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Validation report
On the Comparison of surface ozone in the global (2003-2010) and regional reanalyses (2011) over Europe.

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Contents

Abstract

MACC-II (Monitoring Atmospheric Composition & Climate) has created a consistent database of the atmosphere for various chemical compounds using state-of-the-art atmospheric modeling. The study is divided in two sections that concentrate on the evaluation of surface ozone over the European domain using ground-based measurements.

The first chapter is an extended analysis on the impact of assimilation in the MACC reanalysis using various statistical indices and temporal cycles for the period 2003-2010. The assimilation improves the bias of the model in all selected regions but some issues in the seasonality of the ozone were tracked and discussed.

The second part aims to investigate the added value of MACC-II regional air quality models compared to the global MACC reanalysis using data for 2011. Both regional and global simulations are assimilated. But it is important to note that the global MACC reanalysis was corrected using only satellite data that do not provide measurements close to the ground. On the other hand, the ensemble of the regional models was assimilated with observational data from the surface and thus the results were considerably improved.

1. Evaluation of the global MACC-II reanalysis with European surface ozone observations, 2003-2010

1.1. Introduction

The evaluation of the MACC-II reanalysis was made using measurements from 255 stations all around Europe, from the EMEP and AIRBASE database, for the period 2003 to 2010. Detailed information about the assimilation techniques can be found in Inness et al., 2013. From the AIRBASE database only rural stations were selected to avoid errors due to high-polluted stations from nearby human activity. A detailed re-classification on the type of stations is discussed in a recent study (Joly and Peuch, 2012). But in the present study the metadata from the AIRBASE database were used for the selection of rural stations. The initial units of the AIRBASE and EMEP measurements (μg/m^{-3}) have been converted to volume mixing ratio (ppbv), with the ideal gas equation using standard conditions for temperature and pressure (20°C and 1013.25hPa). This simplistic conversion of units may produce some small biases, especially for high altitude stations, but temperature and pressure was not available for the EMEP stations. Therefore a universal method for the conversion of units was used for both AIRBASE and EMEP databases.

Evaluation has been performed on a spatial and temporal basis. Observed data from the EMEP and AIRBASE database were available in hourly resolution, in contrast to the model values that were available in 3-hourly intervals. Temporal evaluation was performed on daily and monthly basis but no significant change in the indices was noted. Since many of the stations were not operating continuously, days (months) with missing hourly (daily) values more than 25% were excluded from the statistical analysis.

In order to acquire a more detailed view of model performance, eight European sub-regions have been used as shown in the following map (Figure 1). These regions fit data coverage and avoid overlapping between each sub-region. Surface stations for ozone and the number of stations for each region are also depicted in the map.

Some basic statistical parameters were discussed in Huijnen and Eskes (2012). The same parameters have been calculated to evaluate the performance of each experiment in the present report. These parameters help us to explain the amplitude, correlation and bias of each model in comparison with the observations. Furthermore, the daily and annual cycle of each station and European sub-region was calculated, for a more detailed view of photochemical production-destruction of ozone.

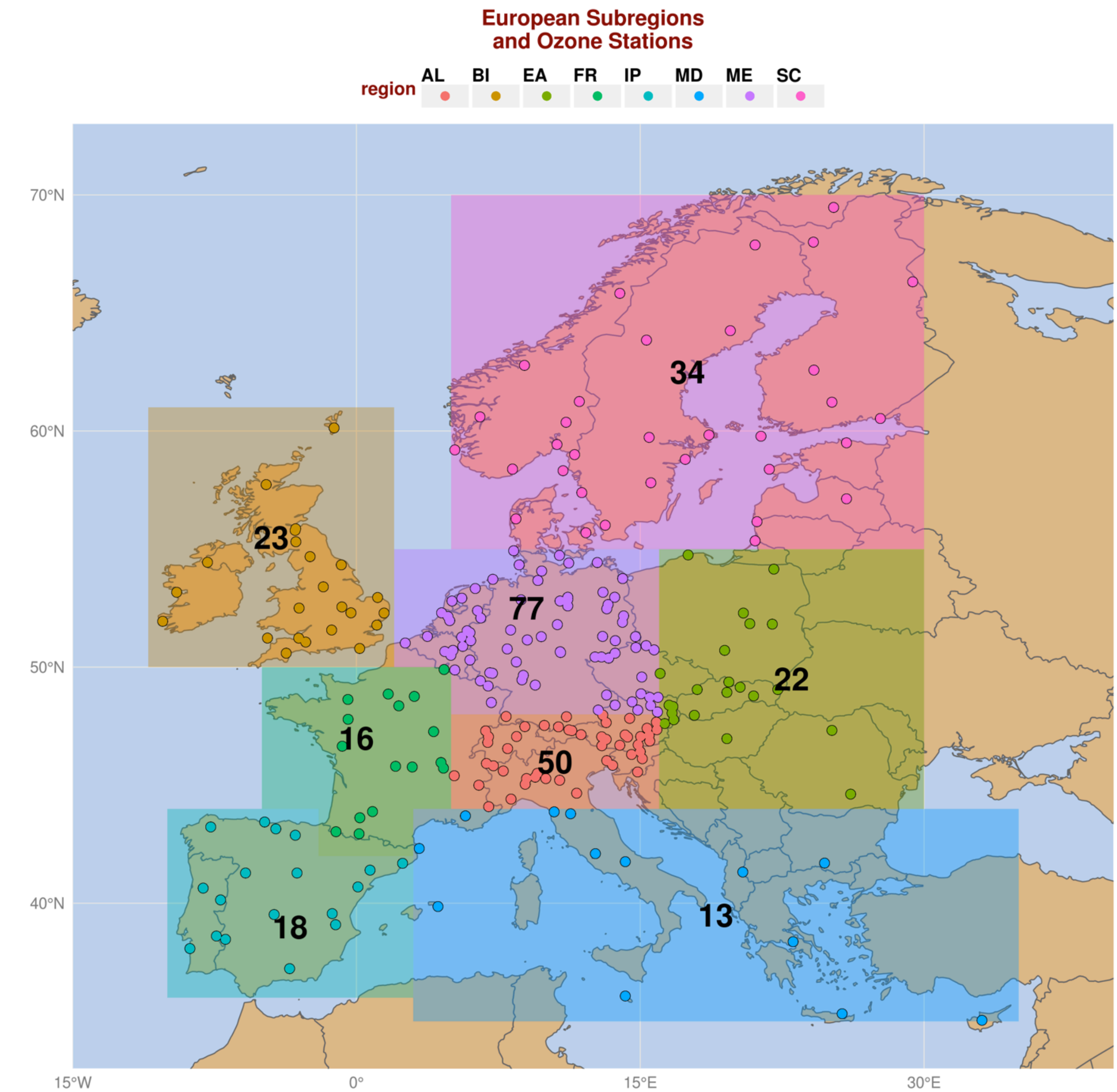


Figure 1. The eight European subregions that were used in the analysis and the corresponding stations of the EMEP and AIRBASE database.

1.2. Basic Validation Metrics

Some of the basic evaluation indices are presented in this chapter. Modified Normalized Mean Bias (MNMB), Fractional Gross Error (FGE) and Pearson Correlation (r) are some major statistics parameters that inform us directly about the performance of each experiment. The formulas used in this study are presented below. Where o and f is observed and model quantities, respectively.

$$MNMB = \frac{2}{N}\sum_i \frac{f_i - o_i}{f_i + o_i} \quad FGE = \frac{2}{N}\sum_i \left|\frac{f_i - o_i}{f_i + o_i}\right| \quad r = \frac{\frac{1}{N}\sum_i (f_i - \bar{f})(o_i - \bar{o})}{\sigma_f \sigma_o}$$

In Figure 2 we can see MNMB for both experiments by region. It is obvious that after the correction from non-assimilated (SA) to assimilated (MRE) experiment, bias generally improves, especially in Mid-Europe, Scandinavia and East-Europe. In some other cases the bias stays unchanged like in the Alps and in Mediterranean with close to zero bias in both experiments. In British Isles the bias simply changes sigh (from -10% to 10%) while in the IP becomes somewhat higher retaining its positive sign.

The Fractional Gross Error (FGE) is an indicator of the overall error in the model performance (Figure 3). Scandinavia and Eastern Europe seems to have the greatest FGE improvement. Overall in Europe the FGE of the model after assimilation decreases by 0.03, as it is indicated by the vertical lines. Furthermore, it is noted that after the assimilation mean FGE levels for all regions are close to 0.38, while before assimilation, the mean FGE varied more. The greatest impact of assimilation is seen over Scandinavia and Eastern Europe, where the bias is improving by a factor of 20% when assimilation processes are included to the system.

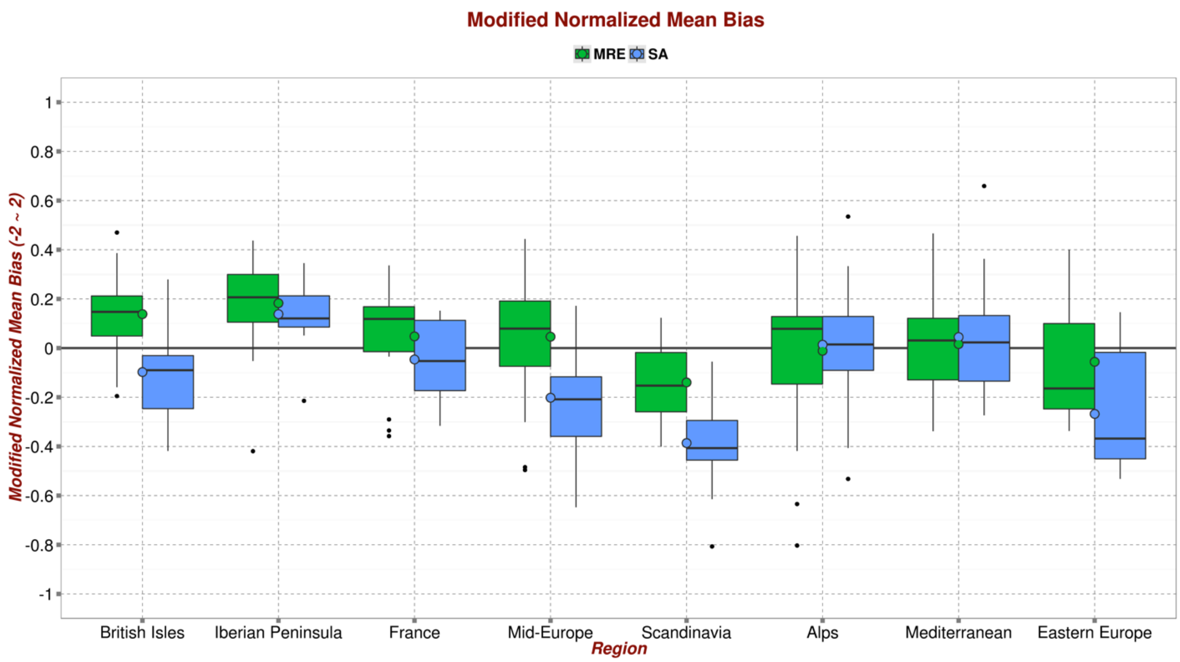


Figure 2. Modified Normalized Mean Bias (MNMB) in box plots for every European subregion. MNMB value was calculated for each station from monthly values. Green and blue colors correspond to MRE and SA experiment respectively. The point next to each box plot shows the mean value of MNMB and grey line on zero value, distinguishes over and under estimation for each region.

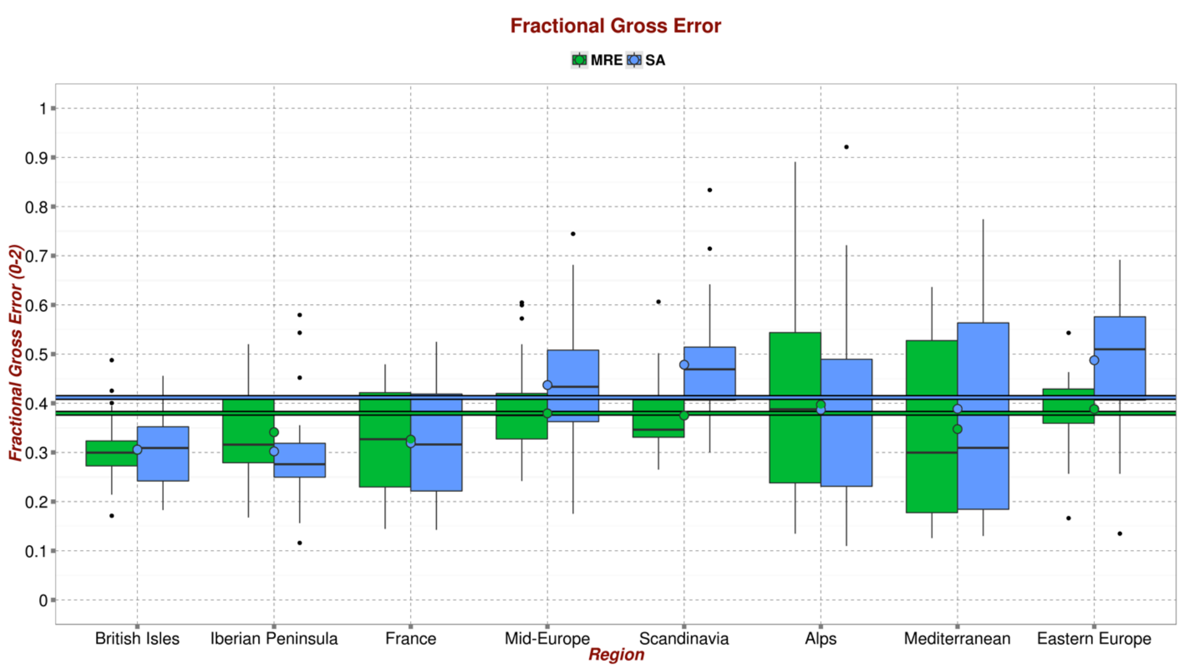


Figure 3. Fractional Gross Error (FGE) in box plots for every European subregion. FGE value was calculated for each station from monthly values. Green and blue colors correspond to MRE and SA experiment respectively. The point next to each box plot shows the mean value of FGE for each region. Vertical lines indicate the mean value of FGE for all regions.

The Pearson correlation is a parameter that corresponds to the degree of linear dependence between two variables. Correlation (**Figure 4**) is slightly reduced in the assimilated experiment (MRE). Scandinavia, Iberian Peninsula and Eastern Europe are the regions where the assimilation tends to reduce correlation the most, by a factor of 0.1. France and Mid-Europe seems to perform better with index values close to 0.60 for assimilated data, unlike Scandinavia were correlation is 0.35 and annual cycle is simulated with a late summer maximum, instead of an early spring maximum. Also **Table 1** shows that the correlation in the monthly analysis is higher compared to the daily in most regions. This is not the case only for the regions British Isles and Scandinavia, which is derived from the monthly values, cannot be reproduced correctly both from the assimilated and the not assimilated experiments. Thus the monthly correlation is very low in these two regions and the daily analysis displays a better score.

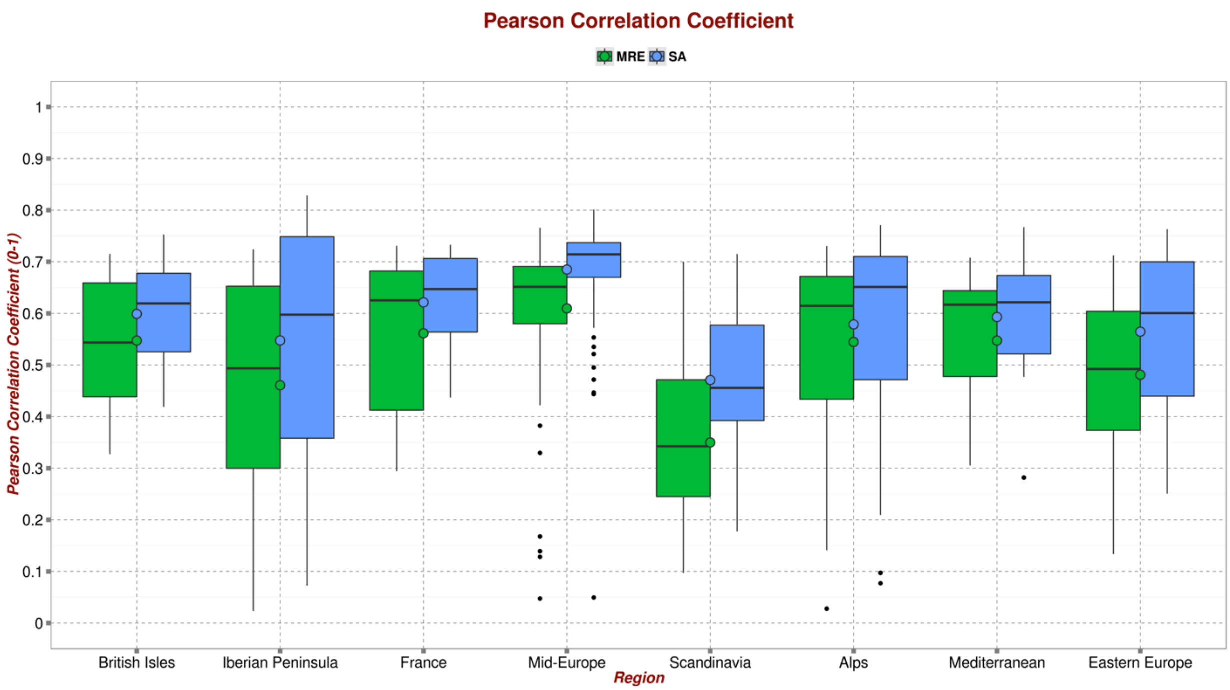


Figure 4. Pearson correlation coefficient (r) in box plots for every European subregion. Correlation coefficient was calculated for each station from monthly values. Green and blue colors correspond to MRE and SA experiment respectively. The point next to each box plot shows the mean value of correlation coefficient for each region.

In the following diagram (**Figure 5**) Pearson correlation is plotted as a function of longitude, latitude and altitude. Correlation coefficients appear in general to be lower close to the north or East parts of Europe (latitudes >50° or longitudes >20°), slightly deteriorating when assimilation is applied.

Altitude seems to have no effect on correlation, every group has values ranging between 0.5 and 0.7 and that reflects that experiments have an equal degree of sensitivity, both for photochemistry processes that are prevailing in low altitude stations and for free tropospheric conditions that are evident in high altitude stations. Although it is important to mention that reductions in correlation between non-assimilated and assimilated data is greater for stations closer to sea level altitude (< 500m) and medium altitude stations (1000m - 1500m).

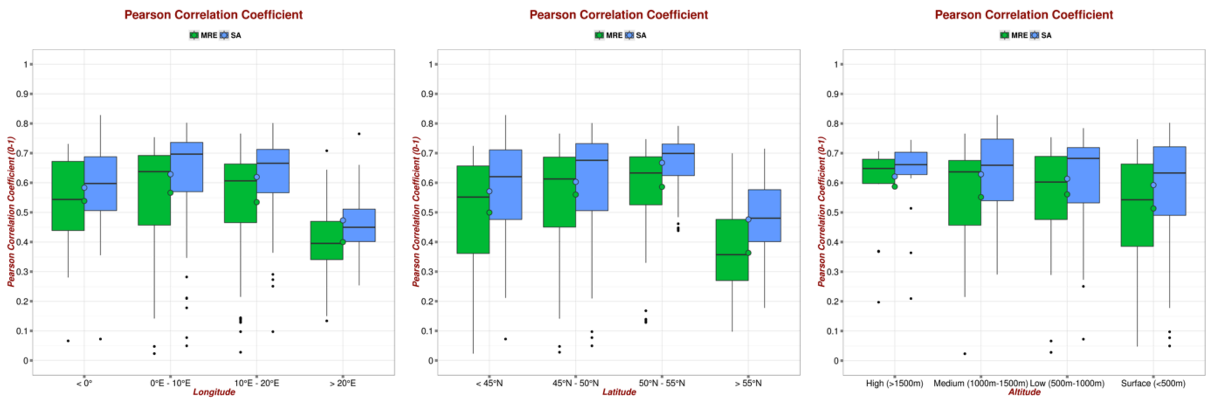


Figure 5. Pearson correlation coefficient (r) in box plots as a function of, longitude, latitude and altitude. Correlation coefficient was calculated for each station from monthly values. Green and blue colors correspond to MRE and SA experiment respectively. The point next to each box plot shows the mean value of correlation coefficient for each group.

Table 1. Mean values of basic statistics parameter for each region. Mean Bias (MB), Root Mean Square Error (RMSE), Modified Normalized Mean Bias (MNMB), Fractional Gross Error (FGE), Pearson Correlation Coefficient (r). Top and bottom panel illustrates results that were calculated from daily and monthly values respectively.

