Interpretation of long-term measurements of radiatively active trace gases and ozone depleting substances (Part 3 of 3)

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5 Nitro Europe Project: Final results and conclusions

5.1 Introduction

This chapter summarises the work done in the final year of the Nitro Europe project and in particular Work Package WP6.2 (*Independent inverse modelling of European N_2O and CH_4 emissions*) and discusses the main results and final conclusions obtained from the work undertaken.

The NitroEurope project was a five-year European project co-ordinated by the Centre for Ecology and Hydrology (CEH), Edinburgh, and considered all parts of the nitrogen cycle, it ended April 2011. The Met Office was involved in a work package to estimate European and national emissions of methane and nitrous oxide through inversion modelling using multi-site observations. Four other groups also participated in this exercise and the emission estimates from each group were inter-compared.

Atmospheric measurements combined with inverse atmospheric models can provide independent top-down estimates of greenhouse gas (GHG) emissions. This is important in particular for N_2O and CH_4 where considerable uncertainties exist in the bottom-up inventories. In the Nitro Europe project, European N_2O and CH_4 emissions have been estimated for the years 2006 and 2007 using 5 independent inverse modelling systems, based on different global and regional Eulerian and Lagrangian transport models. The major objective of this ensemble approach is to provide more realistic estimates of the overall uncertainties of the derived emissions.

To fulfil the requirements of the project, the Met Office NAME Inversion Method is applied using a selection of stations across Europe where measurements of CH_4 and N_2O are available. These include both high-frequency measuring stations as well as flask measurements, for the years 2006 and 2007.

5.2 Setup

The general inversion methodology, as discussed in chapters 3 and 4, is used in this work.

Using the Met Office Lagrangian atmospheric dispersion model NAME, a series of back-runs starting from each station included in the inversion are first performed. From these runs, history maps of integrated concentrations from 0-50m above ground are obtained for 2 hour intervals (the inversions in the previous chapters relate to 3-hour air history maps). These show the air path for the last 13 days prior to reaching the station.

The back runs thus provide the transport model, or dilution matrix D to the equation

$$D\,\bar{e} = \bar{o}' \qquad \ldots(1)$$

where $\bar{e}$ is the desired emission map solution and

$$\bar{o}' = \bar{o} - \bar{b} \qquad \ldots(2)$$

i.e., the deviation of observations $\bar{o}$ from a baseline $\bar{b}$.

A European domain of dimension [-14.63 to 39.13 longitude 33.8 to 72.69 latitude], with 128×144 grid points and a resolution of 0.42°×0.27° in the EW and NS directions respectively was used for the inversion in this work. Note this domain and resolution is different to that used in the inversions discussed in the previous chapters. This domain contains 11 continuous monitoring (CM) and 10 Flask measuring (FM) stations, listed in Table 1. The CM sites have hourly observations, where the FM ones vary but typical they are once a week.

A particular challenge in N_2O measurements is the low signal to noise ratio and significant calibration offsets, which are apparent for measurements from different laboratories. To correct for these calibration offsets, a novel bias correction scheme has been developed [Corazza et al., 2010] and the bias (provided by the NitroEurope project) was applied to the N_2O observational data in this work.

Fundamental to the solution is the choice of the baseline (defined as concentrations representative of the mid-latitude Northern Hemisphere (NH) background). Mace Head (MH) is the obvious choice of station to use for the calculation of baselines and the method for this is described in chapter 3. Using stations across Europe presents a new challenge to the basic methodology, since the MH baseline might not be appropriate or truly representative in all cases. Obvious examples are high altitude mountain stations like Jungfraujoch. Other not so obvious cases include stations in the middle of Europe in relatively polluted areas.

To elucidate baseline concepts as well as other inversion set-up issues, a set of experiments were designed. These are summarised in Table 2. For each experiment, 52 realisations were performed, starting either from random perturbations or a-priori emission maps. For those starting from an a priori, the solutions are also guided by the a priori, i.e. the best fit solution is discouraged (via an extra term in the cost function) from moving too far from the a priori. Statistics (means, uncertainty) were then obtained from these realisations.

ExpY1 uses the MH baseline and a sub-selection of the observation stations, according to whether the MH baseline seems appropriate. To decide upon this, the observations at each station are plotted against the MH baseline. Based on this analysis Jungfraujoch (JJ) and Pic du Midi (PM) were excluded. ExpY1b is identical to ExpY1 apart from the starting point of the inversion and the use of the a priori to guide the solution. Comparison of Y1 and Y1b experiments gives us information about the significance of starting from and being guided by an a-priori. In the experiments that use MH baseline, observations when local contributions are significant have been excluded from the inversion (see chapter 4).

To examine the effect of a 'station fit' baseline on the inversion, baselines were obtained from the TM5 model (personal communication Peter Bergamaschi), for all stations. These were calculated following Rödenbeck et al. 2009. The baselines from TM5 were used in the inversion in ExpY2 which is otherwise identical to ExpY1. Comparison of results between ExpY1 & ExpY2 therefore, gives an insight into the importance and effect on the inversion of the baseline choice.

For methane only, an additional inversion experiment, ExpY3a, was performed, in order to assess the importance of data quantity to the inversion. ExpY3a, uses all stations, except JJ and PM, but only observations in the time windows [12:00-15:00]LT for stations in the boundary layer and [0:00-3:00]LT for mountain sites. These time windows only apply to the high frequency monitoring stations, whereas all flask measurements are used. The baselines are from TM5 and observations when local contributions were significant were included in this case. This type of inversion tries to be as close to as possible to the TM5 model inversion.

Further inversion experiments were performed using all observing stations. All of these use the baselines from TM5 and explore various permutations in the input parameters and the effect these choices have on the inversion.

ExpY2a and ExpY2b use all stations, TM5 baselines and no time window. Their difference is in the suse of an a priori in the inversion process, random vs. a-priori respectively. Comparison between the two provides insight into the effects of the a priori in the inversion. Moreover, comparison of ExpY2a with ExpY2, gives insight into the effect of number of stations used in the inversion.

Finally, ExpY3 and ExpY4 mimic ExpY2a and ExpY2b respectively, apart from using a time window (same as for Y3a) in the selection of observations used in the inversion. A different cost function is used in experiments that use a priori information. Comparison between ExpY3 and ExpY4 shows the effect of using a priori information in the inversion (as was also the case with ExpY2a and ExpY2b). Also, comparison between Y2a and Y3 and between Y2b and Y4, gives insight into the effect of data quantity.

5.3 Results

Analysis of various experiments, during the Nitro Europe project, has shown that:

- ExpY1 and ExpY2 (choice of baseline to otherwise identical simulations) have shown differences in the obtained emissions.
- No real difference between ExpY2 and ExpY2a i.e., exclusion of JJ and PM does not make any significant difference to the results.
- Using time windows (ExpY3, ExpY3a and ExpY4) and thereby limiting the quantity of observational data was shown to have a somewhat detrimental effect to the inversion. The number of observational points used in the inversion influences the structure of the inversion grid (spatial distribution and number of grid points).
- The NAME-Inversion methodology was shown to give similar results from a random start and without any a priori guidance, when sufficient and good quality observational data are used.

Note: Using a-priori to start and also guide the inversion attempts to limit how far the best fit solution diverges from the a priori emissions. Therefore significant errors or biases in the a priori will be detrimental to the solution.

Therefore, in the discussion of results, we concentrate on two main experiments that encompass the fundamentals of input parameters and their effect on the inversion, namely ExpY1 and ExpY2a.

5.3.1 CH_4 emissions

CH_4 emission maps from ExpY1 and ExpY2a are shown in Figure 1, for years 2006 (on the left) and 2007 (on the right). Both whole Europe scale (bottom) and zoom in to Northern Europe (top) views are shown in each case. The overall picture is that both set-ups give rather similar emission maps overall. There is marginally more spread of emissions, especially over the sea, in the Y2a solution. Other differences are mainly near the edges of the domain, where there is not sufficient information and therefore the uncertainty is greater i.e., to the south east of the domain over Turkey. Other areas of difference include the Iberian Peninsula and Mediterranean region.

