	Surface ozone	No
RRF		Fire episode	0.0625x0.125	Surface ozone	No
ORZGN	OSSE, GEO, NO2	PREVAIR ext	0.0625x0.125	Surface ozone	GEO/S4 NO2
OREGN		MACC	0.125x0.25	Surface ozone	GEO/S4 NO2
ORFGN		Fire episode	0.0625x0.125	Surface ozone	GEO/S4 NO2
ORZLN	OSSE, LEO, NO2	PREVAIR ext	0.0625x0.125	Surface ozone	LEO/S5P NO2
ORELN		MACC	0.125x0.25	Surface ozone	LEO/S5P NO2
ORFLN		Fire episode	0.0625x0.125	Surface ozone	LEO/S5P NO2
ORZGF	OSSE, GEO, HCHO	PREVAIR ext	0.0625x0.125	Surface ozone	GEO/S4 HCHO
OREGF		MACC	0.125x0.25	Surface ozone	GEO/S4 HCHO
ORFGF		Fire episode	0.0625x0.125	Surface ozone	GEO/S4 HCHO
ORZLF	OSSE, LEO, HCHO	PREVAIR ext	0.0625x0.125	Surface ozone	LEO/S5P HCHO
ORELF		MACC	0.125x0.25	Surface ozone	LEO/S5P HCHO
ORFLF		Fire episode	0.0625x0.125	Surface ozone	LEO/S5P HCHO
ORZLGN	OSSE, GEO+LEO, NO2	PREVAIR ext	0.0625x0.125	Surface ozone	GEO/S4 NO2 LEO/S5P NO2
ORZNL09	OSSE, GEO 9h only, NO2	PREVAIR ext	0.0625x0.125	Surface ozone	GEO/S4 9h only NO2
ORZNL13	OSSE, GEO 13h only, NO2	PREVAIR ext	0.0625x0.125	Surface ozone	GEO/S4 13h only NO2

Table 5.3.a. The reference runs and OSSE runs conducted with the LOTOS-EUROS model. MACC domain: 15W-35E, 35N-70N; PREVAIR extended domain: 5W-10E, 41N-53N.

Run ID	Run	Domain	Resolution	Species	Assimilation	
					Ground	Satellite
RREC	Reference	MACC	0.2°x0.2°	CO	no	no
RRFC		Fire episode				
ORELC100	OSSE, LEO	MACC	0.2°x0.2°	CO	no	LE0/S5P CO
ORFLC100		Fire episode				
ORELC10	OSSE, LEO CF < 10%	MACC	0.2°x0.2°	CO	no	LEO/S5P CO
RREC	Free run	MACC	0.2°x0.2°	O3	no	no
RRLO (JJA)	LEO	MACC	0.2°x0.2°	O3	no	LE0/S5P O3
RRLGO (JJA)	OSSE, LEO+ GEO	MACC	0.2°x0.2°	O3	no	LEO/S5P+GEO/S4 O3
RRGO (JJA)	OSSE, GEO	MACC	0.2°x0.2°	O3	no	GEO/S4
RRLO	Reference	MACC / Fire episode	0.2°x0.2°	O3	yes	no
ORELO	OSSE, LEO	MACC / Fire episode	O.2°x0.2°	O3	yes	LEO/S5P O3
ORELGO	OSSE, LEO+GEO	MACC / Fire episode	0.2°x0.2°	O3	yes	LEO/S5P+GEO/S4 O3
OREGO	OSSE, GEO	MACC / Fire episode	0.2°x0.2°	O3	yes	GEO/S4
OREGOC (June)	OSSE, GEO	MACC	0.2°x0.2°	O3	yes	Cloudy S4 pixels
ORELOC (June)	OSSE, LEO	MACC	0.2°x0.2°	O3	yes	Cloudy S5P pixels
OREGOR1 (June)	TEST R_obs=1	MACC	0.2°x0.2°	O3	no	GEO/S4

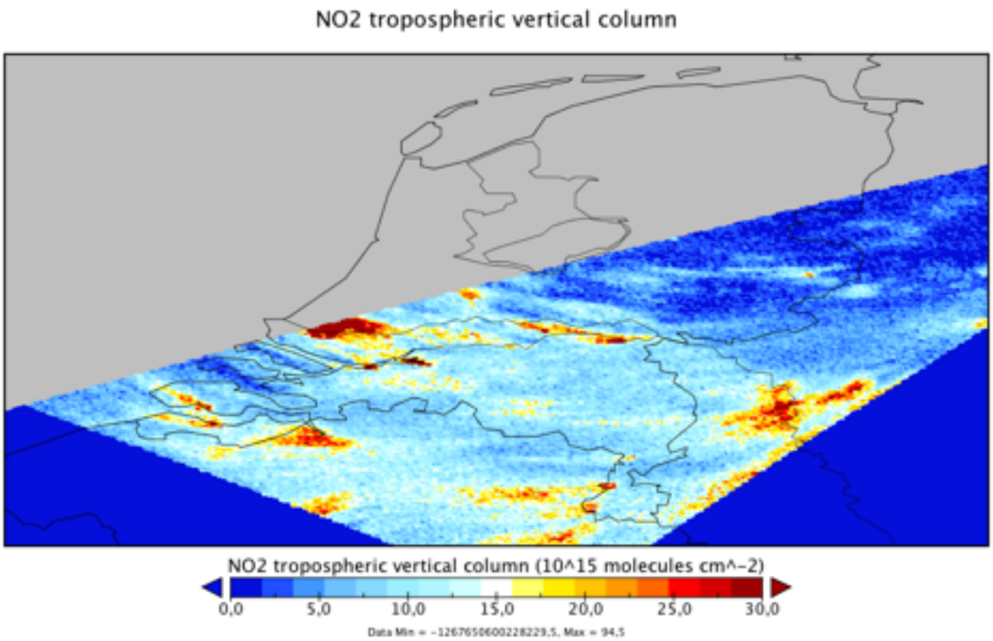
Table 5.3.b: List of runs for MOCAGE, including domain (MACC domain = 15W-35E, 35N-70N), model resolution, species included and synthetic observations assimilated.