	MRE					sa				
Region	MB	RMSE	MNMB	FGE	r	MB	RMSE	MNMB	FGE	r
AL	1.19	14.63	-0.01	0.42	0.48	1.40	14.50	0.01	0.41	0.50
BI	4.98	11.06	0.14	0.31	0.55	-1.43	9.29	-0.10	0.31	0.60
EA	0.01	13.68	-0.06	0.39	0.45	-3.81	13.85	-0.27	0.49	0.53
FR	3.01	12.07	0.05	0.33	0.56	0.87	10.67	-0.05	0.32	0.62
IP	7.57	14.42	0.18	0.33	0.36	5.42	12.66	0.14	0.30	0.38
MD	0.30	13.58	0.02	0.35	0.39	1.20	15.17	0.04	0.39	0.41
ME	1.86	12.08	0.05	0.38	0.61	-2.46	11.48	-0.20	0.44	0.68
SC	-4.17	13.09	-0.20	0.42	0.34	-8.63	13.19	-0.39	0.48	0.47
	MRE					sa				
Region	MB	RMSE	MNMB	FGE	r	MB	RMSE	MNMB	FGE	r
AL	0.59	12.12	-0.04	0.34	0.56	-1.58	11.67	-0.01	0.32	0.58
BI	4.99	9.34	0.13	0.25	0.49	0.94	7.19	-0.08	0.23	0.59
EA	-0.14	10.55	-0.05	0.29	0.58	0.34	11.36	-0.25	0.40	0.62
FR	2.94	9.29	0.05	0.24	0.69	0.92	7.83	-0.04	0.23	0.74
IP	7.43	12.07	0.18	0.28	0.42	0.74	10.05	0.13	0.24	0.46
MD	-0.08	10.62	0.01	0.27	0.44	0.69	13.37	0.03	0.34	0.46
ME	1.81	8.95	0.03	0.26	0.71	0.76	8.87	-0.18	0.33	0.77
SC	-4.24	10.97	-0.18	0.34	0.26	-12.64	11.99	-0.37	0.43	0.40

Table 2. Summary statistics for all the stations. The three quartiles plus the 10% and 90% of the Modified Normalized Mean Bias (MNMB), Fractional Gross Error (FGE), Pearson Correlation Coefficient (r) from monthly data is presented.

	MRE			sa		
Region	MNMB	FGE	r	MNMB	FGE	r
10%	-0.28	0.18	0.10	-0.41	0.15	0.27
25%	-0.14	0.22	0.43	-0.31	0.22	0.54
50%	0.06	0.26	0.67	-0.14	0.32	0.71
75%	0.15	0.33	0.76	0.05	0.42	0.80
90%	0.27	0.42	0.80	0.15	0.50	0.84

In Figure 6, Figure 7 and Figure 8 we can acquire a quick summary of the performance of the two experiments. Modified Normalized Mean Bias for non-assimilated data has only negative values mostly over northern Mid-Europe. Assimilated time series is performing better with values close to zero in most stations. Same effect is evident in FGE map with a Northeast to Southwest improvement of absolute errors in non-assimilated data and a homogeneous improvement in assimilated data. Non-assimilated data have a slightly better performance in correlation compared with the non assimilated.

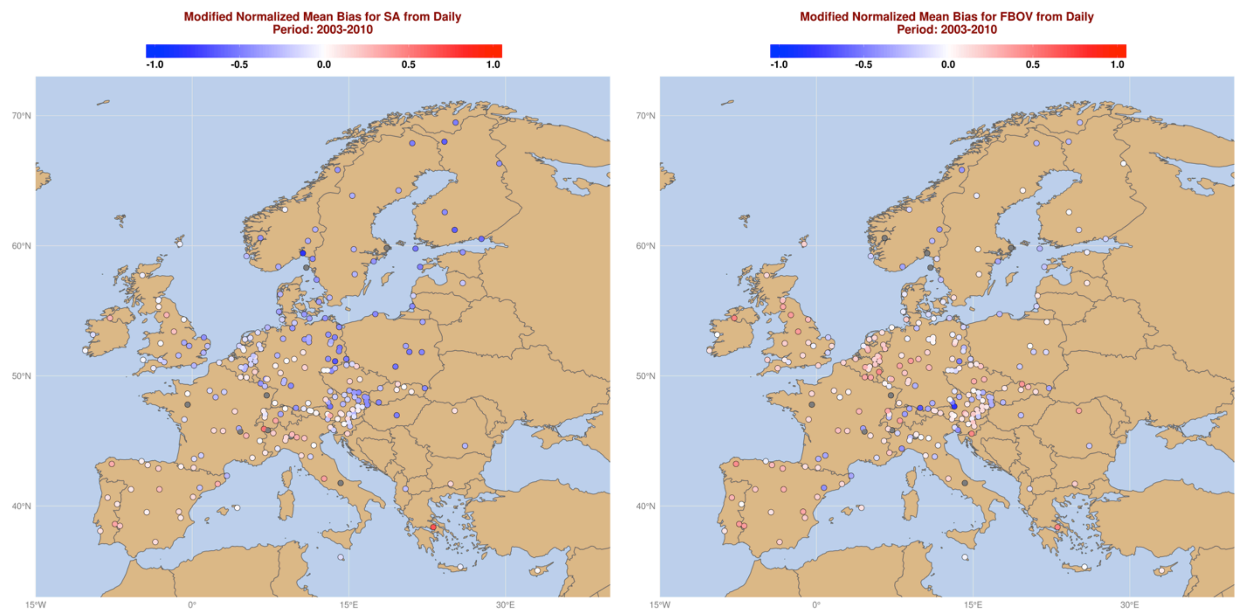


Figure 6. Modified Normalized Mean Bias (MNMB) of each station for SA and MRE experiment. MNMB was calculated using monthly values for each station.

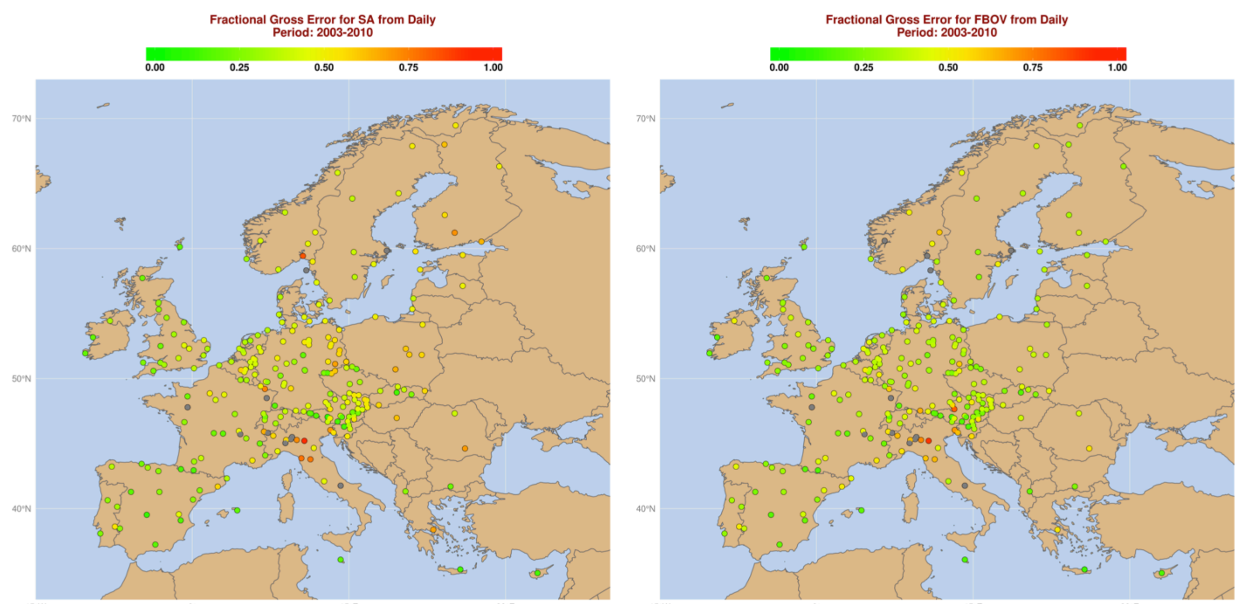


Figure 7. Fractional Gross Error (FGE) of each station for SA and MRE experiment. FGE was calculated using monthly values for each station.

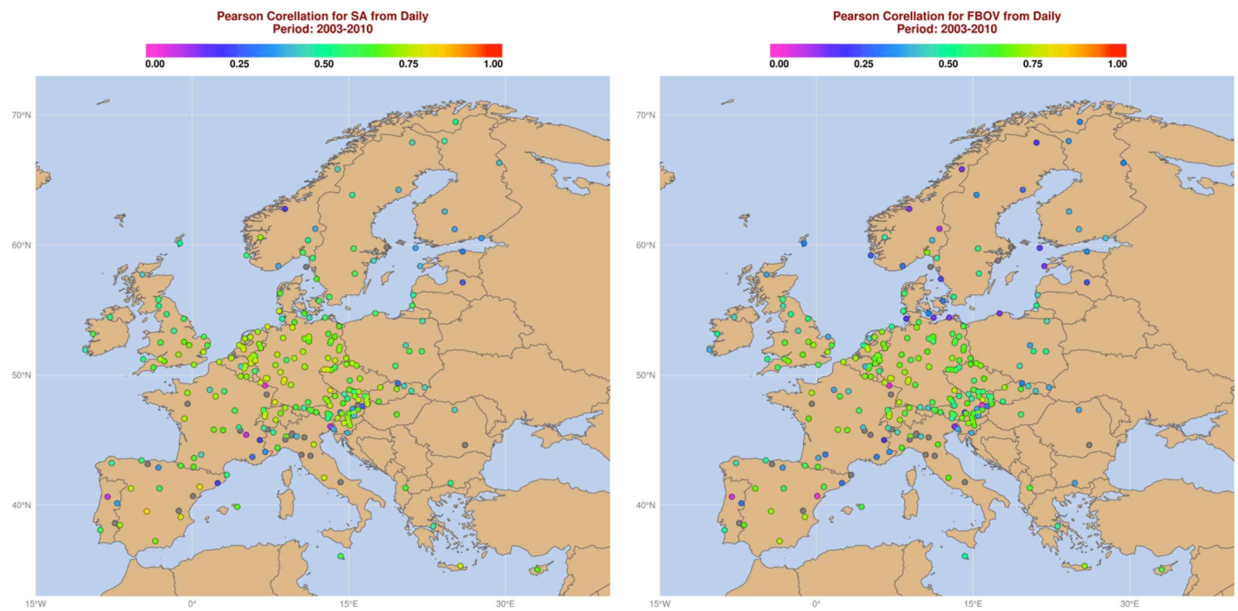


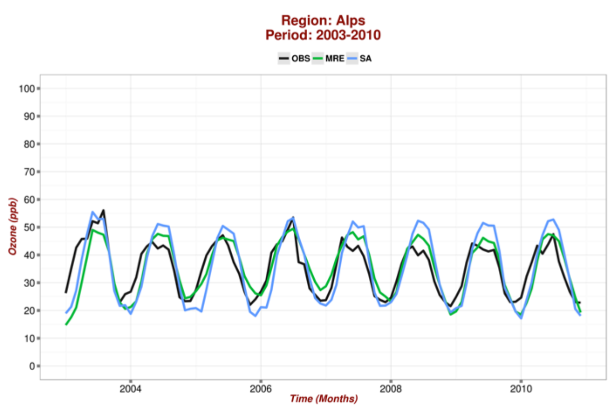
Figure 8. Pearson correlation coefficient (r) of each station for SA and MRE experiment. Correlation coefficient was calculated using monthly values for each station.

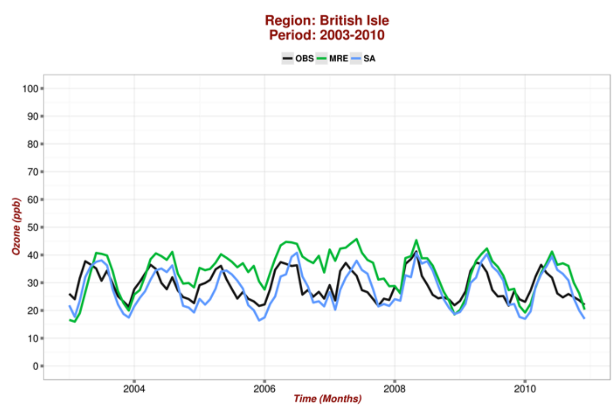
1.3. Time series

The monthly time series of surface ozone varies from region to region due to different chemical processes and regional characteristics (Figure 9). In this section we examine the monthly time series of the two experiments and compare them with the monthly time series derived from the rural EMEP and AIRBASE stations.

In southern regions, like the Mediterranean and Iberian Peninsula, where the annual cycle follows a clear high-summer and low-winter distribution both experiments perform very well. In the winters of the years 2005, 2006 and 2007 the assimilated experiment tends to overestimate the observations in Iberian Peninsula, Mediterranean, France and in other regions in a smaller degree. This overestimation in the winter of certain years is also discussed in the trend analysis of the period 2003-2010.

It is also important to note that in Scandinavia and the British Isles, where the spring maximum cannot be captured by the two experiments, the years 2005, 2006 and 2007 display a noise in the time series that can only be attributed to the assimilation. According to the MACC-II global reanalysis validation report (MACC_VAL_83.5, 2013), the bias of MACC reanalysis is close to zero in the year 2003 and it starts to increase till 2008 where it starts to deteriorate again in the following years. Assimilation data and method used in the MACC reanalysis model were changing over time and therefore some periods may behave better than others. These bias build up during the period 2003-2007 was first indicated in the Innes et al. (2013) and it was corrected from 2008 onward.





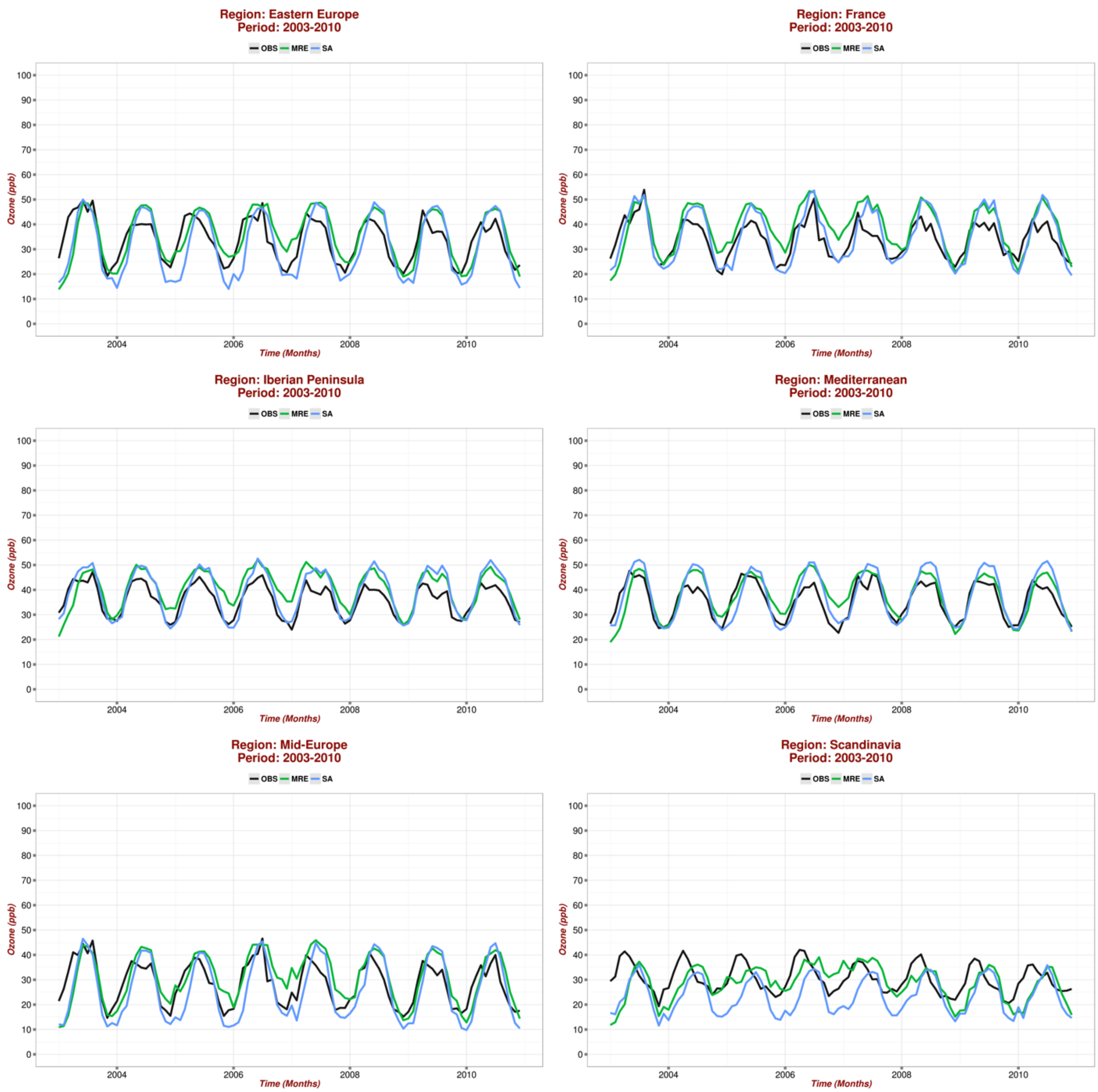
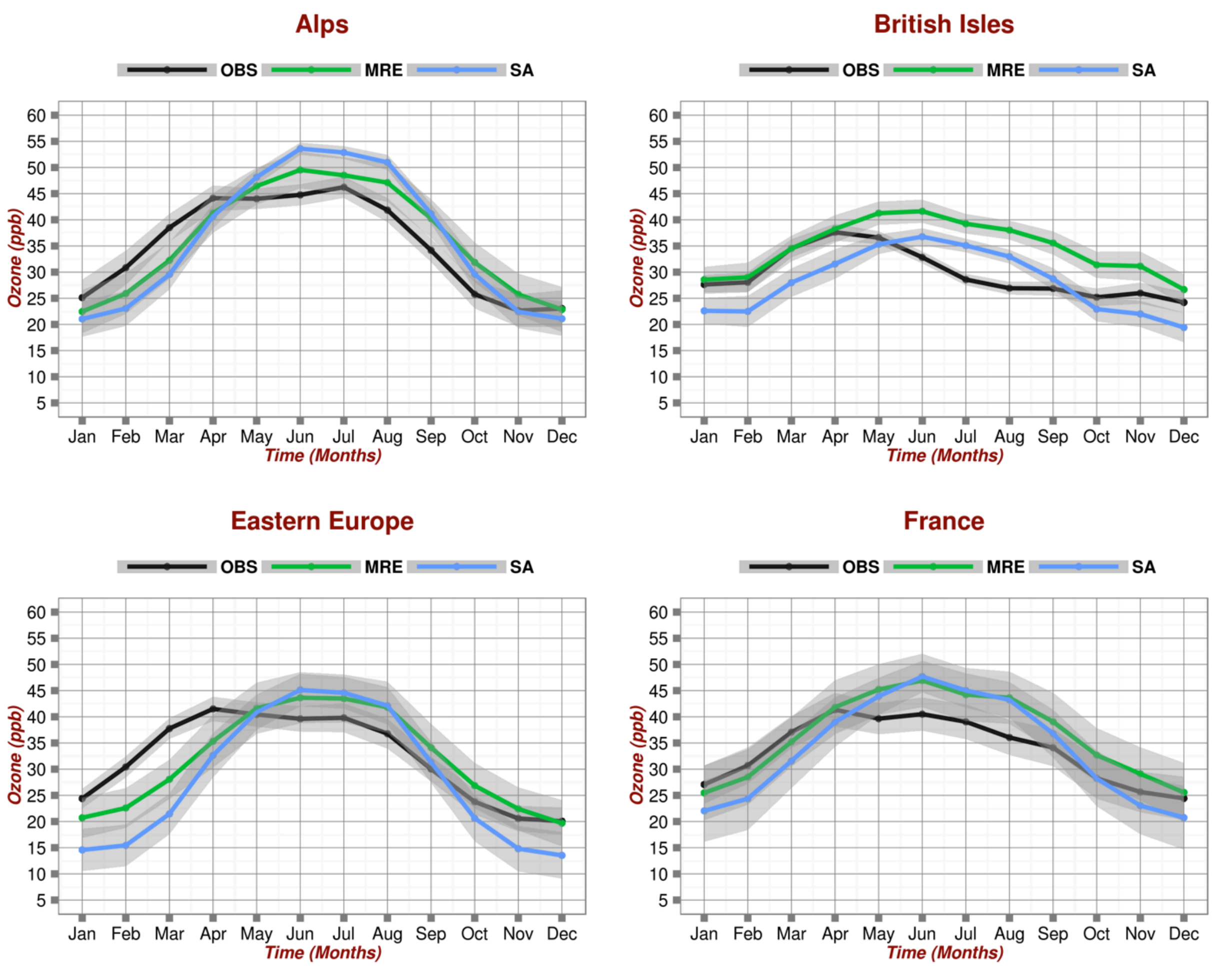


Figure 9. Time series of monthly values for each region. Black, green and blue colors represent the timeseries of observed data, MRE and SA experiment respectively.

1.4. Annual Cycle

The annual cycle for each European region is shown in the following diagrams (Figure 10). Black, Green and Blue colors represent observations (OBS), assimilated reanalysis (MRE) and the non-assimilated reanalysis (SA) experiments respectively.