These differences observed on the emission maps are more clearly observed in the individual country totals shown in Figure 2. ExpY1 and ExpY2a generally give similar country emission values. Exception to this, are Ireland, France, Spain, Portugal, Finland and Greece, with differences between the two solutions in the range of 30% - 50%. Even then though, the differences are within the uncertainty interval of the inversion solutions, defined by the 5^{th} and 95^{th} percentiles. Moreover, these are places with either very low emissions or far away from any observing station and therefore with little influence on the inversion. This reflects on the structure of the resulting grids, shown in Figure 6.

Figure 3 shows the total emissions and uncertainty, concentrating on UK, Ireland, Benelux, NWEU and EU27. There is very little difference between the ExpY2a and ExpY1 solutions, both lie within the uncertainty of the bottom-up UNFCCC inventory. Through these results, it is shown that the MH baseline is applicable for inversions on a European scale when many well inter-calibrated observations are used.

The overall tendency of the ExpY2a to give slightly higher emission estimates than ExpY1 is consistent with the small bias of the TM5 baselines relative to the Met Office baseline. In Figure 4 the MH baseline from the NAME-Inversion Method (MOB) is shown in solid dark blue for 2006 (top) and 2007 (bottom). In the same figure, as a solid pink line, the TM5 baseline for Mace Head, after having been smoothed using the same criteria as in the construction of the MOB, is shown. The MH baseline from TM5 is consistently lower than the MOB (on average by ~ 2ppb). A TM5 baseline is estimated at each station individually (ExpY2a). Figure 44 also shows a sample of the TM5 baselines from several of the other stations, they all show the same tendency i.e., lower in general than the MOB. A lower baseline estimate at each station would result in higher emission estimations from the inversion. Table 4 and Figure 5 show the relative difference between the baselines and the concentrations during pollution episodes. For methane the difference in the baselines is small compared to the difference between the baselines and the concentrations during pollution episodes (less than 8%), therefore the difference in the emission estimates when using the different baselines will be small.

5.3.2 N_2O results

N_2O emission maps from ExpY1 and ExpY2a are shown in Figure 7, for years 2006 (on the left) and 2007 (on the right). Both whole Europe scale (bottom) and zoom in to Northern Europe (top) views are shown in each case. Overall, the two solutions have many similarities. ExpY2a (TM5 baselines) puts significantly more emissions over the Atlantic Ocean on the western side of the domain. Emissions from ExpY2a are generally higher everywhere (total map emissions 20% larger than from ExpY1). The picture is consistent for both years 2006 and 2007.

Looking at the emission estimates for individual countries, Figure 9, the total emissions from ExpY2a are consistently higher than those from ExpY1. This is consistent with the difference between the MOB compared with the baselines from TM5, Figure 11. As for CH_4, the MOB is shown as a solid blue line and the MH baseline from TM5, smoothed in the same way as in the construction of the MOB in solid pink. Unsmoothed TM5 baselines for several of the other European stations are shown as thin lines. There is about 0.5 ppb difference on average between the MOB and the TM5 baselines. As discussed above, this would enhance the emission estimates when using the TM5 baselines compared to those using MOB.

The overall higher emissions obtained from ExpY2a compared with ExpY1 are also shown on the aggregate emissions on a European level, Figure 1010. In this case, the difference between the emissions from ExpY2a and ExpY1 are larger than the uncertainty (5^{th} to 95^{th} percentiles) of the solutions. Given that the uncertainty in the UNFCCC inventory is so large (greater than 100%), both solutions are within the uncertainty range of the bottom-up estimates. Table 4 and Figure 12 show the relative difference between the baselines and the concentrations during pollution episodes. For nitrous oxide the difference in the baselines is significant compared to the difference between the baselines and the concentrations during pollution episodes (more than 33%), therefore the difference in the emission estimates when using the different baselines will be significant.

5.4 Summary discussion

A set of experiments designed to fulfil the requirements of Nitro Europe project and to investigate the sensitivity of the inversion solution to various input parameters, in particular, the choice of baseline, data selection and use of a-priori information to initialise and guide the inversion, are presented.

Quantity of data was shown to be an important factor to the inversion solution, Manning et al., 2010. When only observations were used in specified, strongly restricted, time windows, the inversion grid was coarse and the inversion solution impacted.

Use of a-priori to initialise and guide the inversion, Manning et al., 2010, did not show significant change to the emission solution. Moreover, such an approach discourages divergence from the a-priori information and therefore can be detrimental if the a priori is in error. Using a-priori information was shown to benefit the case when limited data were available. Therefore use of an a priori can potentially be useful when insufficient data are available, however the independency of the result is compromised and in such cases the inversion is adding little to the estimates.

A-priori information is now used at the final stage of post-processing, after the inversion process, to produce more realistically looking emission maps. It does not affect the country total concentrations except in areas like the Iberian Peninsula or around the Mediterranean where the land-sea border is poorly resolved by the inversion grid.

Finally, one of the most interesting results from the Nitro Europe experience relates to the choice of baseline used in the inversion. It has been demonstrated that the MH baseline (from the NAME-Inversion Method, Manning et al., 2011, referred to as MOB) used with a selection of stations scattered across Europe gives realistic emissions on a European level. Site specific baselines benefit areas where the MH baseline is inappropriate e.g. at high altitude stations or at stations on the southern border (if a strong inter-hemispheric gradient is present).

5.5 References

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Table 1: List of high frequency monitoring stations CM, shown in green, & flask type monitoring stations FM, shown in indigo, that are used in the inversion.

ID	Station	lon	lat	alt	CH_4	N_2O
AN	Angus Tower, UK	-3.0	56.6	313+222	✓	✓
BK	Bialystok, Poland	22.8	52.3	160+300	✓	✓
C3	Cabauw, NL	4.93	52.0	-2+120	✓	✓
EG	Royal Holloway, UK	-0.6	51.4	45	✓	×
HY	Hegyhatsal, HU	16.7	47.0	248+96	✓	✓
MH	Mace Head, UK	-9.9	53.3	25	✓	✓
OK	Ochsenkopf, D	11.8	50.1	1185	✓	✓
JJ	Jungfraujoch, Sw	7.98	46.6	3580	✓	✓
PA	Pallas, Finland	24.1	68.0	560	✓	✓
SL	Schauinsland	7.91	47.9	1205	✓	✓
SY	Saclay, France	2.15	48.71	160+7	✓	×
BS	Baltic Sea, Poland	17.2	55.4	28	✓	✓
BR	Begur, Spain	3.23	41.97	13+2	✓	×
CO	Black Sea, Romania	28.7	44.2	3	✓	✓
HB	Hohenpeissenberg, D	11.0	47.8	985	✓	✓
LM	Lampedusa, IT	12.6	35.5	45	✓	✓
IG	Ile Grande, France	-3.58	48.80	20+10	✓	×
PM	Pic du Midi, France	0.14	42.94	2877+10	✓	×
PU	Puy de Dome, France	2.97	45.77	1465+10	✓	×
SI	Shetland, UK	-1.27	59.85	46	✓	✓
OS	Ocean station, Norway	2.0	66.0	5	✓	✓

Table 2: List of experiments performed. Time window: For high frequency stations, use all observations in time windows 12:00-15:00 LT for surface stations & 00:00-03:00LT for mountain stations. For Flask type observing stations, use observations at all times.

CH4 Inversion	
MH baseline (2 experiments)	**TM5 baseline** (6 experiments)
Experiments using all stations except JJ & PM	
Y1: random start, no time window	**Y2**: Like Y1
Y1b: start from a-priori, no time window	**Y3a**: Like Y2, time window
Experiments using all stations	
	Y2a: random start, no time window
	Y2b: start from a-priori, no time window
	Y3: random start, time window
	Y4: start from a-priori, time window

N2O Inversion	**Bias correction from TM5 was applied to all experiments**
MH baseline (2 experiments)	**TM5 baseline** (5 experiments)
Experiments using all stations except AN & JJ	
Y1: random start, no time window	**Y2**: Like Y1
Y1b: start from a-priori, no time window	
Experiments using all stations	
	Y2a: random start, no time window
	Y2b: start from a-priori, no time window
	Y3: random start, time window
	Y4: start from a-priori, time window

Table 3: UK, Ireland and North Europe (NWEU) emissions inventory for CH_4 and N_2O for 2006 and 2007 from various sources. NWEU consists of Ireland, UK, France, Benelux, Germany and Denmark. GHGI stands for Green House Gas Inventory and NAME represents the results obtained from a 3yr inversion using MH as the only observing station.

