ESA project

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

ESA Contract No: **4000105743**

Document on

# $NO_2$ and HCHO OSSE results with LOTOS-EUROS

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<tr><td colspan="4">ABSTRACT:<br>This document contains the OSSE results for the Sentinel 5P and Sentinel 4 NO2 and HCHO observations, using the LOTOS-EUROS model. It includes the results from a delta study identifying the difference in impact of S5 versus S5P with a different overpass time.</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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# Contents

# 1 Introduction

Within the ISOTROP project an OSSE study is performed to quantify the impact of sentinels 4 and 5 observations of $O_3$, $NO_2$, HCHO and CO on air quality analyses. This is done by employing two data-assimilation systems, one around the LOTOS-EUROS model and one around the MOCAGE model. The assimilation systems are used to assess the value of a LEO+GEO satellite observation system for tropospheric composition monitoring and forecast.

In this document we describe the impact of the sentinel 4 (S4) and sentinel 5P (S5P) observations of $NO_2$ and HCHO on the air quality analyses from the LOTOS-EUROS model. The synthetic observations have been produced from the nature run by the MOCAGE model. The satellite products are always assimilated in conjunction with groundbased ozone observations (gb $O_3$). Results are compared to a reference run (RR) with assimilation of gb $O_3$ only or the model run (MR) without any data assimilation.

In the study we have focused on four different domains:

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

and two study periods:

- **Summer** (June - August 2003), with specific two weeks **fire episode** 1-16 August 2003
- **Winter** (November 2003 - January 2004)

The evaluations are focusing on the analyses of $NO_2$, ozone and HCHO where we distinguish between three types of variables:

- **Satellite columns**, where we directly compare the synthetic satellite observations with the collocated (in space and time) values from the model that are convolved with the provided averaging kernels to produce a column value representing the satellite product.
- **Total columns**, where we compare the gridded LOTOS-EUROS $NO_2$ columns (without applying averaging kernels) to the gridded $NO_2$ columns from the nature run. It is unclear if these columnar values are representing the same altitude range and should therefore be considered with care.
- **Surface concentrations,** where we compare gridded LOTOS-EUROS surface concentrations with the surface concentrations from the nature run.

In the sections on $NO_2$ and HCHO we will focus on satellite columns before moving to total columns and surface concentrations. The results for the satellite columns show the most direct impact of the data assimilation; it can provide confidence in the performance of the data assimilation system and is needed to explain the impact of the synthetic satellite observations on surface level concentrations.

We will not show the impact on the modelled CO fields as this impact is not detected in our results.

As the initial objective of the study was to determine the impact of the sentinel 5 with an overpass time in the morning instead of the impact of the sentinel 5P with an overpass in the early afternoon, one of the chapters in this report is devoted to the results of a delta study evaluating the difference in impact of S5 versus S5P observations. For the delta study the runs over the zoom domain are repeated assimilating the S4 observations for 9:00 UTC (hereafter referred to as S4.09) or 13:00 UTC (hereafter referred to as S4.13) only.

## 1.1 Statistical evaluation

The main goal of this report is to provide a quantitative assessment of the added value of S5P and S4 NO2 and HCHO observations on the assimilation analysis at the surface. To achieve this we perform statistical analysis of the LOTOS-EUROS model results in comparison to the Nature run results. The following diagnostics are used:

- Mean bias (MB): $$MB(X) = \frac{1}{N}\sum(X - NR)$$
- Root mean square error (RMSE): $$RMSE(X) = \sqrt{\frac{1}{N}\sum(X - NR)^2}$$
- Temporal correlation ($R^2$): $$R^2 = \left(\frac{\sum(X-\bar{X})(NR-\overline{NR})}{\sqrt{\sum(X-\bar{X})^2\sum(NR-\overline{NR})^2}}\right)^2$$

Where X are the modelled values from RR, CR or AR, NR are the values from the Nature Run, and N is the number of values over which the mean is taken and varies between the different plots.

## 1.2 Assimilation Runs overview

**Table 1 List of reference and assimilation runs for LOTOS-EUROS, including domain, model resolution, species included and synthetic observations assimilated**

| 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 |

# 2 Impact on $NO_2$

## 2.1 Emission increments

As described in the assimilation scheme description document (ASDD), the data assimilation system in LOTOS-EUROS is an active system that defines uncertainties for a number of model parameters, in this case for example NOx and VOC emissions. The assimilation of observations then leads to updates of these input emissions. Such an active data assimilation system is especially useful when looking at applications such as emission inversions and air quality forecasts as it does not only update the state of the atmosphere (e.g. $NO_2$ concentrations) but also the driving input parameters.

Figure 1 shows the average NOx and VOC emission increments for the summer period over the zoom domain for the different assimilation runs. It can be seen that the assimilation of groundbased ozone observations generally leads to a decrease of the NOx emissions over most of the domain, while the effect on VOC emissions is more mixed. The assimilation of S4 $NO_2$ observations has some large additional impact over some areas of the domain. For example the NOx emissions over the centre of the Netherlands are increased, while the NOx emissions over the shipping route in the English channel are decreased. The VOC emissions show a clear increase just West of Marseille. The impact of the S5P observations on the NOx emissions on average is smaller than the impact of the S4 observations, which is expected as the S4 has a higher temporal resolution, thus more observations. But still in the VOC emission increments a clear additional impact of the S5P observations can be seen, different from the S4 impact. What is also visible is the smaller impact length scale of the satellite observations (smaller scale structures visible) in comparison to the length scale set for the groundbased observations, as described in the ASDD document. The box-shaped offset centred over France which is visible in several figures in this report is caused by the Prevair domain using different meteorology than the area surrounding this domain (see ASDD document section 4.3.1).

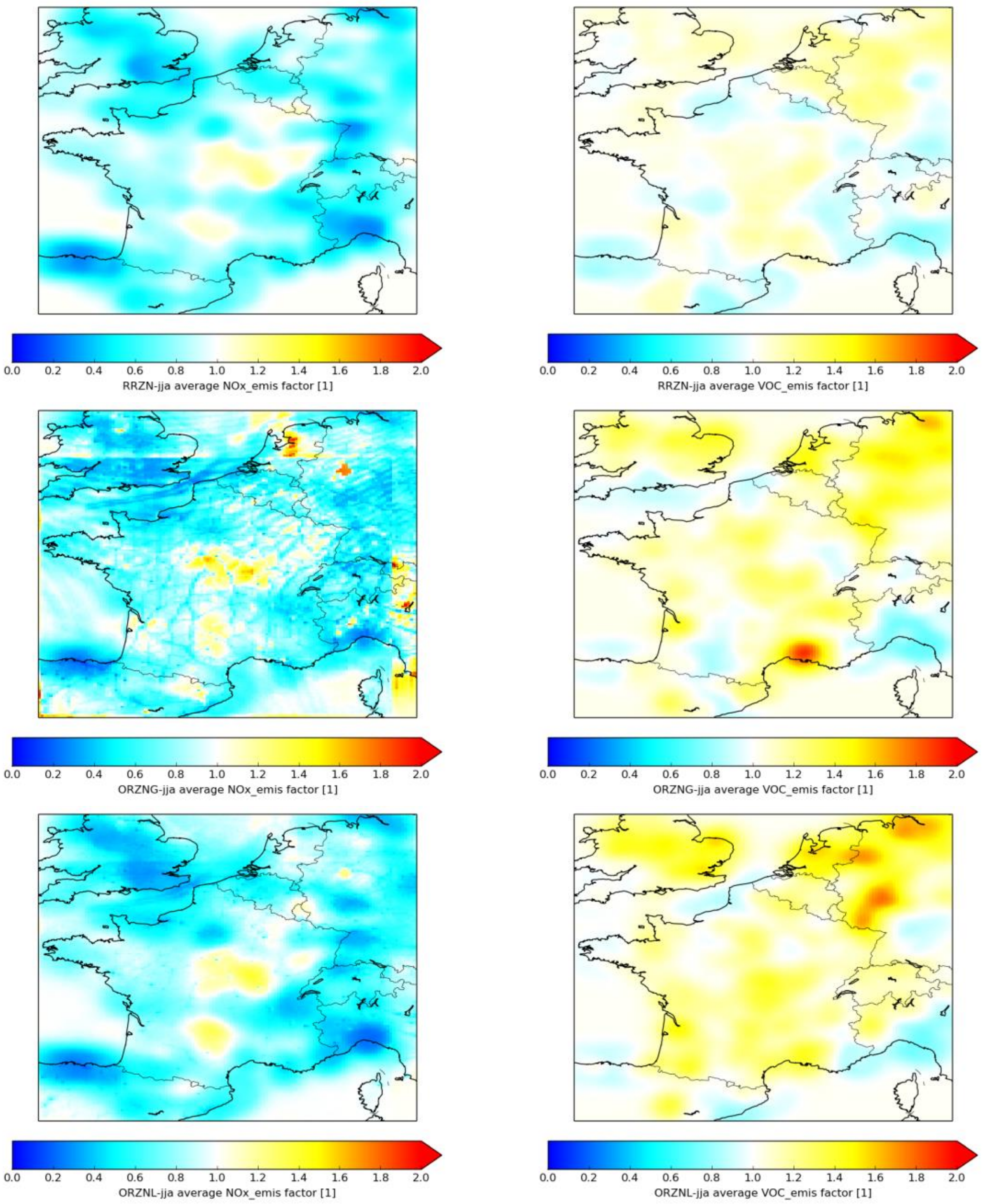


**Figure 1 Zoom domain, summer period: average $NO_x$ (left column) and VOC (right column) emission increments for RR (top panels), AR with S4 $NO_2$ observations (middle panels) and AR with S5P $NO_2$ observations (bottom panels).**

The added value of these emission increments will be evaluated by looking at the impact of the increments on modelled NO2 concentrations in the following sections.

## 2.2 Impact on satellite NO2 columns

Figure 2 shows the averaged synthetic sentinel 4 (S4) and Sentinel 5P (S5P) $NO_2$ observations at 14h over Europe during the summer period, versus the results from the MR run and the assimilation run. Ideally we would like to have had the RR (assimilation of gb $O_3$) results here instead of MR (model run without assimilation), but this would have required simulation of satellite columns from all possible instruments during the RR; a standard model run is however always performed together with an assimilation and therefore simulations of the satellite instruments with a MR are available. It can be seen that the MR shows higher $NO_2$ columns mainly over the Benelux and the UK as compared to the synthetic observations from the nature run. An extensive analysis of the differences between NR and

MR has been provided in the ISOTROP Nature runs description document. Differences are mainly due to the different emissions used and the different temporal profiles of the emissions in both models. When assimilated in combination with the ozone groundbased observations both the S4 $NO_2$ column observations as well as the S5P $NO_2$ column observations are able to largely close the gap between the modelled fields and the synthetic observations. This can also be seen in Figure 3 where the statistical parameters bias, RMSE and temporal correlation versus the synthetic observations are plotted. The positive bias and RMSE are largely reduced, while the temporal correlation is increased considerably over a large part of the domain. Over area's with low $NO_2$ columns the temporal correlation remains low, since here the noise in the satellite retrievals is much higher than the absolute values, and the temporal correlation is effectively close to zero.

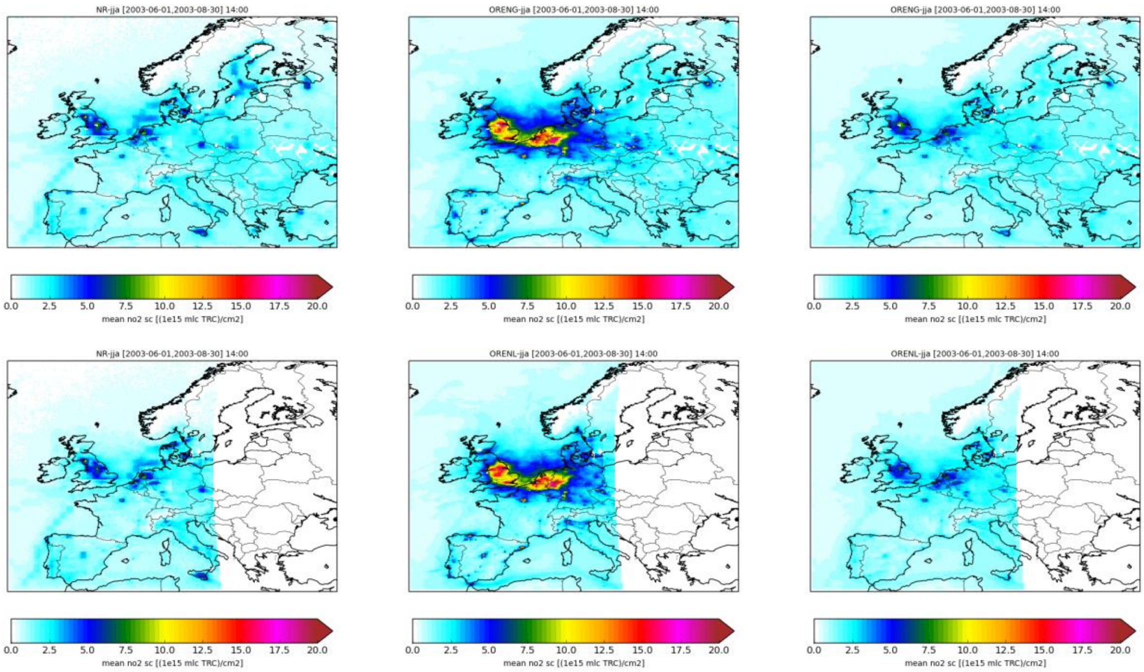


**Figure 2 Europe-summer period averaged synthetic $NO_2$ columns at 14h (left) and collocated convolved $NO_2$ columns from Model Run (middle) and Assimilation run (right) for $O_3$ gb + S4 $NO_2$ (top) and $O_3$ gb+ S5P $NO_2$ (bottom).**

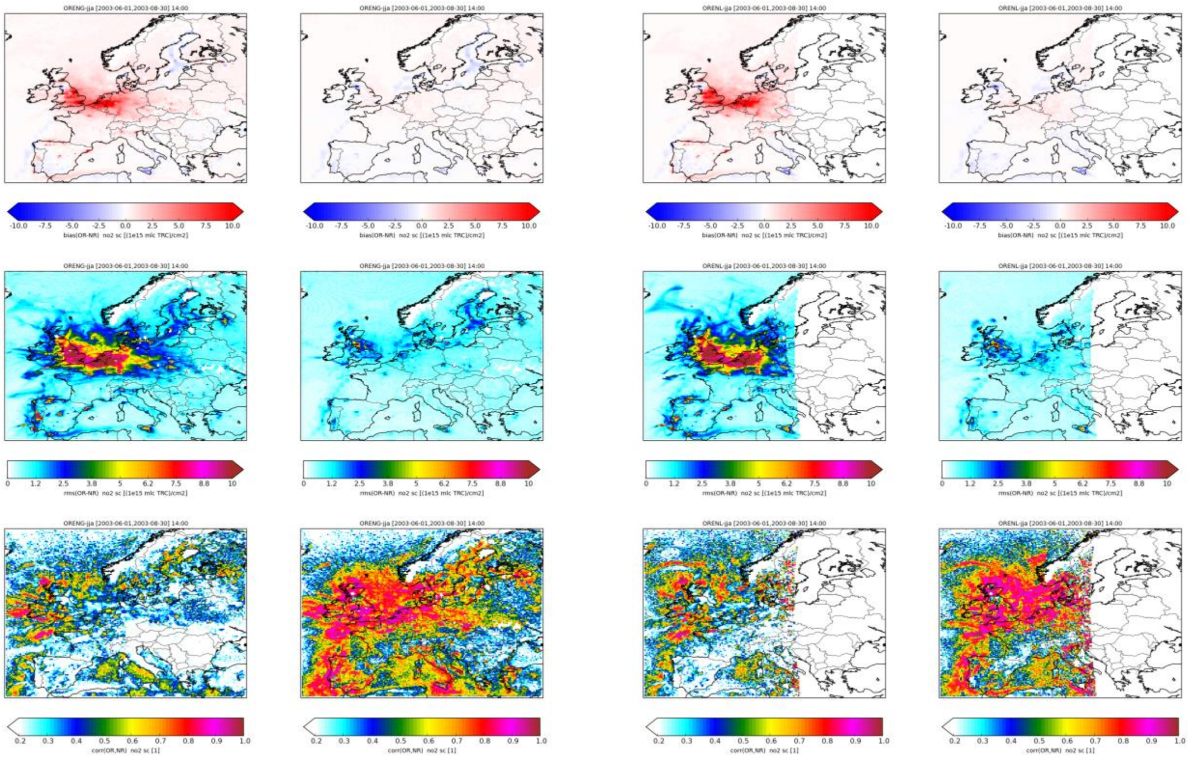

**Figure 3 Europe – summer period: Bias (top row), RMSE (middle row) and correlation (bottom row) with synthetic observations for $O_3$ gb+S4 $NO_2$ (left two columns) and $O_3$ gb+S5P $NO_2$ (right two columns). First column for each instrument is from Model Run without data assimilation, second column is from assimilation run with assimilation of synthetic observations.**

Figure 4 and Figure 5 show the results for the winter study period. It can again be seen that the satellite observations from both S4 and S5P decrease the positive biases and the RMSE, and increase the temporal correlation. In the winter period the modelled values also show some large negative biases over large parts of the domain, these biases are decreased through assimilation of the satellite $NO_2$ columns however to a much lesser extent than the decrease in the positive biases. In Eastern Europe the modelled values are lower than the observations and the observations are quite high. As the error in the observations is a relative error, a high column also means a high error leading to a smaller impact of the observations. Near the Eastern boundary of the model domain adjustment of the model is also hard as the concentrations are can be largely influenced by the boundary conditions in case of Eastern winds.