The annual cycle in each region differs due to the different processes of production and destruction of ozone. The annual cycle of the EMEP and AIRBASE surface ozone observations over similar European sub-regions is described elsewhere in detail (Akritidis et al. 2013). The impact of assimilation on surface ozone is mostly towards the increase of non-assimilated (SA) surface ozone in the cold period of the year, reducing thus the model negative bias.



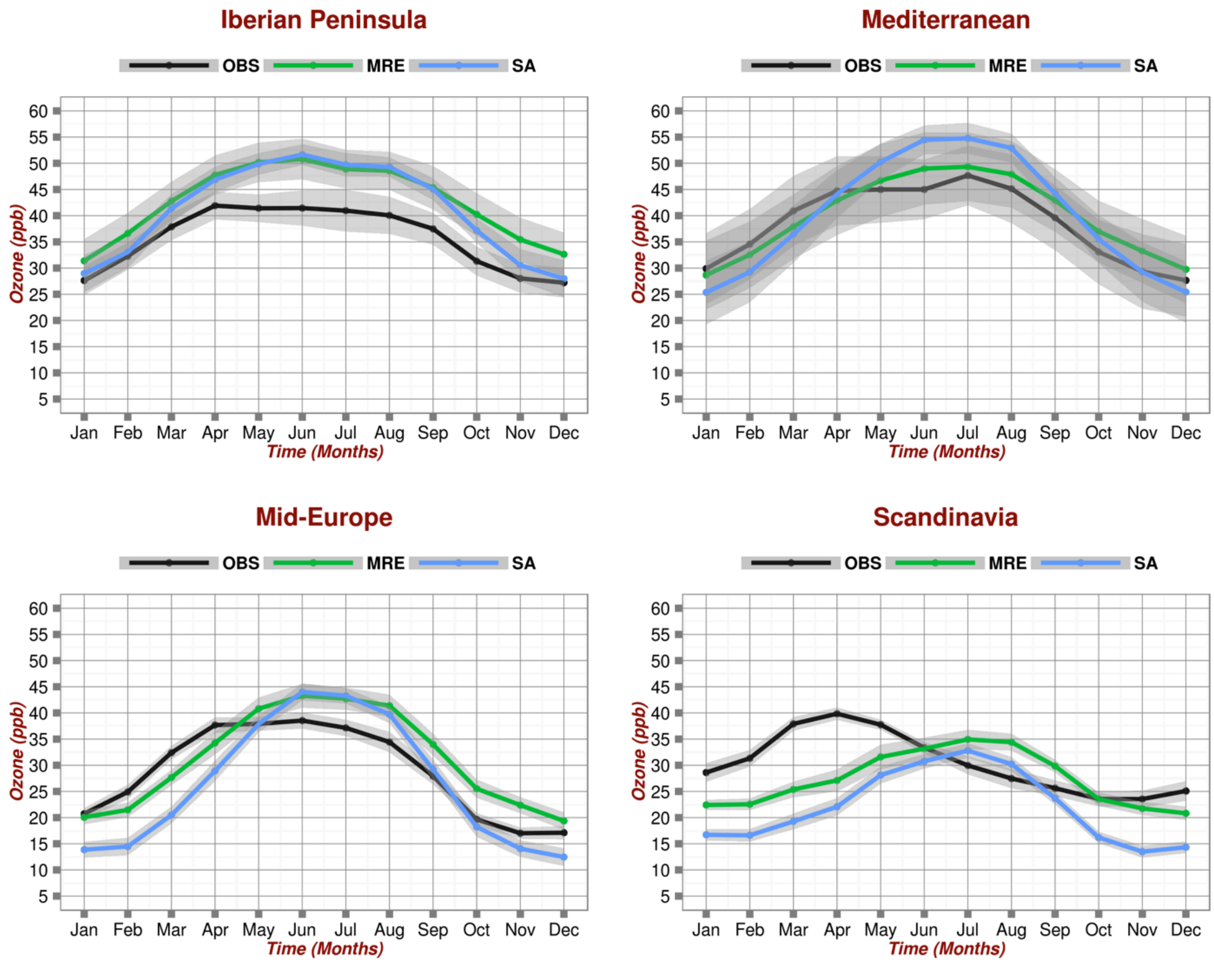


Figure 10. Annual cycles of O_3 for each European region. Black, green and blue colors represent the annual cycles of observed data, MRE and sa experiment respectively. Grey area distinguishes the 95% confidence level of the mean. Annual cycles have been calculated using monthly data of each station.

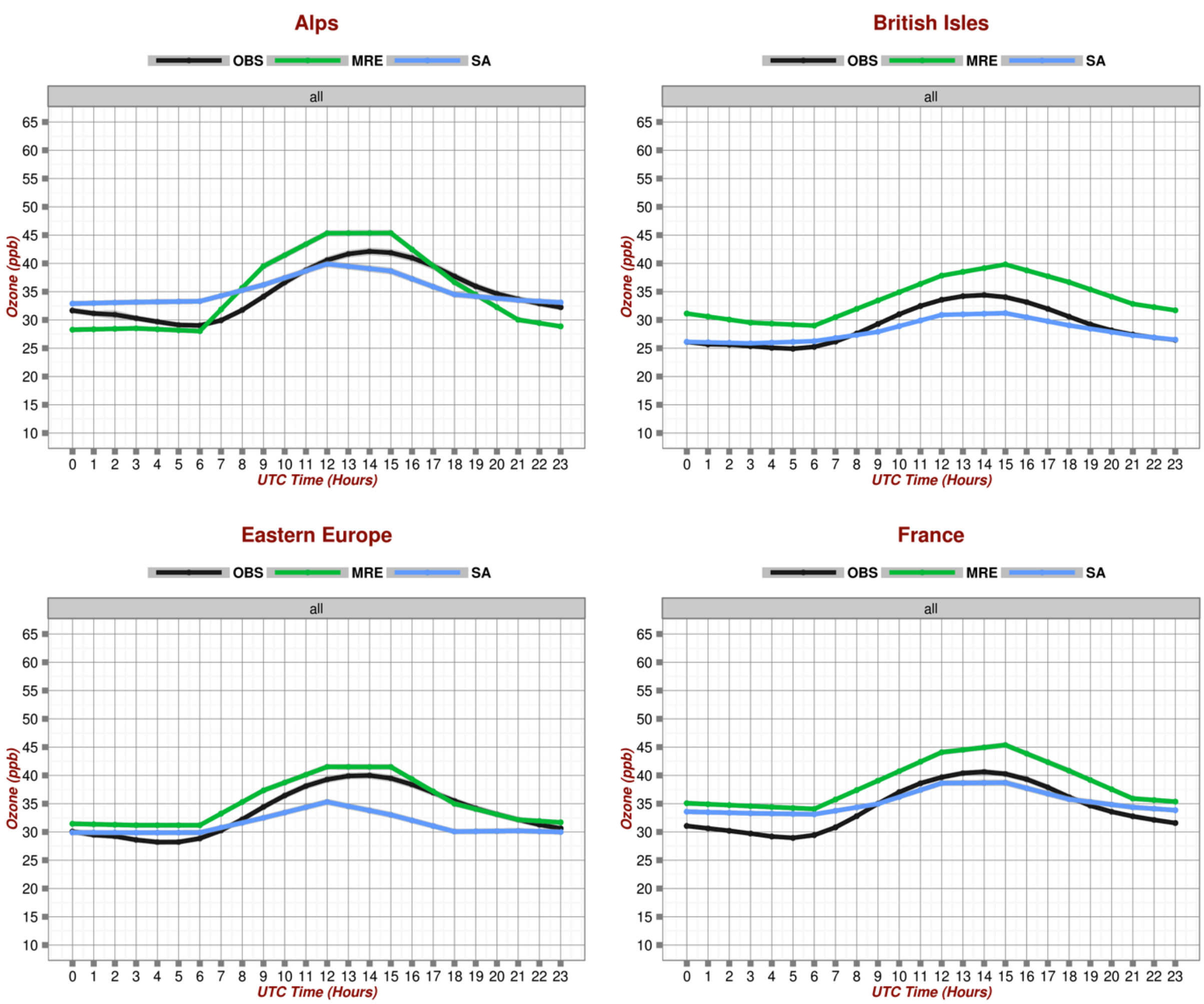
However the model annual cycle, fails to capture the early spring maximum, over northern Europe. If we examine the higher latitude sub-regions like Scandinavia or Eastern Europe, it seems that both experiments cannot reproduce the early spring maximum. These results are consistent with the MACC-II global reanalysis validation report (MACC_VAL_83.5, 2013), where in Europe the northern and central latitudinal regions were missing the annual spring maximum too (high negative bias during March and April). On the contrary, southern European sub-regions, like Iberian Peninsula and Mediterranean seems to reproduce the observational annual cycle and the broad Spring-Summer maximum better. In the Alps, where the majority of stations is established in high altitudes, the model is not capable to simulate the secondary spring maximum in April. Same results can be observed in previous studies that were conducted for larger European regions using only measurements from the EMEP database. The ability of the model to reproduce the annual cycle correctly decreases as the latitude increases (Benedictow et al. 2013; Inness et al. 2013).

It is evident from the discussion above that photochemical production of surface ozone, is driving a stable annual cycle in all regions, with winter minimum and summer maximum. The Spring maximum owes its origin to photochemical processes in the free troposphere of the northern mid-latitudes and to procedures of vertical transfer with stratospheric intrusion into the free troposphere (Monks 2000; Roxanne 2004). These processes are probably underestimated and maybe some development has to be made for regions of high latitudes (Scandinavia) and high altitudes (Alps), where vertical transfer sometimes plays a significant role in the variability of surface ozone. But in order to acquire safe conclusions, the effect of vertical transport needs to be validated and examined with meteorology measurements for each station. Also it is noted that most of the errors in air quality models is introduced by bias in emissions, boundary conditions and meteorological drivers (Solazzo et al. 2012).

1.5. Diurnal Cycle

As shown in Figure 11 the daily cycle in most regions is reproduced correctly. The daily cycle was calculated using values from all seasons. All observations were set in UTC time. Corrections from non-assimilated data to assimilated increase daytime concentration more than it does for night-time, creating a sufficient daily range that it is closer to reality. This pattern appears clearly in regions with a higher diurnal range, such as the Mediterranean, France, Mid-Europe and Alps. Yet, in some cases assimilation is increasing diurnal range more than necessary, as we will present in the next chapter (1.6. Diurnal Range).

In central mainland Europe (France, Mid-Europe and Alps), MRE data is overestimating observation in daytime and especially from 12:00 to 17:00 UTC time where ozone reach its maximum value due to photochemistry (van Loon et al. 2007). In these hours, photochemistry production peaks, due to the abundance of sunlight.



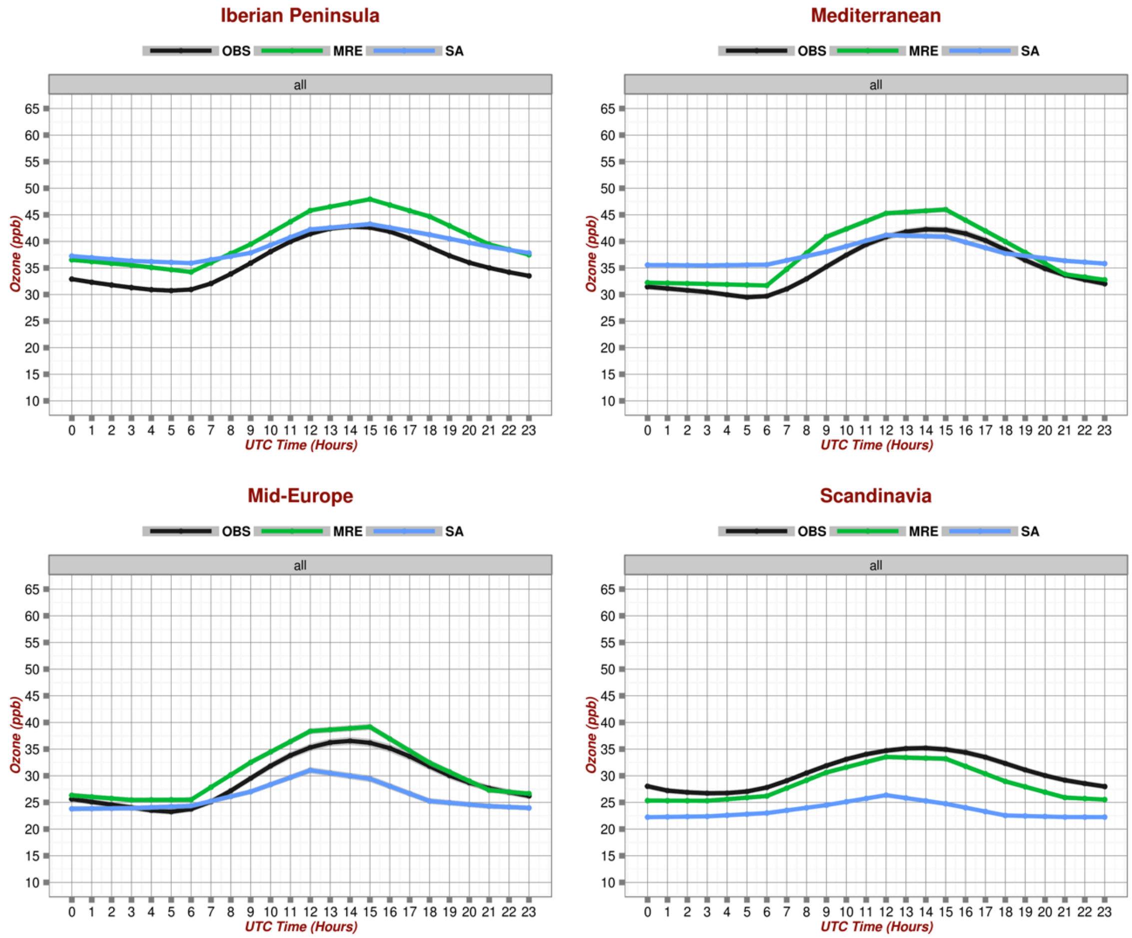
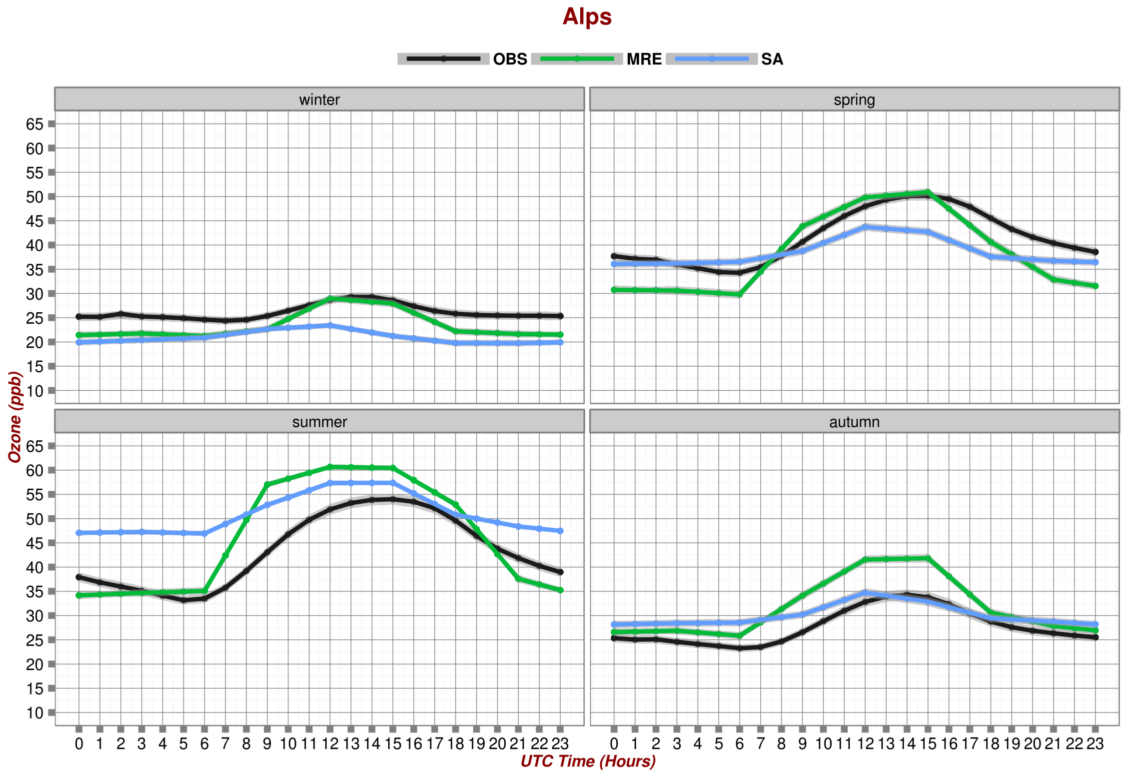
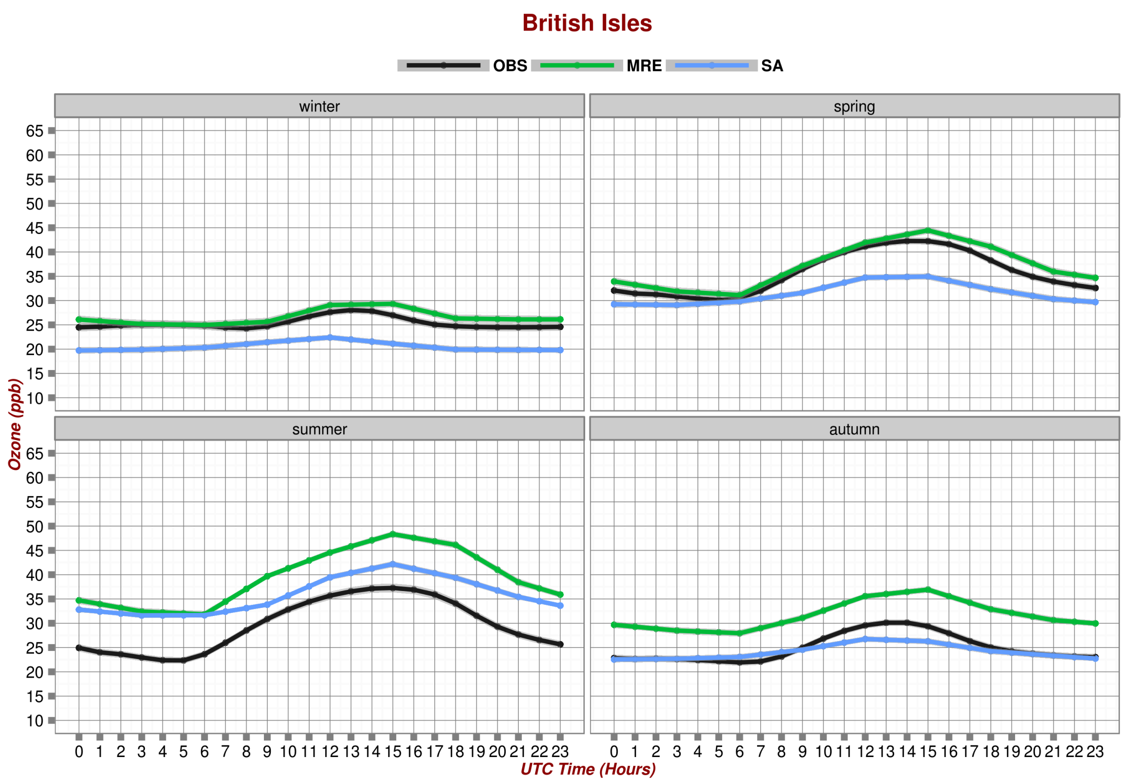
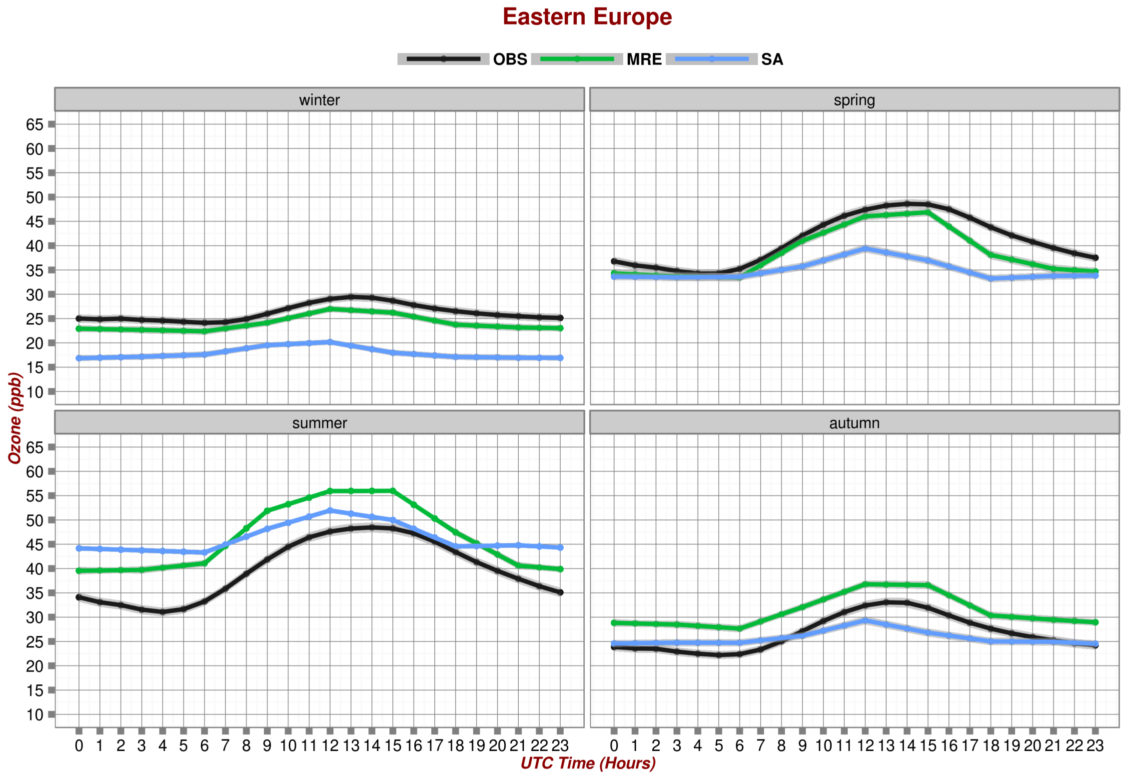