Gg/yr		**ExpY1**	**ExpY2a**	**EDGAR**	**GHGI**	**NAME**
CH_4	UK 06	2123.8	2305.5	3485.5	2401.9	
	UK 07	2016.8	2187.9	3485.5	2330.4	
	Ire 06	269.6	377	769.2	632.3	
	Ire 07	233.4	331.6	769.2	617.2	
	NWEU 06	8353.6	9513.9	13362.7	9127.8	
	NWEU 07	7913.3	9168.5	13363.4	8961.5	
N_2O	UK 06	82.3	121.5	61.2	112.8	
	UK 07	69.6	124.8	61.2	110.6	
	Ire 06	22.9	36.7	15.1	27.2	
	Ire 07	23.6	33.5	15.1	25.9	
	NWEU 06	497.3	665	348.3	625.1	
	NWEU07	441.3	623.9	348.3	622.7	

Table 4: Difference between the MOB and average pollution episodes (Mace Head only) and the TM5 baselines at different stations for CH_4 (top) and N_2O (bottom). Values are in ppb.

CH_4					
Pollution - MOB	MH	AN	BK	C3	OK
2006	50.7				
2007	45.1				
MOB – TM5					
2006	3.9	3.4	6.1	6.5	10.8
2007	2.7	1.4	3.7	4.0	8.1

N_2O					
Pollution -MOB	MH	AN	BK	C3	OK
2006	0.75				
2007	0.76				
MOB – TM5					
2006	0.24	0.26	0.32	0.29	0.32
2007	0.30	0.30	0.34	0.32	0.36

CH_4

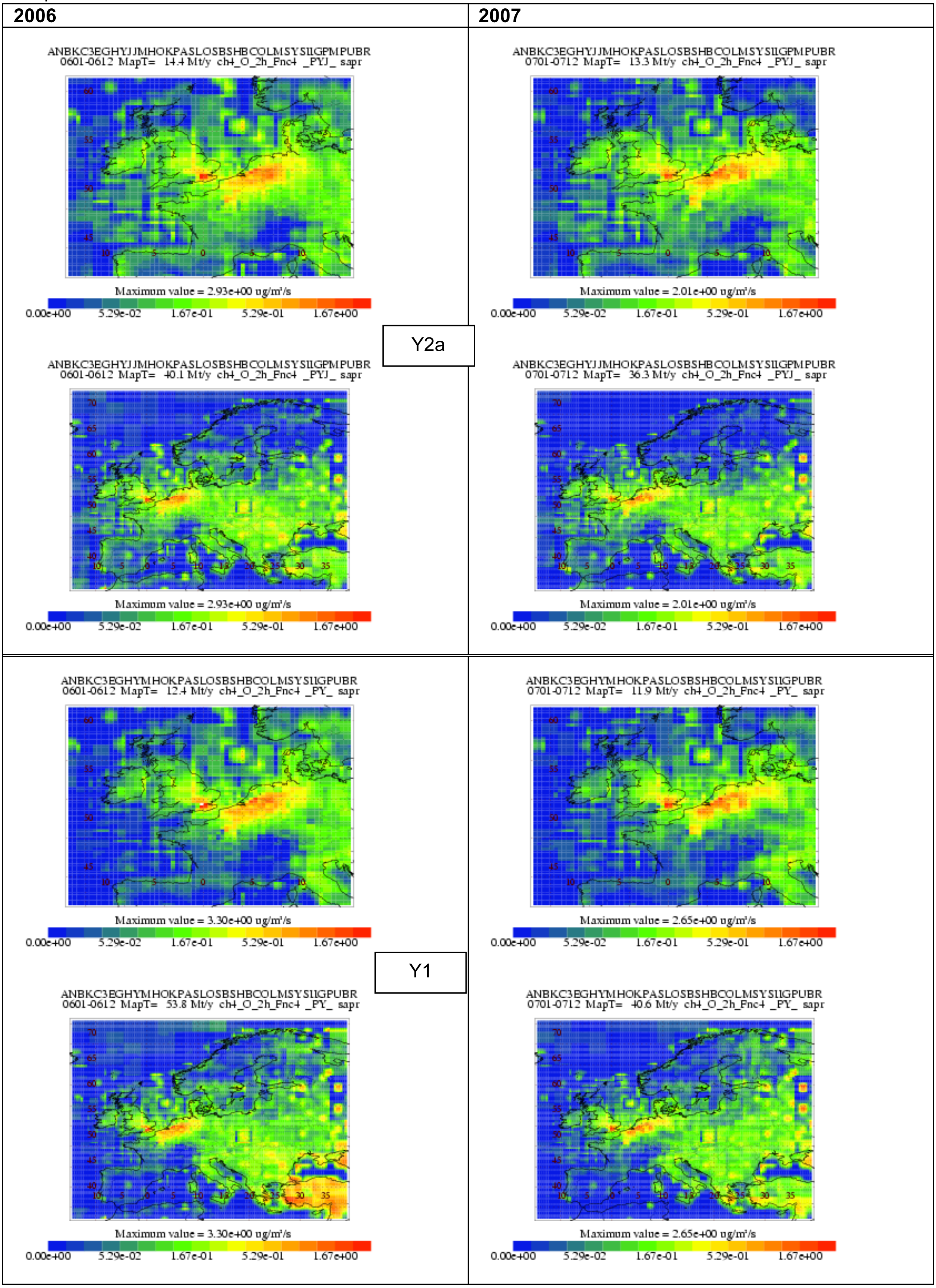


Figure 1: Emission maps for CH_4, obtained from ExpY2a and ExpY1 inversions.

CH_4

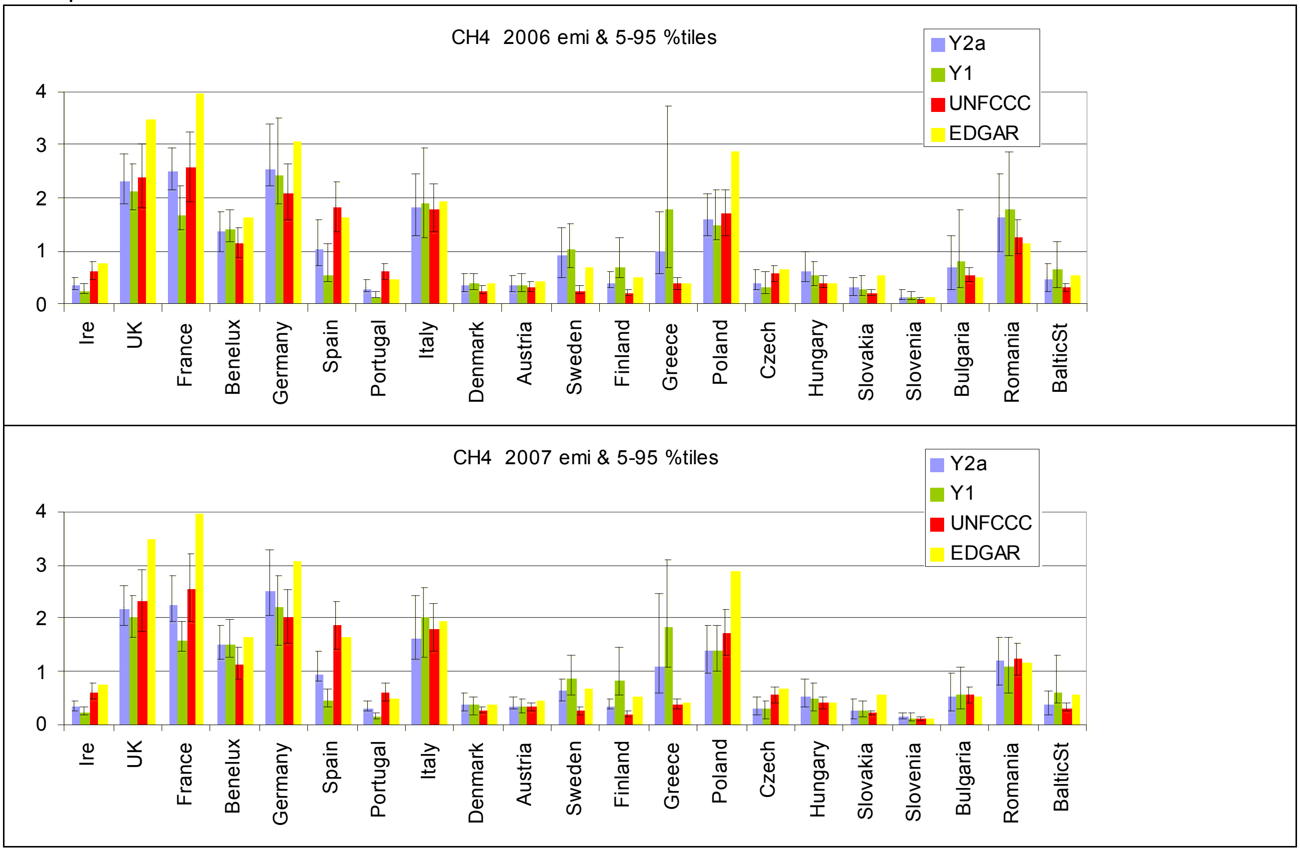


Figure 2: Individual country total emissions and uncertainty (5 and 95 percentiles), for CH_4.

