Interpretation of long-term measurements of radiatively active trace gases and ozone depleting substances

Quarterly report

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Authors:
Alistair J. Manning, Maria Athanassiadou (Met Office),
Simon O'Doherty, Aoife Grant, Dickon Young (University of Bristol),
Richard G. Derwent (rdscientific),
Peter Simmonds

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1 Executive Summary

Monitoring of atmospheric concentrations of gases is important in assessing the impact of international policies related to the atmospheric environment. The effects of control measures on chlorofluorocarbons (CFCs), halons and HCFCs introduced under the 'Montreal Protocol of Substances that Deplete the Ozone Layer' are now being observed. Continued monitoring is required to assess the overall success of the Protocol and the implication for atmospheric levels of replacement compounds such as HFCs. Similar analysis of gases regulated by the Kyoto Protocol on greenhouse gases will likewise assist policy makers.

Since 1987, high-frequency, real time measurements of the principal halocarbons and radiatively active trace gases have been made as part of the Global Atmospheric Gases Experiment (GAGE) and Advanced Global Atmospheric Gases Experiment (AGAGE) at Mace Head, County Galway, Ireland. For much of the time, the measurement station, which is situated on the Atlantic coast, monitors clean westerly air that has travelled across the North Atlantic Ocean. However, when the winds are easterly, Mace Head receives substantial regional scale pollution in air that has travelled from the industrial regions of Europe. The site is therefore uniquely situated to record trace gas concentrations associated with both the mid-latitude Northern Hemisphere background levels and with the more polluted air arising from Europe.

The observation network in the UK has been expanded to include three additional stations; Angus Tower near Dundee, Tacolneston near Norwich and Ridge Hill near Hereford. Tacolneston is under construction and is now operational. Ridge Hill became operation March 2012. Angus Tower, run by Edinburgh University, has been making measurements since late 2005. This report will focus on the progress on setting up the Tacolneston site and on the modelling side, developing emission estimates for each Devolved Administration (DA). The Met Office inversion modelling system used is called to as InTEM (Inversion Technique for Emission Modelling).

1.1 Publications

Derwent, R.G., Simmonds, P.G., O'Doherty, S., Grant, A., Yates, E.L., Manning, A.J., Utembe, S.R., Jenkin, M.E., Shallcross, D.E. 'Seasonal cycles in short-lived hydrocarbons and halocarbons in baseline air masses arriving at Mace Head, Ireland', Submitted to Atmospheric Environment, June 2012.

Kim, J., Fraser, P., Mühle, J., Li, S., Manning, A.J., Trebler,A., Stohl, A., Ganesan, A., Krummel, P., Steele, P., Saito, T., Park, S., Kim, S-K., Park, M-K., Arnold, T., Harth, C., Salameh, P., Yokouchi, Y., Weiss, R., Prinn, R., Kim, K-R. 'Emissions of Tetrafluoromethane and Hexafluoroethane: Balancing Anthropogenic Budgets from Atmospheric Measurements'. Submitted to JGR, July 2012.

Arnold, T., Harth, C.M., Mühle, J., Salameh, P.K., Kim, J., Manning, A.J., Ivy, D.J., Steele, P., Petrenko, V.V., Severinghaus, J.P., Baggenstos, D., Weiss, R.F., 'Nitrogen trifluoride global emissions estimated from revised and updated atmospheric measurements'. Submitted to Proceedings of the National Academy of Sciences, July 2012

1.2 Meetings

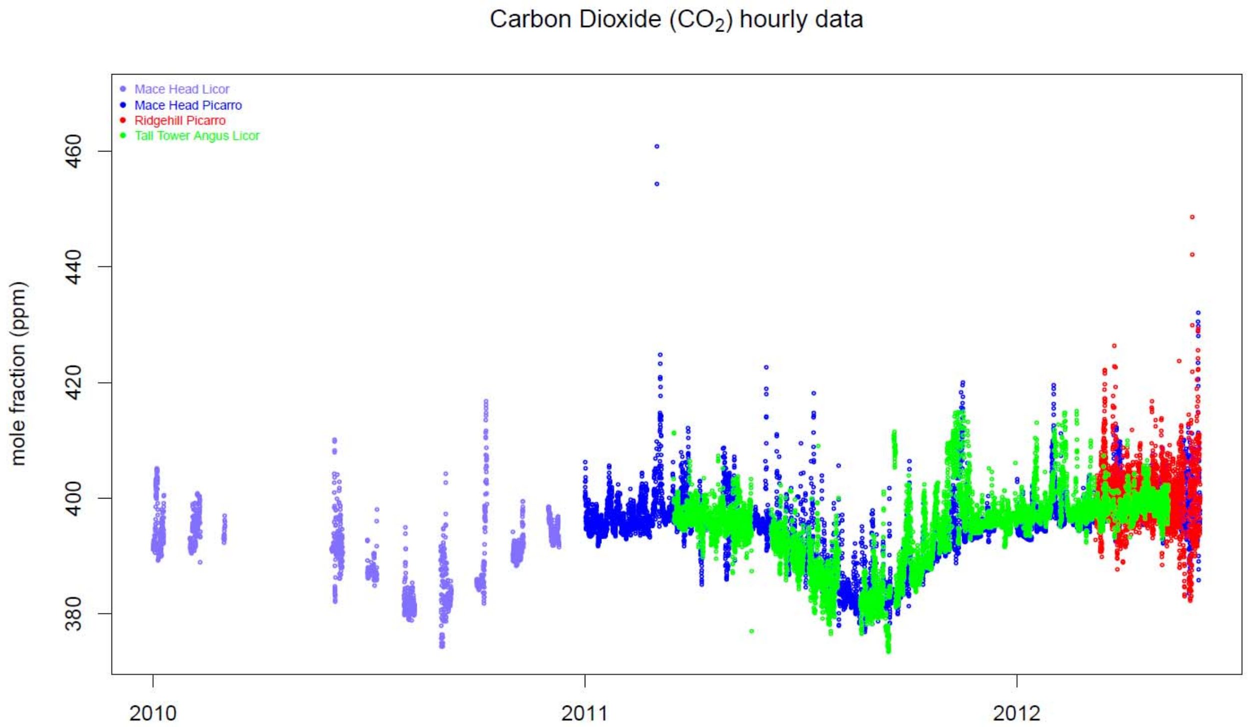
AGAGE meeting, Boulder, Colorado, USA (May 2012)