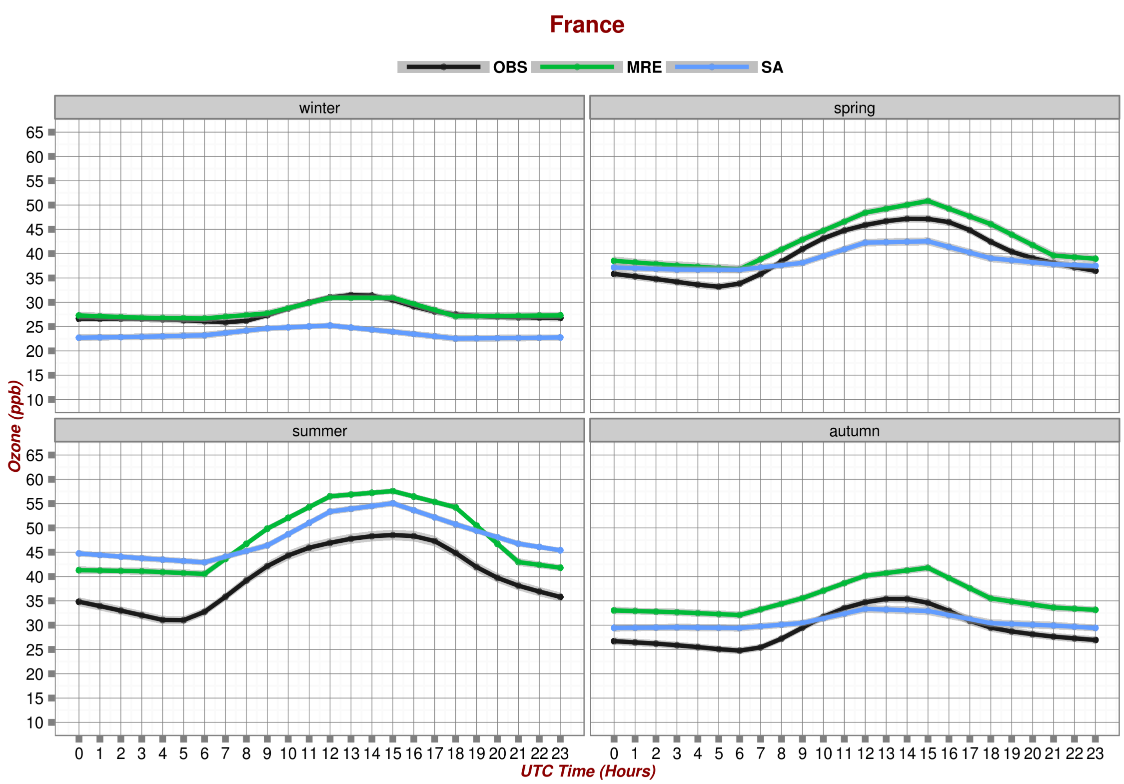
Figure 11. Diurnal cycles of O_3 for each European region. Black, green and blue colors represent the diurnal cycles of observed data (OBS), the MACC reanalysis experiment (MRE) and not assimilated (SA) experiment respectively. Grey area distinguishes the 95% confidence level of the mean. Diurnal cycles have been calculated using hourly data of each station.

Since daily production is modulated by solar intensity, it is important to evaluate the performance of the model by season (Figure 12). Results show that in all regions, winter is the season where the daily cycle is captured more accurately in the assimilated experiment. For example the winter daily cycle of Mediterranean and France is captured almost with zero biases by the MACC reanalysis model. This performance in the daily cycle by a global model of a coarse resolution is really astonishing, since the assimilation does not include surface observational data in the correction. On the other hand, summer poses more difficulties, since both production and destruction processes are more intense. In Mid-Europe for example, assimilation improves the performance of the model in winter and spring. But in autumn and especially in summer assimilation has the opposite effect. Overall the effect of assimilation in summer is positive because it creates higher diurnal amplitude, but the downside is that it increases the bias of ozone during the day.


Alps
OBS
MRE
SA
winter
spring
summer
autumn
Ozone (ppb)
UTC Time (Hours)


British Isles
OBS
MRE
SA
winter
spring
summer
autumn
Ozone (ppb)
UTC Time (Hours)


Eastern Europe
OBS
MRE
SA
winter
spring
summer
autumn
Ozone (ppb)
65
60
55
50
45
40
35
30
25
20
15
10
0 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 20 21 22 23
UTC Time (Hours)


France
OBS
MRE
SA
winter
spring
summer
autumn
Ozone (ppb)
65
60
55
50
45
40
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30
25
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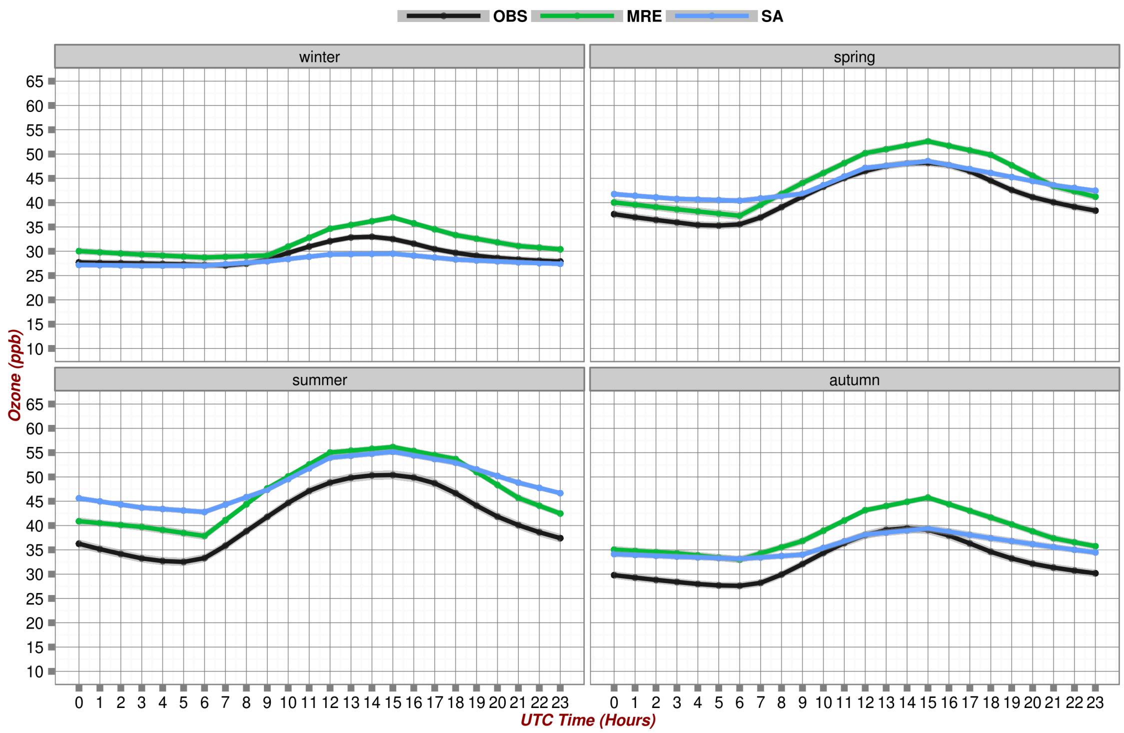
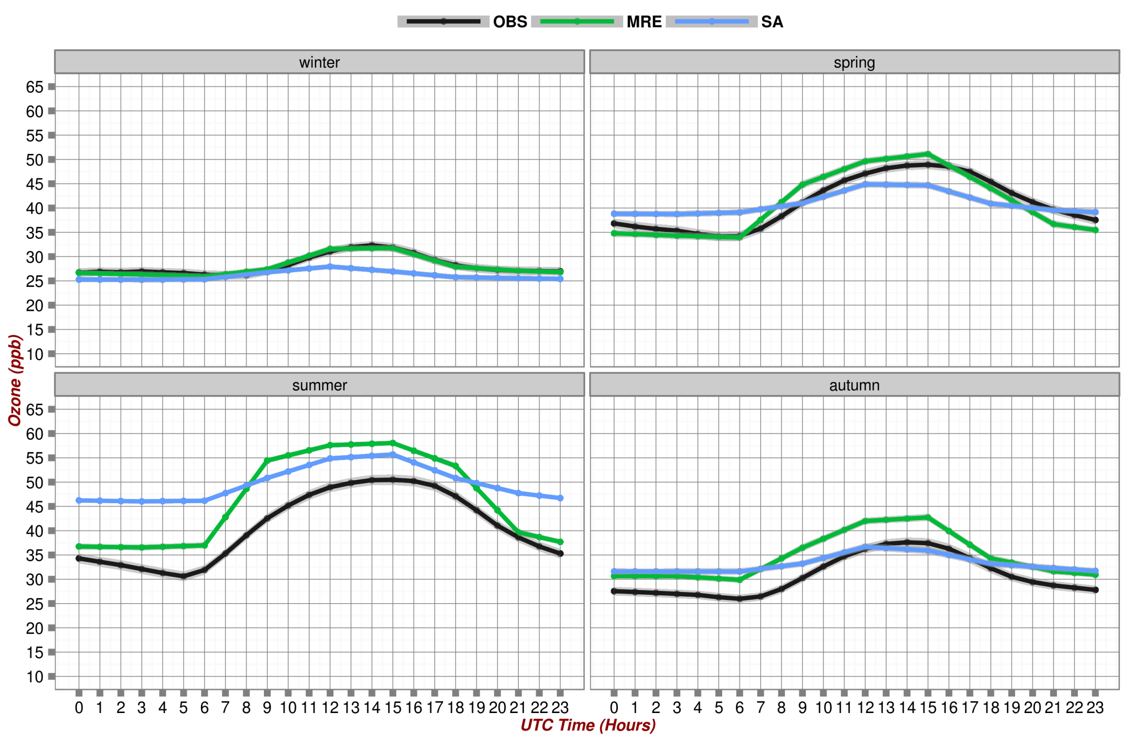
0 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 20 21 22 23
UTC Time (Hours)

Iberian Peninsula



Mediterranean



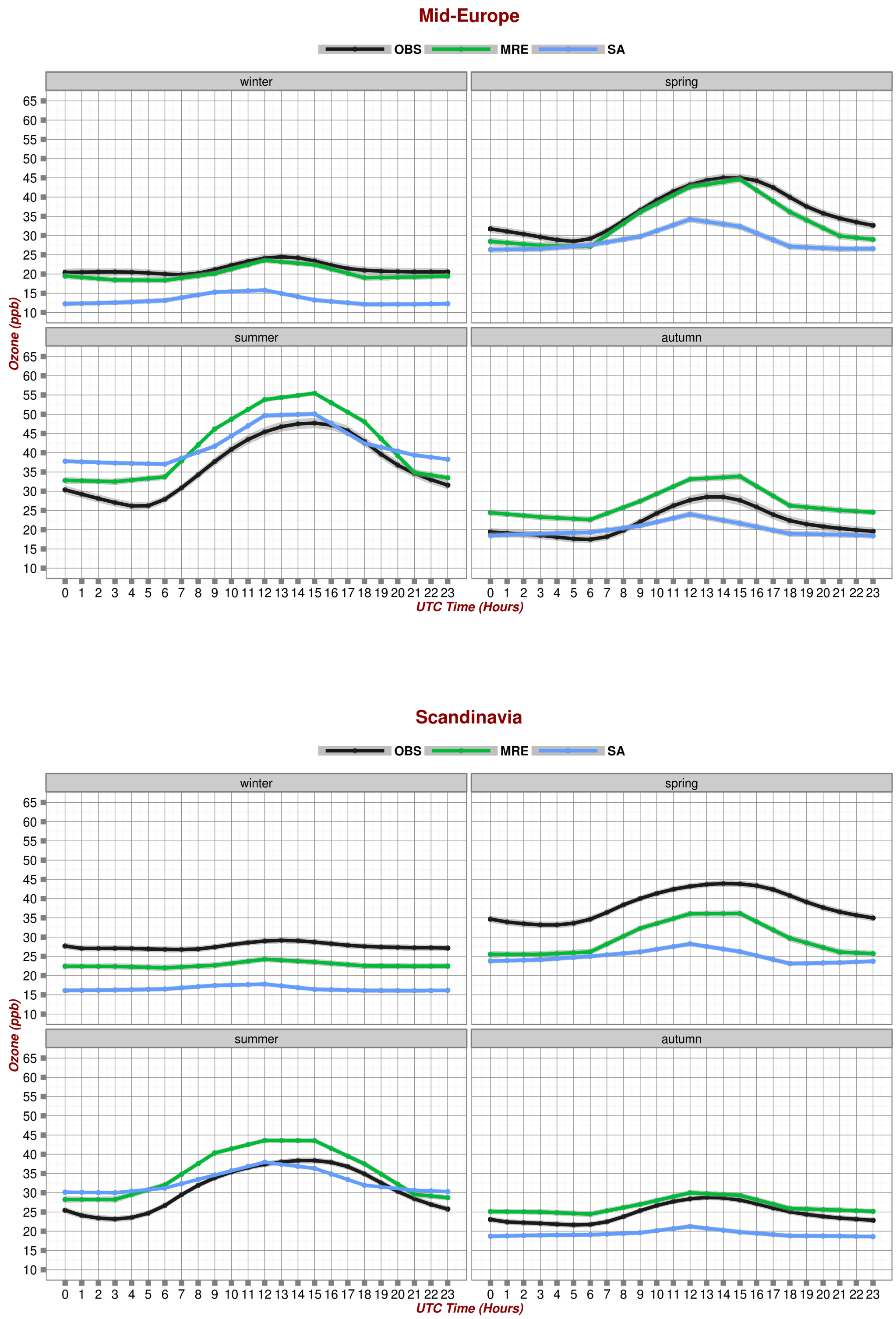


Figure 12. Diurnal cycles by season. Black, green and blue colors represent the diurnal cycles of observed data (OBS), the MACC reanalysis experiment (MRE) and not assimilated (SA) experiment respectively. Grey area distinguishes the 95% confidence level of the mean. Diurnal cycles have been calculated using hourly data of each station.

1.6. Diurnal Range

The diurnal range is the difference between the maximum and minimum values of ozone observed in one day. It is always positive and it relates to photochemical activity of ozone in the atmosphere. In the present analysis it was calculated by the subtraction of the maximum and minimum value for each day-station, and then calculating the averaged value for each region-season. In Figure 13 diurnal range is presented by region. In most cases the underestimation of the mean value of the diurnal range is evident for both experiments. In MRE there are regions like Alps or France, where the mean of the diurnal range matches the mean of the observations, but that doesn't necessarily mean that the model has a perfect performance in those regions, because the spread of the values is greater than the one on the observational data. The non-assimilated experiment displays a very low diurnal range, which means that photochemistry procedures are under-graded in that case. The greatest improvement, as far as the mean of the diurnal range, is observed in Alps.

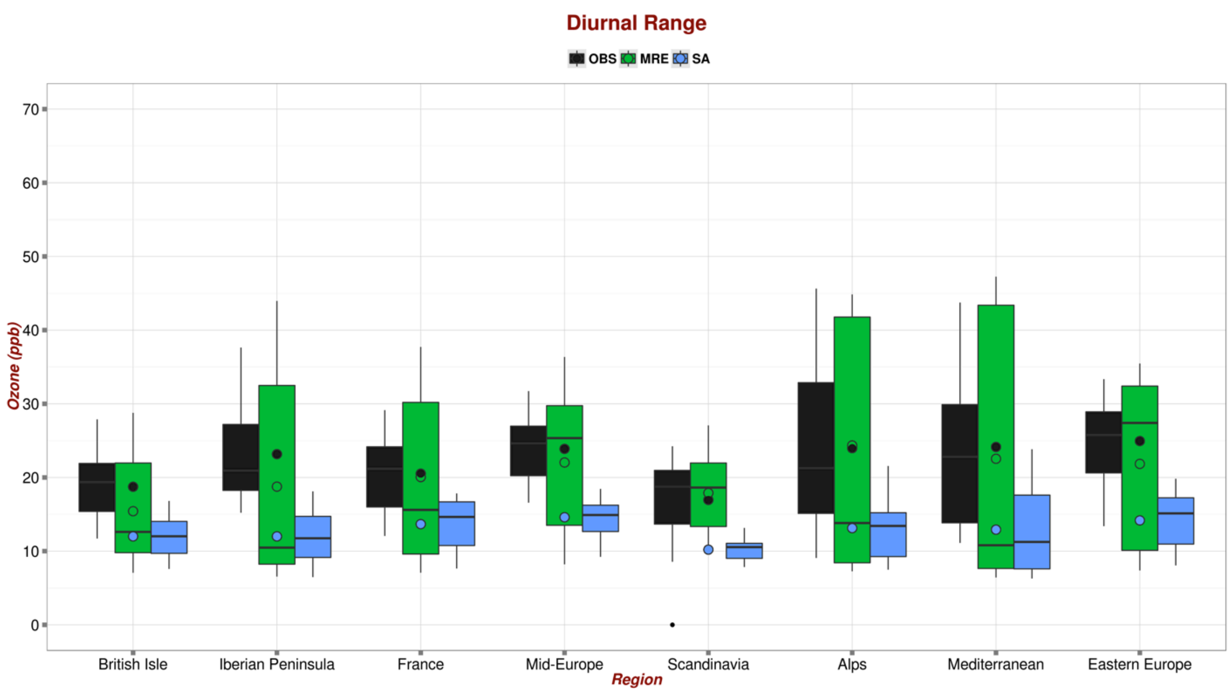


Figure 13. Diurnal range in box plots by region. Black, green and blue colors represent the diurnal range of observed data, MRE and sa experiment respectively. The points in the center of the box plot show the mean value of diurnal range for the observed data and the two reanalysis experiments.

Figure 14 illustrates diurnal range by season. We can see that the greatest spread and higher values are evident in spring and summer, when photochemistry production is maximized due to the abundance of sunlight. Overall assimilation enhances the diurnal range, by about 10ppbv (SA to MRE), reproducing therefore, more successfully the observations. But still, over most regions, the diurnal range remains under-estimated even after the assimilation. Another issue is the amplitude of variation in the diurnal range after the assimilation, which appears over most sub-regions overestimated (MRE). Especially over the Alps, despite the excellent agreement between the MRE and OBS mean amplitude of diurnal cycle, the presentation of percentiles indicates a greater dispersion of model diurnal ranges. On the contrary, before assimilation (SA), the model has lower amplitude in variation of the diurnal range.

Further investigation is necessary to evaluate and quantify the differences between night (minimum) and day (maximum) ozone concentrations in different time frames (by season) and the role of data assimilation in ozone diurnal range.