The negative biases are also seen over the Atlantic and North Sea. In these areas the model does not have any NOx emission sources except for the shipping emissions and therefore the system will not be able to increase the $NO_2$ values through changing emissions.

The square box which can be seen in the synthetic data is a feature of the nature run from which they have been derived, which combined results from a low resolution European run with a high resolution zoom.

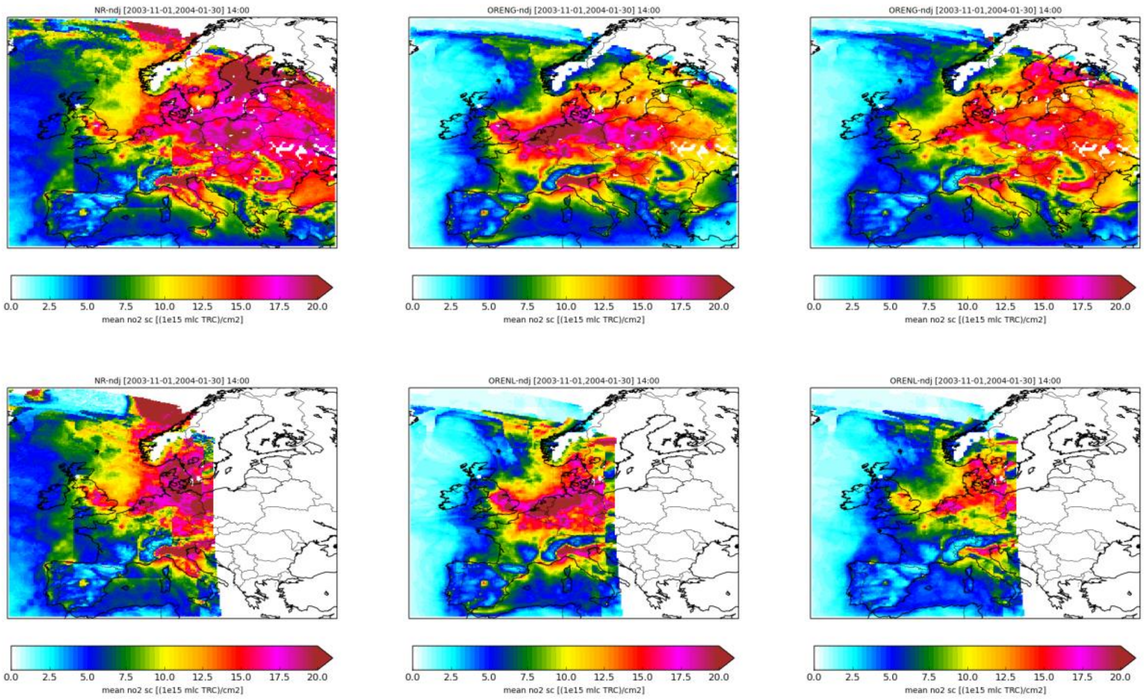


**Figure 4 Europe-winter period averaged synthetic $NO_2$ columns at 14h (left) and collocated convolved $NO_2$ columns from Model Run (middle) and Assimilation run (right) for $O_3$ gb+S4 $NO_2$ (top) and $O_3$ gb+S5P $NO_2$ (bottom).**

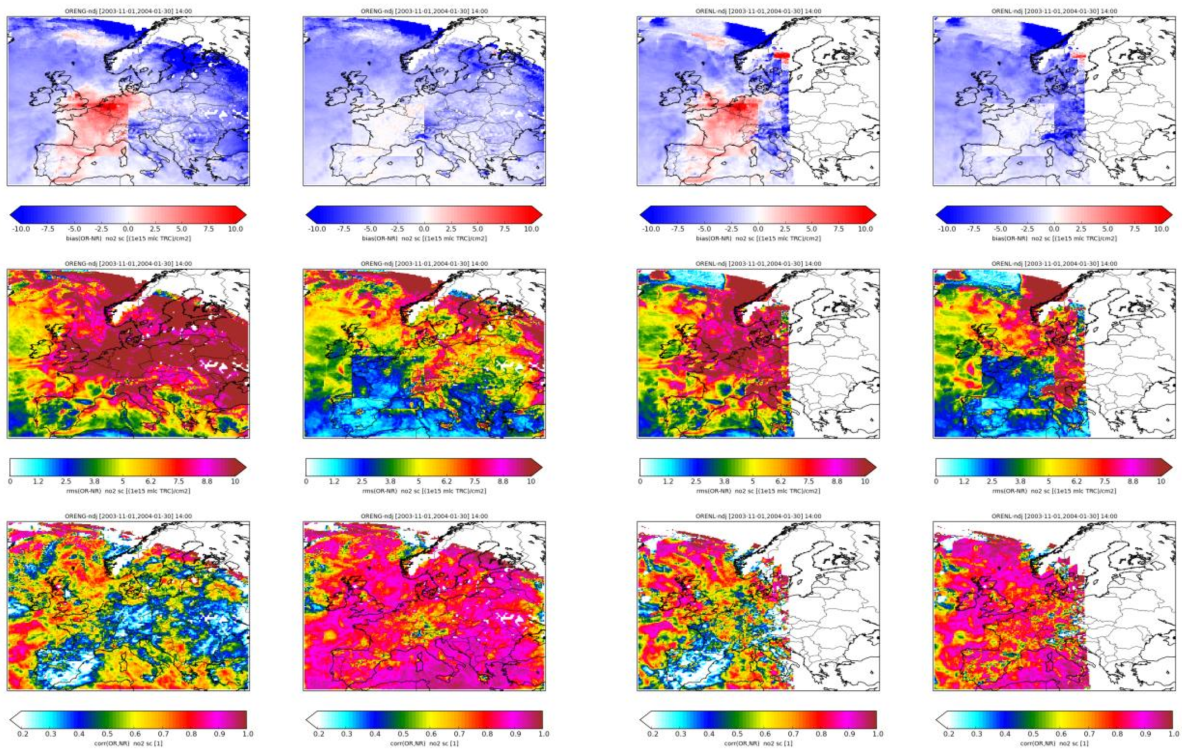


**Figure 5 Europe – winter period: Bias (top row), RMSE (middle row) and correlation (bottom row) with synthetic observations for $O_3$ gb+S4 $NO_2$ (left two columns) and $O_3$ gb+S5P $NO_2$ (right two columns). First column for each instrument is from Model Run without data assimilation, second column is from assimilation run with assimilation of synthetic observations**

The results for the satellite columns provide evidence that the data assimilation system is working as expected and able to move the modelled fields closer to the observations. Both the S4 and S5P observations provide a positive impact on the modelled satellite $NO_2$ columns for the European domain. The same holds for the other investigated domains. Figure 6 shows the statistics for the satellite $NO_2$ columns as function of hour of the day for the zoom domain over the summer study period. The Sentinel 5P satellite only comes over once a day and therefore only values around these overpass times are available and plotted for S5P. The impact of the S4 and S5P instruments on the bias, RMSE and temporal correlation is similar around 14h while the impact of S5P is slightly lower before and after that time due to fewer observations.

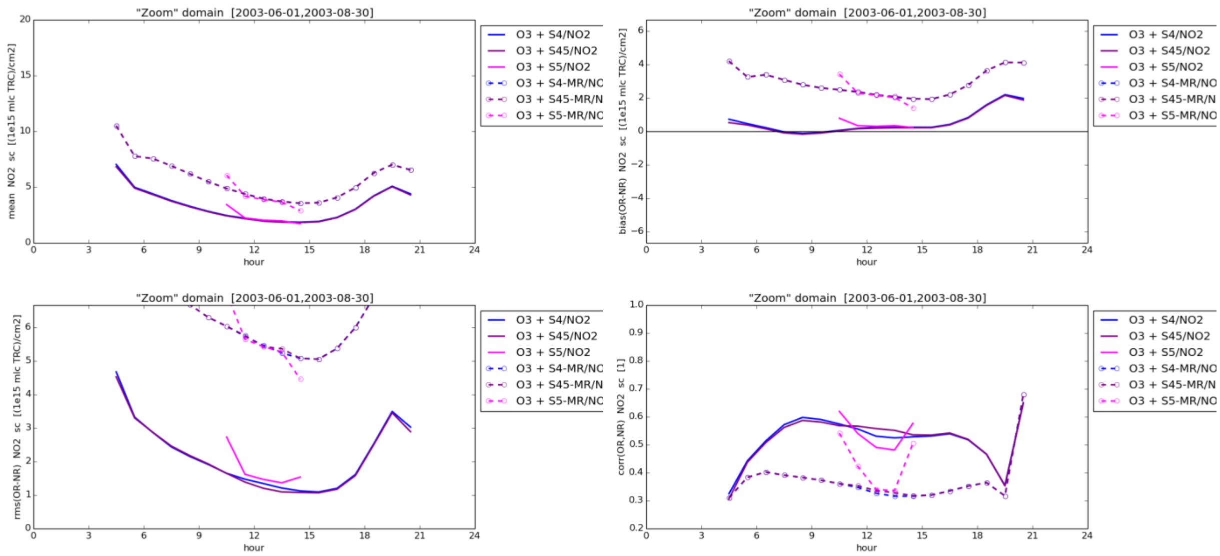


**Figure 6 Zoom domain – summer: satellite $NO_2$ column (top left), bias (top right), RMSE (bottom left) and correlation (bottom right) without assimilation (Model Runs, dashed lines) and with assimilation (solid lines) of $O_3$gb+S4 (blue), $O_3$ gb+S5P (pink) or $O_3$gb+S4 and S5P (purple) $NO_2$ observations.**

## 2.3 Impact on total $NO_2$ columns

The potential benefit of the high temporal resolution of the S4 observations can not easily be visualized by looking at the satellite column results, as these only take into account the results at overpass times of the satellites. For this reason we also have evaluated the analysed total $NO_2$ columns at all times during the day and over the entire grid. Figure 7 shows the same statistical results as Figure 6 but now for the summer total $NO_2$ columns over the zoom domain as function of time of the day.

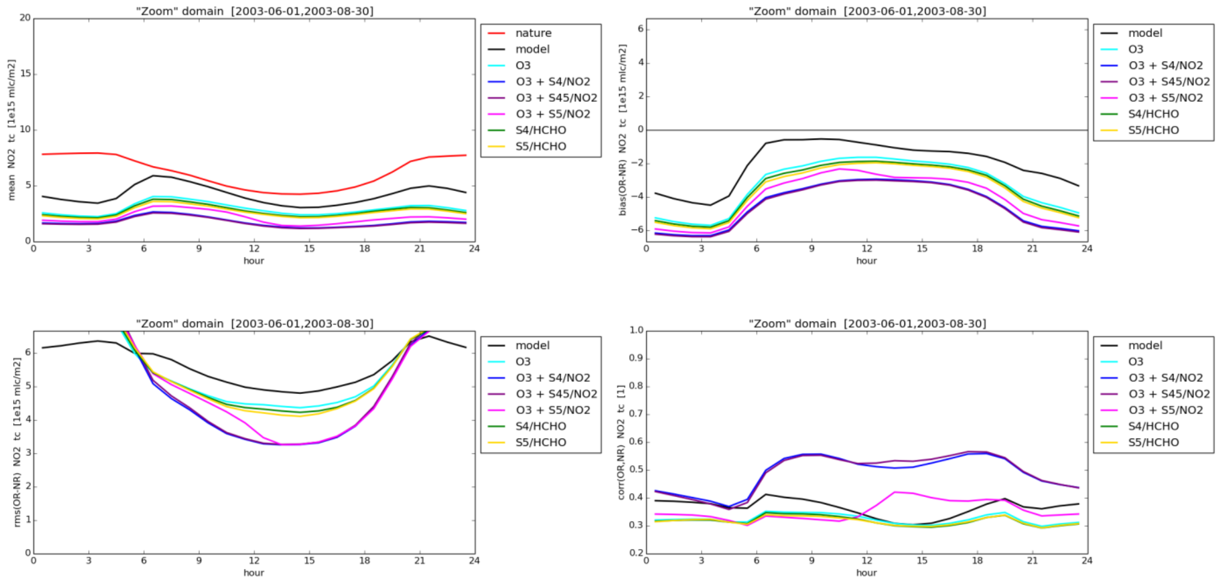


**Figure 7 Zoom domain – summer: $NO_2$ column (top left), bias (top right), RMSE (bottom left) and correlation (bottom right) prior (black line) and after assimilation of observations (colored lines, $O_3$ gb+S4 $NO_2$ (blue), $O_3$ gb+S5P $NO_2$ (pink) or $O_3$gb+S4 and S5P $NO_2$ (purple)).**

The first thing we want to discuss is the negative average bias in total $NO_2$ columns over this domain before data assimilation while we have seen a mainly positive bias in the satellite $NO_2$ columns over the same domain. Figure 8 shows the total and satellite columns from nature run and from the LOTOS-EUROS model without data assimilation. It can be seen that the difference between total and satellite column is much larger for the nature run than for the LOTOS-EUROS model used for the data assimilation. This can be explained from the higher model top of the MOCAGE model used for the Nature Run; the total column therefore includes a stratospheric contribution on top of the tropospheric column represented by the synthetic satellite observations. The LOTOS-EUROS model has a much lower top however, and what is put out as 'total' column is effectively only the tropospheric contribution.

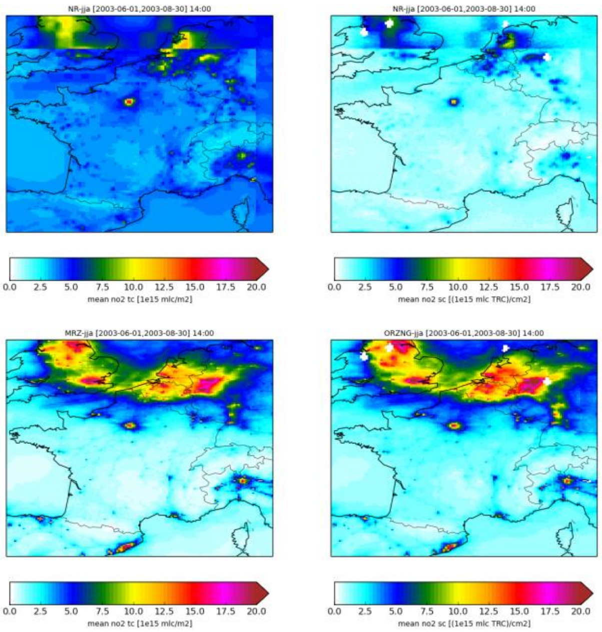


**Figure 8 Zoom domain – summer, 14h average: total $NO_2$ columns (left column) and satellite tropospheric $NO_2$ columns (right column), from either 'nature' (top row, MOCACE output left and synthetic data on right), or from assimilation model (LOTOS-EUROS, bottom row) run without data assimilation.**

Figure 7 illustrates that the assimilation of groundbased ozone observations in this case increases the bias and decreases the correlation in the total $NO_2$ column. As can be seen in Figure 9 the surface ozone observations do have a positive impact on the bias in surface ozone and $NO_2$ concentrations, unfortunately this does not translate into a smaller bias in the $NO_2$ column over the entire domain. The positive biases in the total columns decrease while the negative biases do not increase. In the next section where we look at the impact on surface concentrations we will elaborate a bit further on this relation between surface and total $NO_2$.