6 Spin-off

The ISOTROP products have in the past years been used for several other projects and studies. The ISOTROP results have been discussed at several conferences and scientific publications have been written.

6.1 Use of ISOTROP datasets and software for other projects and proposals

- The high-resolution ISOTROP Nature Run simulation with MOCAGE was used in the paper by de Laat et al. (2014). These runs were useful to estimate variability of CO along the flight path, important to understand space-based vs. aircraft observations.
- The ISOTROP team has provided support to the L2 preparations for Sentinel 4. The high-resolution simulations of LOTOS-EUROS, combined with TM5 have been used by the retrieval teams involved. Synthetic observations simulated within ISOTROP have been made available to the S4 L2 teams.
- TNO is exploring small (satellite) instruments that can measure NO2 with very high resolution (TROPOLITE concept). The ISOTROP synthetic observations software will be used to simulate synthetic observations for these instruments.
- Météo-France/CNRS-GAME and NILU have used results from the S4 ozone OSSE for the studies done for an EE9 mission concept called MAGEAQ.
- The software to produce the ISOTROP synthetic observations has been adapted to simulate observations at even higher resolution (1 km). These simulations have been used in a satellite mission concept for EE9, called NITROSAT. The figure below shows an example of an orbit overpass over the Netherlands and the corresponding NO_2 simulated observations with 1km footprints.



6.2 Publications

- During the project, the ISOTROP team has prepared a review paper on atmospheric chemistry OSSE studies (Timmermans et al., 2015).
- The CO OSSE results for S5P have been submitted (Abida et al., accepted for ACP, 2016)
- Two more papers are planned: one describing the ozone OSSE, and one describing the combined NO2 and HCHO OSSE, both for S4 and S5P.

6.3 Presentations at workshops

- ISOTROP partners have participated in the first OSSE workshop, held at ECMWF, in October 2012. The ISOTROP project plans were presented and discussed.
- Henk Eskes has presented the ISOTROP project during a visit in Japan, May 2013.
- ISOTROP was presented orally at ESA Living Planet Symposium, September 2013.
- The CO OSSE results were presented by Jean-Luc Attié at the AGU of December 2014.
- Henk Eskes presented the ISOTROP project results at the CEOS ACC-11 meeting, ESRIN, 28-30 April 2015.
- Henk Eskes presented the ISOTROP project results at the AGU in December 2015.
- Four ISOTROP team members participated in the second OSSE workshop, Reading, 9-11 November 2016. A session was devoted to ISOTROP results, with the following presentations:
    - An OSSE to Study the Impact of Sentinel S4, S5P and S5 Spaceborne Observations on Air Quality Data Assimilation Systems (H. Eskes, KNMI)
    - Benefit of future S4-UVN and S5P ozone measurements: an ISOTROP study (W. Lahoz, NILU)
    - The impact of Sentinel 4 and 5P observations of NO2 (and HCHO) on air quality analyses, Results and limitations from the ISOTROP study (R. Timmermans et al., TNO)

7 Recommendations

ISOTROP was an ambitious project, including OSSE studies for 4 compounds for both geostationary and polar platforms. The project has mainly focussed on demonstrating the impact of the observations on the model concentrations and on the degree in which the Nature Run simulations could be recovered based on the synthetic satellite datasets. As such we see the ISOTROP results as a first step and there is room for complementary studies to answer the five science questions in more detail. The OSSE setups can be further optimised to answer important questions like how much the satellite observations can help to improve our understanding of air pollutant concentrations in the planetary boundary layer, to quantify long-range transport, to improve emission fluxes, and to improve our understanding of chemical composition and processes in the troposphere.