CH_4

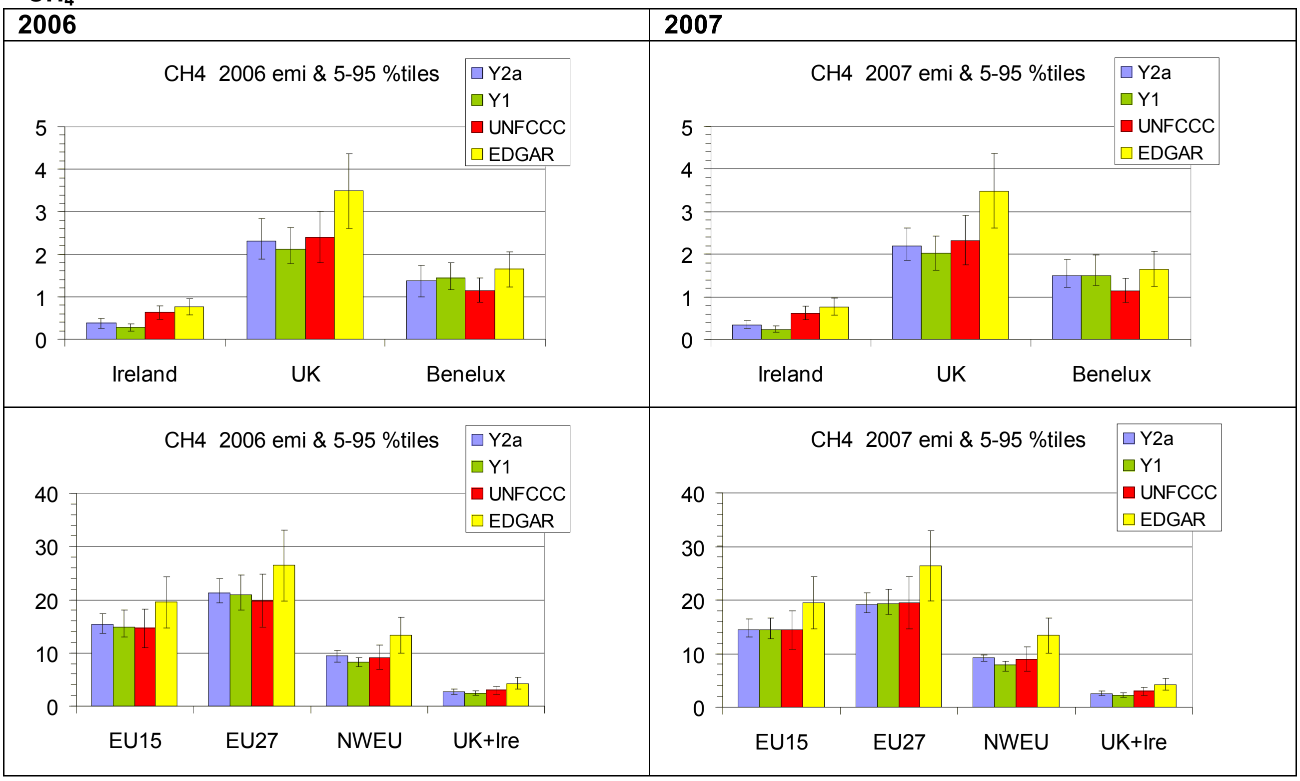


Figure 3: Emission totals and uncertainty (5 and 95 percentiles), for CH_4.

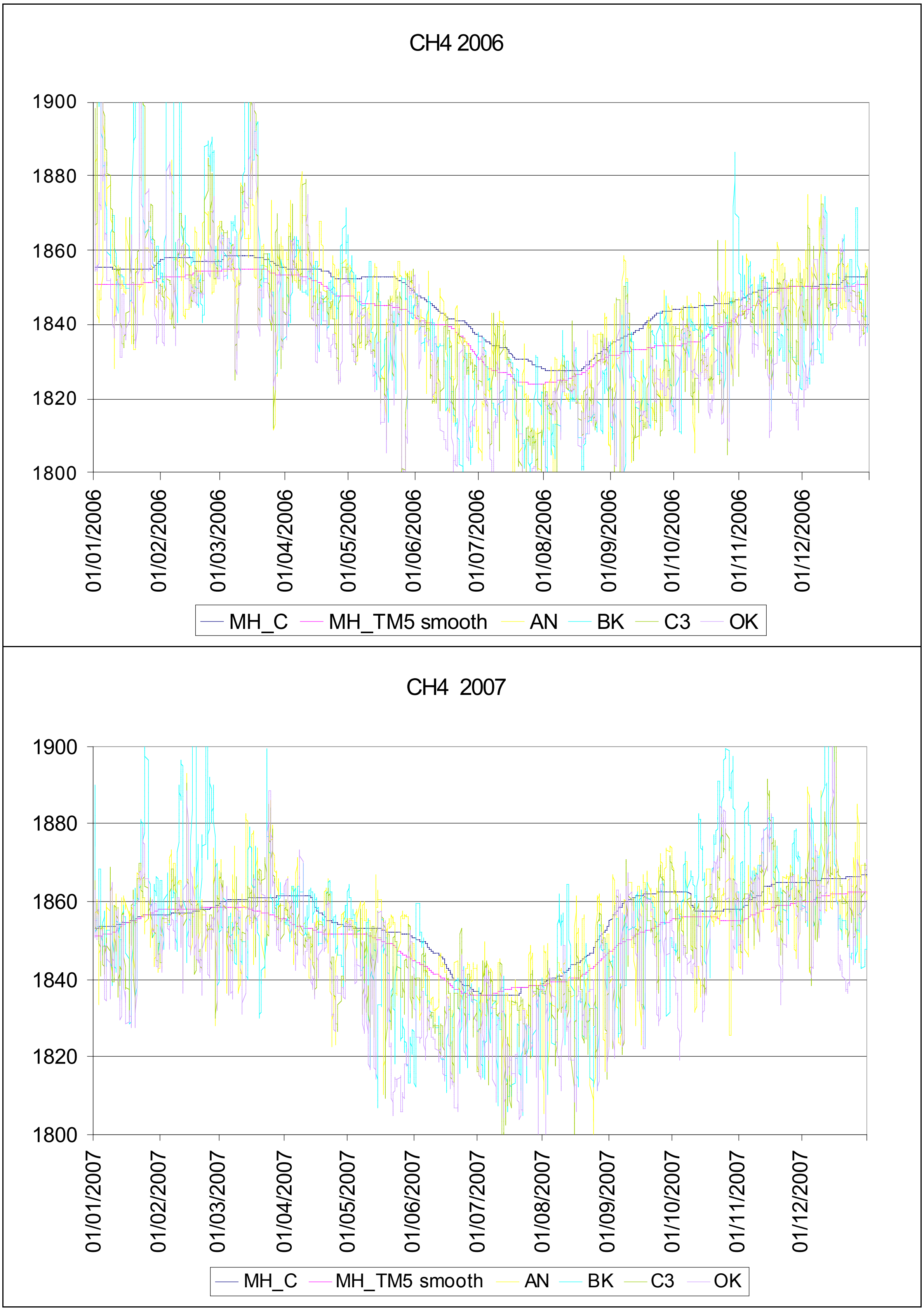


Figure 4: Mace Head (Met Office) baseline compared to TM5 baselines at various stations across Europe for CH_4.