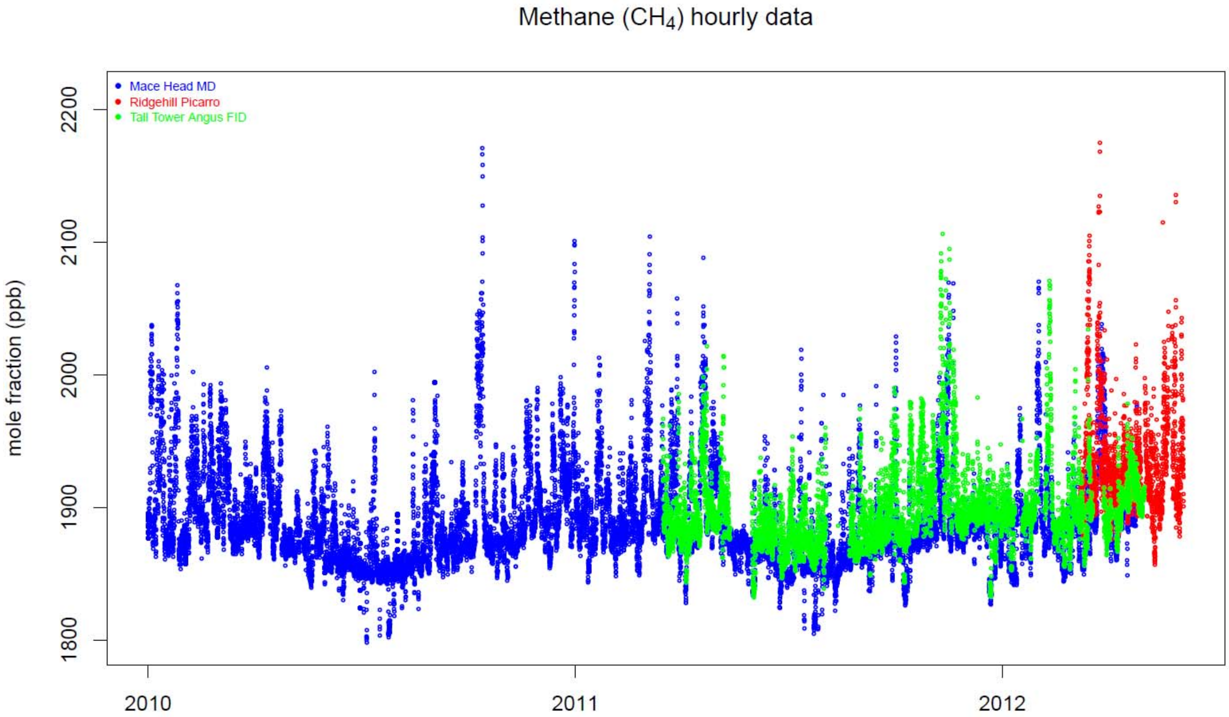
DECC progress meeting, London (June 2012)

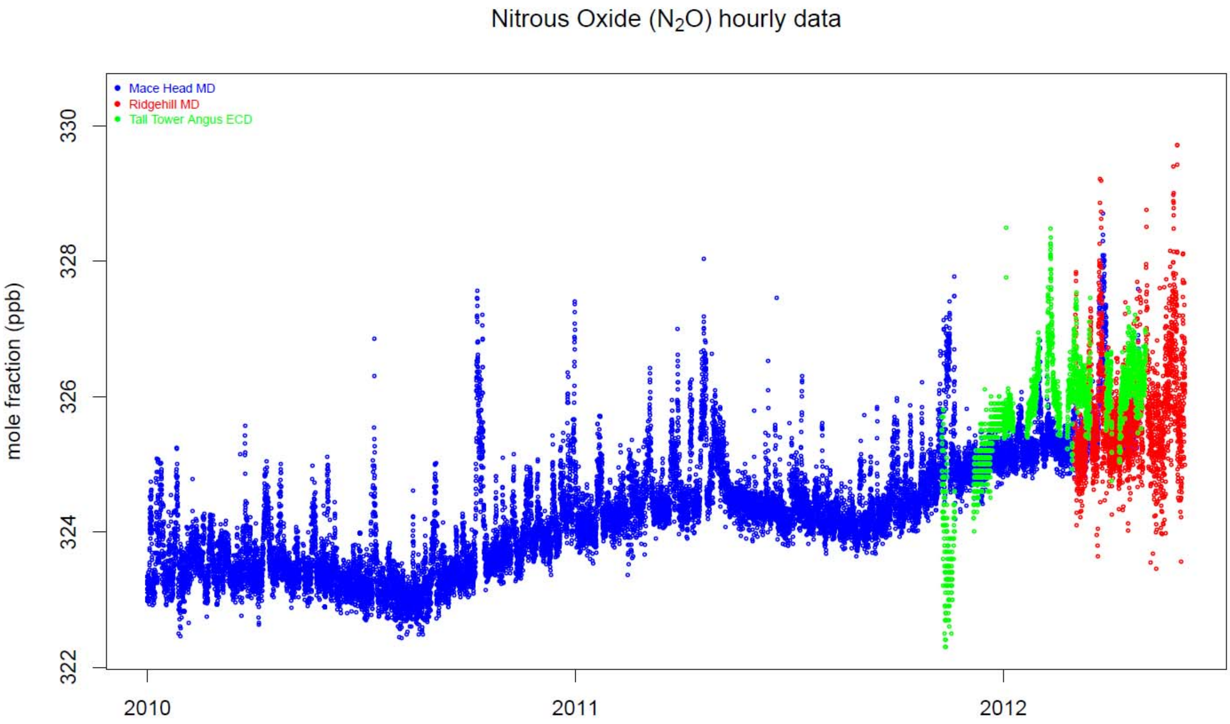
2 Update on Three UK sites

2.1 Angus Tower

The GC-ECD at Angus Tower (TTA) which measures N_2O and SF_6 was previously operating with problems (and a very high background signal). The ECD detector was replaced in December 2012. The new detector gave a low background and data quality improved for N_2O and SF_6 for a time. However recent N_2O data does not appear to overlay well with Mace Head measurements (Figure 1) indicating there may be a problem with these data. This could be caused by possible chromatographic problems or problems with peak integration but it could also be due to contamination of the air sample line with algae. SF_6 data overlays well with Mace Head data, which is a little unexpected as it is separated on the same column as N_2O and measured on the same detector as N_2O. CH_4 data overlays well with Mace Head but the magnitude of pollution events is smaller at Angus than Mace Head. Possible reasons for this are that the line is contaminated with algae which may consume CH4 (as for N_2O). Or because Angus samples are taken from 185 m up the tower and at this height the pollution events are diluted. These possibilities are currently being investigated. CO_2 data overlays well with Mace Head data.