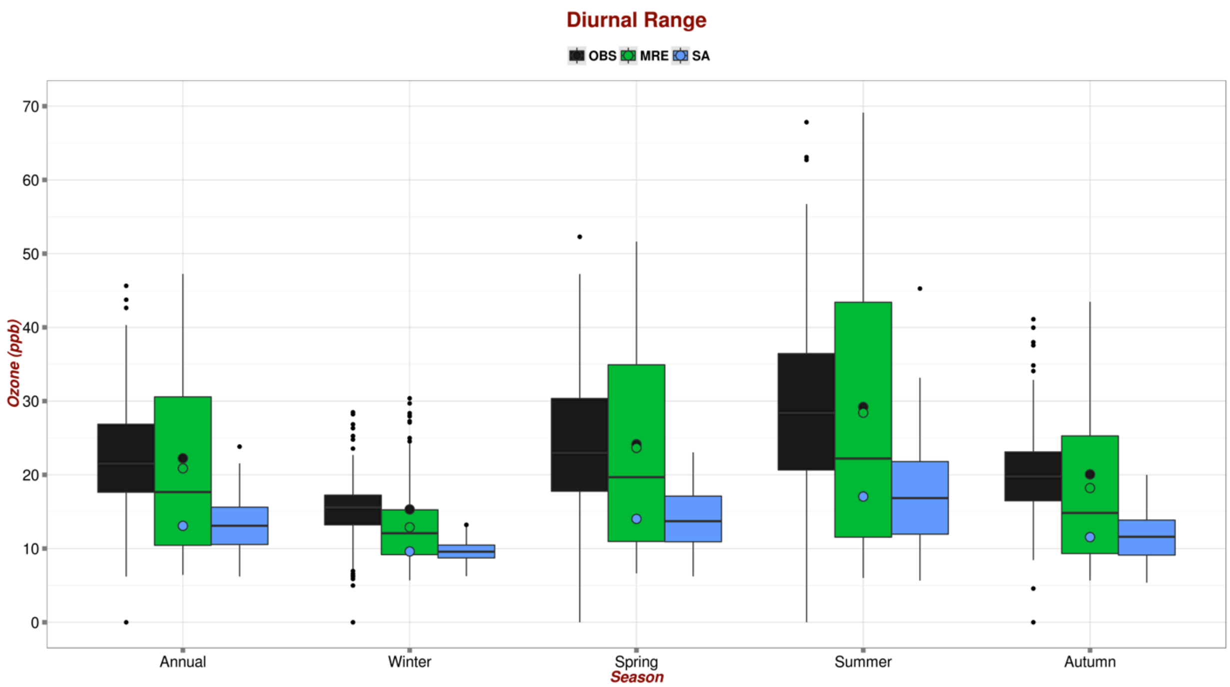
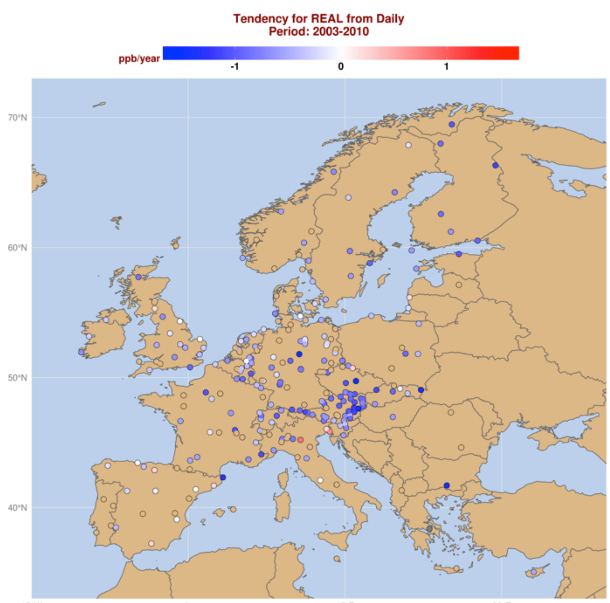


Figure 14. Diurnal range in box plots by season, averaged over the whole European domain. Black, green and blue colors represent the diurnal range of observed data, MRE and sa experiment respectively. The points in the center of the box plot shows the mean value of diurnal range for the observed data and the two reanalysis experiments.

1.7. Trends

The following maps (Figure 15) present ozone trends for observed data and the two experiments. Due to the short period examined, units were set in ppb/year. Stations with more than 25% of missing data have been excluded from this analysis. Observed data show a clear negative trend of ozone concentration all around Europe and especially in central Europe. The trend is about -1ppb per year in most cases. Probably this is explained with the negative trends of ozone precursors that it was observed in the last two decades (Colette et al. 2011; Derwent 2004; E. Gerasopoulos et al. 2005; Rouïl et al. 2009). It is worth mentioning, that the selected stations from the EMEP and AIRBASE database are classified as rural and they are located away from any nearby human activity. Different results may derive, especially in trends analysis, if we examine urban or sub-urban stations (Colette et al. 2011) or use a different classification method of the ground-based stations (Joly and Peuch, 2012).

Both SA and MRE experiments tend to have both negative and positive trends, ranging between -0.5ppb to 0.5ppb per year. The SA experiment does exhibit a stronger positive trend over some stations in eastern Mediterranean and northern Germany. MRE is performing better with more negative trends, but still those values are way smaller compared with the observed data.



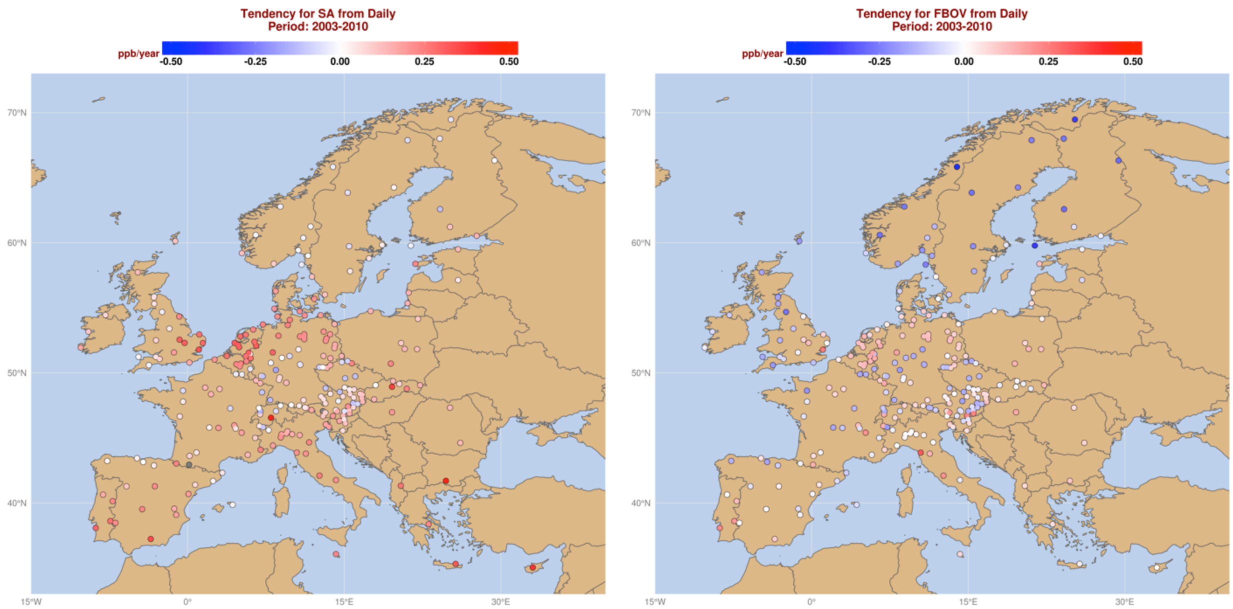


Figure 15. Trend (ppb/year) of each station for observed data, assimilated (MRE) and non assimilated (sa) experiments. Beige color represents stations that were excluded from this analysis.

It is noted that the build-up bias in the period 2003-2007, as stated in the MACC-II global reanalysis validation report (MACC_VAL_83.5, 2013), probably affects the trend of the MACC reanalysis experiment. A different approach in the trend analysis was conducted, comparing the mean values between the periods 2003-2004 and 2009-2010. In Figure 16 the difference between these periods is illustrated for each region. The two experiments cannot reproduce the high negative observational difference and results indicate a similar behavior with the previous trend analysis for both experiments. Therefore we can conclude that increasing bias in the period 2003-2007 in the MACC reanalysis model does not significantly change the trend of MRE during the period 2003-2010.

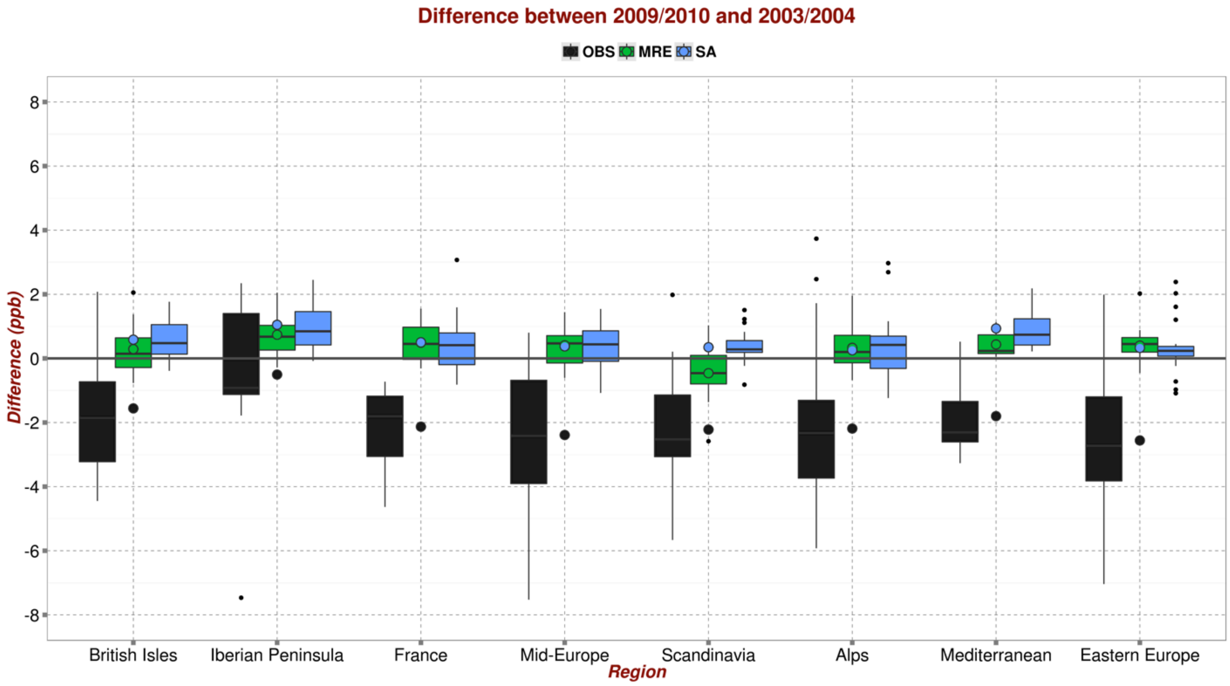


Figure 16. Difference of mean ozone values between the periods 2009/2010 and 2003/2004 by region.

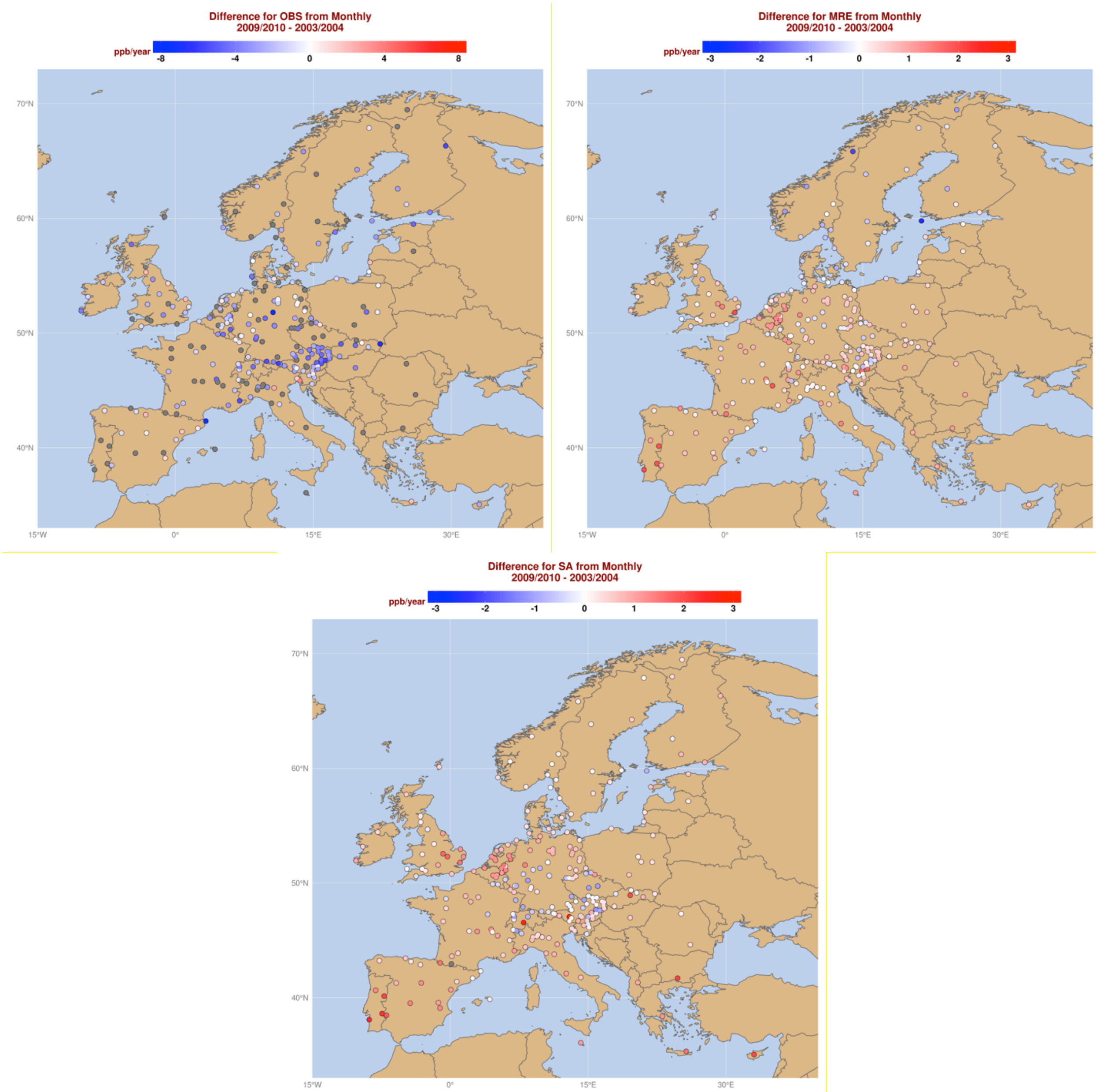


Figure 17. Difference of surface mean value between the periods 2009/2010 and 2003/2004 for all the stations. It is noted that the limits in the color scale is different between the observations and the two experiments.

Even if the degree of the tendency between observed data and model outcome is different, it is important to test positive and negative trends and which stations have the same slope in observed and model data. In the following maps (Figure 16) green color corresponds to stations where observed slope and model slope were both positive and negative, unlike red colors where the slope between the observations and the model differs. Assimilated data seems to have a great improvement in all regions, where more than the half of not corresponded trends between observed and not assimilated data, shifted after the correction process.

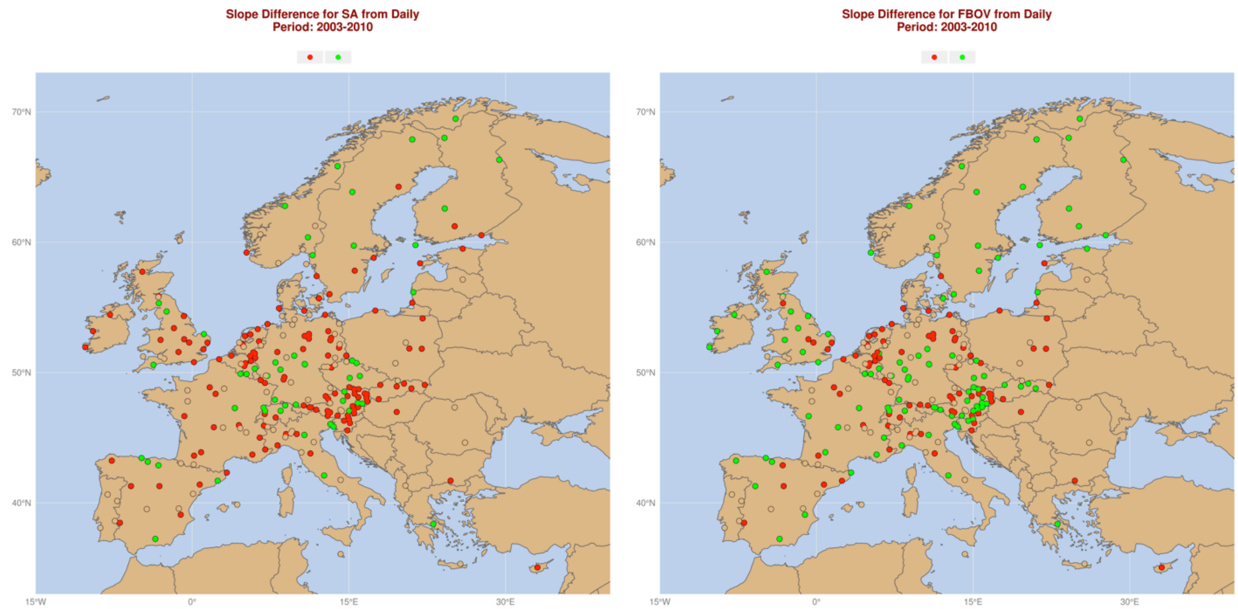


Figure 16. Slope agreement between observed data and the assimilated (MRE) and non assimilated experiments. Red and green color represent agreement and disagreement in slope.

2. Comparison of global and regional reanalysis over Europe for the year 2011

2.1. Introduction

In this section we evaluate the ensemble of the regional chemistry models using observations from the AIRBASE and EMEP database and compare it with the global reanalysis experiment MRE for the year 2011. In total 96 rural stations have been used. The analysis was mainly focused between the ensemble of regional models and the global reanalysis experiment MRE in an attempt to justify the benefits of higher resolution experiments. It should be noted that the ensemble of the regional models was assimilated also with ground-based observations, while MRE was only assimilated with satellite data that do not provide measurements close to the ground. Therefore when we compare those two experiments with the EMEP and AIRBASE ground base observational data, we have to keep in mind that the ENS experiment has a great advantage.

2.2. Basic Validation Metrics

Basic validation metrics were calculated for each region and in most cases the ensemble analysis scored better than the MRE experiment. In Figure 19 modified normalized mean bias and fractional gross error is presented for each region. The majority of the stations in both experiments underestimate surface ozone. In the regions Alps, France, Mediterranean and Scandinavia mean ozone is underestimated by 10% to 15% in the MRE experiment. Contrary in the regions British Isle and Iberian Peninsula surface the mean ozone is overestimated by 15% to 20%. Pearson correlation varies from 0.8 to 0.9 in most regions except the British Isles and Scandinavia where is lower than 0.55. Due to the different selection of the stations, MRE score may differ from the previous analysis, were 255 stations were selected.

The regional ensemble analysis underestimates ozone in Eastern Europe by 20% and in the remaining regions less than 10%. The large bias in Eastern Europe is not a product of just one station, but from a number of stations that are located mainly in Croatia as shown in Figure 21. The same negative bias is evident also in the MRE experiment but in the ENS model intensifies. Furthermore, if we examine the MNMB of the British Isles we can see that the positive bias of the MRE is converted to a close to zero bias in the high resolution ensemble model. Pearson correlation, which was derived from monthly values for each station, is close to and in most regions higher than 0.9 in the ensemble reanalysis (Figure 20).

Table 3 displays the MNMB, FGE and R for the ensemble of the regional models and the MRE experiment by region. Values marked with bold, indicate the model that had the best score in each region. The ensemble analysis exhibit very good scores in the three statistic parameters and in most cases outperforms the MRE experiment. The strong point of ENS against MRE is temporal correlation where it has very high values in all regions, even in British Isles and Scandinavia. As shown in previous studies, the use of all the available models or the most skillful models probably will not create the most skillful ensemble. A selection of models that minimize the ensemble error is the best choice (Solazzo et al. 2012). More information about the performance of each ensemble member can be found in the EVA report.

Table 2. Mean values of basic statistics parameter for the Ensemble of the regional models and the MACC reanalysis by region. Modified Normalized Mean Bias (MNMB), Fractional Gross Error (FGE), Pearson Correlation Coefficient (R) respectively. Statistics have been calculated from monthly values. Bold values indicate which model performed better in each region.