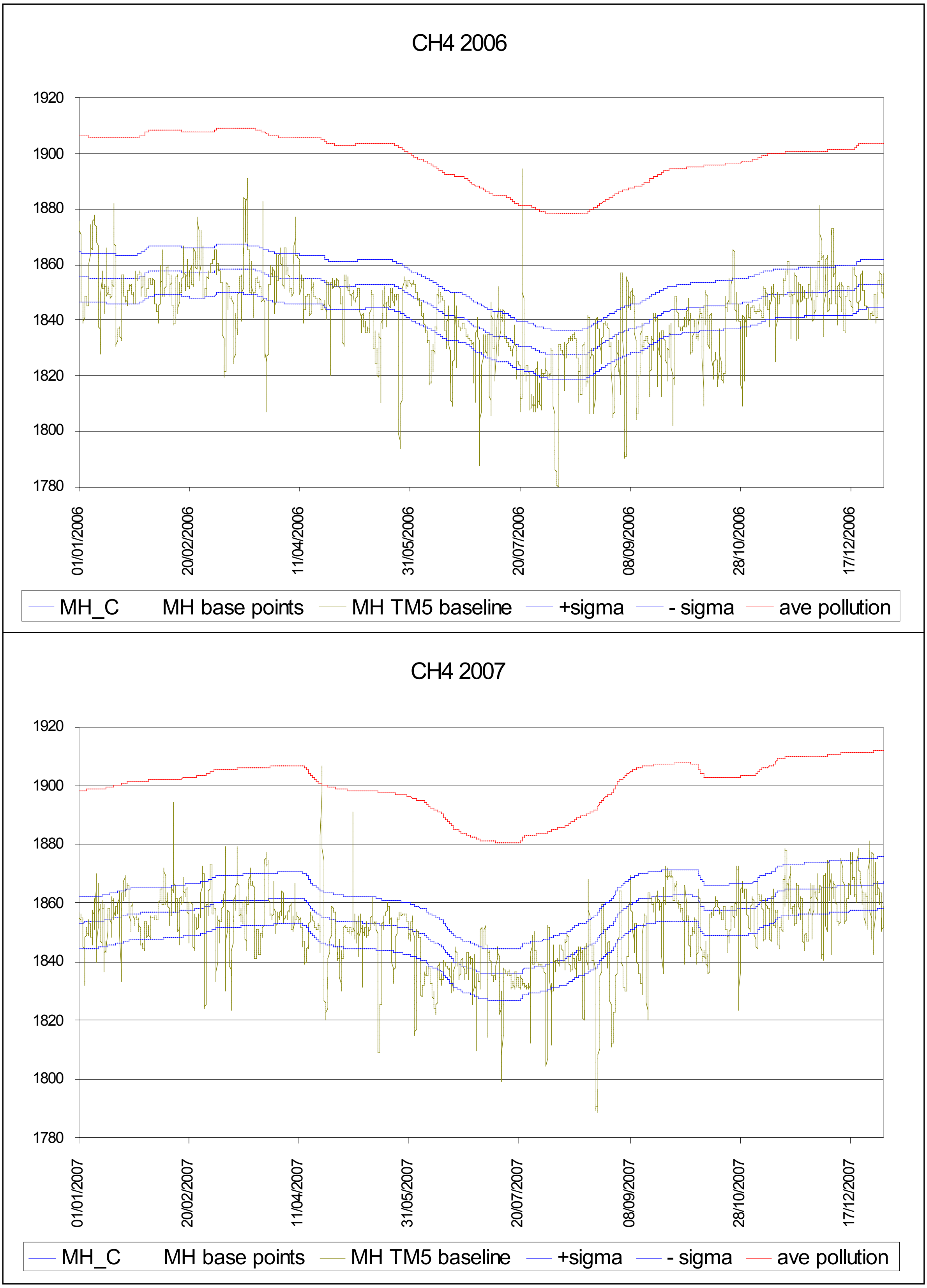


Figure 5: Comparing the Met Office baseline (MOB) at Mace Head (blue with 1 standard deviation uncertainty) with the unsmoothed TM5 baseline at Mace Head (green) for 2006 (top) and 2007 (bottom) for CH_4. The red line shows the average elevation in concentration seen in pollution episodes.

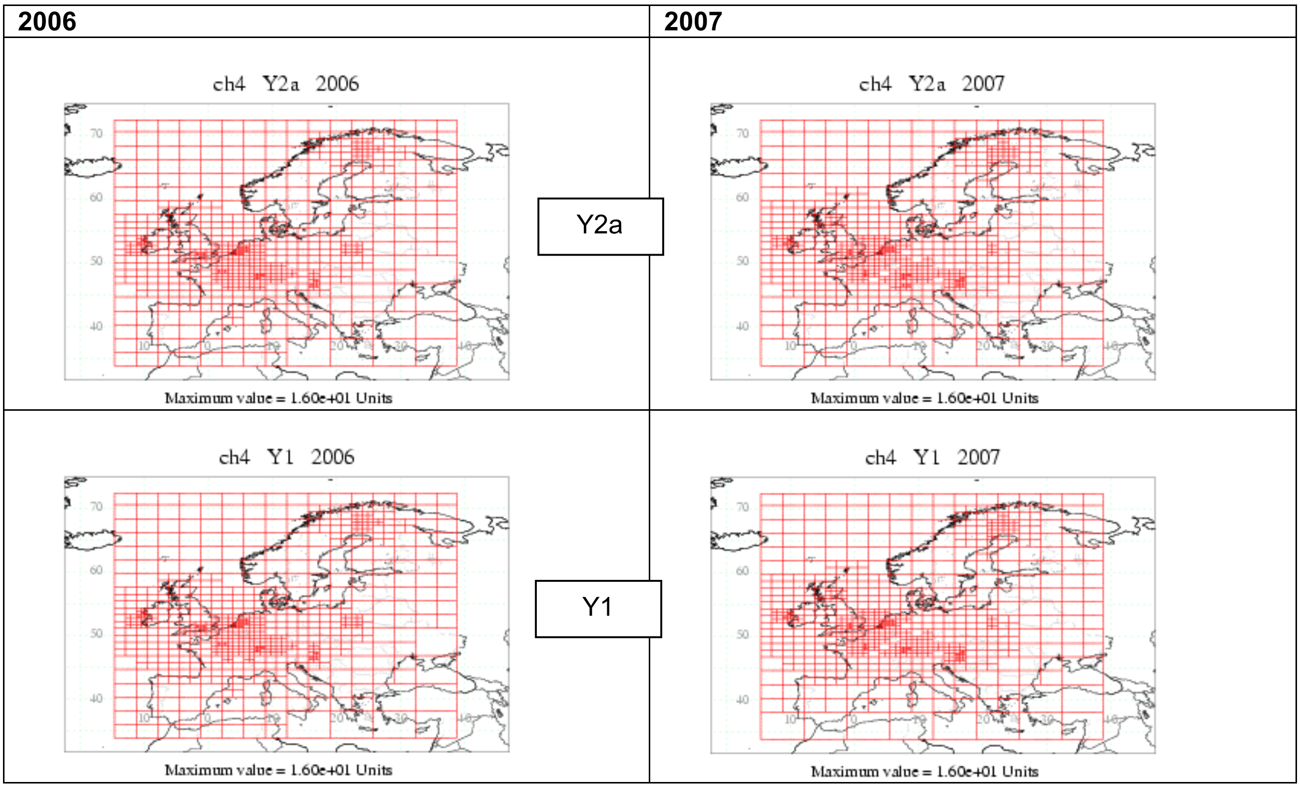


Figure 6: Inversion grids for CH_4 ExpY2a (top) and ExpY1 (bottom).