Methane (CH_4) hourly data
Mace Head MD
Ridgehill Picarro
Tall Tower Angus FID
mole fraction (ppb)
1800
1900
2000
2100
2200
2010
2011
2012


Nitrous Oxide (N_2O) hourly data
Mace Head MD
Ridgehill MD
Tall Tower Angus ECD
mole fraction (ppb)
322
324
326
328
330
2010
2011
2012

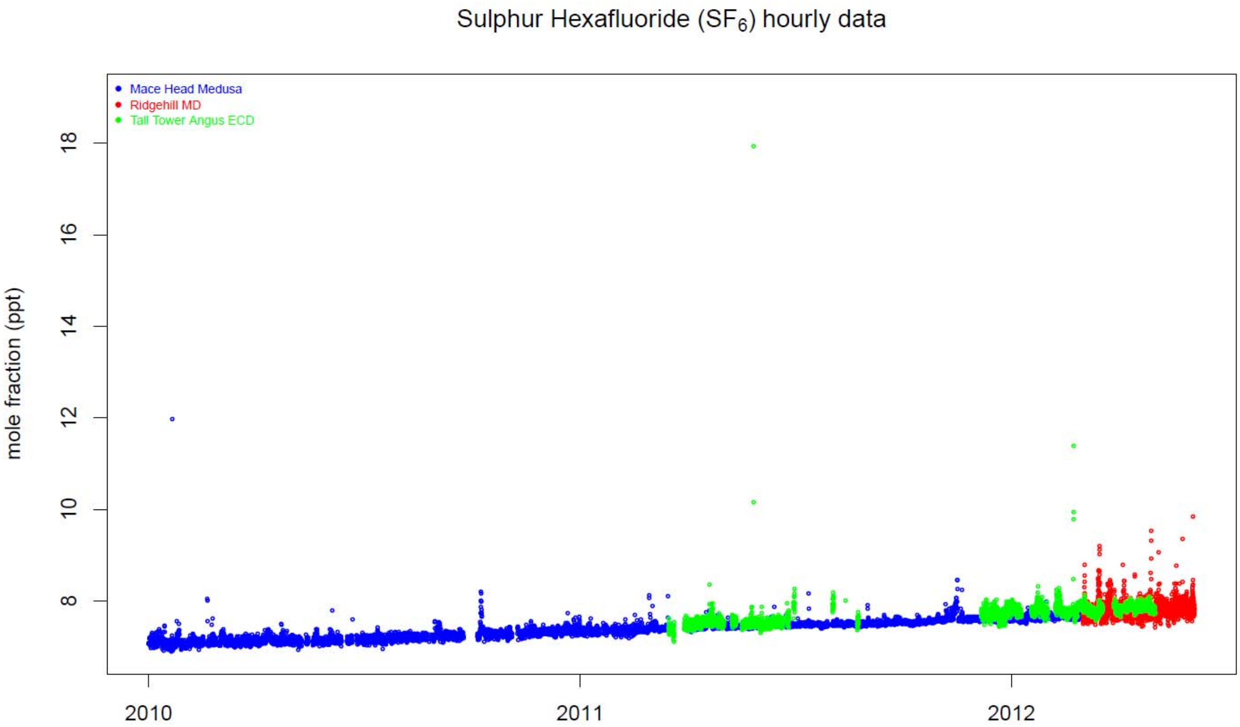


Figure 1: Angus data (green), Mace Head data (blue) and Ridge Hill data (red) for CO_2, CH_4, N_2O and SF_6. N_2O at Angus is not currently useable due to instrumental issues.

2.2 Tacolneston

Tacolneston will be operational by August 2012.

Timeline of events at Tacolneston:

- 18th 19th Oct: Instrument training for UEA
- 27th Oct: Installation site meeting
- 8th Nov: Inlet line offer received and signed
- 21st Nov.: Inlet lines installed on tower (Figures 2 and 3)
- 12th Dec: Initial planning drawings
- 19th Dec: Instruments moved into mobile lab (Figure 4)
- 10th-11th Jan: Medusa training for UEA at UoB
- 13th Jan: Planning drawings confirmed
- 29th Jan: Panning application submitted
- 27th March: Planning application approved
- 17rd May: Build offer received (23rd May revised Build Offer received)
- 25th May: Build offer returned
- 31st May: Teleconference re install
- 7th June: Licence agreement signed
- 21st June: Pre-start on site meeting for civil works/craning of mobile lab
- 16th-20th July: Mobile Lab plinth built, electricity supply and lines lead along gantry from tower to Mobile.
- 20th July: Mobile lab lifted in place by crane.
- 25th July: Instruments set-up.
- w/c 20th August: Internet installed into Mobile Lab and data transfer/communications set-up

All instruments were set-up and running on the 25th of July. The internet line was installed on the 25th of July but there has been several problems with its activation. BT have assured us this will be connected by the end of the week commencing 20th of August so data transfer from Tacolneston can occur.

Figure 2: The 100 meter line installed in the tower at Tacolneston. The attachment of the air sampling line to the stainless steel cup is shown on the left and two of the air sampling lines running down through cleats is shown on the right.

Figure 3: The three measured air sampling lines coiled up on the day of installation with the riggers in the background preparing the proprietary cleats for attaching the tubing at 1 m intervals (left). The right hand image shows the new tall tower at Tacolneston on the left, where our sample lines are installed, with the obsolete historic tower in the centre and the smaller but still operational tower on the far right.

Figure 4: The Medusa (left) and MD (right), installed and running samples in the Mobile Lab.

2.3 Ridge Hill

The Cavity Ring Down Spectrometer (CRDS) has generally been running well at Ridge Hill. The CRDS was installed on the 23rd of February. The CRDS began running air from the two tower heights 45 m and 90 m alternating every 30 minutes and running the target tank at 7 hour intervals to asses for instrumental drift. The cavity vacuum pump broke on the 7th of March and was replaced. Later this month a large portion of the target tank was lost so the sequence was changed to run this tank less frequently (every 20 hours). Automatic data transfer to ICOS was established along with the return transfer of calibrated data.