Region	**MNMB**		**FGE**		**R**	
	ENS	**MRE**	**ENS**	**MRE**	**ENS**	**MRE**
British Isles	**-0.06**	0.15	**0.14**	0.25	**0.87**	0.52
Iberian Peninsula	**0.01**	0.18	**0.15**	0.21	**0.89**	0.78
France	**-0.09**	-0.13	**0.13**	0.21	**0.97**	0.89
Mid-Europe	-0.07	**-0.01**	**0.14**	0.22	**0.97**	0.84
Scandinavia	**0.00**	-0.12	**0.08**	0.33	**0.90**	0.34
Alps	**-0.08**	-0.10	0.25	**0.23**	**0.96**	0.87
Mediterranean	**0.04**	-0.07	**0.08**	0.24	**0.97**	0.79
Eastern Europe	-0.19	**-0.07**	0.22	**0.21**	**0.96**	0.82

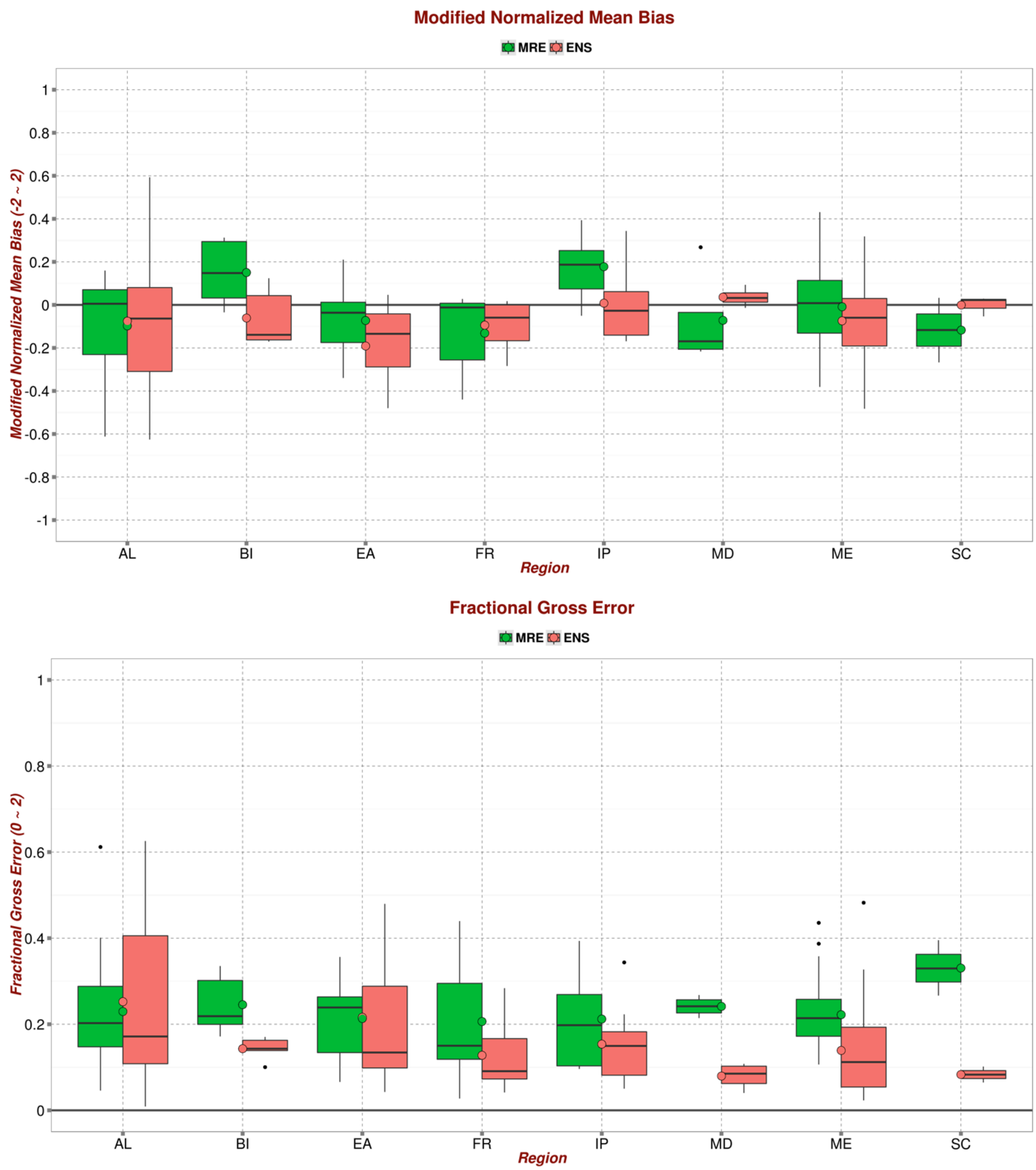


Figure 17. Modified Normalized Mean Bias (MNMB) and Fractional Gross Error in box plots for every European subregion. MNMB and FGE values were calculated for each station from monthly values. Green and red colors correspond to MRE experiment and ensemble analysis respectively. The point next to each box plot shows the mean value of MNMB and FGE. The grey line on zero value, distinguishes over and under estimation for each region.

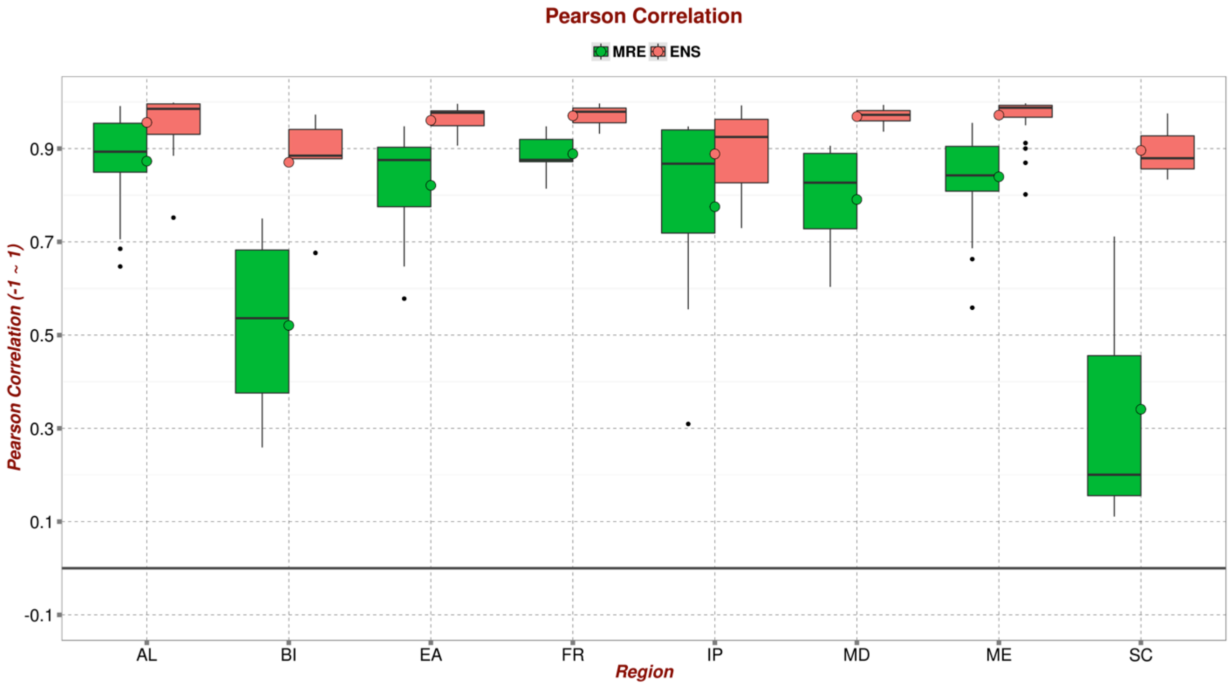
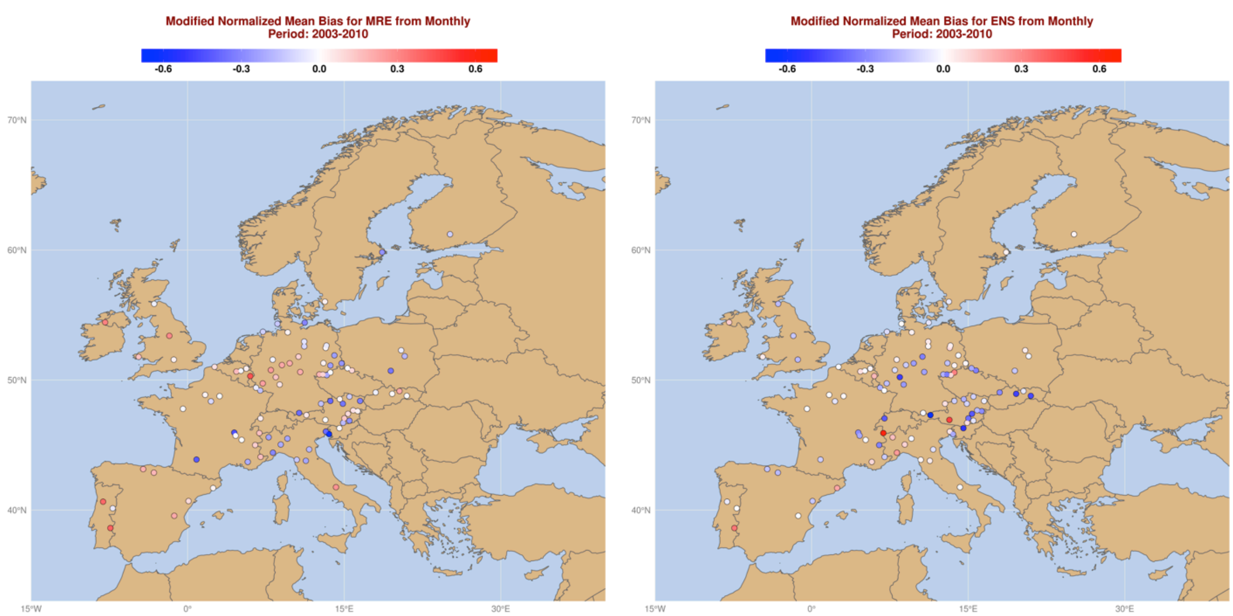


Figure 18. Pearson correlation coefficient (R) in box plots for every European subregion. R values were calculated for each station from monthly values. Green and red colors correspond to MRE experiment and ensemble analysis respectively. The point next to each box plot shows the mean value of MNMB and FGE.

Figure 21. Modified Normalized Mean Bias (MNMB), Pearson Correlation (R) and Fractional Gross Error (FGE) for each station for ENS and MRE experiments. All the statistics were calculated using monthly values for each station.



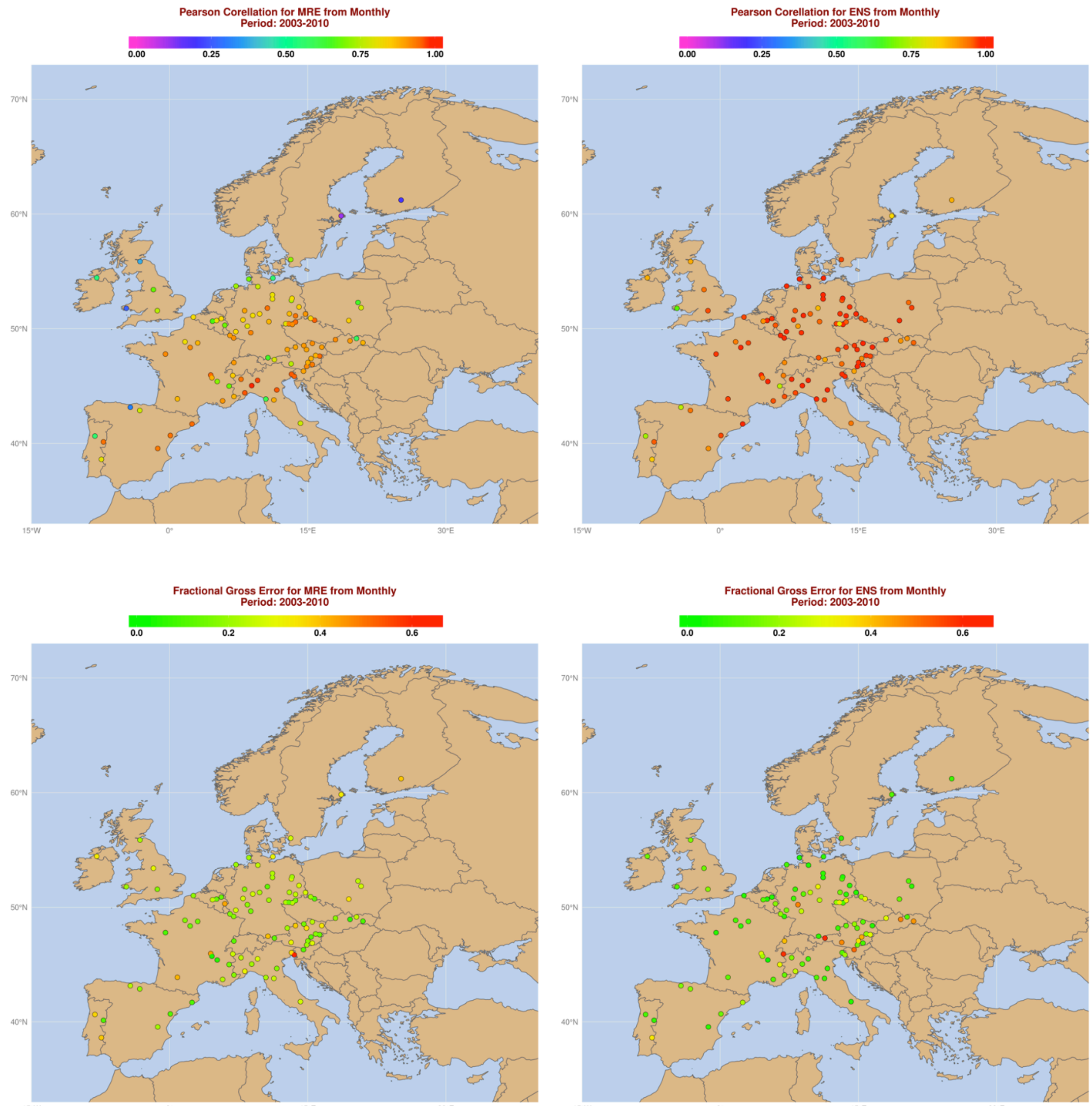


Figure 21. The Modified Normalized Mean Bias (MNMB), the Fractional Gross Error (FGE) and the Pearson Correlation (R) of each station for SA and MRE experiment. All statistics were calculated using monthly values for each station.

2.3. Annual Cycle

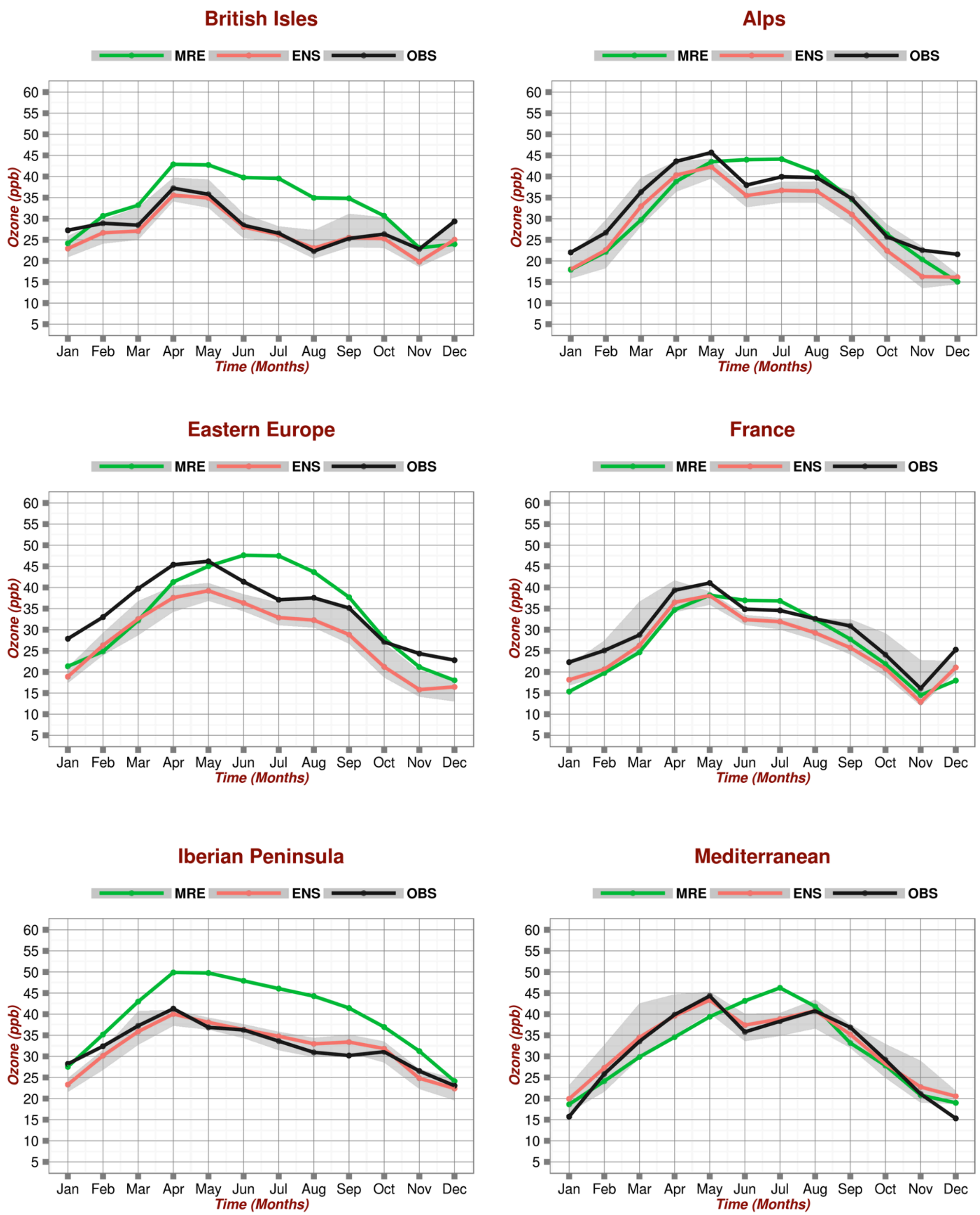
In Figure 22, the annual cycle for each region is presented. It is visible that the annual cycle of the ensemble experiment, which is derived from all the regional climate models, does in fact greatly improve the annual cycle in all regions and solves some limitation of the global-low resolution MRE experiment. The higher spatial resolution of the ensemble plays an important role in this outcome, but the assimilation with ground based observational data is most probably responsible for this improvement. In order to derive safe conclusion more research should be done in the future, comparing non-assimilated ENS and assimilated ENS experiments.

The overall behavior of MRE experiment in the previous section, where rural 255 stations have been available in the analysis, was a steady and almost smooth cycle in all regions, with a broad maximum in late spring and summer and a minimum in the winter and autumn. With this selection some new features are produced, for instance the annual cycle of MRE reproduces the spring maximum in the months April-May in the British Isles. Furthermore the observed annual cycle in Iberian Peninsula in this analysis displays a maximum in April, which is captured by the MRE model. The overall annual cycle in both cases is not captured very well, but at least an improvement in the temporal reproduction of the maximum values for MRE is detected. Nevertheless there are still some issues in Scandinavia, where the annual cycle cannot be reproduced.

On the contrary, the ensemble analysis captures the different patterns of annual cycle in all regions, such as the clear spring maximum, which is evident in some regions or the dual maximum during spring and summer over the Alps and Mediterranean. The benefit of the high-resolution regional model is most evident in Scandinavia where MRE experiment scored a very low correlation and could not reproduce the annual cycle correctly.

High ozone concentration levels are displayed over Mediterranean where ozone concentrations, especially in summer, are elevated due to the anticyclonic condition that lead to intense photochemical production and through horizontal transportation from continental Europe (Gerasopoulos et al. 2006; Kalabokas et al. 2008). Recently Zanis et al. (2014) indicated that the dominant mechanism causing a free tropospheric ozone pool in summer over Eastern Mediterranean is the downward transport from the upper troposphere and lower stratosphere, a process which may have important influence on near surface summer ozone levels.The Alps also exhibit high concentration levels due to the high elevation (Pay et al. 2010).