N_2O

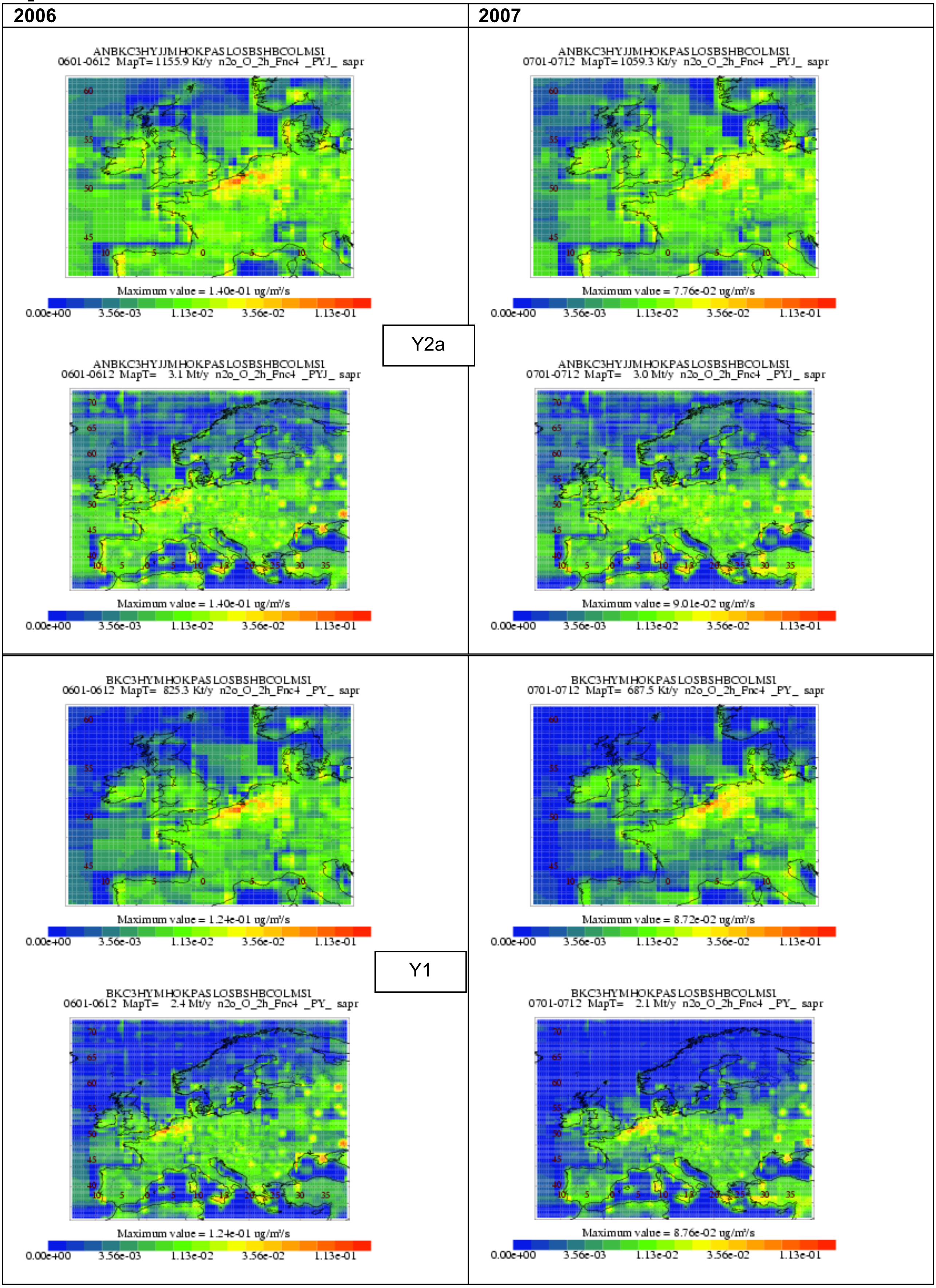


Figure 7: Emission maps for N_2O, obtained from ExpY2a and ExpY1 inversions.

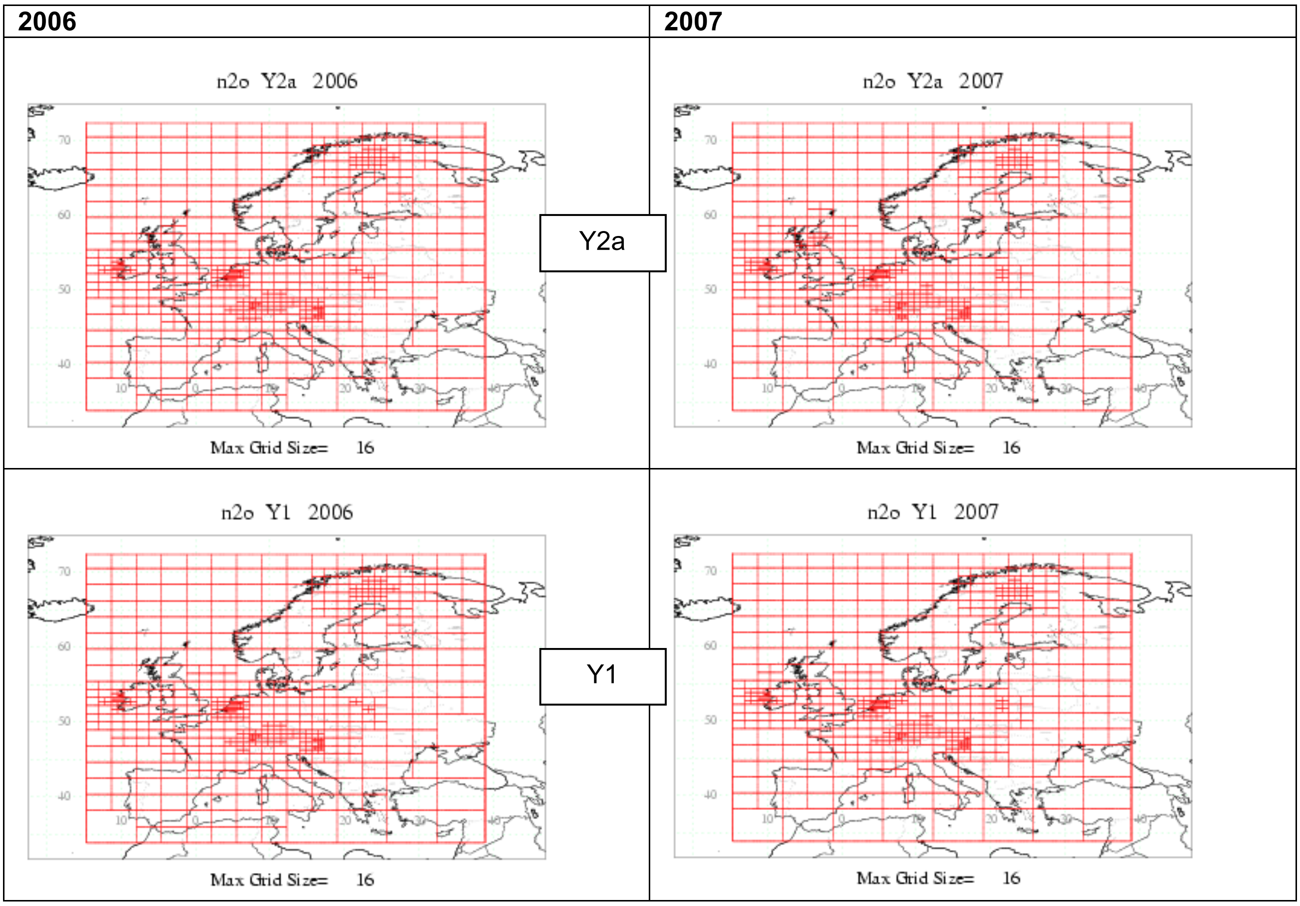


Figure 8: Inversion grids for N_2O ExpY2a (top) and ExpY1 (bottom).