The multiple detector (MD) at Ridge Hill has generally been running well since it's installation on the 24th of February however the precision, particularly for N_2O measurements needed to be improved for data to be used in emission estimation
During very heavy rain at the end of April water the water trap for the 90 m line filled to capacity and water was sucked into the air sample line. This entered the air pump module for the MD and caused problems. The sampling set-up was modified to prevent the re-occurrence of this but precision on the instrument dropped due to the water contamination even after its removal.
The size of sample collected and analysed on this instrument was 3 ml. To improve the precision of measurements a 8 ml sample was fitted on the 11th of July. Extra insulation was also fitted around the ECD detector as the ECD is very sensitive to lab temperature fluctuations. And as the lab is not air-conditioned temperature fluctuations were causing part of the ECD problems. These two changes improved N_2O precision dramatically. Precision on concurrent standards went from 0.1-0.2% daily precision to 0.03% - 0.06% daily precision. SF_6 precision did not improve so dramatically as it is adjacent to a large oxygen peak and minimising this peak may sacrifice the precision of N_2O data. SF_6 daily precisions has improved from 1% to between 0.5 and 1.0%.

3 Regional Emission Estimates – Devolved Administrations

Previously the grid used in the inversion system, InTEM, as set up to be regular, see Figure 5. In that formulation the core basic grid (25 km resolution), referred to as a 1x1 grid, is combined with adjacent grids to form 2x2, 4x4, 8x8, etc. grids based on the amount of information the receptor (measurement location) 'sees' of each grid. In that way the grids used in the inversion system that are more distant from the receptor are larger than those close by. Although mathematically elegant, this solution has difficulty in defining the actual borders of different countries. In order to estimate emissions at the DA level a new method has been developed that preserves the actual (to the resolution of the basic grid resolution) country borders but that still ensures the contribution from each amalgamated grid in the inversion system is similar, i.e. each amalgamated grid provides a similar level of information to the receptor.

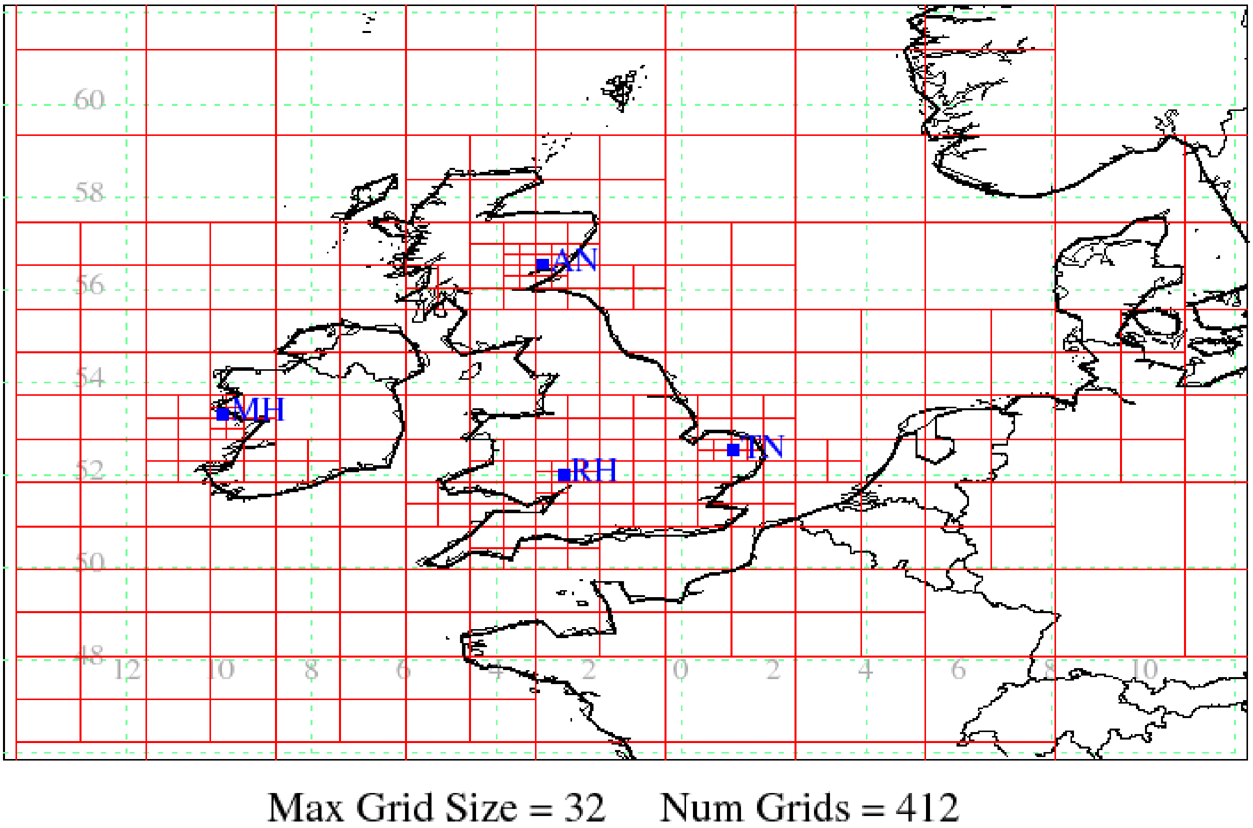


Figure 5: Traditional method for amalgamating the basic core grids based on the amount of information the receptor 'sees'. The basic grids (1x1) are grouped into 2x2, 4x4, 8x8, 16x16 and 32x32 amalgamated grids for use in the inversion system (InTEM).

The new system requires the inversion domain to be split into different core regions, these regions will not be amalgamated but can be further divided, see Figure 6. The contribution each region makes to the receptor is calculated by considering all of the NAME air history maps for the receptor when observations are available. A region is split into two if its contribution is above a certain threshold. This process is repeated until either all grids in the region are 1x1 or all sub-regions contribute less than the threshold. Each time a split is required, a split in both the x and y directions are considered, the direction which preserves a more equal balance of the number of grids and air history contribution is chosen. In this manner some of the larger regions are sub-divided many times (i.e. near Mace Head), others, like SE Europe, are potentially left as large single entities. The number of basic grids in each amalgamated grid can range anywhere from 1 to over 3500, previously they were restricted to one of 1,4,16, 64, 156, 1024.