The spring maximum still remains an open issue for the global MACC reanalysis model. Although we observe some improvement in the British Isles, as far as the temporal detection of the maximum values, in the other regions the problem still persists. Since this spring maximum most probably is derived from larger scale processes, the MRE should reproduce it. In order to assess this issue various actions can be taken. It is important to check the NOx levels in order to evaluate the chemical conditions that control photochemical ozone production. Furthermore, the role of vertical transport should also further investigated by evaluating ozone profiles



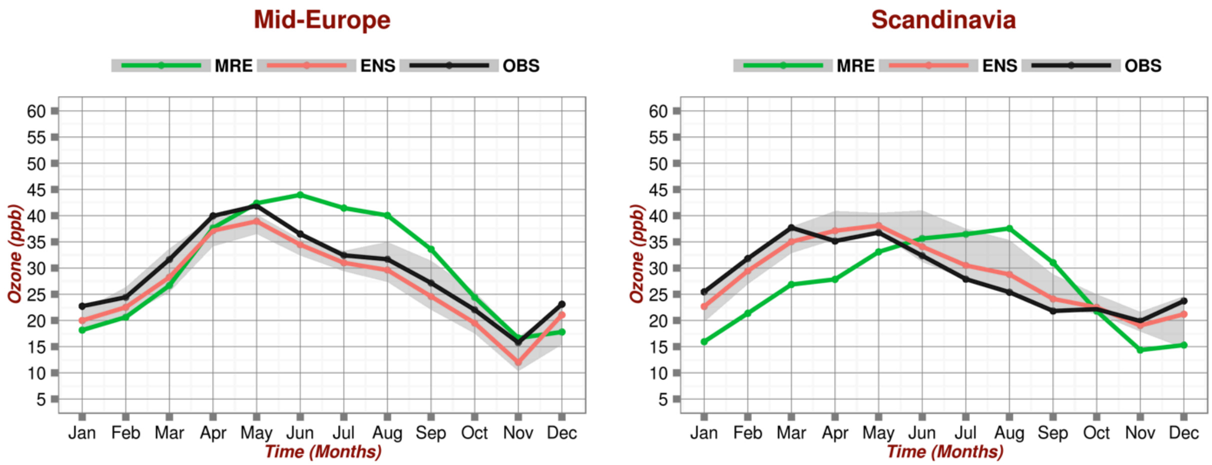
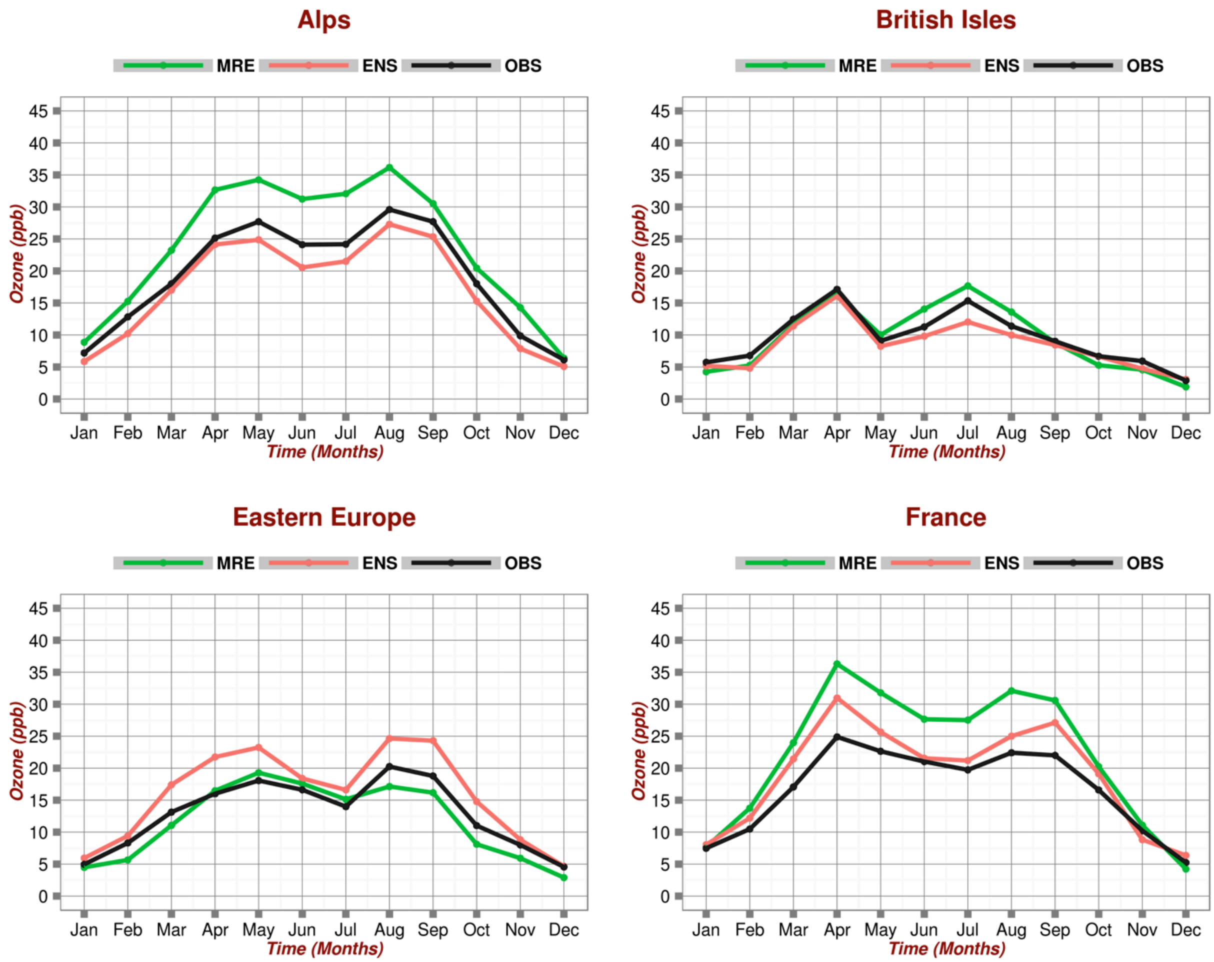


Figure 22. Annual cycles of O3 for each European region. Black, green and red colors represent the observed data, MRE experiment and the ensemble analysis of the regional climate models respectively. Grey area distinguishes the amplitude of the regional models. Annual cycles have been calculated using monthly data.

2.4. Diurnal Range

The importance of the daily range and its relation with ozone photochemical production has already been highlighted in the previous analysis. In Figure 23 the daily range of the mean diurnal cycle of each region is illustrated. High values indicate periods where the photochemical production of ozone and processes of dry deposition are increased. These processes are maximized in the warm period of the year as shown in the graphs. In the Alps, France and Mediterranean the MACC reanalysis is overestimating the diurnal range in all months, while in Scandinavia and Eastern Europe an underestimation is observed. In the annual cycle of the diurnal range there is a pattern that shows that ENS displays a better correlation compare to MRE. Even in cases like Mediterranean where ENS underestimates diurnal range a lot, it follows the variability of the observations.

The daily range can also help as assess the issues of the annual cycle in the northern regions. For example in Scandinavia the maximum of annual cycle in March-April-May is not characterized by high values of diurnal range, therefore we can conclude that photochemical production most probably does not play a major role in the spring maximum of this region. Maybe some other processes that increase surface ozone in spring are involved. While in British Isle the April-May maximum in the annual cycle agrees with the distinctive April maximum in the daily range annual cycle. These conclusions should be treated with caution, because the analysis includes only one year, 2011.


Alps
British Isles
Eastern Europe
France
MRE
ENS
OBS
Ozone (ppb)
Time (Months)
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec
0 5 10 15 20 25 30 35 40 45

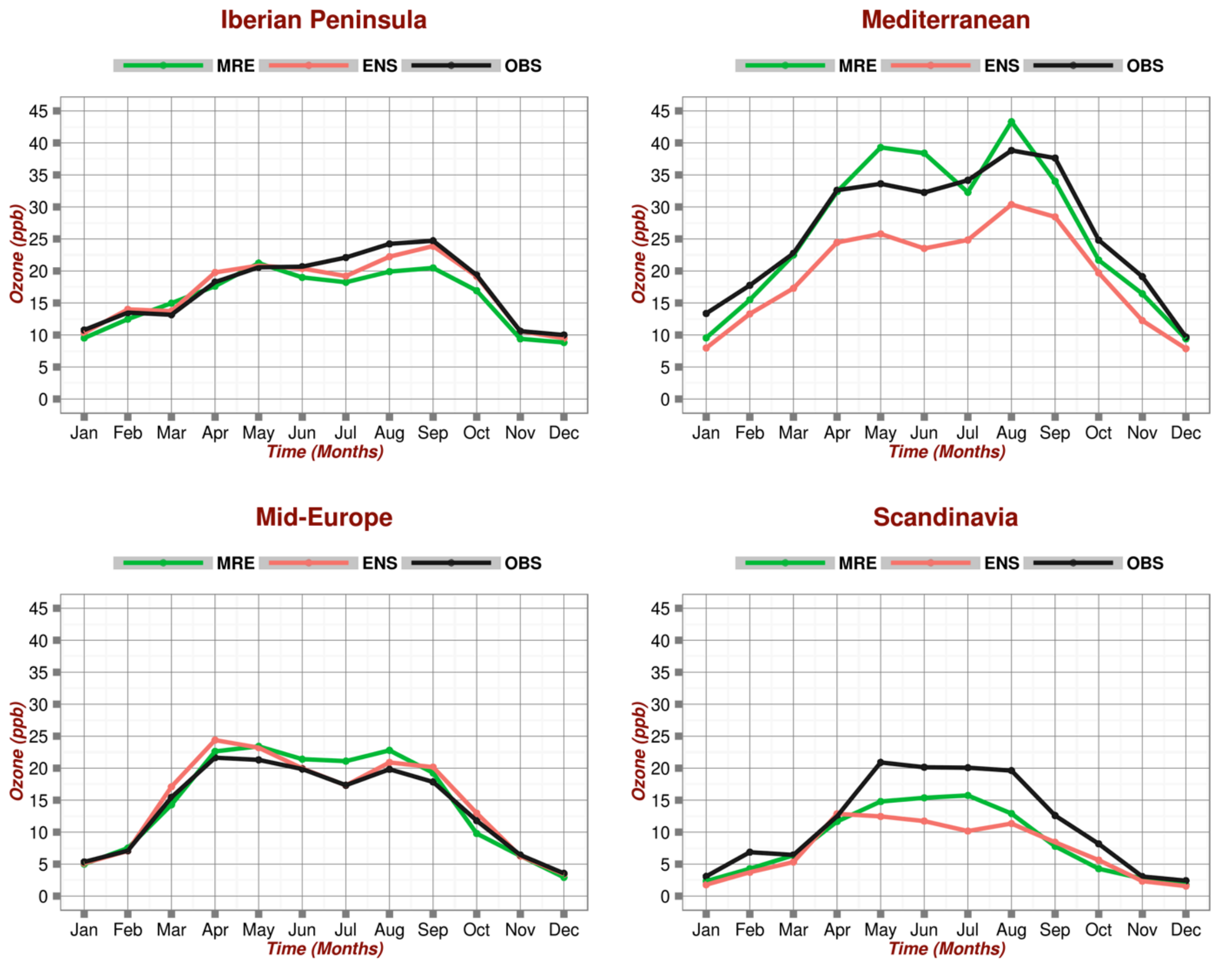


Figure 23. The Diurnal cycles of O3 for each European region by season. Black, green and red colors represent the diurnal cycles of observed data, MRE and ensemble analysis respectively. Grey area distinguishes the amplitude of the regional models. Diurnal cycles have been calculated using hourly data of each station.

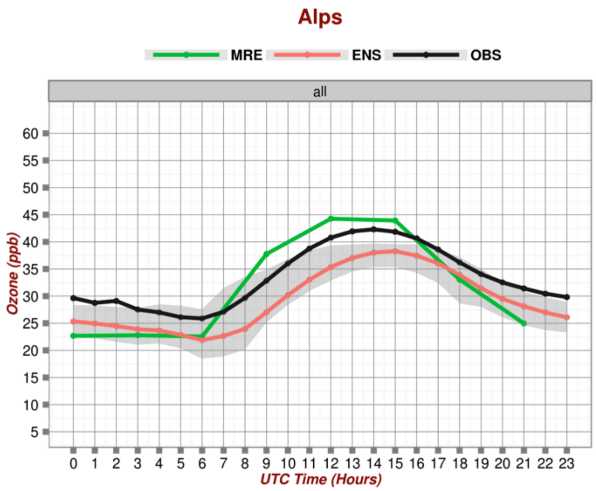
2.5. Diurnal Cycle

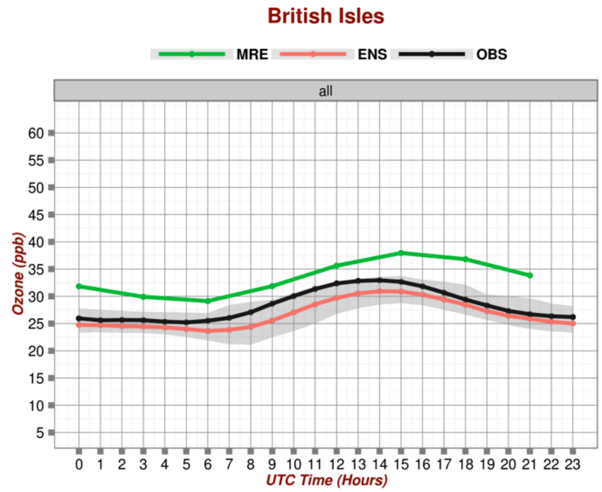
The daily cycle of the observed data, the MRE experiment and the ensemble of the regional models are illustrated in Figure 23. The models performance for surface ozone concentration varies seasonally, so the diurnal cycle was studied also by season in Figure 24. In winter and in regions with a fairly small diurnal range, like Scandinavia, the MRE experiment follows the diurnal cycle more accurately. Photochemical activity in those cases is quite low and the atmosphere more stable. These conditions enhance the ability of the models to capture the diurnal cycle.

The ensemble analysis reproduces the annual cycle of the observed data to a higher degree. A great challenge for the ensemble of the regional models lies in Mediterranean during spring and especially in summer, where they cannot capture the fast reduction of surface ozone concentration just before the dawn. Most probably this deepening owes its origin to a faster rhythm of dry deposition during summer in a global scale (Auvray and Bey, 2005) and especially over Eastern Mediterranean (Gerasopoulos et al. 2006).

The advantage of the regional ensemble is evident mostly in summer, where the ENS reproduces the correct daily cycle and amplitude. On the other the MRE has a very high bias during the day in all regions (same behavior as in the previous analysis Figure 12) and a very high overestimation over all hours in British Isles, Eastern Europe and Iberian Peninsula. In winter the differences between MRE and ENS is reduced but still we can conclude that the ensemble of the regional models outperforms the global MACC reanalysis in most regions.

As it was stated before, the assimilation of the ENS includes ground based observational data and MRE does not. So the differences in the two databases cannot be attributed only in the spatial resolution of the regional models, but mostly in the different methods and data used for the assimilation.


Alps
MRE
ENS
OBS
all
60
55
50
45
40
35
30
25
20
15
10
5
Ozone (ppb)
0 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 20 21 22 23
UTC Time (Hours)


British Isles
MRE
ENS
OBS
all
60
55
50
45
40
35
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15
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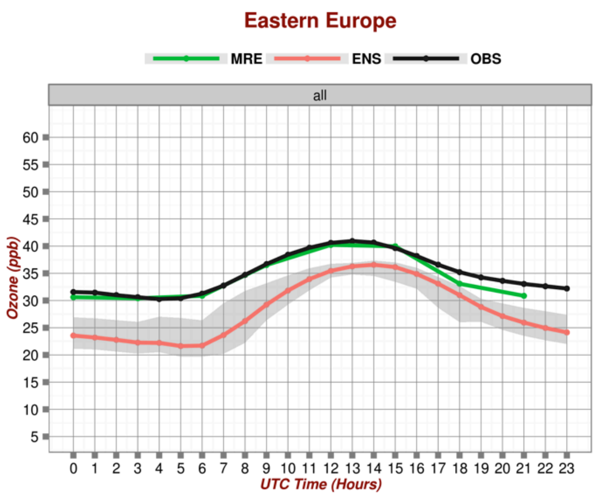
5
Ozone (ppb)
0 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 20 21 22 23
UTC Time (Hours)


Eastern Europe
MRE
ENS
OBS
all
60
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40
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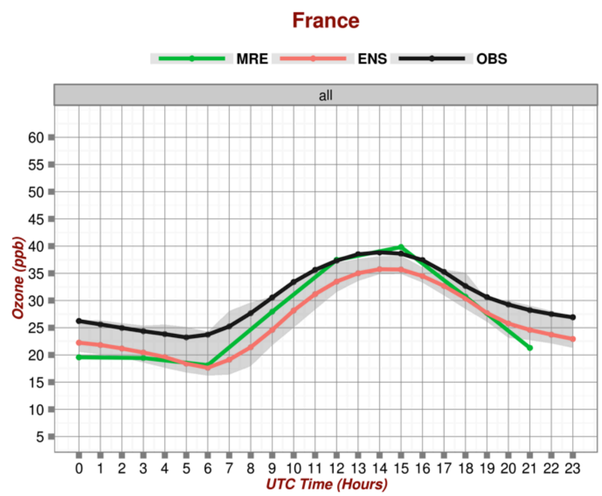
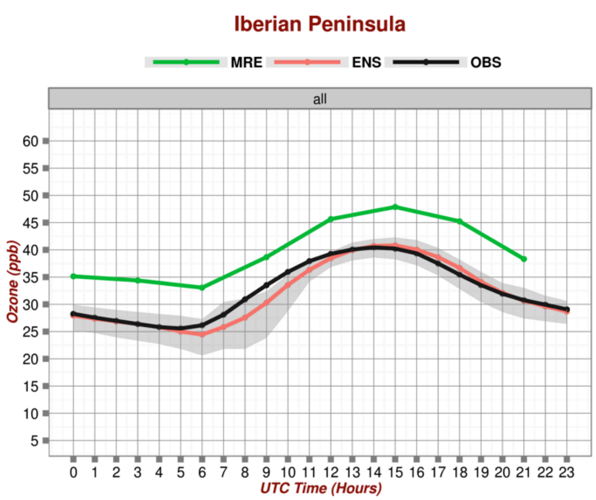
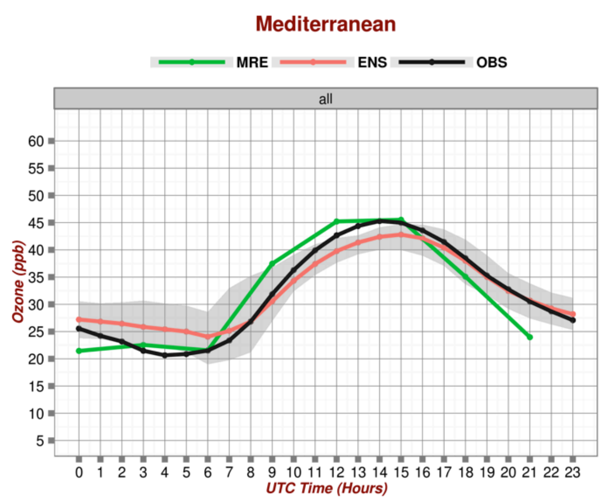
10
5
Ozone (ppb)
0 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 20 21 22 23
UTC Time (Hours)


France
MRE
ENS
OBS
all
60
55
50
45
40
35
30
25
20
15
10
5
Ozone (ppb)
0 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 20 21 22 23
UTC Time (Hours)


Iberian Peninsula
MRE
ENS
OBS
all
60
55
50
45
40
35
30
25
20
15
10
5
Ozone (ppb)
0 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 20 21 22 23
UTC Time (Hours)


Mediterranean
MRE
ENS
OBS
all
60
55
50
45
40
35
30
25
20
15
10
5
Ozone (ppb)
0 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 20 21 22 23
UTC Time (Hours)