N_2O

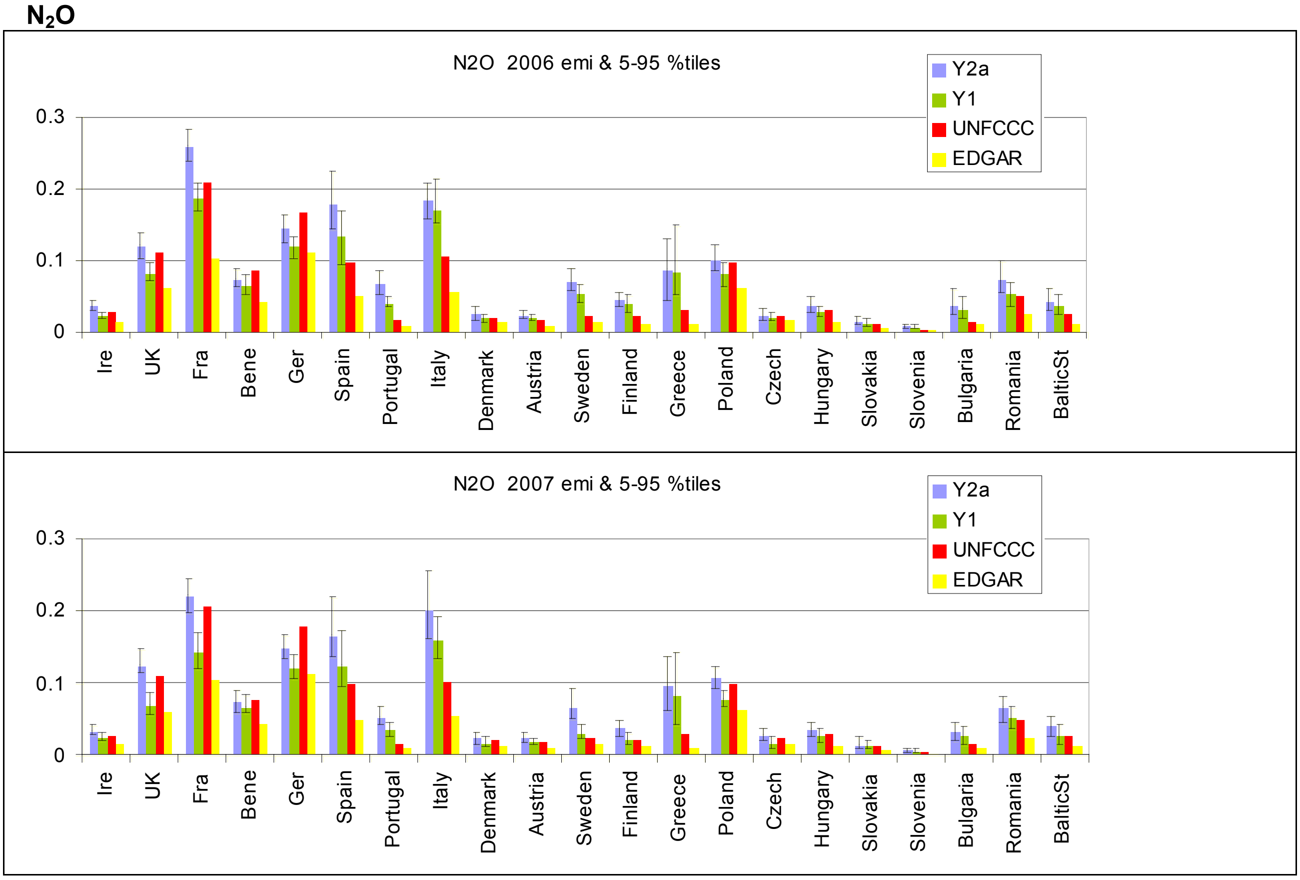


Figure 9: Individual country total emissions and uncertainty (5 and 95 percentiles), for N_2O.

N_2O

2006	2007

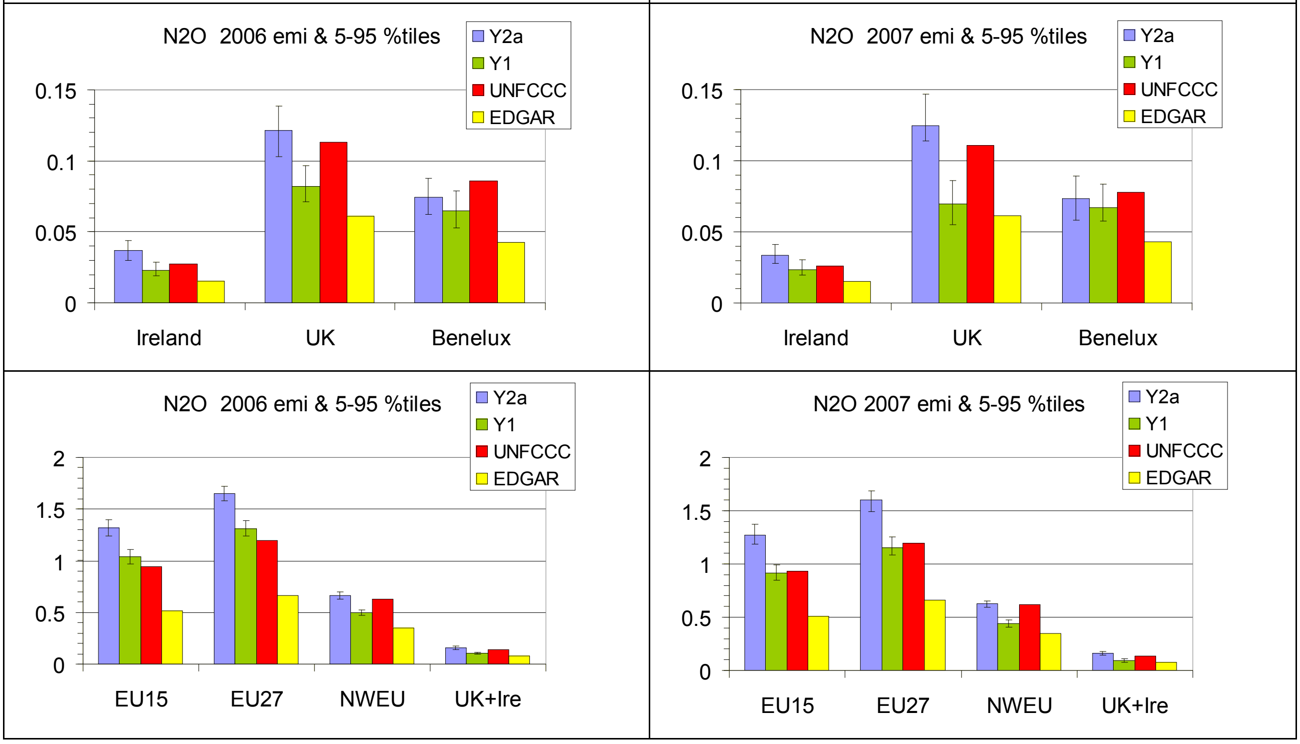
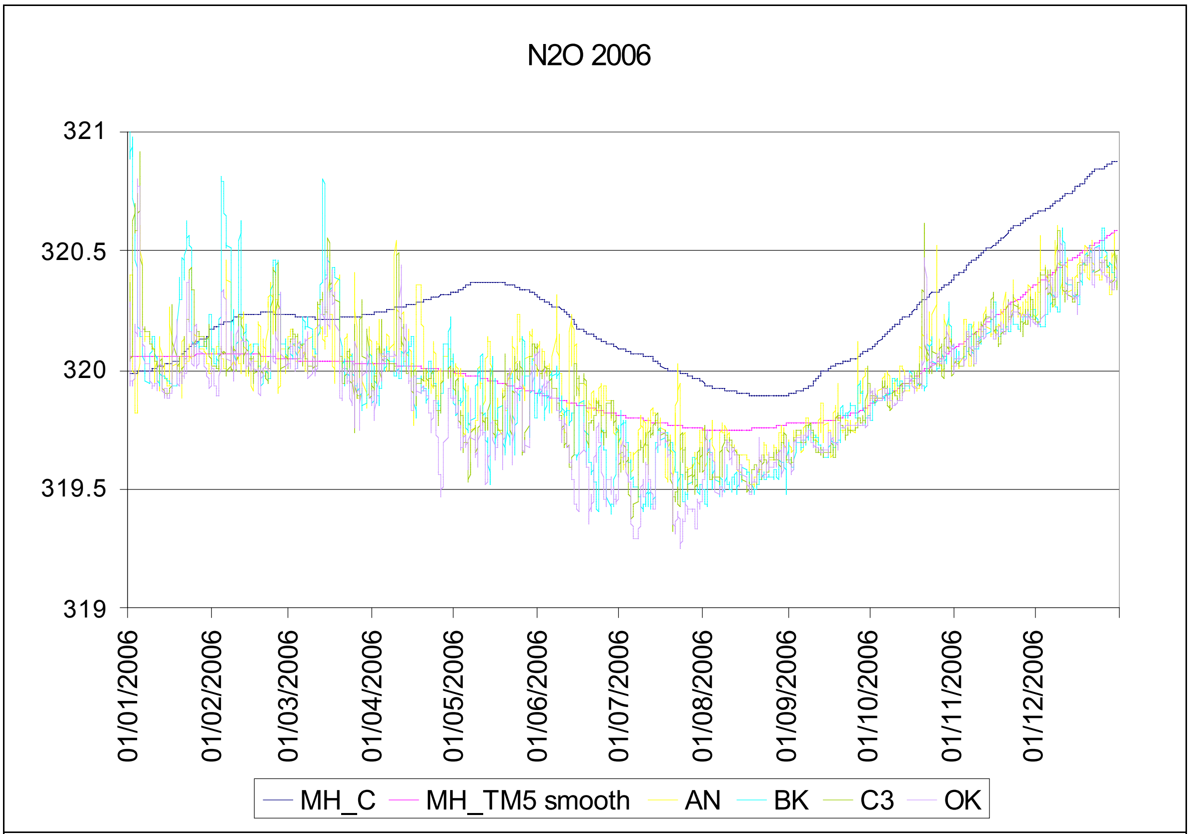


Figure 10: Emission totals and uncertainty (5 and 95 percentiles) for N_2O.