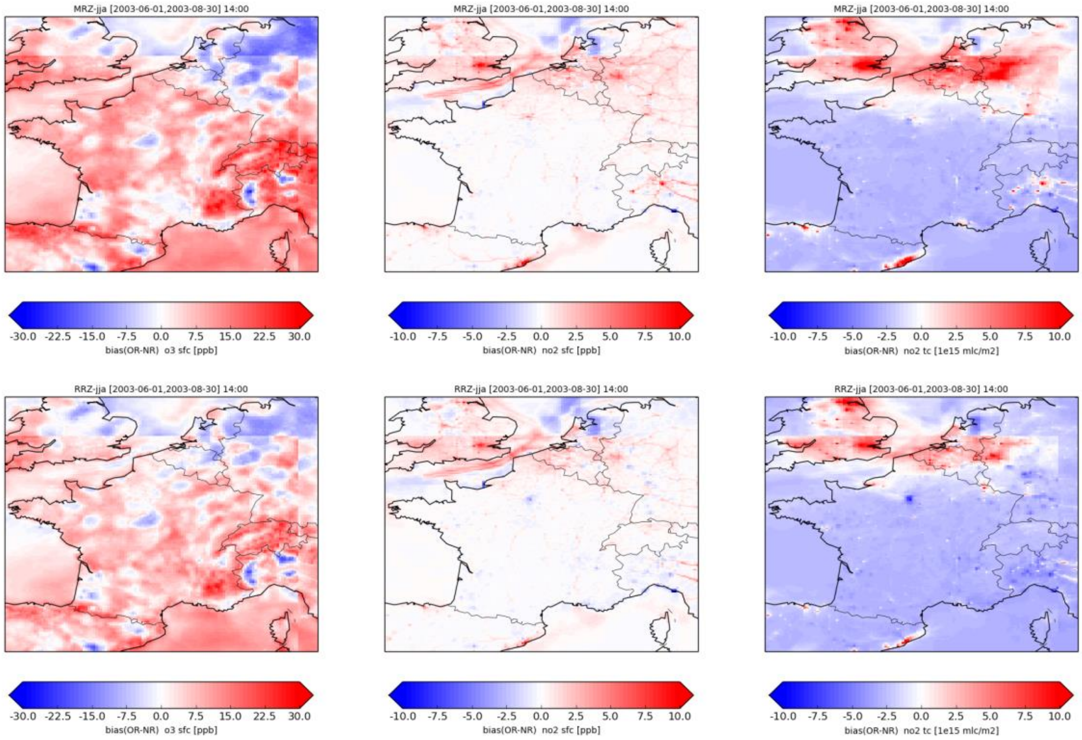


**Figure 9 Zoom domain – summer, 14h average: bias in surface ozone concentrations (left column), surface $NO_2$ concentrations (second column), total $NO_2$ columns (third column) before (top row) and after (bottom row) assimilation of groundbased ozone observations.**

The negative bias is further increased through assimilation of the satellite data; Remembering the satellite column results, we have seen for the summer period that the satellite no2 columns are mostly overestimated by the model. The assimilation will decrease the NOx emissions to remove this overestimation, leading to smaller total $NO_2$ columns in the model. This explains the more negative bias in the total columns for this case.

Nevertheless assimilation of $NO_2$ column observations improves the RMSE and correlation, for the geostationary S4 during entire day and for the S5P from around overpass time until nighttime, clearly pinpointing the added value of geostationary observations. Thus in the morning the value of the S4 observations is larger than the value of the S5P observations. In the afternoon, both satellite observations sets are able to reduce the average RMSE over the domain and study period in the same amount, note that this does not mean the modelled fields are exactly the same at every time and place as it is an average value. This is also visible as the correlation improvement is not similar for both instruments. While the RMSE is a measure on the absolute differences between two datasets largely influenced by locations and times with large deviations between the datasets, the correlation measure provides information on the ability to reproduce temporal variations. The improvement in correlation from S5P is about half of the improvement from S4. It could be that the S4 improves the correlation in areas with low NO2 concentrations, here small variations in concentrations do not lead to a large difference in RMSE but can lead to improvements in correlation. For applications where the aim is to produce the best average representation of the state of the atmosphere, datasets that lead to a similar reduction in RMSE have comparable value. For applications where the representation of the temporal variability plays an important role, for example when running forecasts using emission changes seen in the past, the impact on temporal correlation should also be taken into account.

Combined assimilation of both S4 and S5P data slightly improves the temporal correlation around the overpass time of S5P.

Figure 7 also shows the impact of the assimilation of HCHO columns from the two different sentinels on the $NO_2$ columns, in both cases this impact is small.

The benefit of the high temporal resolution of the S4 observations is even clearer looking at the fire domain for a 2 week period (1-16 August 2013) containing large wildfires as shown in Figure 10. While the correlation using S5P observations clearly improves after overpass time, the correlation using S4 observations already improves after sunrise when the observations become available.

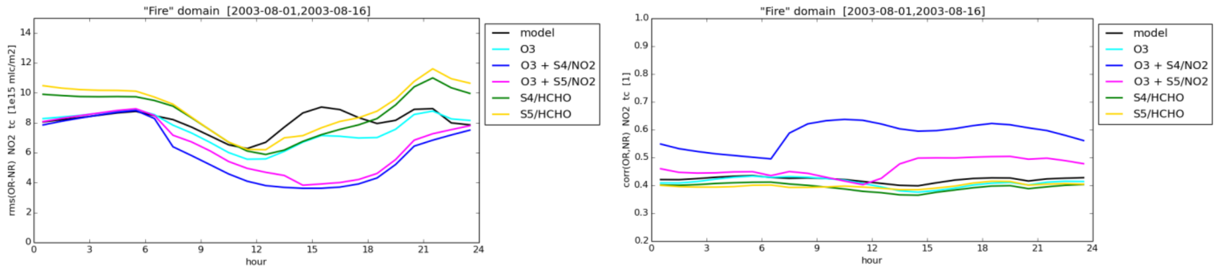


**Figure 10 Fire domain – 1-16 August 2013: RMSE (bottom left) and correlation (bottom right) prior (black line) and after assimilation of observations (colored lines, $O_3$ gb+S4 $NO_2$ (blue), $O_3$ gb+S5P $NO_2$ (pink) or $O_3$ gb+S4 and S5P $NO_2$ (purple)).**

## 2.4 Impact on surface $NO_2$

As the main interest for air quality lays in the health relevant surface concentrations, it is valuable to investigate the impact of the satellite observations on the analysed fields at the surface. Figure 11 and Figure 12 show the bias with the nature run before and after assimilation of different observation data for the summer and winter period respectively. In the summer we can see a large advantage of the satellite observations. Due to their coverage the satellite observations are able to decrease the positive bias over the shipping tracks where no ground based ozone observations are available to constrain the emissions.

In the winter period the surface ozone observations mainly decrease the positive biases in the surface $NO_2$ concentrations. This effect is in some regions counteracted through the additional assimilation of the satellite $NO_2$ observations. In the winter period the modelled satellite columns are too low over the Eastern part of the domain (see Figure 5). The assimilation of satellite observations will induce increases of NOx emissions in this region. However in some parts of this region with underestimated $NO_2$ columns (around German-Austrian border, Po valley) the surface $NO_2$ concentrations are correctly modeled or even overestimated. In these areas the increase of the NOx emissions will thus induce or even increase the positive bias. This contradiction between the bias in satellite columns and bias in surface concentrations is due to different $NO_2$ profiles in the nature run and LOTOS-EUROS. It is thus crucial that $NO_2$ profiles are correctly modeled and the difference between modelled and nature run profiles should be analysed to correctly assess OSSE results.

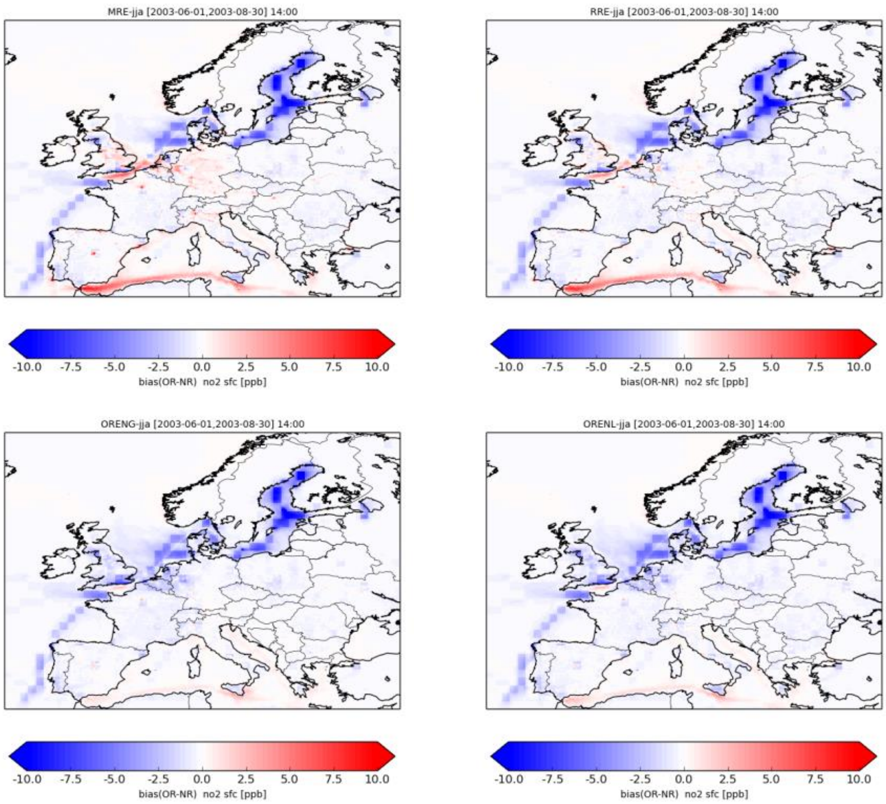


**Figure 11 Europe – summer- 14h average: Bias in $NO_2$ surface concentration between LOTOS-EUROS and nature run before assimilation (top left), and after assimilation of gb $O_3$ (top right), gb $O_3$+S4 $NO_2$ (bottom left), or gb $O_3$ +S5P $NO_2$ (bottom right).**

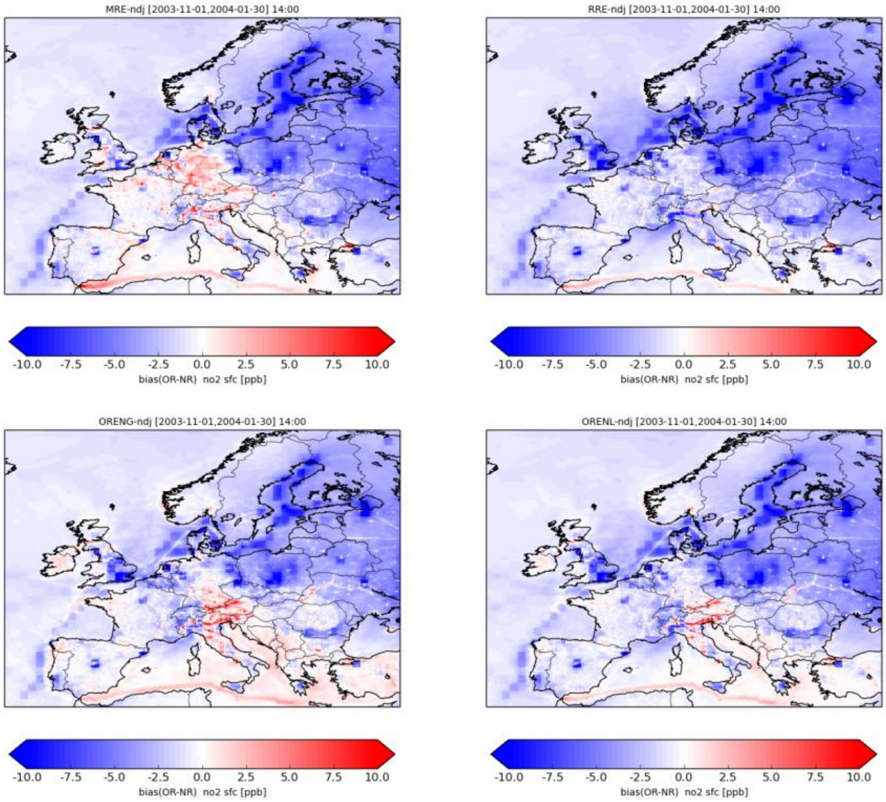


**Figure 12 Europe – winter- 14h average: Bias in $NO_2$ surface concentrations between LOTOS-EUROS and nature run before assimilation (top left), and after assimilation of gb $O_3$ (top right), gb $O_3$+S4 $NO_2$ (bottom left), or gb $O_3$ +S5P $NO_2$ (bottom right).**

When the bias in surface concentrations and satellite columns agree the satellite observations are able to positively impact the bias in the surface concentrations as can be seen during the fire episode from 1-16 August 2003 over the Iberian peninsula shown in Figure 13. The overestimation in the South-Western corner of Portugal is decreased through the assimilation of the satellite observations. The S4 observations have more impact than the S5P observations. Another nice example where the satellite observations are able to decrease the bias is presented in Figure 14. The MR overestimates the surface $NO_2$ concentrations over the traffic highways. It can be seen that especially the S4 observations are able to a large extend to reduce this positive bias.