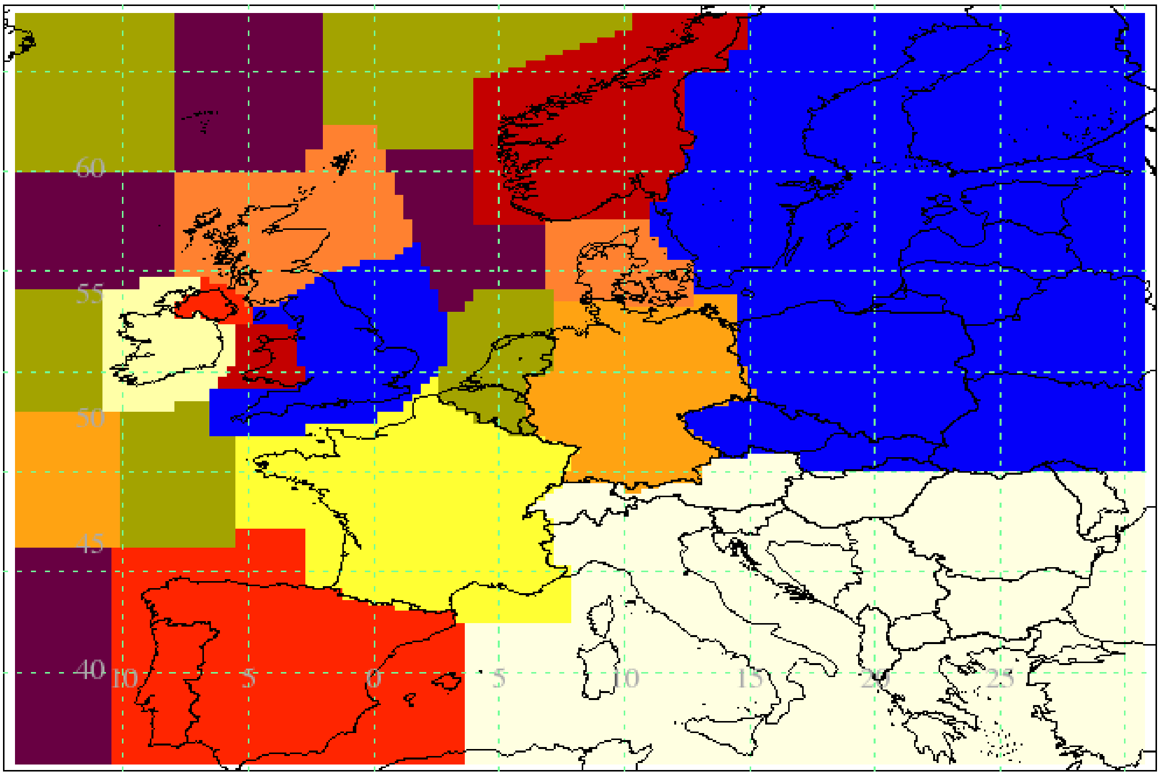


Figure 6: Core regions defined by the user. These regions cannot be amalgamated but can be sub-divided.

Figure 7 shows for HFC-134a for the period Oct 1994 – Sep 1997 the final grid resolution using a threshold of 17 s/m to be achieved at least 120 times. 120 times represents 10 days (the air history maps are 2-hourly so all observations are interpolated to 2-hours), 17 s/m is an arbitrary value estimated through comparison with the original grid amalgamation system.

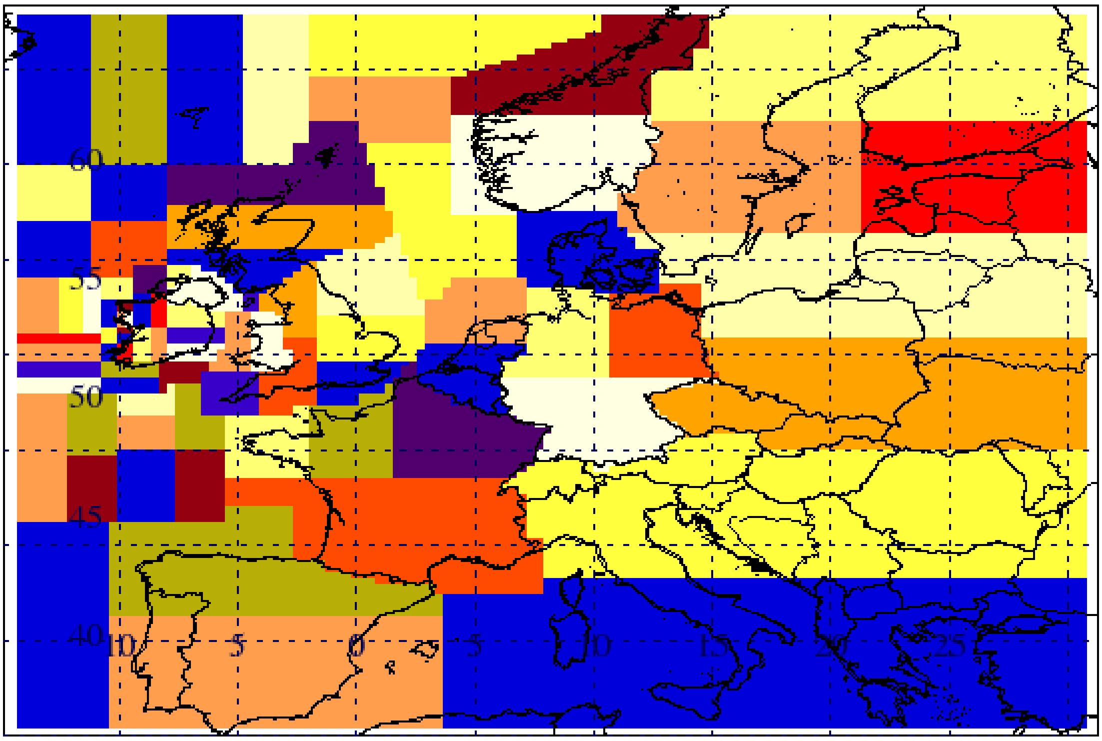


Figure 7: The distribution of the amalgamated grids for a 3-year (1995-1997) inversion using HFC-134a observations. The colour of each amalgamated grid is randomly determined to differentiate between adjacent grids.