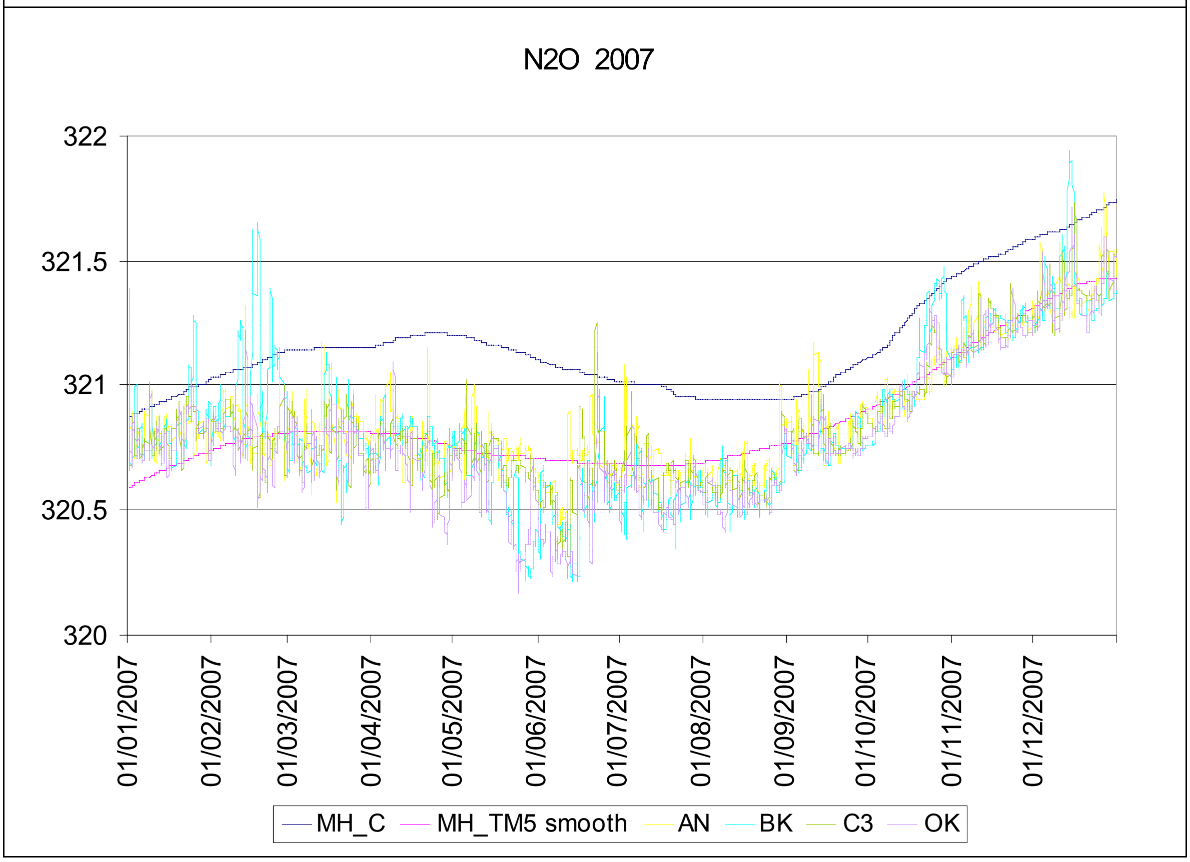


Figure 11: Mace Head (Met Office) baseline compared to TM5 baselines at various stations across Europe for N_2O.

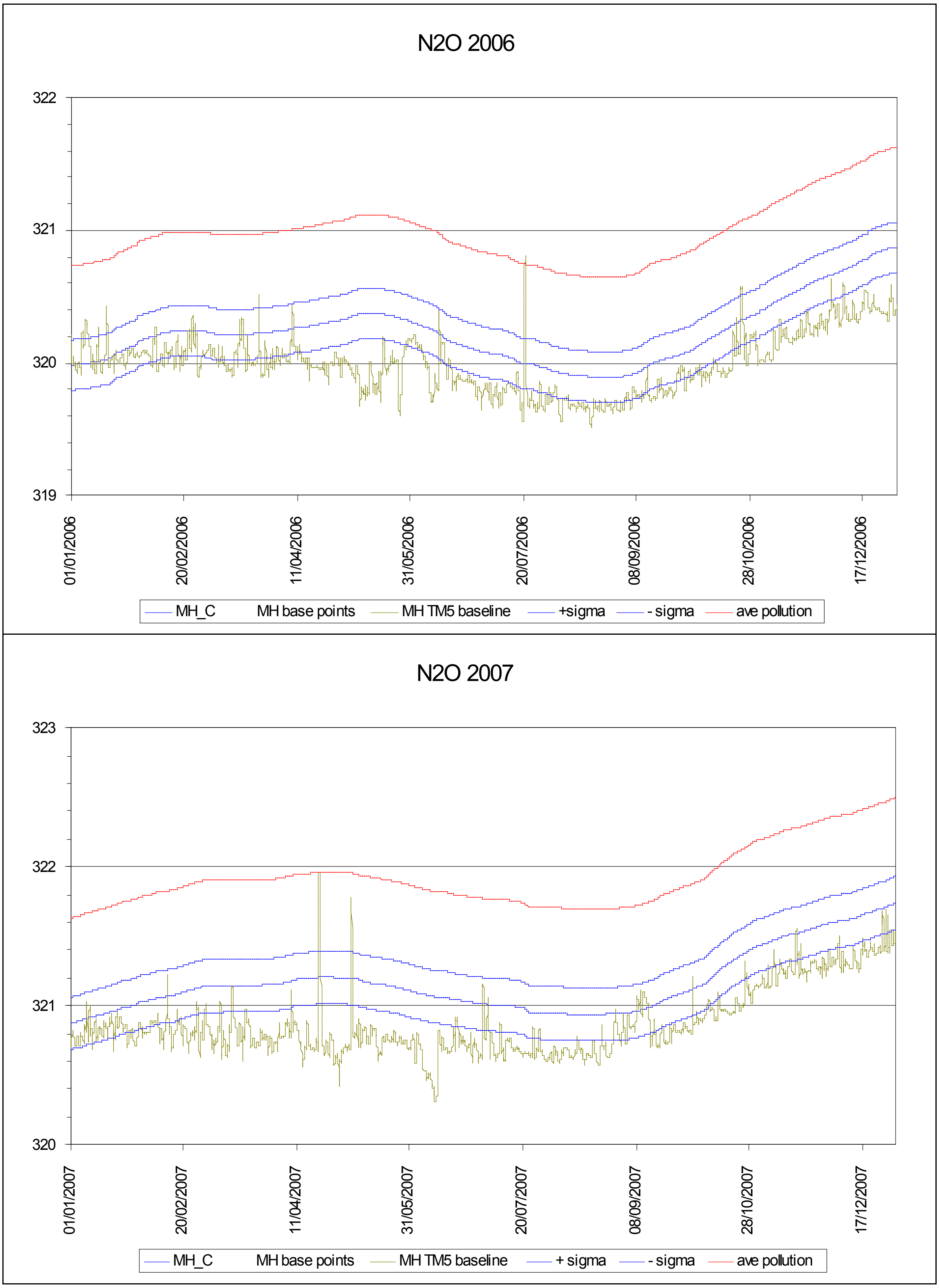


Figure 12: Comparing the Met Office baseline (MOB) at Mace Head (blue with 1 standard deviation uncertainty) with the unsmoothed TM5 baseline at Mace Head (green) for 2006 (top) and 2007 (bottom) for CH_4. The red line shows the average elevation in concentration seen in pollution episodes.