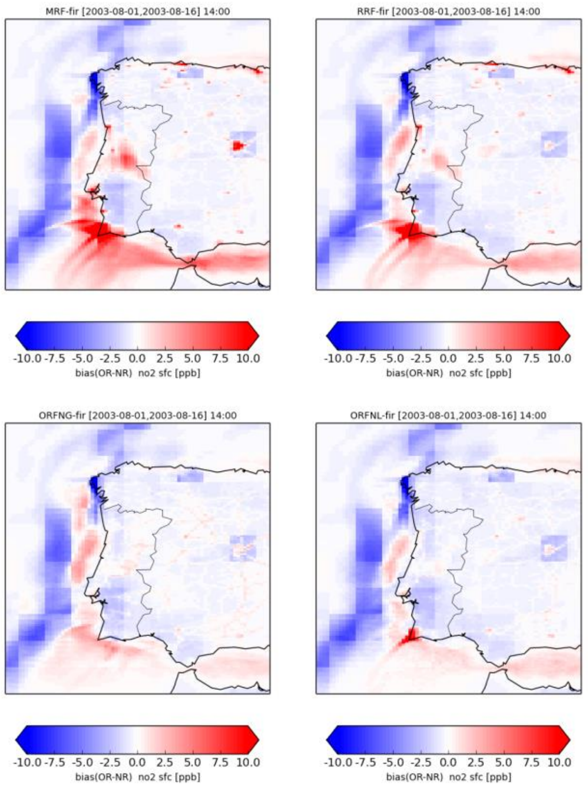


**Figure 13 Fire domain – 1 to 16 August 2003, 14h average. Bias with nature run surface $NO_2$ before assimilation (top left), and after assimilation of gb $O_3$ (top right), gb $O_3$+S4 $NO_2$ (bottom left) or gb $O_3$ +S5P $NO_2$.**

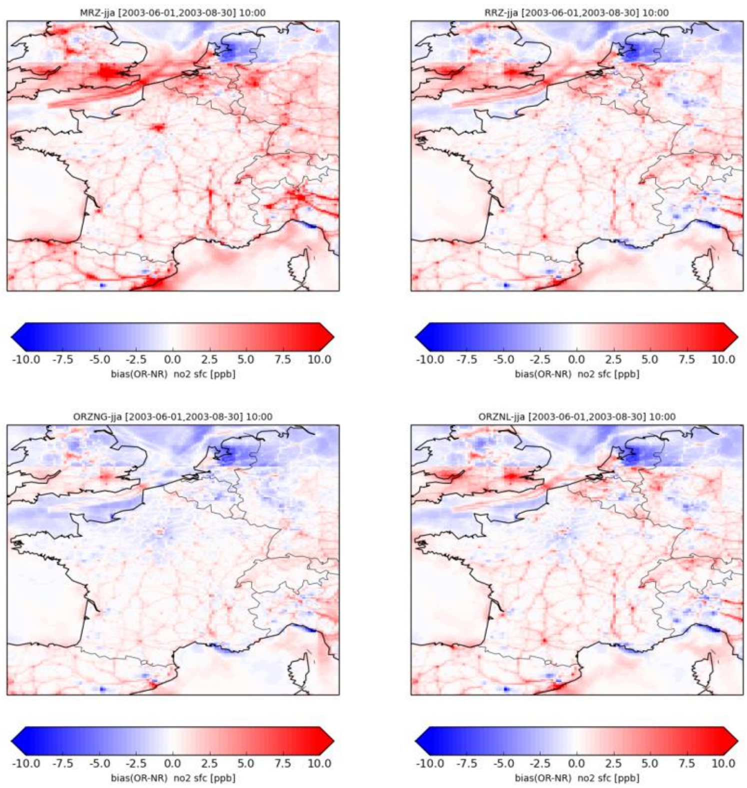


**Figure 14 Zoom domain – summer, 14h average. Bias with nature run surface $NO_2$ before assimilation (top left), and after assimilation of gb $O_3$ (top right), gb $O_3$+S4 $NO_2$ (bottom left) or gb $O_3$ +S5P $NO_2$.**

Although the impact of the satellite observations on the bias might not always be positive, the observations do tend to improve the RMSE and temporal correlation in most cases as compared to the run with assimilation of ozone ground observations only. Figure 15 shows the statistics for the zoom domain and summer period. Again it can be seen that the observations from the geostationary satellite have a positive impact on the RMSE and temporal correlation throughout the entire day while the observations from the low-earth orbiting satellite with one overpass a day have an impact starting from the overpass time. The combined assimilation of both sentinel 4 and 5P data does not improve the results from the S4 only.

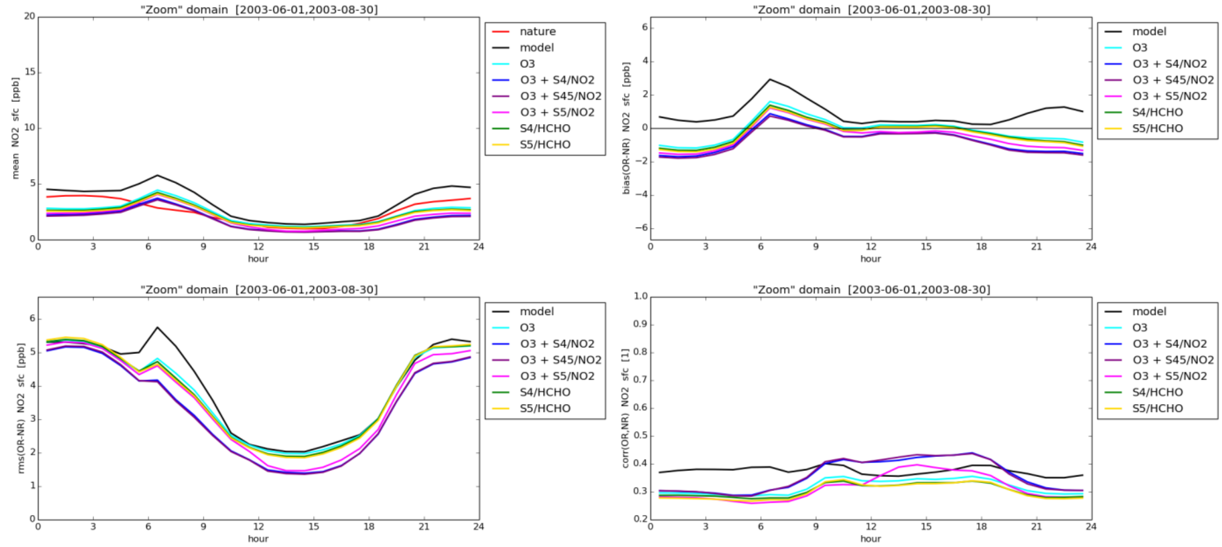


**Figure 15 Zoom domain – summer: surface $NO_2$ concentrations (top left), bias (top right), RMSE (bottom left) and correlation (bottom right) with nature run concentrations prior (black line) and after assimilation of observations (colored lines, $O_3$ gb+S4 $NO_2$ (blue), $O_3$ gb+S5P $NO_2$ (pink) or $O_3$gb+S4 and S5P $NO_2$ (purple)).**

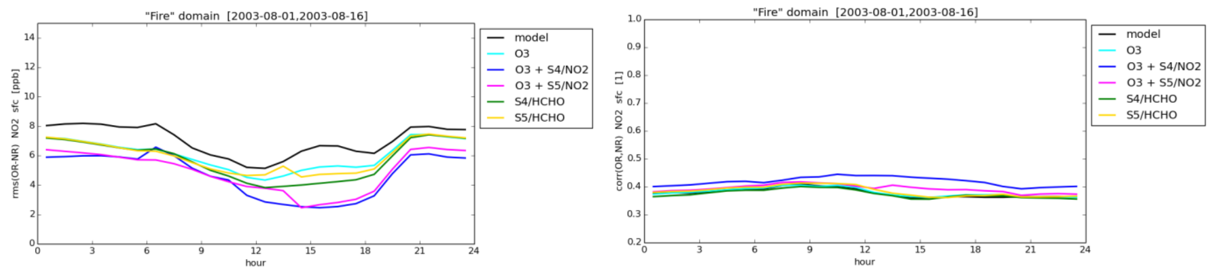


**Figure 16 Fire domain – 1-16 August 2013: RMSE (bottom left) and correlation (bottom right) with nature run surface $NO_2$ prior (black line) and after assimilation of observations (colored lines, $O_3$ gb+S4 $NO_2$ (blue), $O_3$ gb+S5P $NO_2$ (pink) or $O_3$ gb+S4 and S5P $NO_2$(purple)).**

For the fire episode the impact of the data assimilation on the RMSE is most clear (see Figure 16). Again the impact of S4 observations is largest but comparable to the impact of the S5P observations at the S5P overpass time. For this case we can also see that the S4 HCHO column observations decrease the RMSE.

# 3 Impact on HCHO

The HCHO product is much less developed as the $NO_2$ product and has a small signal to noise ratio. This means a large number of observations is needed to cancel out the noise in the product, and high HCHO concentrations are needed to get a consistent picture. To this reason we will show results for the fire domain and the two week period of 1-16 August 2003 as this is a period with wildfires over the Iberian Peninsula and high concentrations of HCHO, showing a clear signal in the satellite products. For the other domains, we see that the averaged modelled HCHO columns and surface concentrations do not show an improvement (rather a detoriation ) when assimilating HCHO satellite observations.

## 3.1 Emission increments

Figure 17 shows the average NOx and VOC emission increments for the fire episode over the Iberian Peninsula for the different assimilation runs. It can be seen that the assimilation of groundbased ozone observations leads to a decrease of the NOx emissions over some high source regions. while the effect on VOC emissions is smaller. The assimilation of satellite HCHO observations does not largely influence the NOx emissions, but is shown to have a substantial additional impact on the VOC emissions. The added value of these emission increments will be evaluated by looking at the impact of the increments on modelled HCHO fields in the following sections.

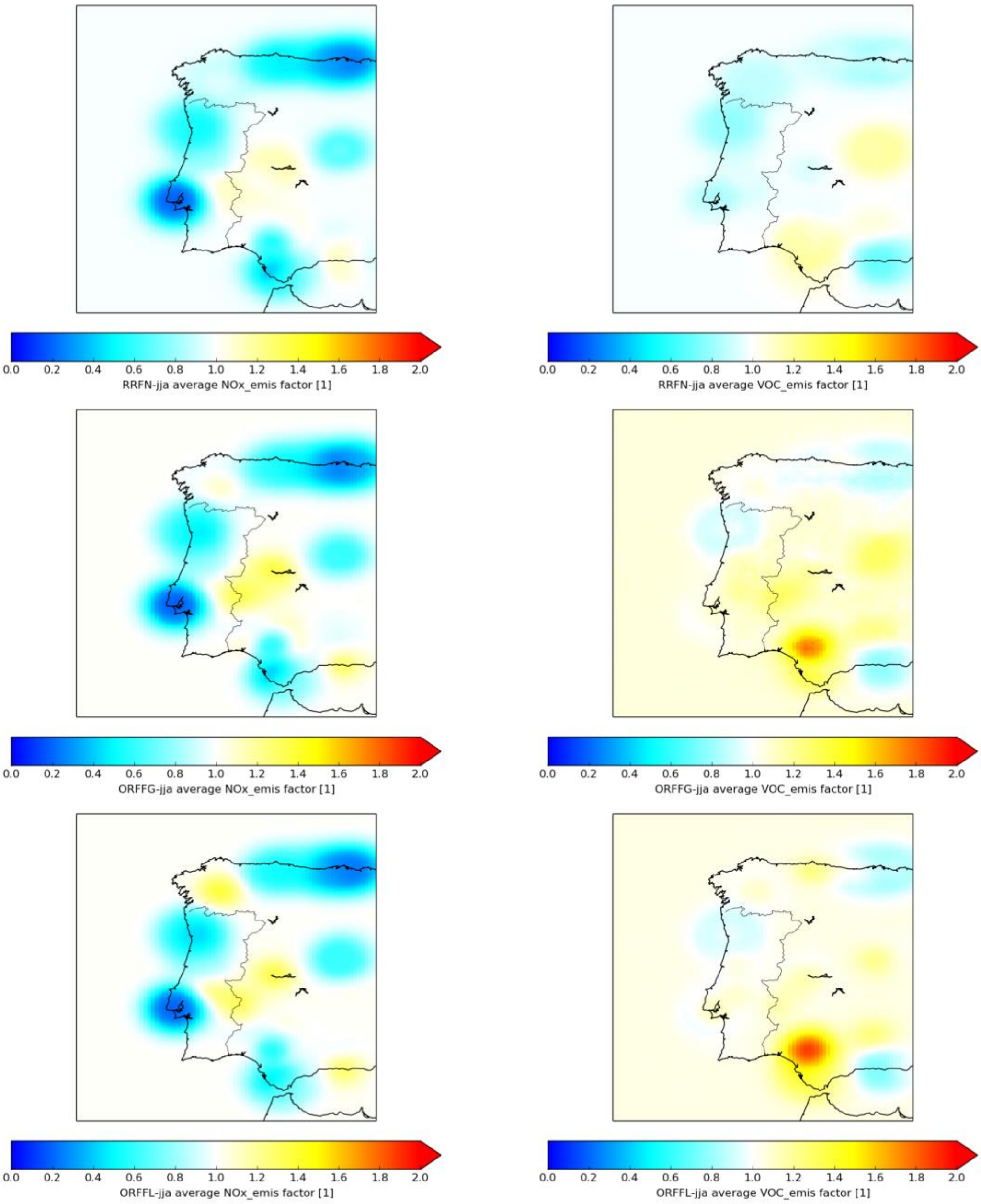


**Figure 17 Fire domain, 1-16 August 2003 averaged $NO_x$ (left column) and VOC (right column) emission increments for RR (top panels), AR with S4 HCHO observations (middle panels) and AR with S5P HCHO observations (bottom panels).**

## 3.2 Impact on HCHO satellite column

Figure 18 shows the averaged synthetic sentinel 4 (S4) and Sentinel 5P (S5P) HCHO observations at 14h over the fire domain for 1-16 August 2003, versus the results from the MR run and the assimilation run. It can be seen that the Model Run shows higher HCHO columns over some hot spots in Portugal as compared to the synthetic observations from the nature run. When assimilated in combination with the ozone groundbased observations both the S4 HCHO column observations as well as the S5P HCHO column observations are decreasing the values over these hotspots to get in better agreement with the synthetic observations. This can also be seen in Figure 19 where the statistical parameters bias, RMSE and temporal correlation versus the synthetic observations are plotted. In these plots the influence of the high amount of noise in the satellite products is visible, but especially the high positive biases and RMSE over the hotspots are decreased. The number of observations used in the assimilation runs during this two week fire event over the fire domain are 2.377.836 and 247.168 for the S4 and S5P instrument respectively. Note these are not the original amount of pixels but the amount of model gridcel averages as used in the data assimilation.

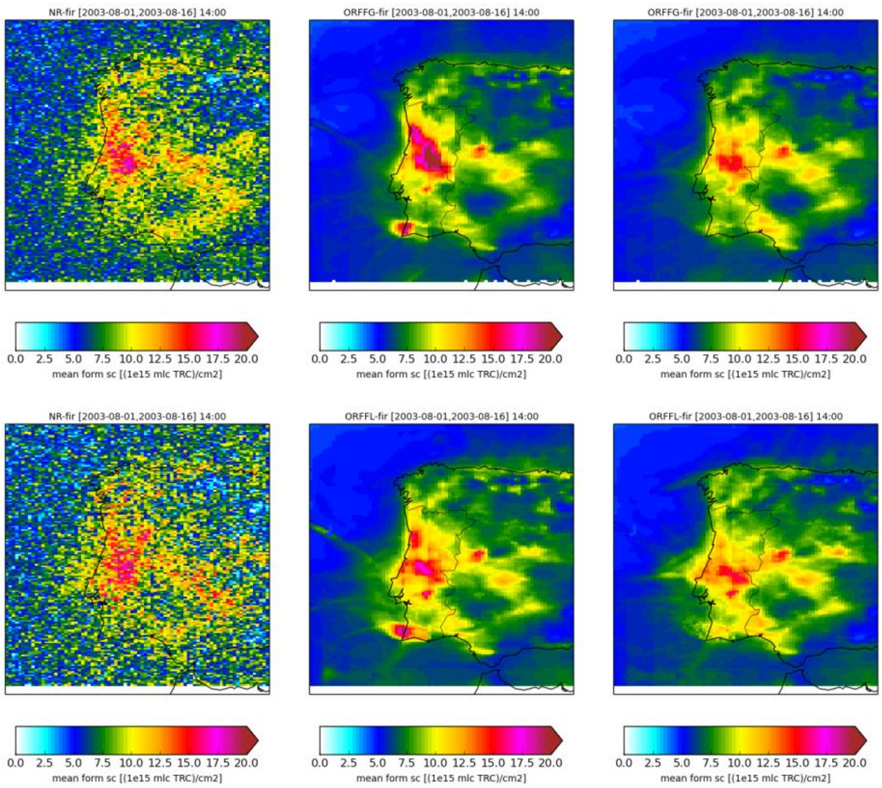


**Figure 18 Fire domain- 1-16 August 2003 averaged synthetic HCHO columns at 14h (left) and collocated convolved HCHO columns from Model Run (middle) and Assimilation run (right) for $O_3$ gb + S4 HCHO (top) and $O_3$ gb + S5P HCHO (bottom).**