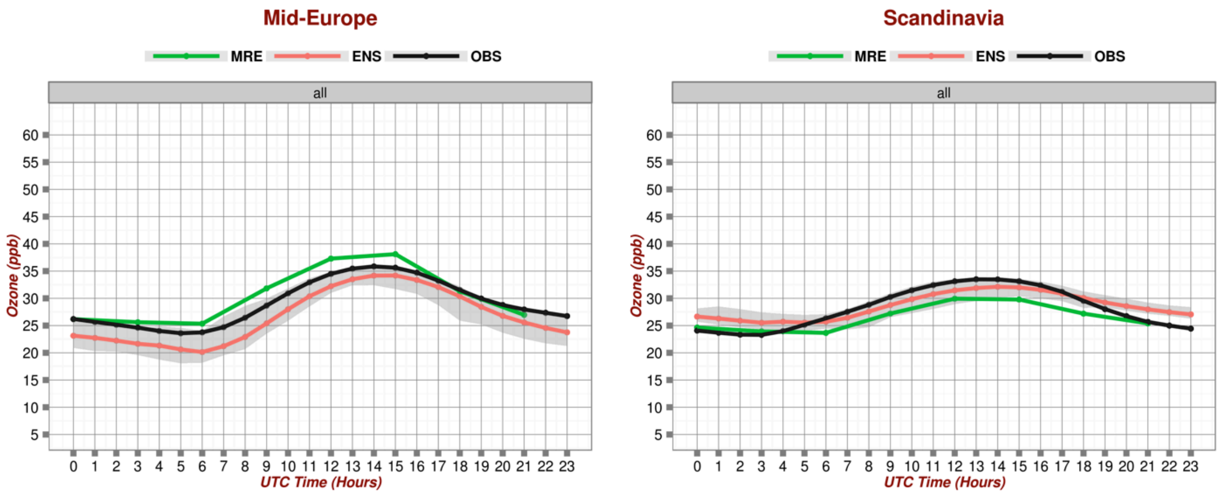
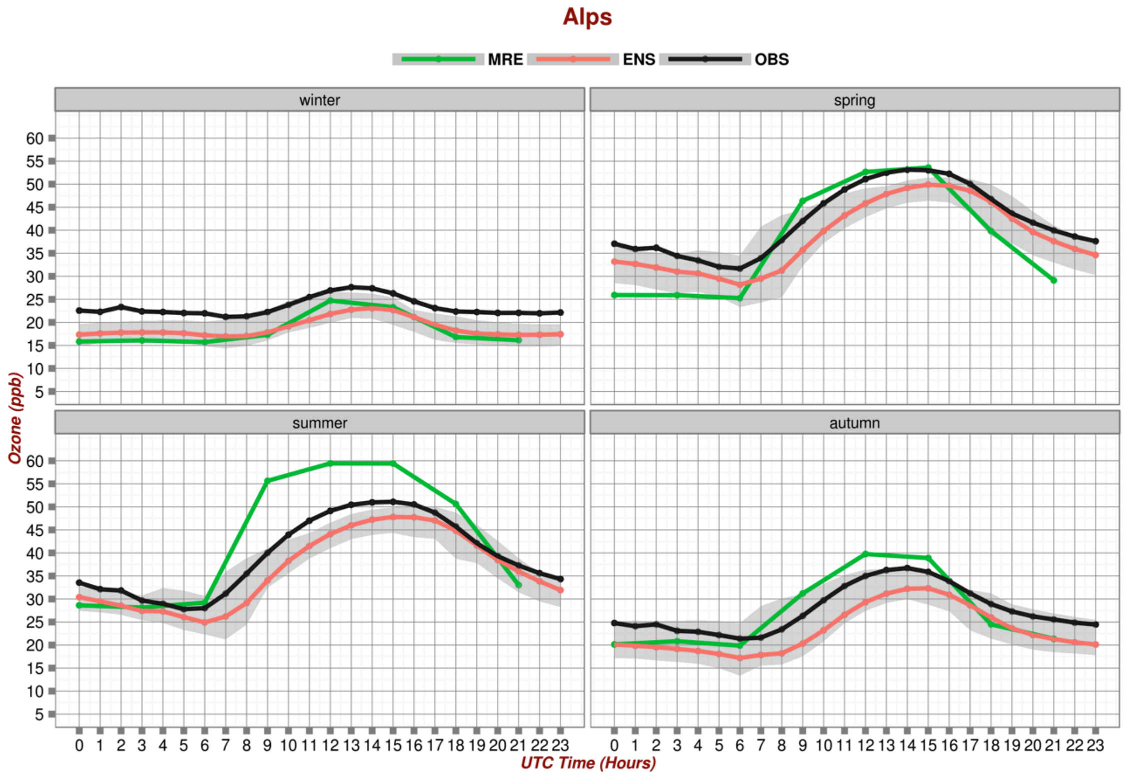
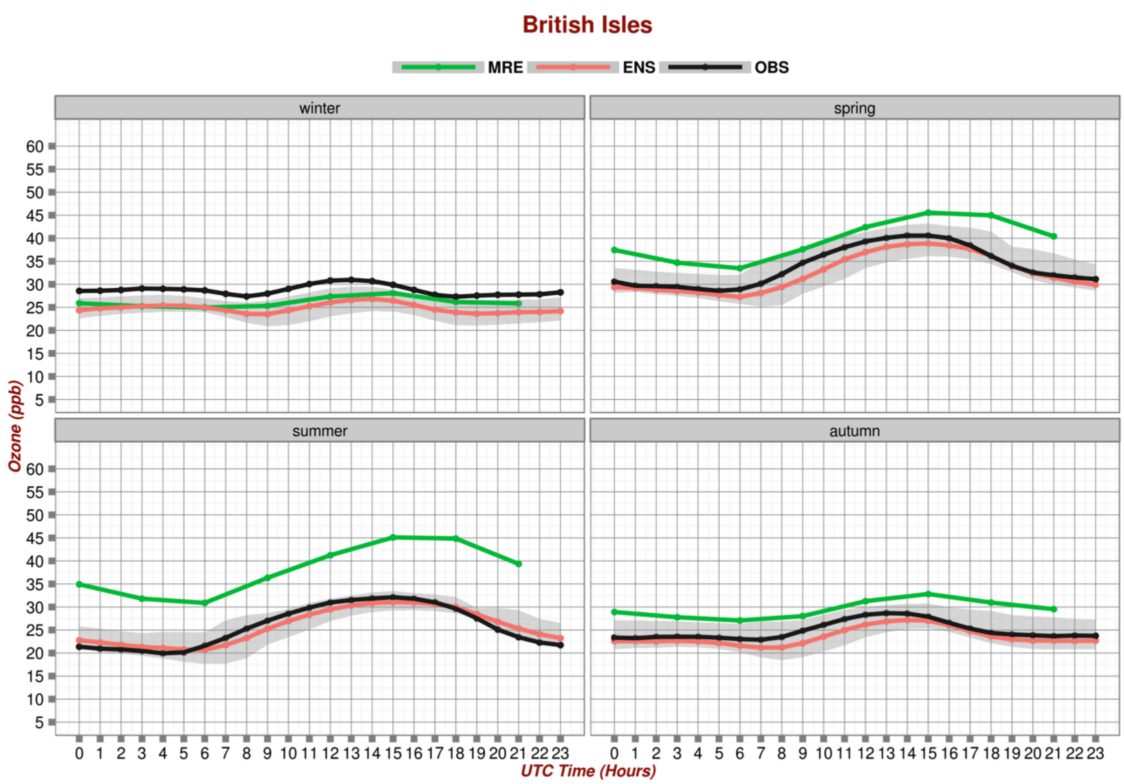
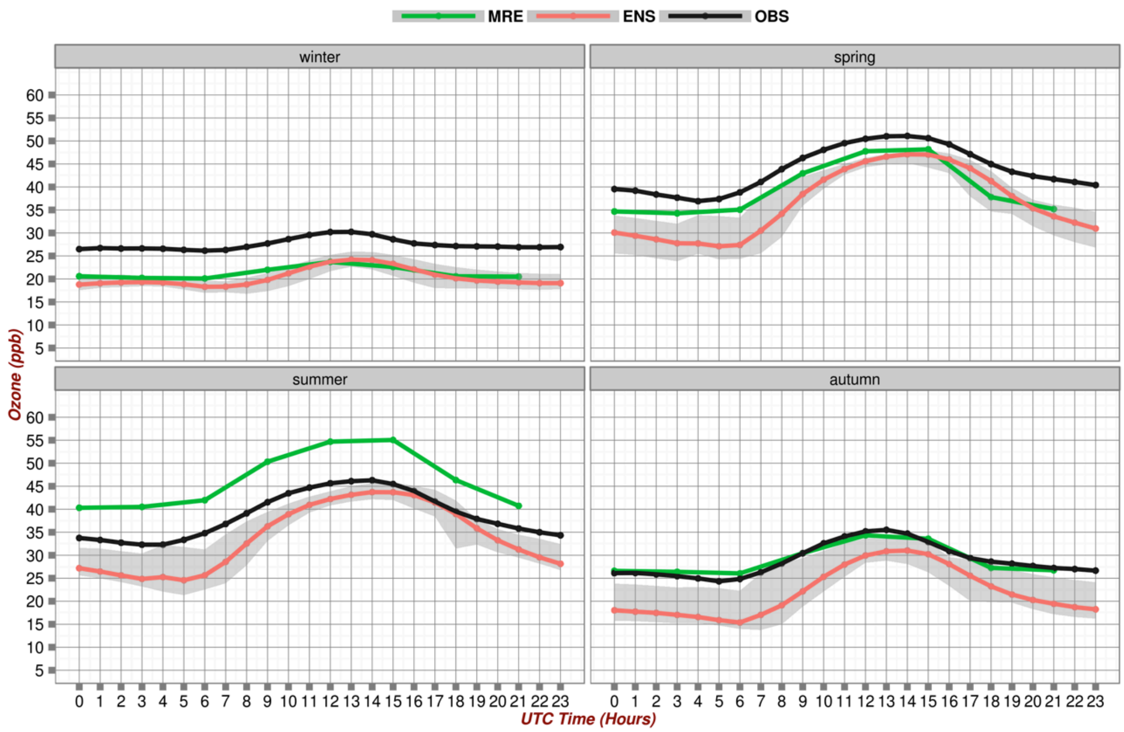


Figure 24. Diurnal cycles of O3 for each European region. Black, green and red colors represent the diurnal cycles of observed data, MRE and ensemble analysis respectively. Grey area distinguishes the amplitude of the regional models. Diurnal cycles have been calculated using hourly data of each station.

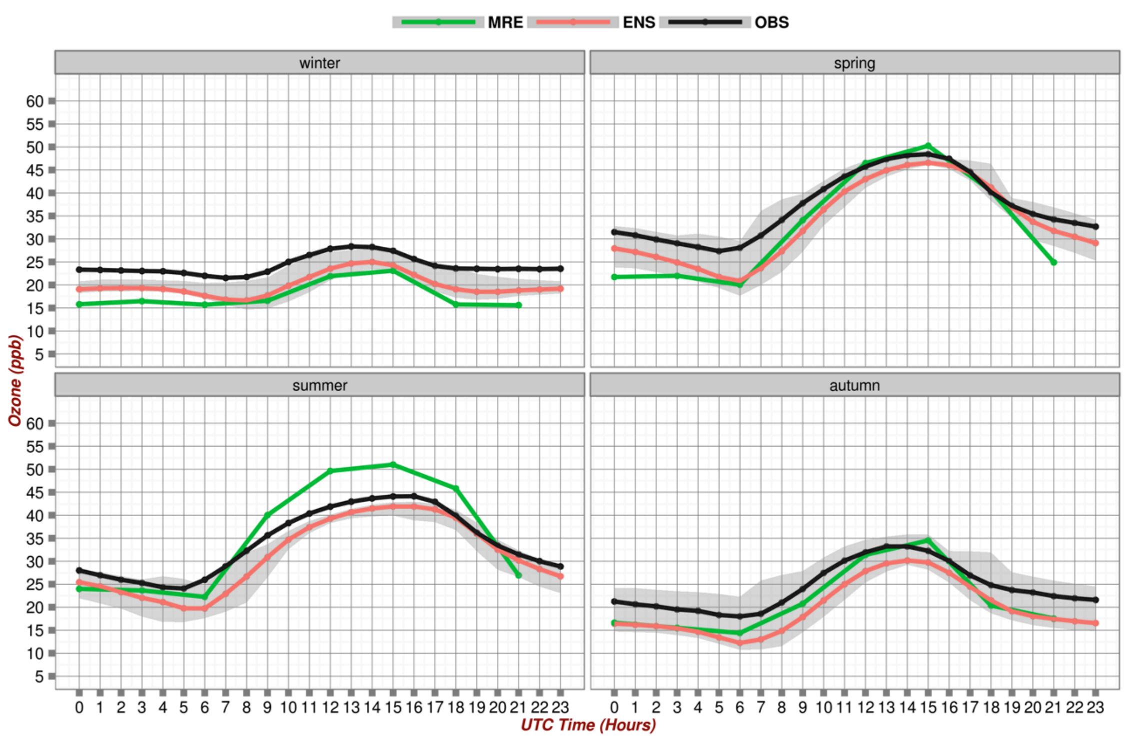

Alps
MRE
ENS
OBS
winter
spring
summer
autumn
Ozone (ppb)
UTC Time (Hours)


British Isles
MRE
ENS
OBS
winter
spring
summer
autumn
Ozone (ppb)
UTC Time (Hours)

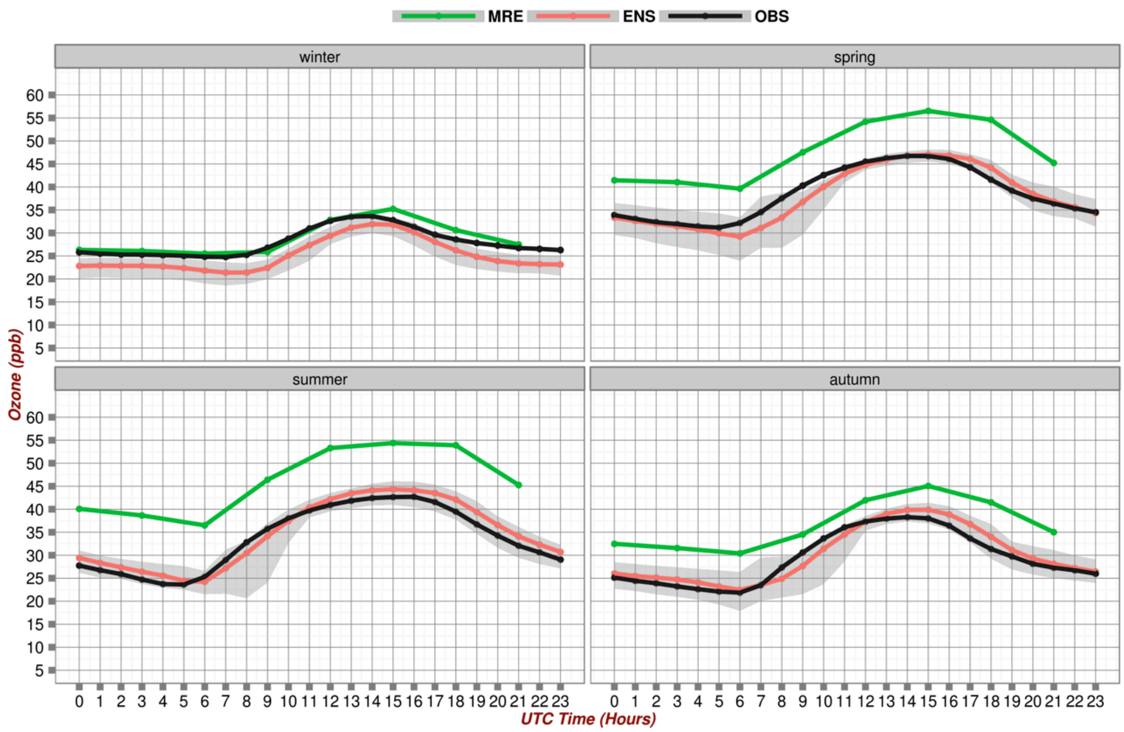
Eastern Europe



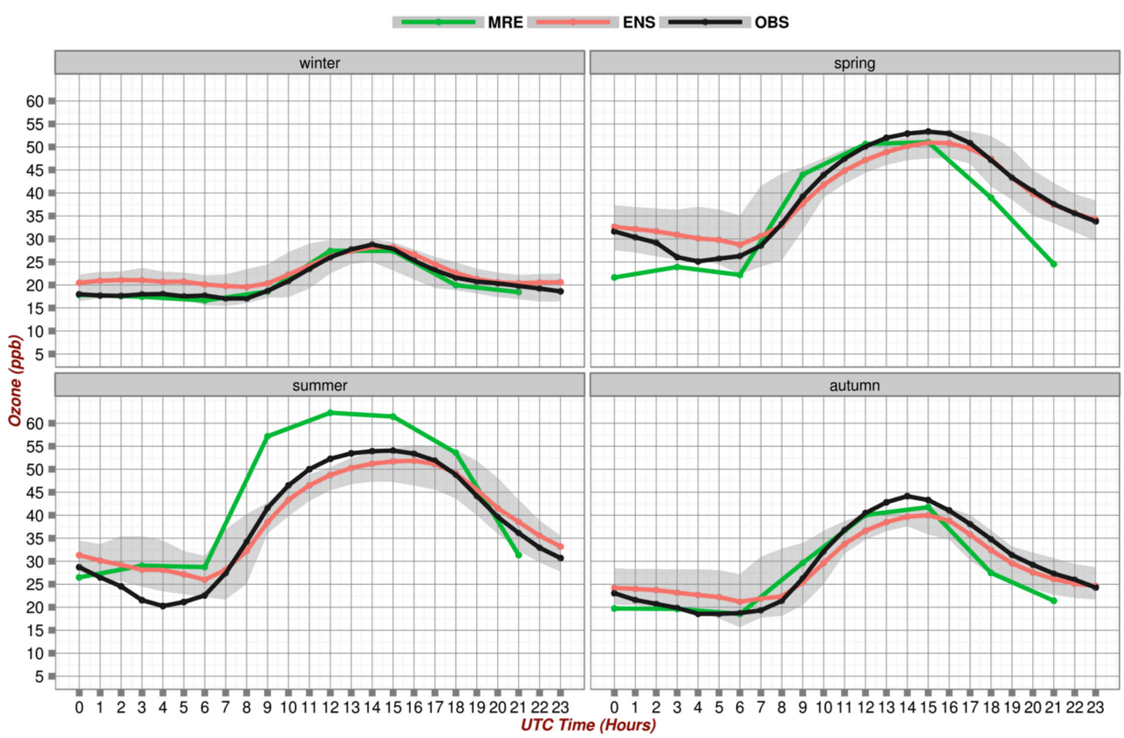
France



Iberian Peninsula



Mediterranean



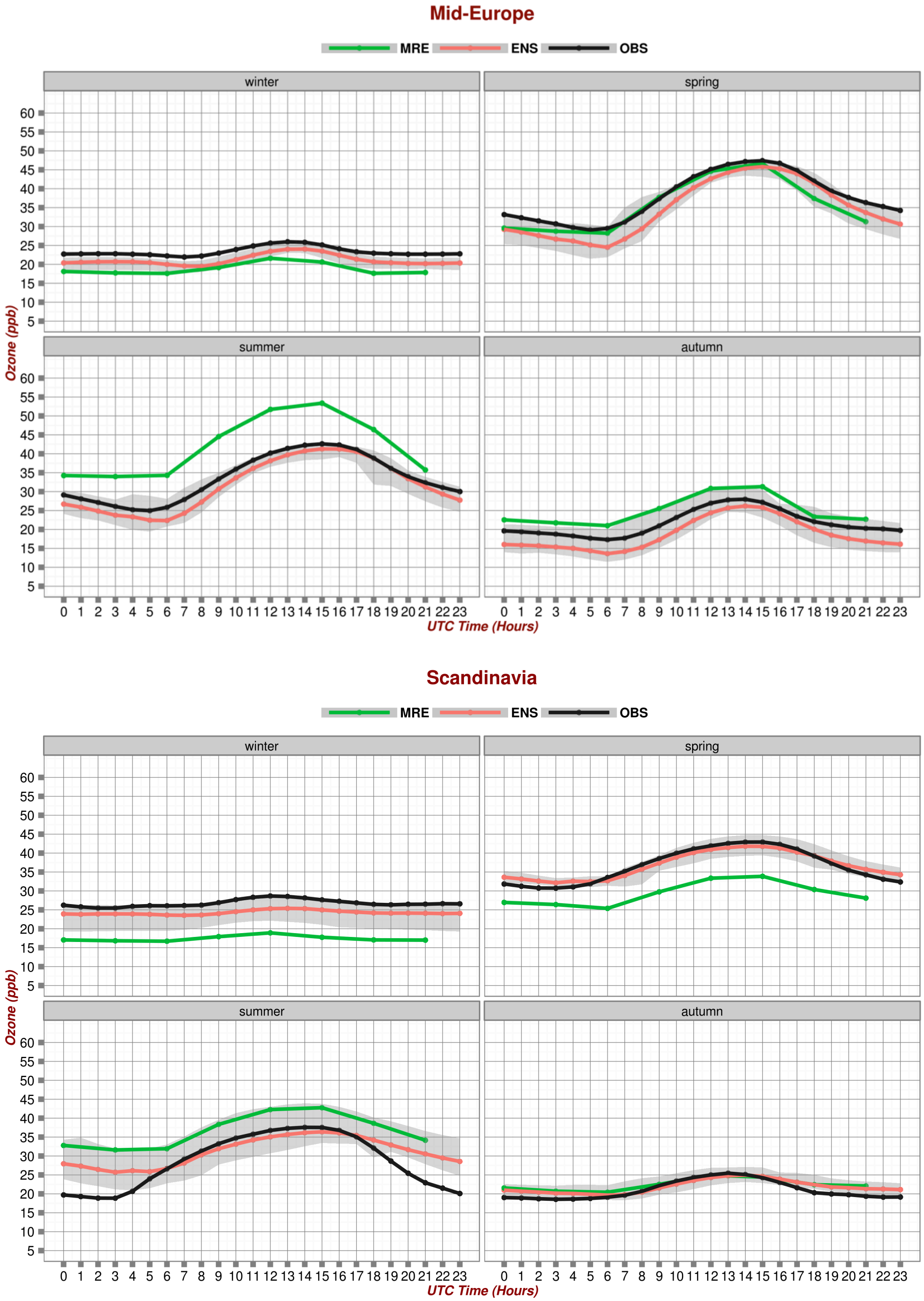


Figure 25. Diurnal cycles of O3 for each European region by season. Black, green and red colors represent the diurnal cycles of observed data, MRE and ensemble analysis respectively. Grey area distinguishes the amplitude of the regional models. Diurnal cycles have been calculated using hourly data of each station.

Conclusions

Evaluation of the global MACC-II reanalysis with European surface ozone observations, 2003-2010

- A general reduction of the negative bias in the assimilated experiment (MRE) over most regions while correlation is slightly deteriorating.
- The assimilation does not impact the annual cycle of surface ozone, exhibiting for both experiments over all European sub-regions a broad late spring-summer maximum.
- Observational data suggest that the annual cycle has different characteristics over each region, with early spring or spring-summer bimodal maxima, which are not reproduced by MACC-II reanalysis.
- The use of assimilation seems to reduce and improve the amplitude of the annual cycle with respect to the observations in most sub-regions.
- In all regions the MRE experiment underestimates the observed diurnal cycle and it can capture the daily cycle in winter more accurately than summer. Furthermore in summer both experiments overestimate ozone concentrations.
- In all regions the underestimation of diurnal range is evident for both experiments, while the variation and the mean value in the MRE experiment is increased.

Comparison of global and regional reanalysis over Europe for the year 2011

- The ensemble of the regional models (ENS), reduces the bias in most regions compare to the MACC reanalysis global model (MRE). Also the temporal correlation is very high in the ensemble experiment even in northern regions.
- Although the MACC global reanalysis could not reproduce the annual spring maximum in Eastern Europe and Scandinavia in the previous analysis (validate with 255 rural stations), the regional analysis reproduces the annual ozone maximum in April-May in the British Isles.
- The ensemble model can reproduce the annual cycle of surface ozone in all regions very accurately. It should be noted once again the regional models have been assimilated with surface observations while MRE was not.
- The daily cycle is reproduced more accurately in most regions-seasons by the ensemble of the regional models. The advantage of the regional ensemble is evident mostly in summer, where the ENS reproduces the correct daily cycle and amplitude, while in winter both experiments have very low biases.

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