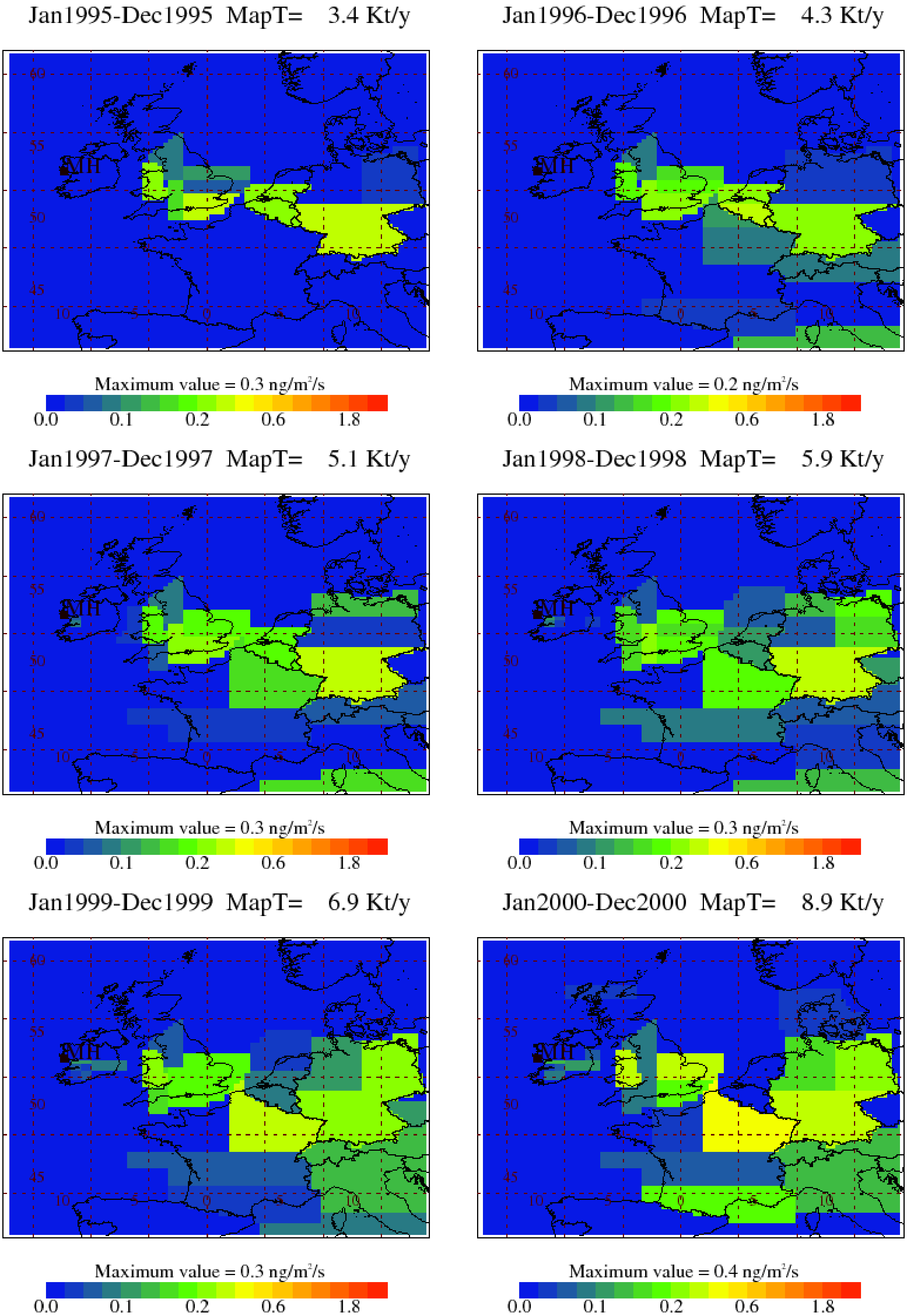


Figure 8: Annual average emission estimates for HFC-134a 1995-2000 inclusive.

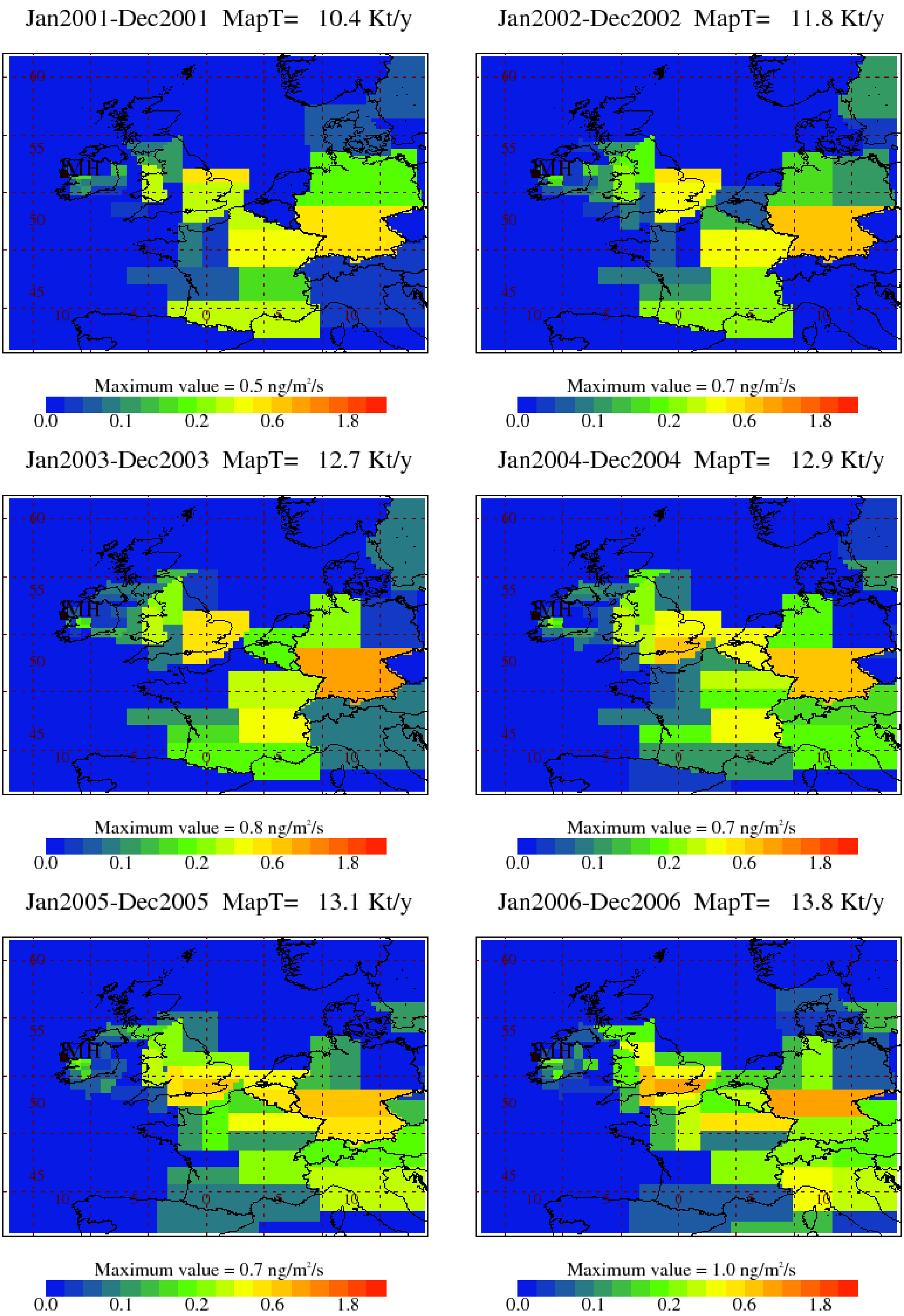


Figure 9: Annual average emission estimates for HFC-134a 2001-2006 inclusive.