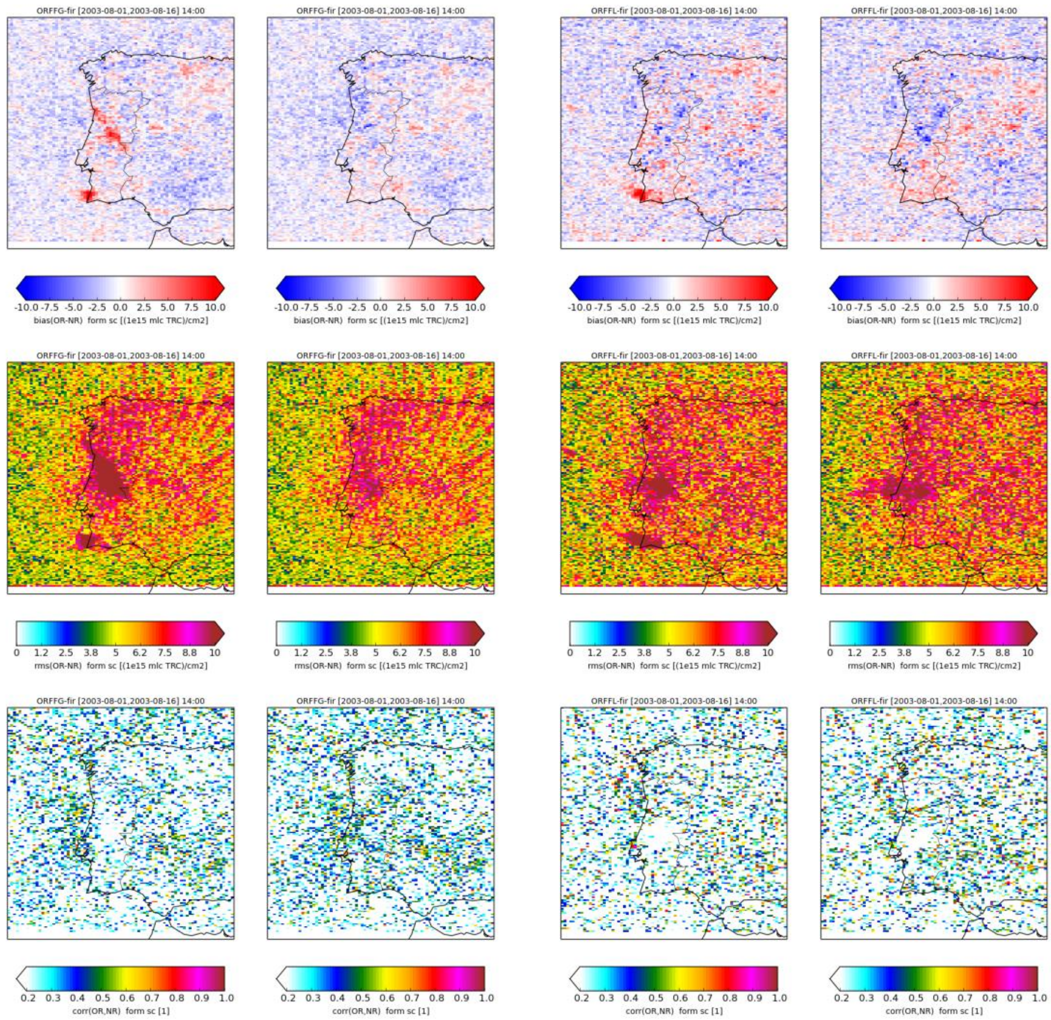


**Figure 19 Fire domain – 1-16 August 2003: Bias (top row), RMSE (middle row) and correlation (bottom row) with synthetic observations for $O_3$ gb + S4 HCHO (left two columns) and $O_3$ gb + S5P HCHO (right two columns). First column for each instrument is from Model Run without data assimilation, second column is from assimilation run with assimilation of synthetic observations.**

## 3.3 Impact on total HCHO column

When looking at the temporal evolution of the statistics in Figure 20 largest impacts are seen on the RMSE. Both the S4 and S5P observations decrease the RMSE as compared to the reference run. The best improvement is seen when assimilating the S4 HCHO observations, probably due to the higher temporal resolution and thus the availability of observations throughout the entire day. The same conclusion can be drawn from the bias and RMSE maps shown in Figure 21 and Figure 22**.**

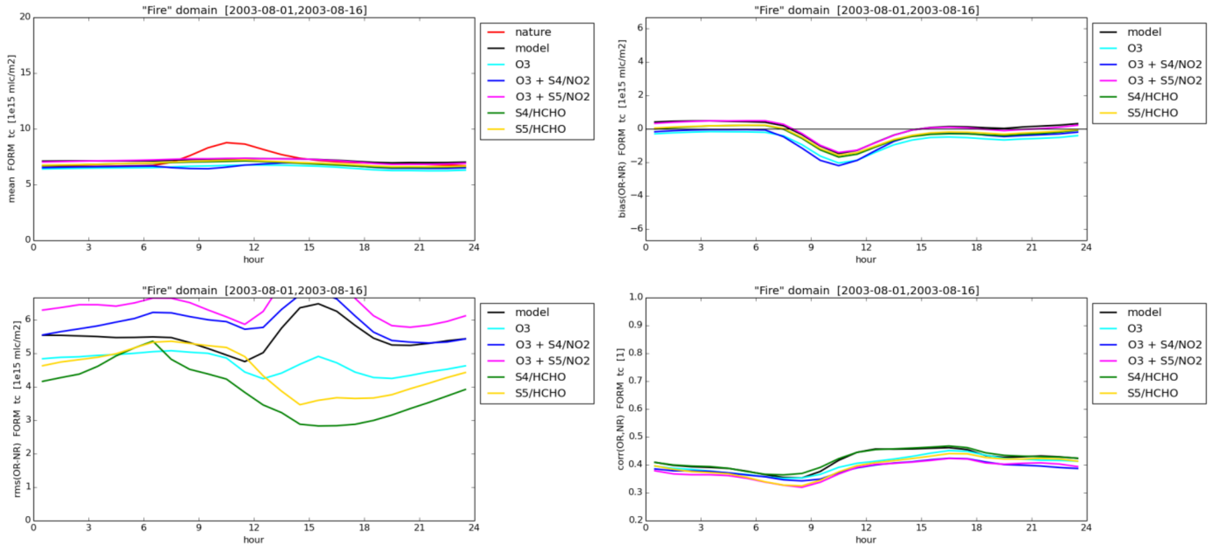


**Figure 20 Fire domain – 1-16 August 2003: HCHO total columns (top left), bias (top right), RMSE (bottom left) and correlation (bottom right) with nature run total HCHO columns prior (black line) and after assimilation of observations (colored lines, $O_3$ gb + S4 HCHO (green), $O_3$ gb + S5P HCHO (yellow)).**

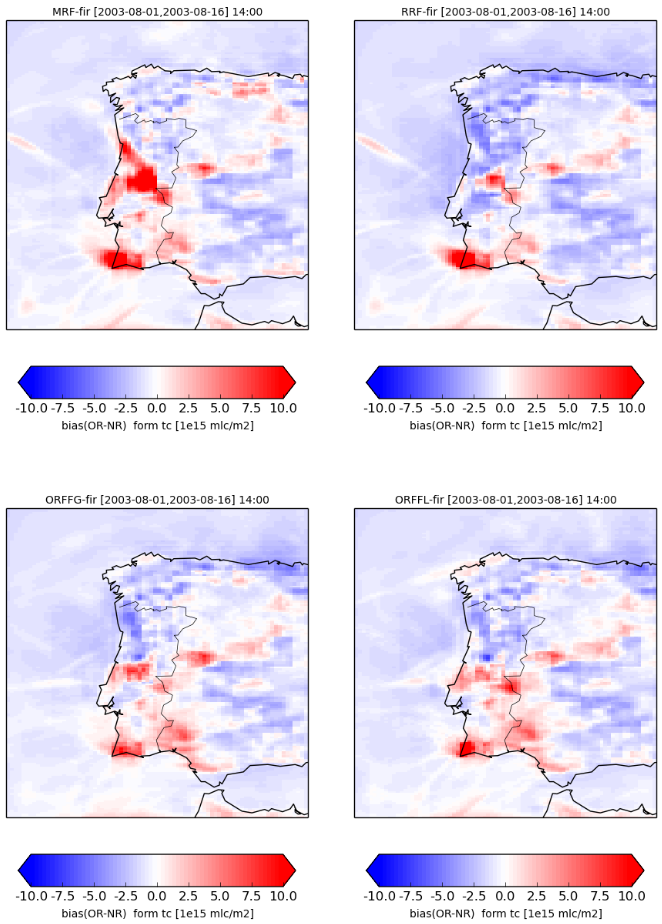


**Figure 21 Fire domain – 1 to 16 August 2003, 14h total HCHO column. Bias with nature run before assimilation (top left), and after assimilation of gb $O_3$ (top right), $O_3$ gb + S4 HCHO (bottom left) or $O_3$ gb + S5P HCHO (bottom right).**

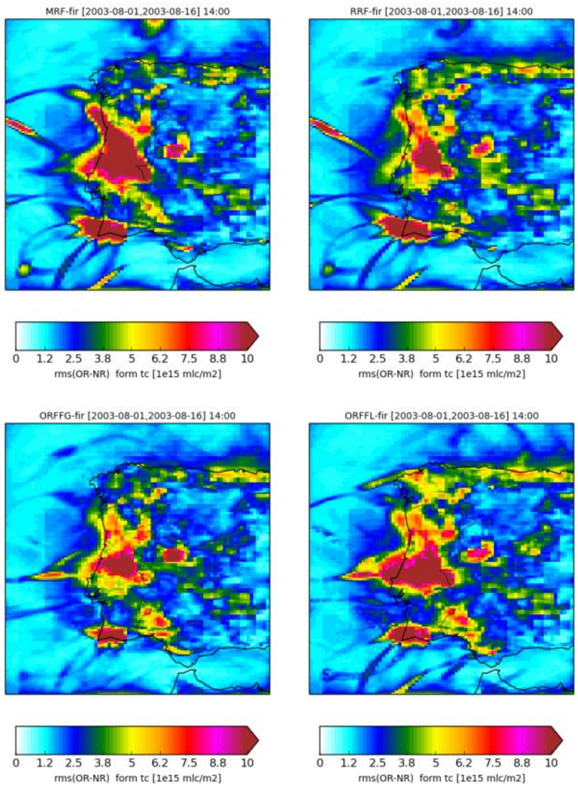


**Figure 22 Fire domain – 1 to 16 August 2003, 14h total HCHO column. RMSE with nature run before assimilation (top left), and after assimilation of gb $O_3$ (top right), $O_3$ gb + S4 HCHO (bottom left) or $O_3$ gb + S5P HCHO (bottom right).**

## 3.4 Impact on surface HCHO concentrations

The positive effect of the satellite HCHO columns on the modelled HCHO columns during this fire event is also translated into an improvement of the HCHO surface concentrations, most clearly visible in the RMSE shown in Figure 23.

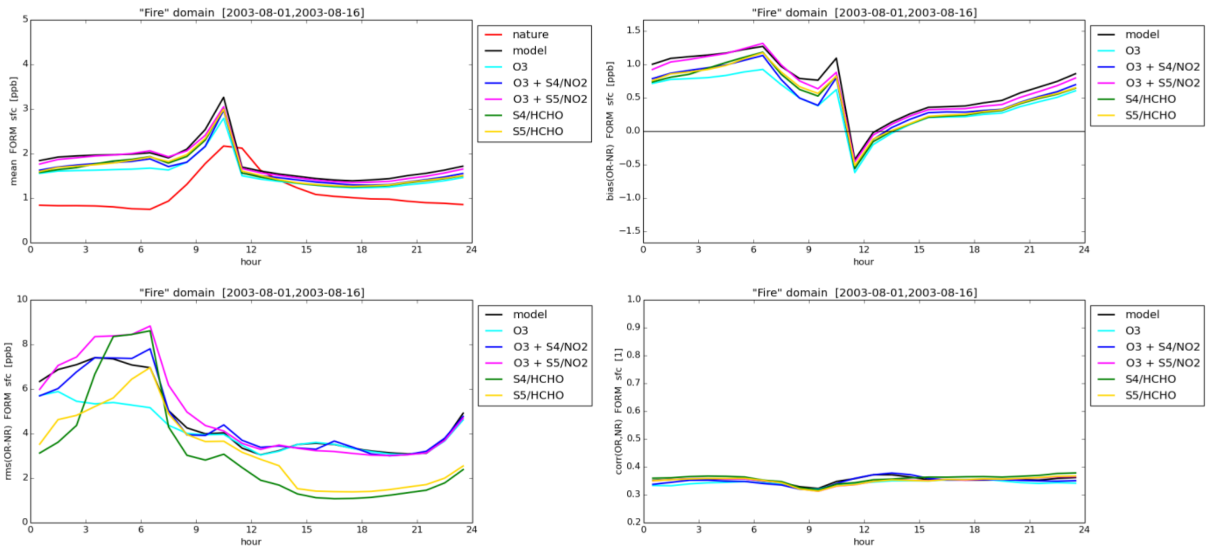


**Figure 23 Fire domain – 1-16 August 2003: HCHO surface concentrations (top left), bias (top right), RMSE (bottom left) and correlation (bottom right) with nature run prior (black line) and after assimilation of observations (colored lines, $O_3$ gb + S4 HCHO (green), $O_3$ gb + S5P HCHO (yellow)).**

# 4 Impact on $O_3$

In this section we present the impact of the assimilated datasets on the surface ozone concentrations.

Figure 24 and Figure 26 show the results for the summer period over Europe. The assimilation of ground based ozone observations on average decreases the bias, RMSE and improves the correlation. The additional assimilation of satellite observations does on average not show an impact on the surface ozone concentrations, although over the North Sea for example the RMSE does decrease further through the assimilation of the S4 and S5P observations. The same conclusions can be drawn from the results for the winter period shown in Figure 25 and Figure 27. The impact of the satellite observations on surface NO2 concentrations seems not to translate to a similar impact on surface ozone observations. If the errors in modeled $O_3$ concentrations would be equally influenced by the same errors (e.g. in emission inputs) as the  errors in the modeled $NO_2$ concentrations, then one would expect that the improved emissions would lead to improvements in both $NO_2$ and $O_3$. However $O_3$ is a much longer lived species than $NO_2$ and variability in ozone is largely influenced by changes in biogenic emissions. In this study uncertainties in biogenic emissions are not taken into account in the active data assimilation set-up, thus assimilation of NO2 observations can not induce changes in the biogenic emissions. This means that an improvement in $NO_2$ does not necessarily mean an improvement in ozone. See also the discussion on the limiting factors of the assimilation performance in next section.

