

Introduction

Hydrofluorocarbons (HFCs) are partially fluorinated hydrocarbons, typically used as refrigerants and propellants both industrially and domestically. Owing to the phase-out of CFCs and HCFCs under the Montreal protocol, the atmospheric abundances of these species are rising dramatically. Long atmospheric lifetimes and strong IR absorption profiles make them potent greenhouse gases.

Using a top-down approach, atmospheric observations of these species can be combined with a particle dispersion model to infer national emissions. Accurate top-down estimates are required for verification of the UK's greenhouse gas inventory, submitted to the UNFCCC as a stipulation of the Kyoto Protocol. The UK's existing HFC measurement network comprises two surface sites; here, we add aircraft measurements to enhance the network density.



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Emission estimates and characterisation of HFCs measured over the United Kingdom

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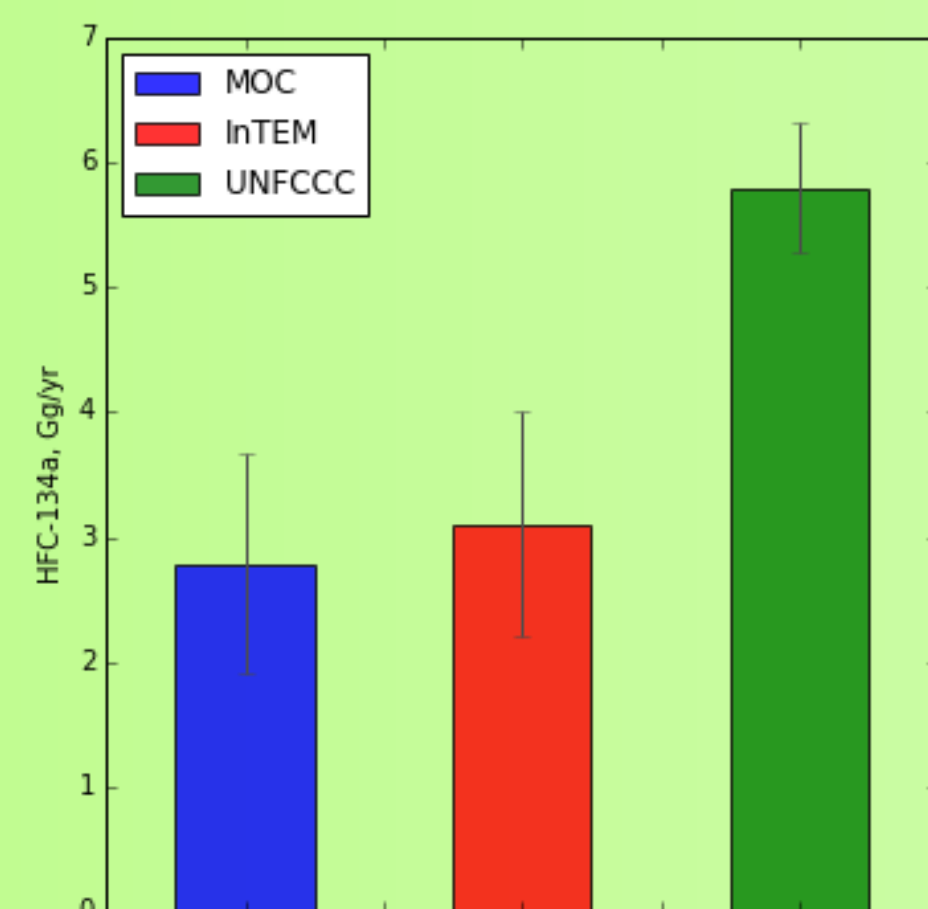


Figure 4: UK 2014 HFC-134a flux estimate comparison. In blue, the result of the current study using MOC, in red, output from the Met Office' inverse modelling system, InTEM (using UK surface observations), and in green, an extrapolated estimate taking from the national inventory report and submitted to the UNFCCC (2015 submission).

Conclusions & Further Work

UK HFC emissions have been characterised using 172 whole air samples collected during 2014/15 aboard the FAAM research aircraft. Significant enhancements were observed down wind of the UK for six selected HFCs. We use the MOC method to estimate a UK HFC-134a flux of 2.74 ± 0.45 Gg yr⁻¹ for 2014, which agrees well with a previous top-down estimate but poorly with the national inventory. Further work is planned to combine our observations with an inverse modelling framework², which will allow national emissions to spatially resolved.