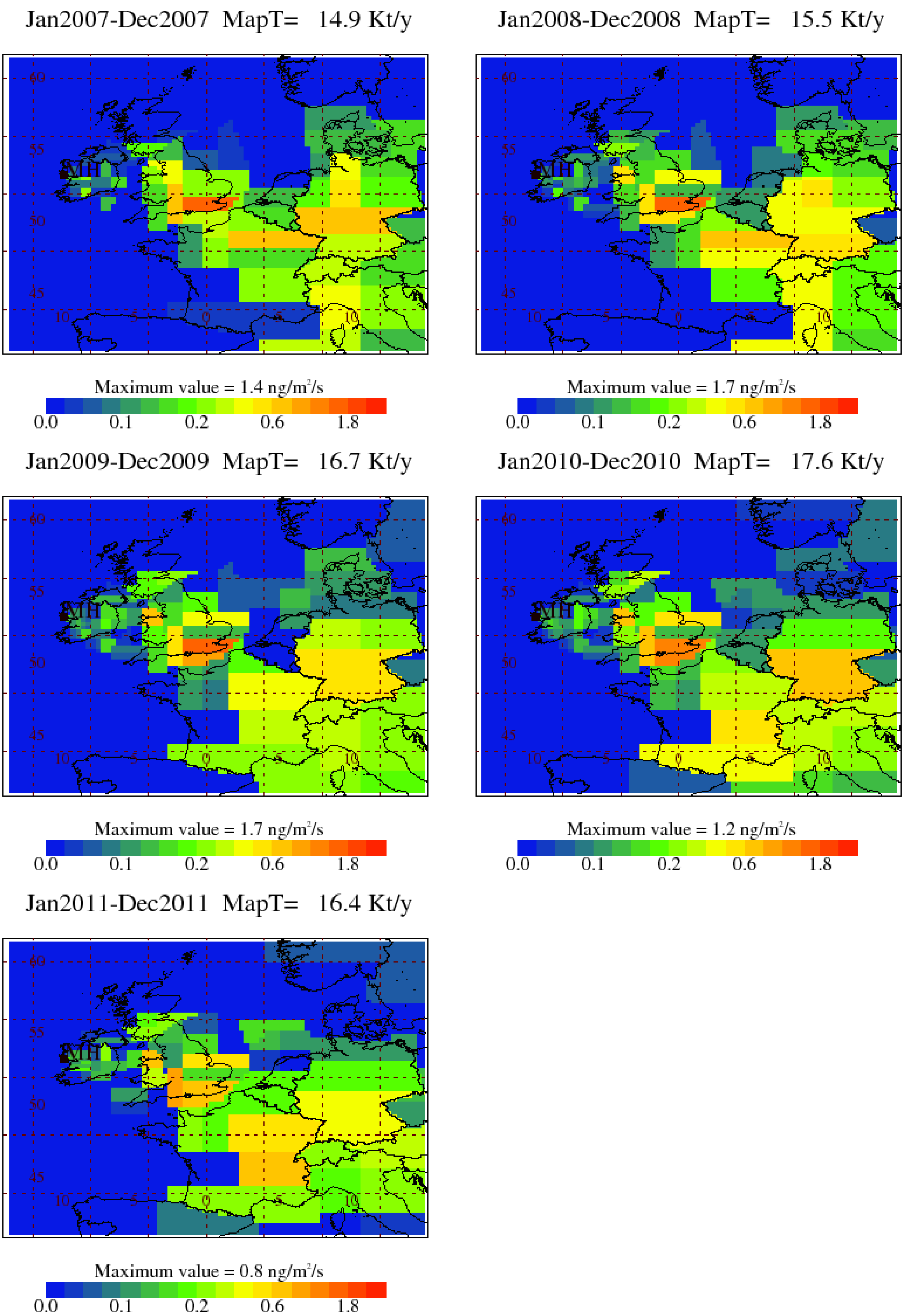


Figure 10: Annual average emission estimates for HFC-134a 2007-2011 inclusive.

Figures 8-10 show the InTEM (inversion) results for HFC-134a for each year from 1995 – 2011. The clear growth in emissions across Europe is clearly shown. The Irish, UK, UK + Ireland and North West European (NWEU) annual estimated emissions are shown in Figure 11.