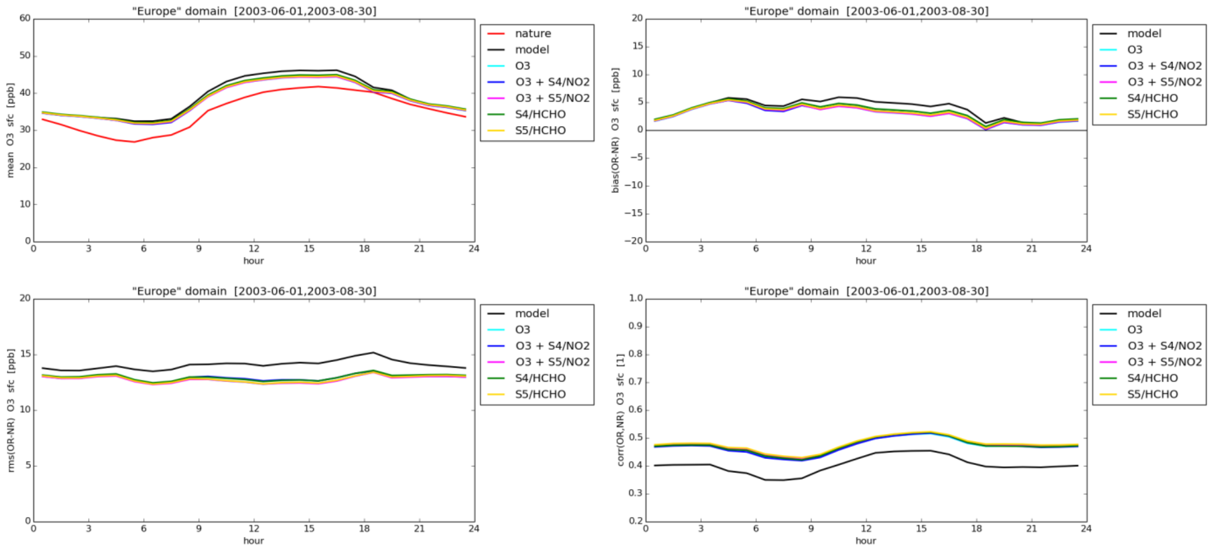


**Figure 24 Europe – summer: $O_3$ surface concentrations (top left), bias (top right), RMSE (bottom left) and correlation (bottom right) with nature run prior (black line) and after assimilation of observations (colored lines).**

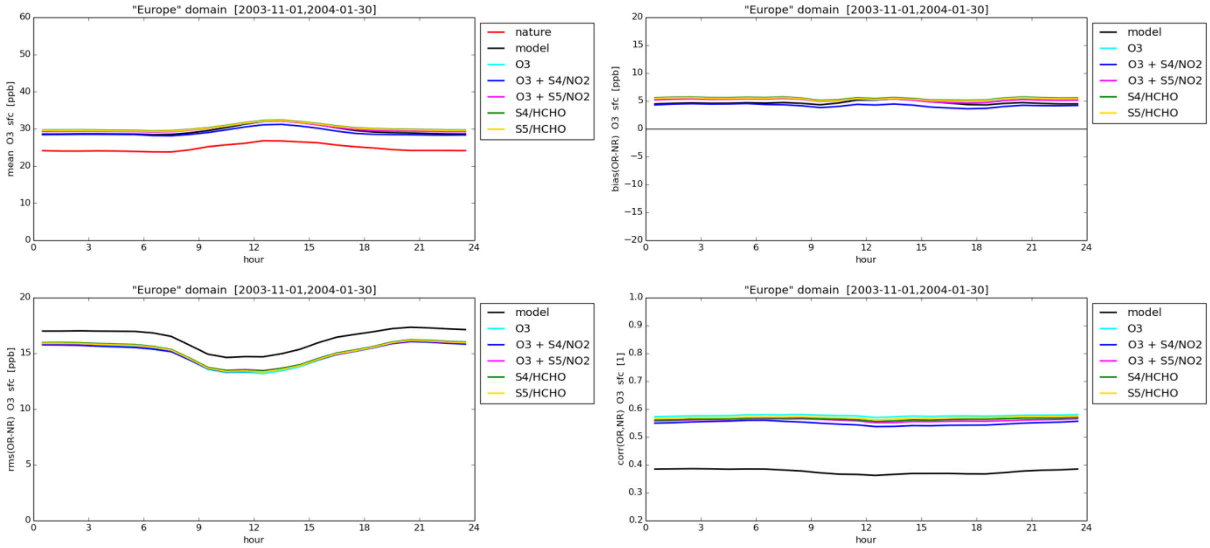


**Figure 25 Europe – winter: $O_3$ surface concentrations (top left), bias (top right), RMSE (bottom left) and correlation (bottom right) with nature run prior (black line) and after assimilation of observations (colored lines).**

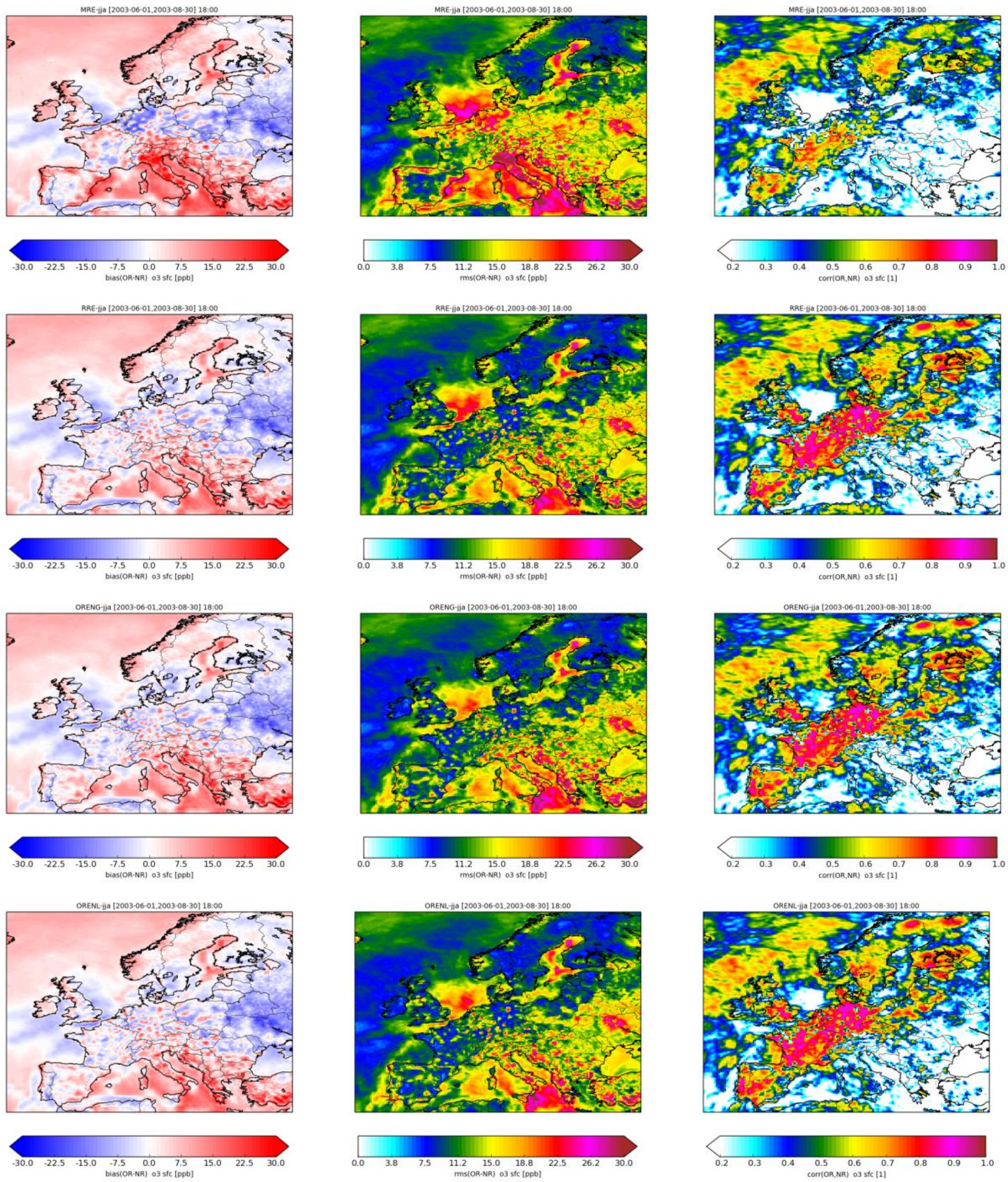


**Figure 26 Europe – summer: $O_3$ surface concentrations: Bias (left column), RMSE ( middle column) and temporal correlation with nature run surface $O_3$ concentrations from MR (top row), RR (second row), AR with gb $O_3$ + S4 $NO_2$ (third row), AR with with gb $O_3$ + S5P $NO_2$ (fourth row).**

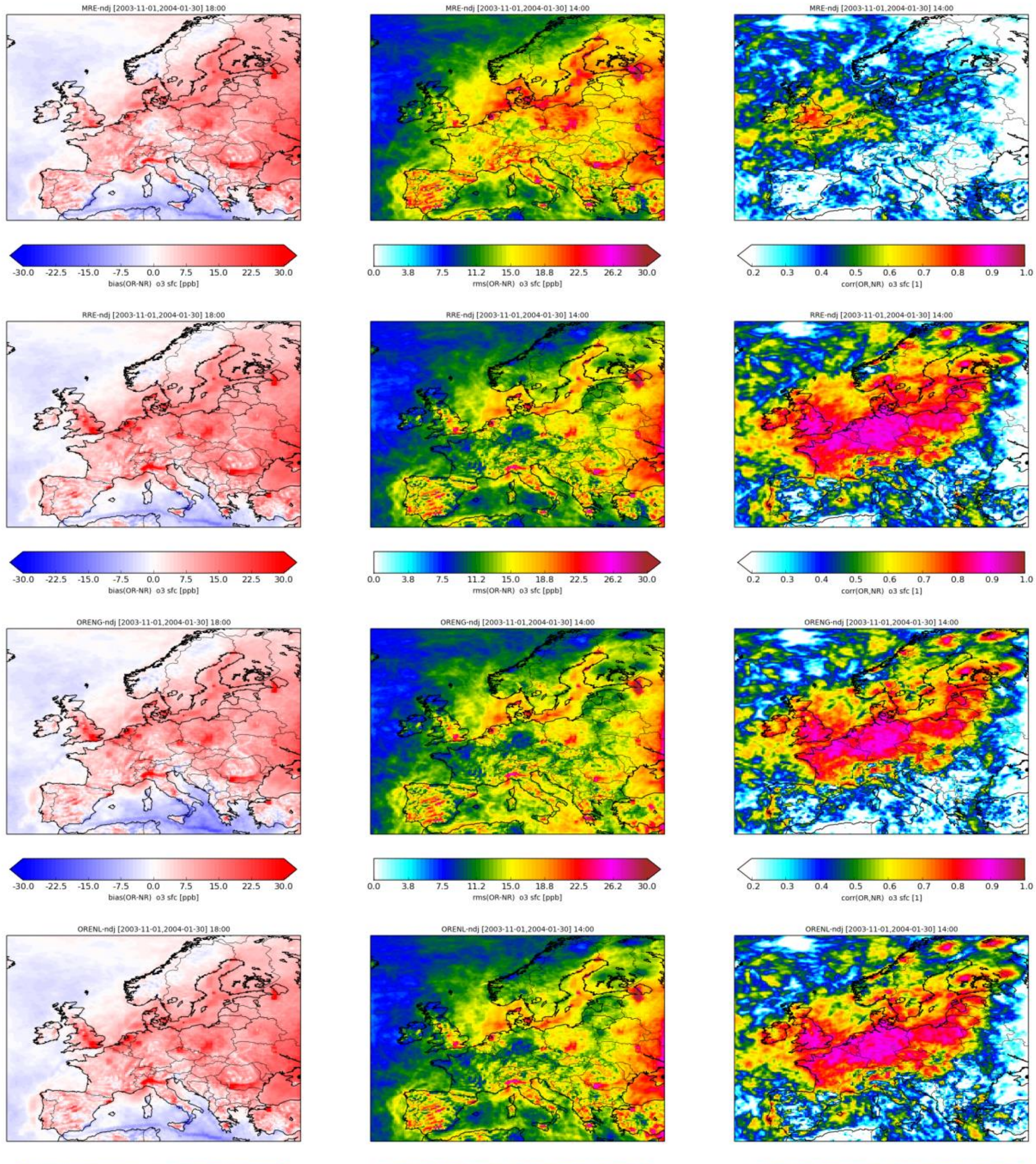


**Figure 27 Europe – winter: $O_3$ surface concentrations: Bias (left column), RMSE ( middle column) and temporal correlation with nature run surface $O_3$ concentrations from MR (top row), RR (second row), AR with gb $O_3$ + S4 $NO_2$ (third row), AR with with gb $O_3$ + S5P $NO_2$ (fourth row).**

The other domains show the same effect, for example for the Paris domain in summer shown in Figure 28. Here the decrease in bias and RMSE, and increase in correlation is even larger than over the entire European continent.

For the fire episode (see Figure 29) some small differences can be seen between the impacts of the different satellite observations. The HCHO observations seem to slightly decrease the RMSE over this domain and time period.

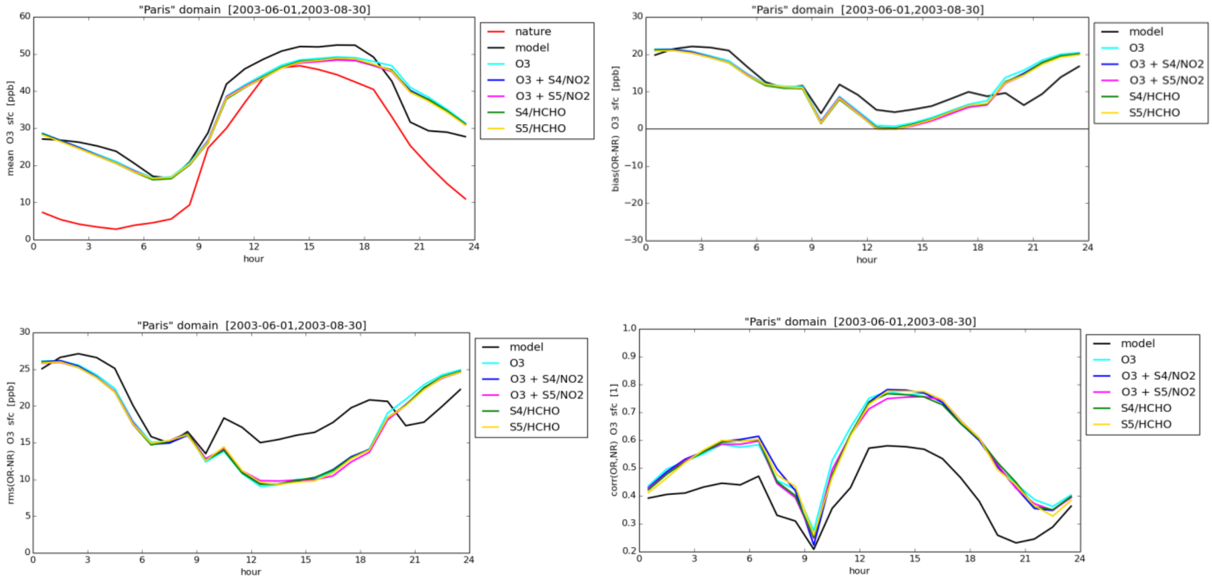


**Figure 28 Paris – summer: $O_3$ surface concentrations (top left), bias (top right), RMSE (bottom left) and correlation (bottom right) with nature run prior (black line) and after assimilation of observations (colored lines).**

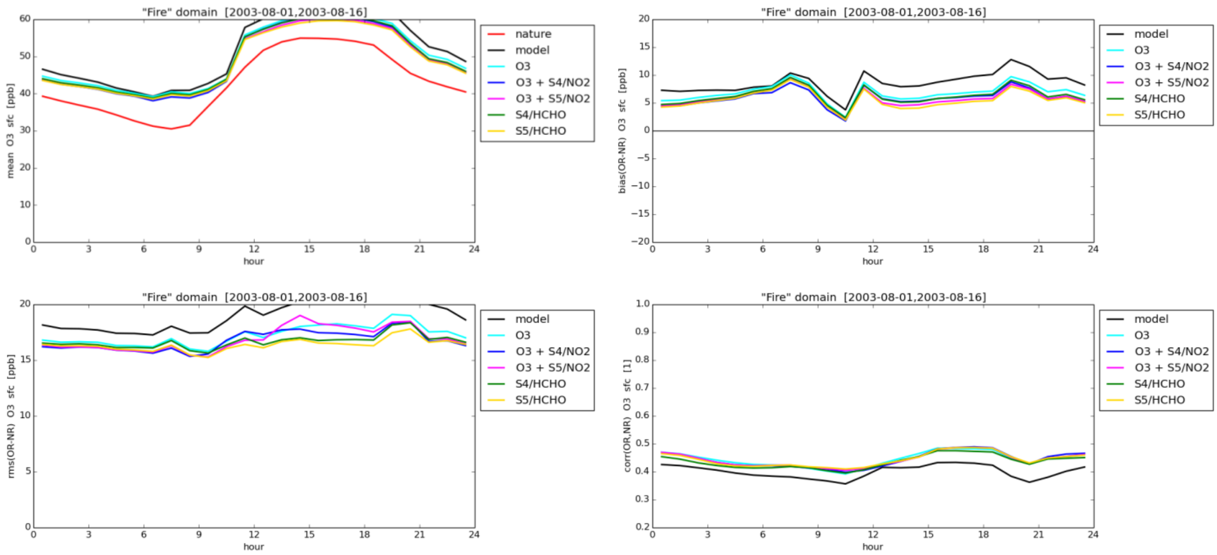


**Figure 29 Fire domain – 1 – 16 August 2--3: $O_3$ surface concentrations (top left), bias (top right), RMSE (bottom left) and correlation (bottom right) with nature run prior (black line) and after assimilation of observations (colored lines).**

# 5 Delta study on the impact of S5 versus S5P observations

In this chapter we present the results of the delta study evaluating the difference in impact of S5 (morning) versus S5P (afternoon) observations. For the delta study the runs over the zoom domain are repeated assimilating the S4 observations for 9:00 UTC (hereafter referred to as S4.09), as surrogate for the Sentinel 5 morning overpass data. In addition new runs are also performed assimilating the S4 observations for 13:00 UTC (hereafter referred to as S4.13) only. The latter runs are compared to the S5P assimilation runs to check the validity of using the Sentinel 4 single hour data as surrogate for Sentinel5 data.