In this context there are a number of recommendations and suggestions for possible follow-up studies:

- **International collaboration.** There is a lot of attention internationally for performing OSSE experiments for atmospheric chemistry. In particular, OSSEs are discussed internationally in the context of CEOS and the constellation of geostationary platforms for air pollution monitoring. Further international collaboration would be advised and can be sought in this respect. For instance, models, synthetic observations and Nature Runs could be shared with American and Asian teams, and results should be discussed internationally at events such as the OSSE workshop held in Reading, November 2016.
- **Ozone OSSE**. The ISOTROP ozone OSSE was limited to observations derived from the UV, but this by itself is of limited use for air quality and boundary layer composition. A clear recommendation is the combined use of observations from various wavelength ranges, UV, Infrared and possibly the visible part of the spectrum. Several studies have already proved that such combinations will largely increase the information content in the troposphere, and will add information to the lowest few kilometres. An example would be the combination of observations from Sentinel 4 and IRS.
- **Meteorology**: The future satellite monitoring capabilities will have an order of magnitude resolution increases compared to present-day observations. In ISOTROP we decided to focus on 2003 because of the heat wave and fire conditions. A drawback of this approach was that the available meteorological datasets are relatively coarse, and do not reflect the amount of detail resolved by the satellites. For future studies we recommend the use of more modern meteorological analyses at resolutions, which are achievable today. This will give a more realistic impression of the impact of the future observations for e.g. emission inversions.
- **Spatial resolution of the clouds and albedo maps**. The ISOTROP synthetic observations are based on relatively coarse resolution albedo maps and coarse resolution model clouds. Furthermore, we find differences between the ISOTROP cloud simulations and observations by OMI, which observes on average larger cloud fractions. Future OSSE studies could improve upon these

aspects by using a high-resolution meteorological driver, and by using e.g. albedo maps from MODIS or similar instruments.

- **Treatment of clouds and aerosols in OSSE**. The approach we have followed is to compute synthetic satellite observations of cloud cover and height by using the cloud information for the meteorological drivers. Future OSSEs could investigate other observation-based approaches, for instance by using geostationary cloud observations. In the ISOTROP approach observations are produced for all fractional cloud covers. An alternative approach would be to start with cloud filtering, and to retrieve only the clear-sky observations, accounting more explicitly for aerosols. Such alternative approaches have different error budgets and may result in different impacts on air quality models. Understanding the impacts of such choices is important.
- **Multi-species OSSE**. The LOTOS-EUROS simulations clearly show that observations of one species impact the others. The chemistry of ozone, CO, NO2 and HCHO is tightly linked. In ISOTROP we investigated the impacts of the observations individually. A full impact study of S4/S5/S5P data, however, should assimilate all these observations simultaneously. This is a clear recommendation for future OSSE studies.
- **Formaldehyde.** Formaldehyde observations are special, because of the large uncertainty levels, which is largely of a random noise-like character. The LOTOS-EUROS system is not optimised to deal with such information, and the impact seen is rather small. It is a clear challenge and recommendation to develop other assimilation strategies (e.g. a 4D-Var with a long time window) or pre-processing strategies for the observations (spatial/temporal averaging) to exploit noisy data and to extract the information contained. Because of the large number and high density of observations we are confident that systems will be developed that can deal with the data efficiently.
- **Formaldehyde retrievals.** The synthetic observations for formaldehyde are based on typical slant column errors of 1.2e16 molecules/cm^2. (Source: S5P/TROPOMI HCHO ATBD, 2015) However, more recent results for e.g. OMI suggest that this number is conservative, and 0.8e16 molecules/cm^2 may be achievable/realistic by optimising the DOAS fitting procedure (results obtained in the European QA4ECV project, Isabelle de Smedt private communications). This suggests that a larger impact of the observations may be foreseen than what is found in the ISOTROP HCHO OSSE.
- **CO observations above the ocean**. Carbon monoxide observations above cloud-free ocean are very noisy due to the low surface albedo and not very useful. However, the cloud-covered oceans have a good signal. It is advised to study such cases in more detail and to better quantify the uncertainties, and finally to set up an ocean OSSE.
- **Sensitivity to OSSE components**. The outcome of the OSSE experiments is a result of all the details of the components of the entire OSSE system: the synthetic observation characteristics and uncertainty estimates, the assimilation approach, treatment of the observations in the assimilation, and modelling details. In general, it is good that OSSE experiments are repeated by other groups to test the results and the OSSE components. In particular, the impact of the assimilation approach is an interesting aspect to study. The Nature Runs in ISOTROP have been assessed, but mainly at the surface. For instance, for the ozone and NO_2 OSSE the realism of the vertical profiles is important and additional studies could be defined accordingly.

- **Emissions:** Improving emissions top-down by assimilating satellite observations is a study by itself. In ISOTROP we have applied the Ensemble Kalman filter, which adjusts emissions to improve the concentrations. We could show that the NO_2 observations were efficiently assimilated, which adjusted the emissions, which in turn brought the model results in closer agreement with the Nature Run. However, model uncertainties especially concerning chemical processes influence the emission estimates. A multi-species assimilation may be an efficient approach to reduce uncertainties in the model state and processes, and in this way improved the emission estimates. This could be a topic for future OSSEs.
- **Efficient interface to ozone observations:** One of the innovations of ISOTROP is the delivery of the ozone profile information in the form of leading eigenvectors of the radiative transfer code. This represents a very efficient and convenient interface between the retrievals and data assimilation systems. The use of this approach is a clear recommendation for future OSSEs, but also for the delivery of new reprocessing optimal estimation datasets of existing satellite instruments (not only ozone profiles).

8 References

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