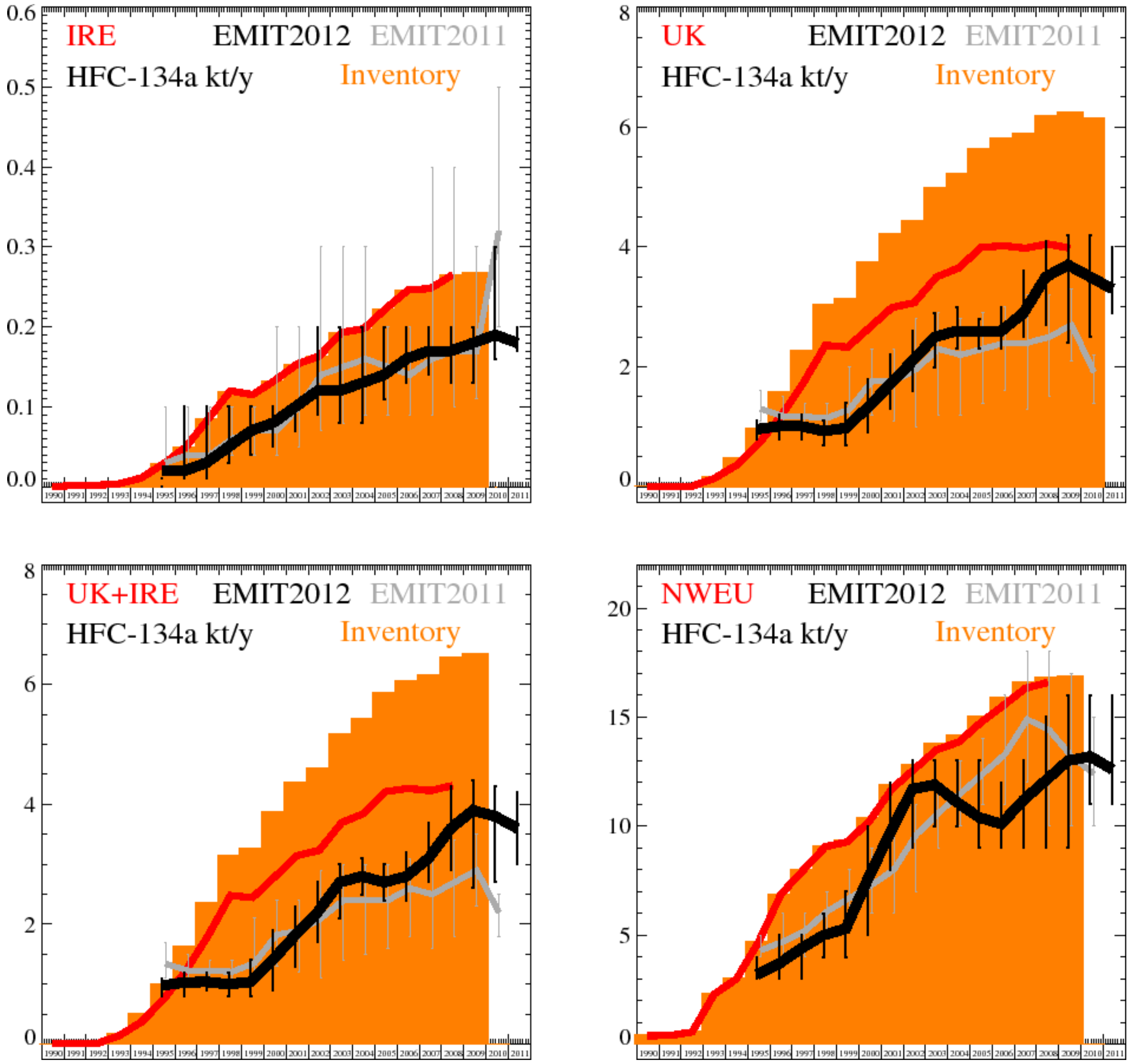


Figure 11: InTEM emission estimates of HFC-134a for IRE, UK, IRE+UK and NWEU. Uncertainty bars represent the 5th and 95th percentiles. Orange bars represent UNFCCC emission estimates from the 2012 inventory. Grey (InTEM) and red (UNFCCC) are emissions as estimated last year.

The new grid regions allow a clean definition of each Devolved Administration (DA). Figure 12 shows the annual estimates for HFC-134a for Scotland, Northern Ireland, Wales and England. Given a population split of 5.2M (Scotland), 1.8M (Northern Ireland), 3M (Wales) and 51.5M (England), the relative magnitudes are broadly consistent across the 4 DAs with population. The relative uncertainties of the emissions from the DAs are predictably larger compared to the whole UK.

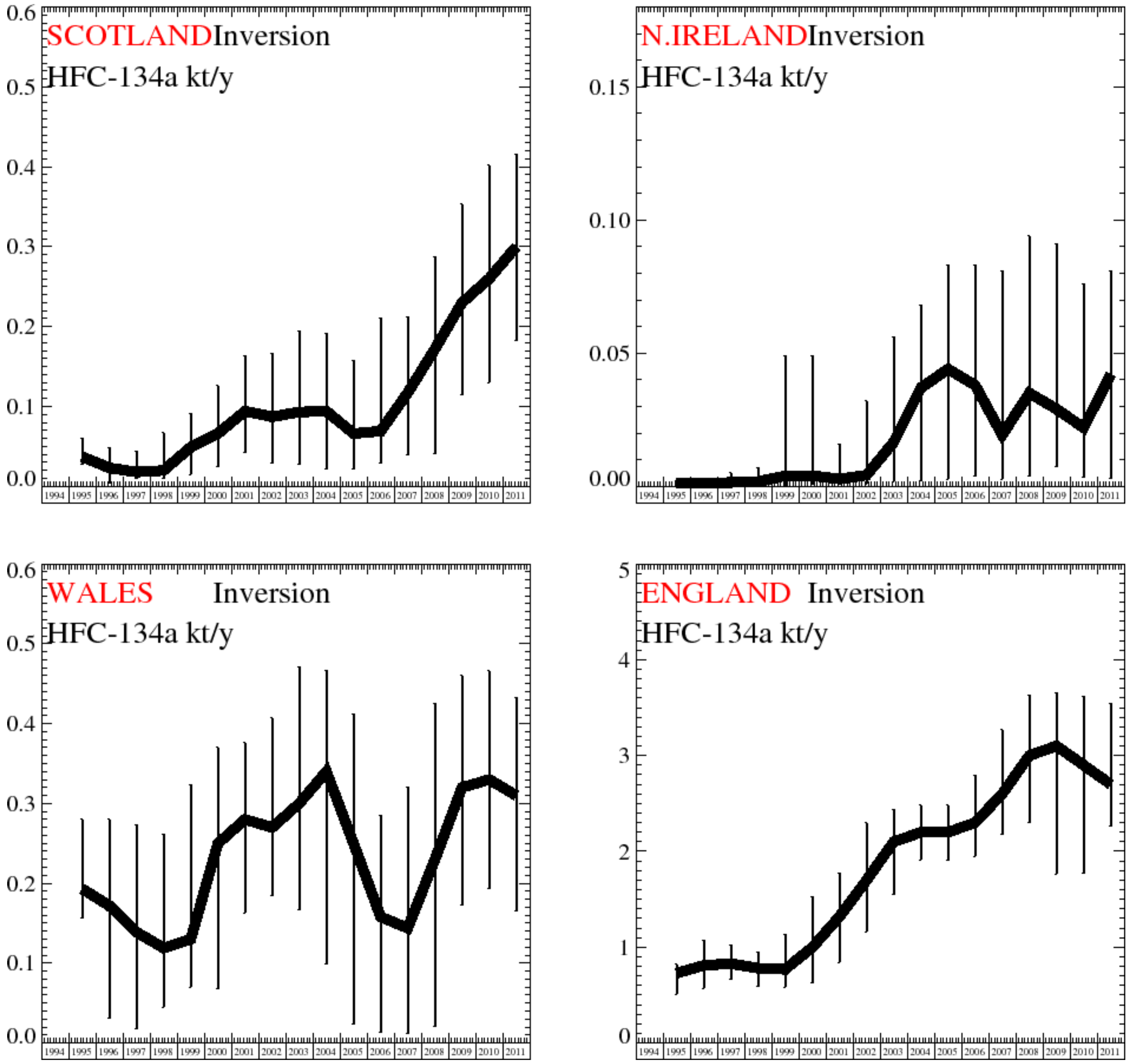


Figure 12: InTEM emission estimates of HFC-134a for Scotland, Northern Ireland, Wales and England. Uncertainty bars represent the 5th and 95th percentiles.