## 5.1 Validity of delta study: S5P versus S4.13 impact

Figure 30 shows the impact of assimilating either the Sentinel 5P observations (top panels) or the Sentinel 4.13 observations (bottom panels) on the modelled satellite $NO_2$ columns at 14h or 13h in the afternoon, just after overpass; the 14h is used for the S5P since these were used in the prior part of the report too. When assimilated in combination with the ozone groundbased observations both the S5P $NO_2$ column observations as well as the S4.13 $NO_2$ column observations are able to largely close the gap between the modelled fields and the synthetic observations. This is also visible in Figure 31 showing the bias, RMSE and correlation with the synthetic datasets before and assimilation of either S5P or S4.13 simulated observations. The impact of both satellite datasets is similar, providing evidence that the S4.13 dataset can be used as surrogate for S5P data and thus the S4.09 dataset can be used as surrogate for S5 observations. Further prove is shown in the figures in next section for the impact on total $NO_2$ columns and on surface $NO_2$ observations.

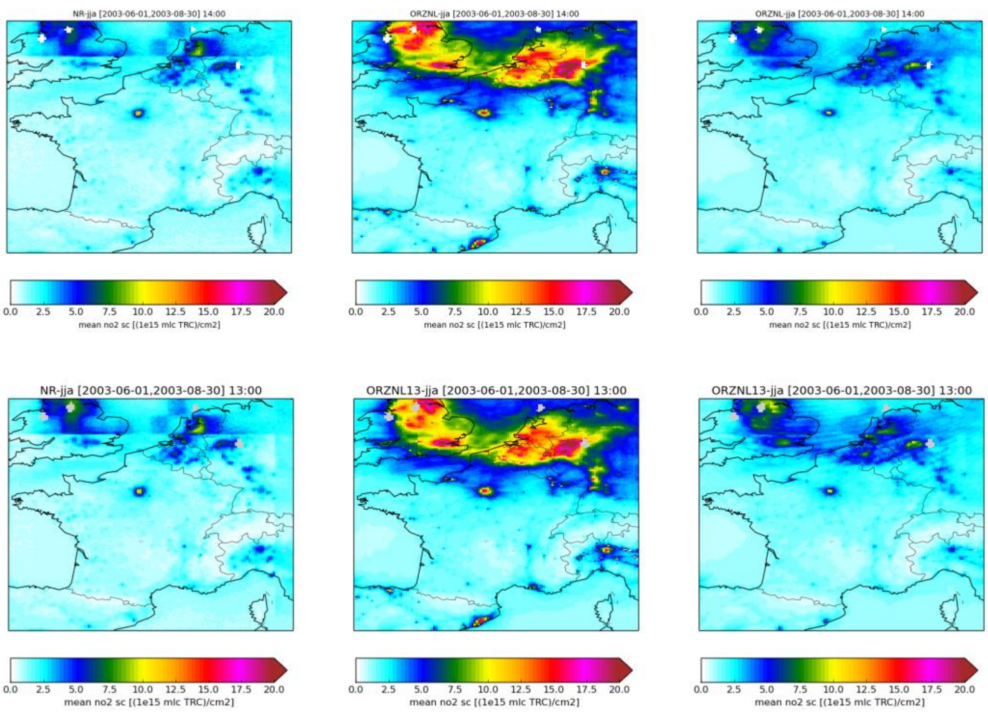


**Figure 30 Zoom-summer period averaged synthetic $NO_2$ columns and collocated convolved $NO_2$ columns from Model Run (middle) and Assimilation run (right) for $O_3$ gb + S5P $NO_2$ (top, at 14:00) and $O_3$ gb+ S4.13 $NO_2$ (bottom, at 13:00).**

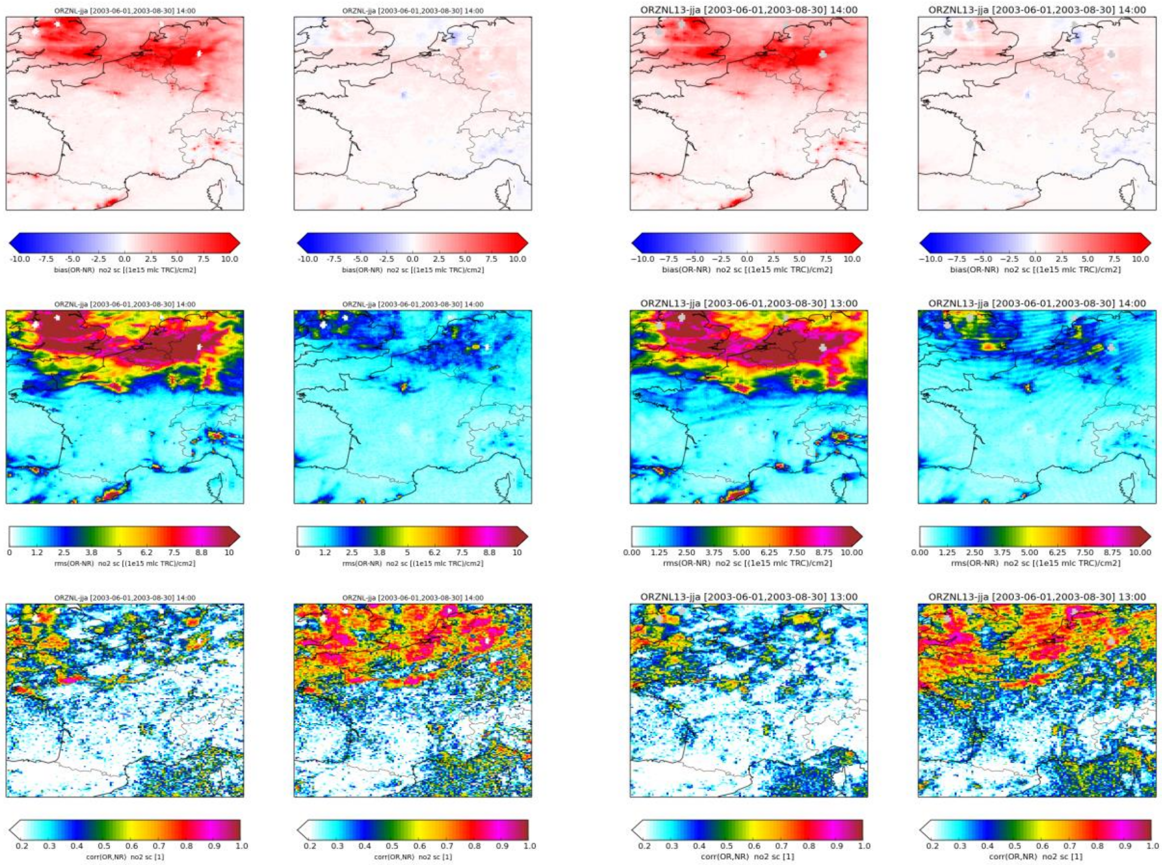

**Figure 31 Zoom – summer period: Bias (top row), RMSE (middle row) and correlation (bottom row) with synthetic satellite columns for $O_3$ gb+S5P $NO_2$ (left two columns) and $O_3$ gb+S4.13 $NO_2$ (right two columns). First column for each instrument is from Model Run without data assimilation, second column is from assimilation run with assimilation of synthetic observations.**

## 5.2 S5P versus S5 impact: S5P versus S4.09 impact

In previous section we concluded that the S4.09 (09:00 image) observations can be used as surrogate for Sentinel 5 (morning overpass) data. Below we will present the results of the comparison between the impact of S5/S4.09 data and the impact of the S4 and S5P data. Again we split the results in three sections: the impact on satellite $NO_2$ columns, the impact on total $NO_2$ columns and the impact on the modelled surface $NO_2$ concentrations.

### 5.2.1 Impact on satellite $NO_2$ columns

Figure 32 shows the impact of assimilating either the Sentinel 4 observations (top panels) or the Sentinel 5/4.09 observations (bottom panels) on the modelled satellite $NO_2$ columns at 10h or 9h, just after overpass time. The 10h time for S4 is used to have the same figures as before.

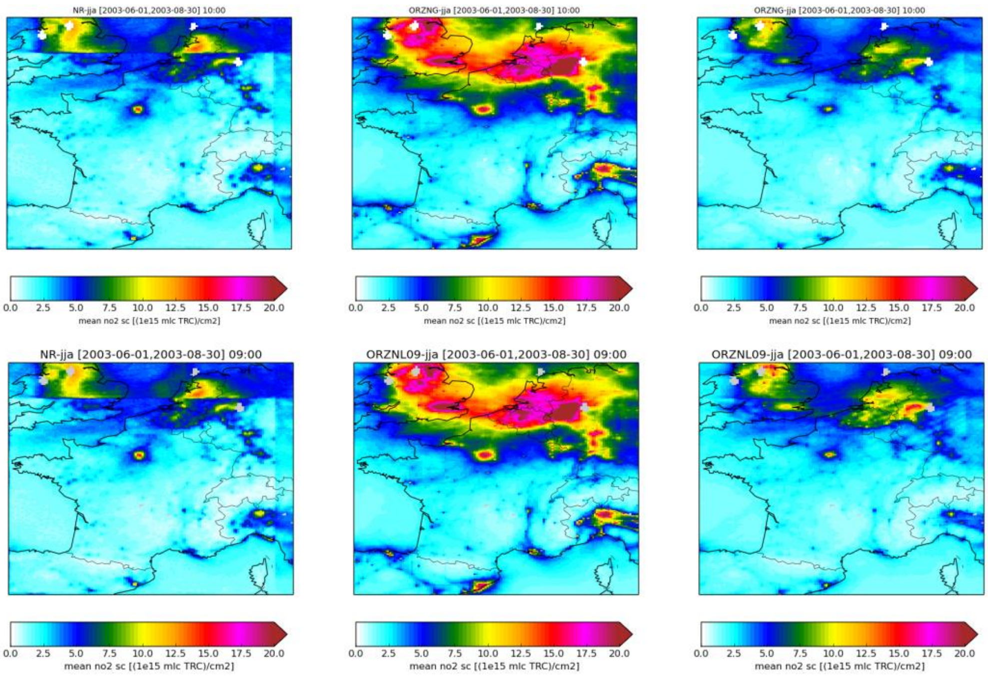


**Figure 32 Zoom-summer period averaged synthetic $NO_2$ columns after overpass (left) and collocated convolved $NO_2$ columns from Model Run (middle) and Assimilation run (right) for $O_3$ gb + S4 $NO_2$ (top) and $O_3$ gb+ S5 (S4.09) $NO_2$ (bottom).**

When assimilated in combination with the ozone groundbased observations both the S5 (S4.09) $NO_2$ column observations as well as the S4 $NO_2$ column observations are able to largely close the gap between the modelled fields and the synthetic observations. This can also be seen in Figure 33 where the statistical parameters bias, RMSE and temporal correlation versus the synthetic observations are plotted. The positive bias and RMSE are largely reduced, while the temporal correlation is increased considerably over a large part of the domain. Over areas with low $NO_2$ columns the temporal correlation remains low, since here the noise in the satellite retrievals is much higher than the absolute values, and the temporal correlation is effectively close to zero. Over some areas **t**he impact of the S5 (S4.09) seems to be smaller than the impact of the S4 observations, e.g. the RMSE over the Ruhr area is reduced less, and the correlation over the English Channel is less increased. This is probably due to the availability of observations from the S4 before the selected hour 10:00, while S5 (S4.09) only has observations at its overpass time of 9:00 h. However in general we can say that the impact of both datasets on the modelled 'satellite' NO2 columns is comparable., which is the same conclusion as was drawn for the S5P versus S4 data. The same conclusion can be drawn from the results in the winter period as can be seen in Figure 34.

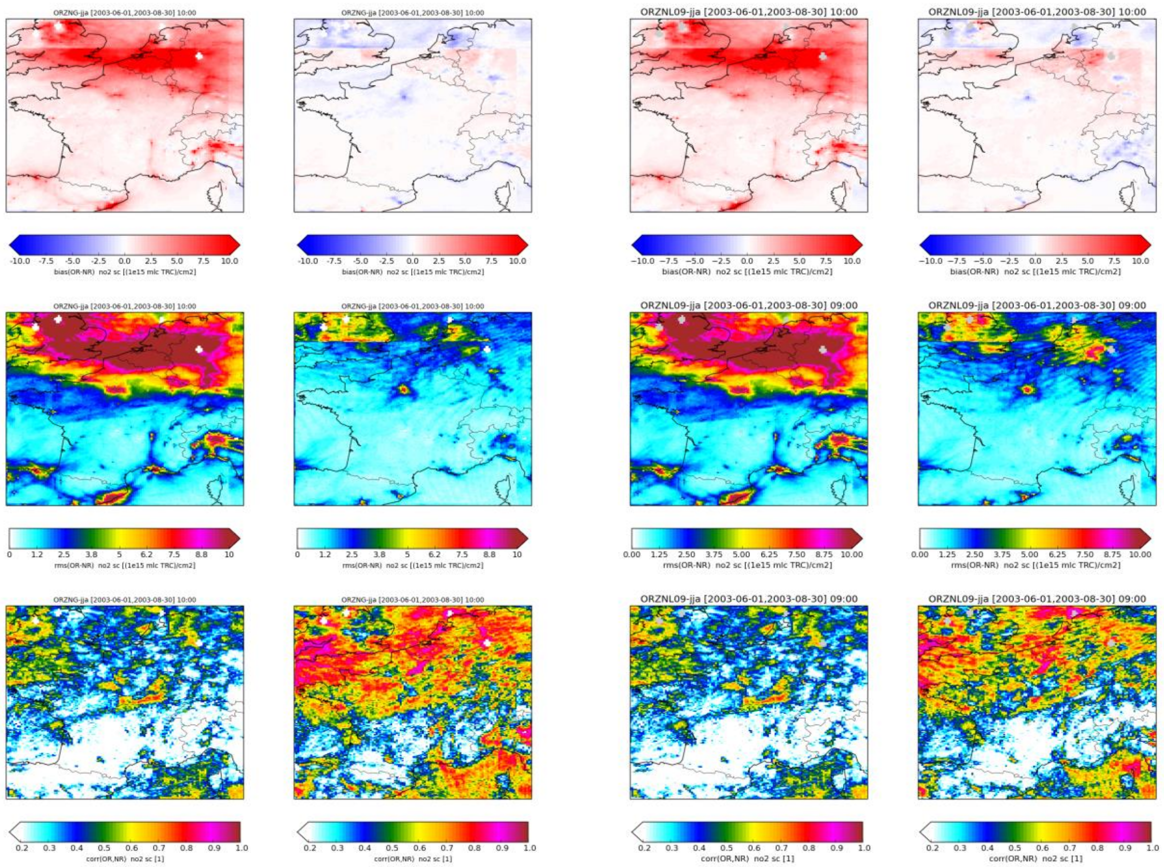


**Figure 33 Zoom – summer period: Bias (top row), RMSE (middle row) and correlation (bottom row) with synthetic observations for $O_3$ gb+S4 $NO_2$ (left two columns) and $O_3$ gb+S5 (S4.09) $NO_2$ (right two columns). First column for each instrument is from Model Run without data assimilation, second column is from assimilation run with assimilation of synthetic observations.**

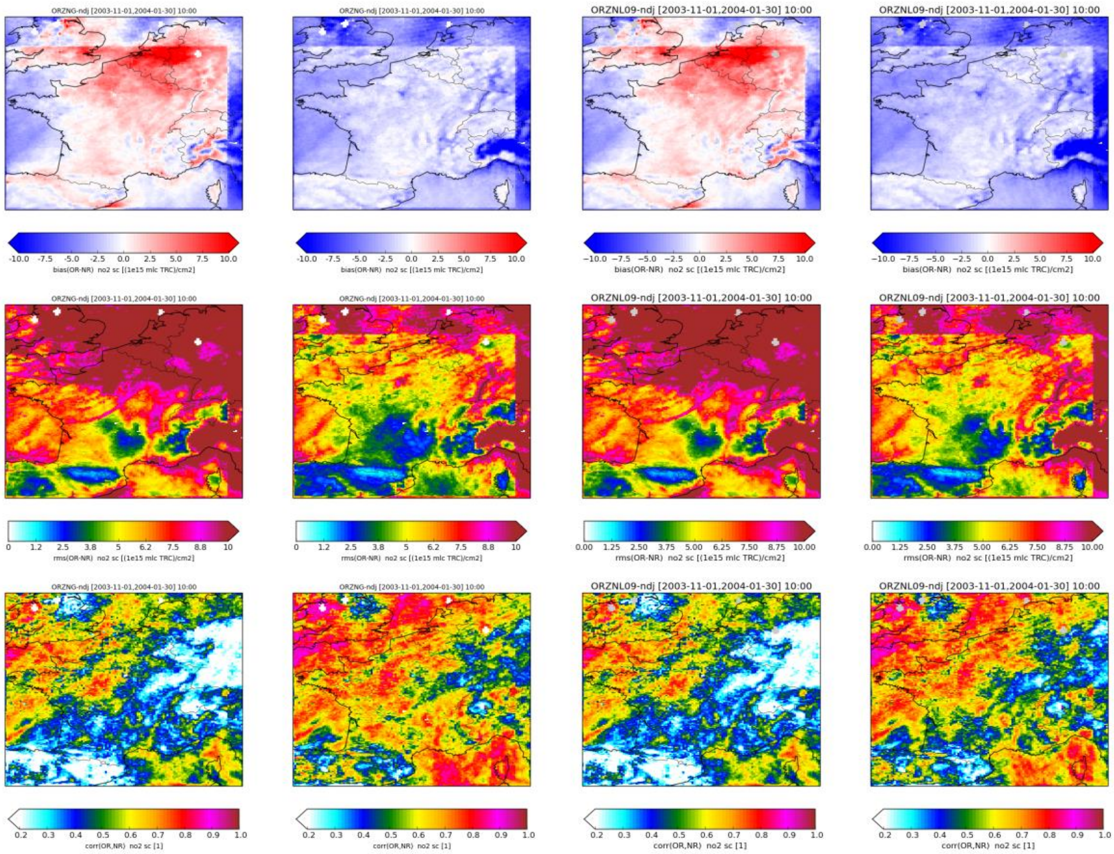


**Figure 34 Zoom – winter period: Bias (top row), RMSE (middle row) and correlation (bottom row) with synthetic observations for $O_3$ gb+S4 $NO_2$ (left two columns) and $O_3$ gb+S5 (S4.09) $NO_2$ (right two columns). First column for each instrument is from Model Run without data assimilation, second column is from assimilation run with assimilation of synthetic observations.**

### 5.2.2 Impact on total $NO_2$ columns

Figure 35 shows the statistics for the satellite $NO_2$ columns as function of hour of the day for the zoom domain over the summer study period.

The bias is increased through assimilation of the observations, this is explained in Chapter 2. Nevertheless assimilation of $NO_2$ column observations improves the RMSE and correlation, for the geostationary S4 during entire day and for the S5 (S4.09) and S5P/S4.13 from around overpass time until nighttime, clearly pinpointing the added value of geostationary observations. Again we see that the results for S5P and S4.13 are very similar, except for the fact that S4.13 includes only 13h observations leading to an impact from that time on, while the S5P dataset also contains observations from ~11h to ~15h (local overpass times of 13:30 can be somewhere between 11 and 15 UTC) resulting in an impact starting at 11h.

The assimilation of S5 observations has a similar effect as the assimilation of the S5P observations but at a different time of the day.

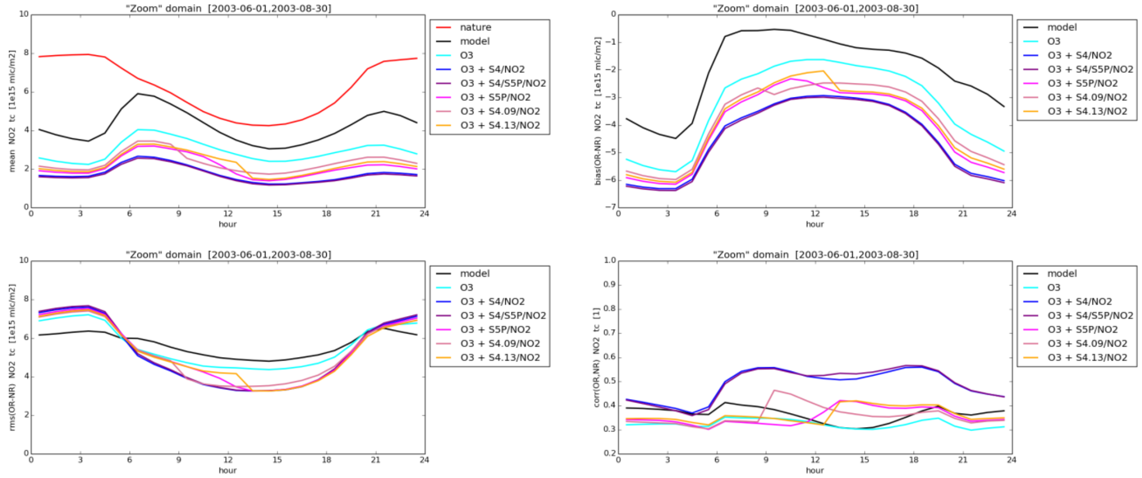


**Figure 35 Zoom domain – summer: $NO_2$ column (top left), bias (top right), RMSE (bottom left) and correlation (bottom right) prior (black line) and after assimilation of observations (colored lines, $O_3$ gb+S4 $NO_2$ (blue), $O_3$ gb+S5P $NO_2$ (pink), $O_3$gb+S4 and S5P $NO_2$ (purple), $O_3$ gb+S4.09 $NO_2$ (light pink) or $O_3$ gb+S4.13 $NO_2$ (yellow)).**

### 5.2.3 Impact on surface $NO_2$ concentrations

Figure 36 shows the statistics for the zoom domain and summer period as function of the hour of the day. Again it can be seen that the observations from the geostationary satellite have a positive impact on the RMSE and temporal correlation throughout the entire day while the observations from the low-earth orbiting satellite with one overpass a day have an impact starting from the overpass time. The combined assimilation of both sentinel 4 and 5P data does not largely improve the results from the S4 only.

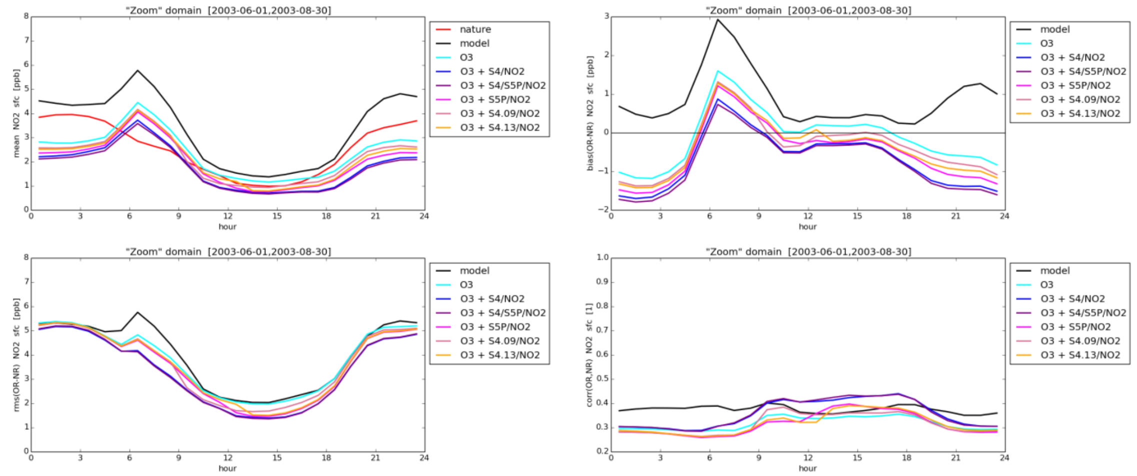


**Figure 36 Zoom domain – summer: surface $NO_2$ concentrations (top left), bias (top right), RMSE (bottom left) and correlation (bottom right) with nature run concentrations prior (black line) and after assimilation of observations (colored lines, $O_3$ gb+S4 $NO_2$ (blue), $O_3$ gb+S5P $NO_2$ (pink) , $O_3$gb+S4 and S5P $NO_2$ (purple) ), $O_3$ gb+S4.09 $NO_2$ (light pink) or $O_3$ gb+S4.13 $NO_2$ (yellow)).**

## 5.3 S5P versus S5 value for emission estimates

In the previous section we showed that the effect of the assimilating S5 is similar to the effect of assimilating S5P observations, except that the impact is seen at different times of the day. Largest impacts can be seen right after the different overpass times of the instruments.  As emission inversion is one of the growing application areas of satellite data we would like to know whether this dependency on time of the day will influence the usability of Sentinel 5 versus Sentinel 5P data. One expects that the morning S5 observations are influenced more by the traffic emissions that peak in the morning while the afternoon S5P observations are influenced more by the industrial emissions which remain more or less constant during the day.

For the purpose of the ESA GlobEmission project, a comparison has been made between what is observed in NO2 tropospheric columns measured by OMI and by GOME-2. The OMI overpass is at 13:30, the same as S5P, and GOME-2 overpass is at 9:30, as is the case for S5. For both instruments, a source-apportionment run with the model has been done to identify from which sector (e.g. traffic, powerplants) and from when (e.g. 1, 2, or 3 hours before observation) the NO2 in the column originates. In this study, the relative contribution of each sector to the measured NO2 column observed by the two instruments is very similar, as can be seen in Figure 37 for the Ruhr area in Germany and the Po valley in Italy. From these results we concluded that for use in emission inversion studies GOME-2 and OMI provide similar information. Thus, for emission monitoring it is not expected that S5 will mainly observe traffic related $NO_2$ while S5P mainly observes power plants related $NO_2$ for example.

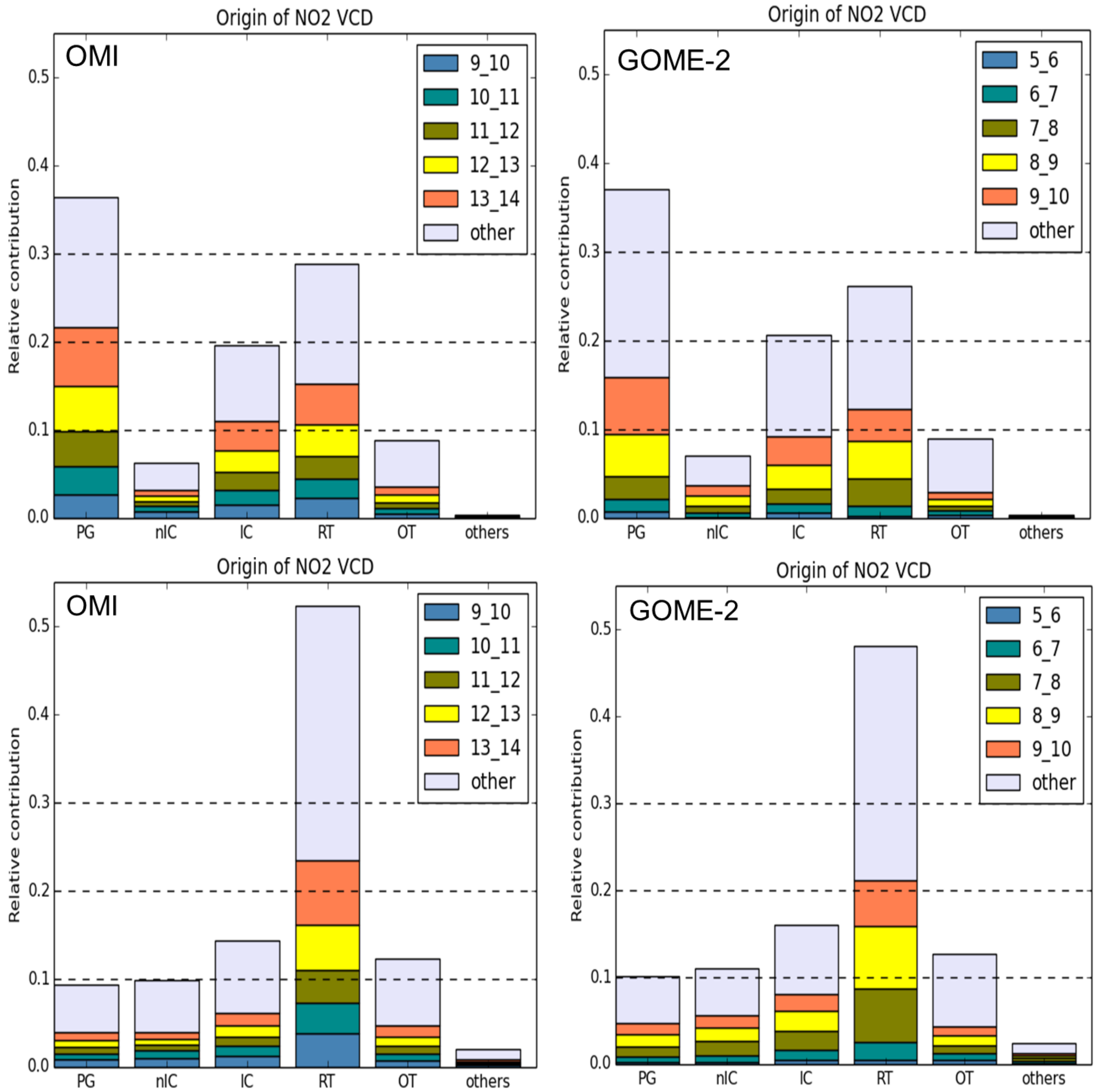


**Figure 37 Relative contribution of different source sectors and emission hours on modelled NO2 columns in the Ruhr area (top row) and Po valley (bottom row) using overpass time and averaging kernels from either OMI (left) or GOME-2 (right). PG: power generation, nIC: non-industrial combustion, IC: industrial combustion, RT: road transport, OT: other transport. Figure produced for the ESA/GlobEmission project.**

# 6 Conclusion and discussion

In this report we have shown the additional impact of the sentinel 4 (S4) and sentinel 5P (S5P) 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.

Both sentinel 4 and 5P $NO_2$ columns have a clear impact on modelled $NO_2$ values. 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.

The HCHO observations do show an added value in case of elevated HCHO values during a wildfire event. 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 sentinel 4 observations from one hour only can serve as substitute for simulated S5P or S5 observations as the impact from the S5P observations is similar to the impact of S4 at 13h observations. The results provide evidence that the above conclusions for sentinel 5P with an afternoon overpass versus the geostationary S4 can be transferred to Sentinel 5 with a morning overpass. 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.

For interpretation of the results the limitations of the assimilation scheme should be kept in mind. The scheme used in this study, as described in the *Assimilation Scheme Description Document*, assumes that the differences between simulations and observations could be explained from uncertainty in a number of selected model parameters (ozone precursor emissions of NOx and VOC, ozone deposition velocity, and ozone top boundary condition). These parameters are all allowed to vary within reasonable bounds. The advantage of this method is that model simulations are always forced by parameters with realistic values; besides, in case of air quality forecasts initiated by an assimilation, the analyzed parameters can be used to force the model .

Although the chosen parameter uncertainty is able to explain a large part of the difference between simulations and observations, not all of it can be explained. Mismatches due to extreme conditions are not captured for example, such as stagnant conditions that sometimes occur in winter and are difficult to describe by a meteorological model, but which have a strong impact on concentrations. The settings of the parameter uncertainty such as spatial and temporal correlation length are also chosen using statistical analysis of simulation-minus-observation mismatches. These are therefore statistical averages which describe over-all results representative for a longer time period, and might not be valid for specific events.

Another limitation of the method is that chemical correlation between tracers is induced by the chosen uncertainty. For example, a change in $NO_x$ emissions will induce correlated changes in $NO_2$ and $O_3$, while the actual uncertainty might have another reason. There is therefore not much freedom to change concentrations in a direction opposite of what is induced by an emission change. For ozone this is to some extend handled by the specification of uncertainty in the ozone-specific parameters for deposition velocity and top boundary condition.