1) Sample Collection and Analysis

Samples were collected during 6 flights in the summer of 2014 and a single East coast transect in May 2015. Of the 172 samples collected, 88% were obtained below 1.5 km. At low altitudes, influence from long distance transport events is considered negligible¹.

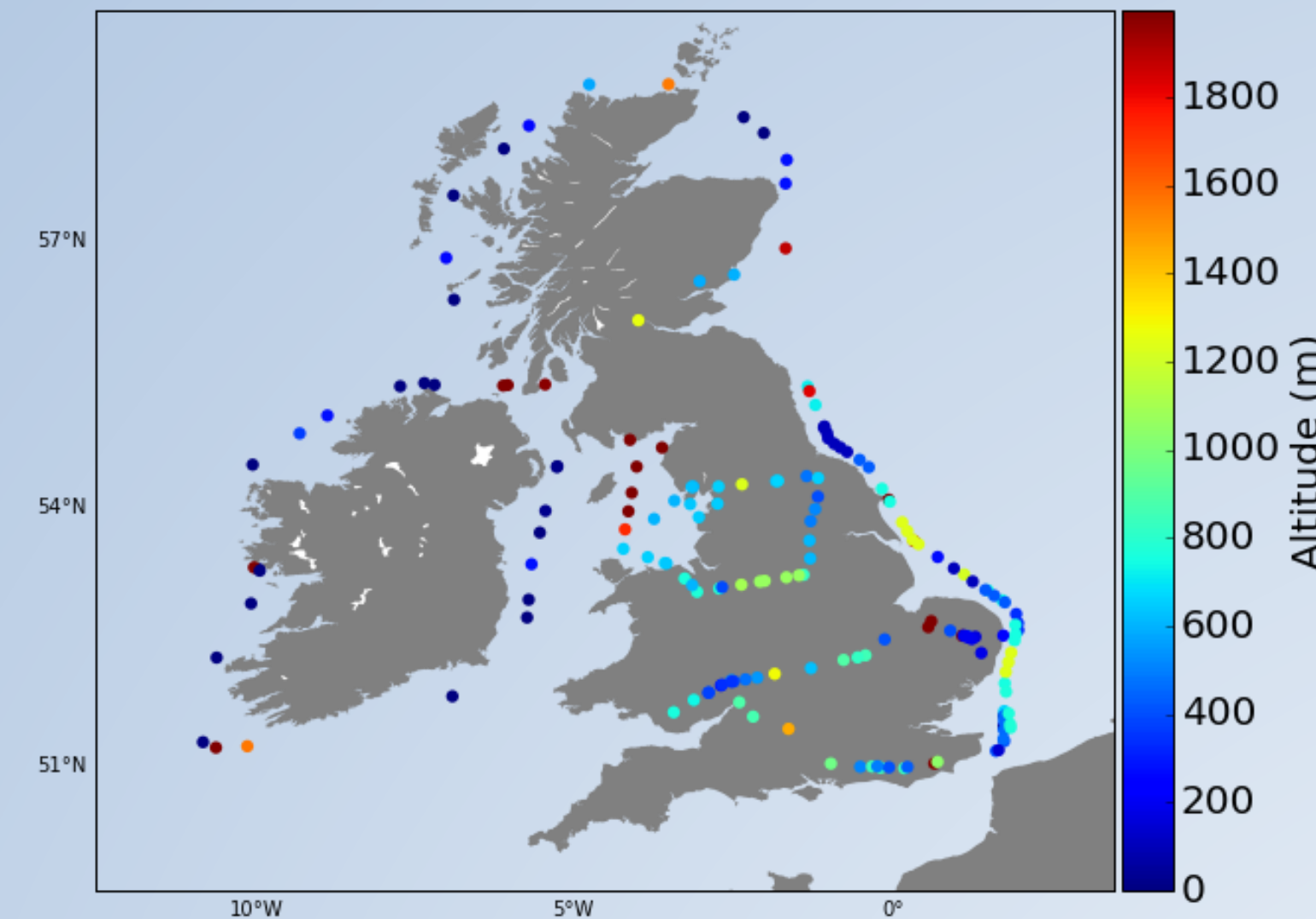


Figure 1: Geographic distribution of Whole Air Samples (WAS) collected above the UK and Ireland. Sample collection points are colour coded by altitude.

Flight criteria

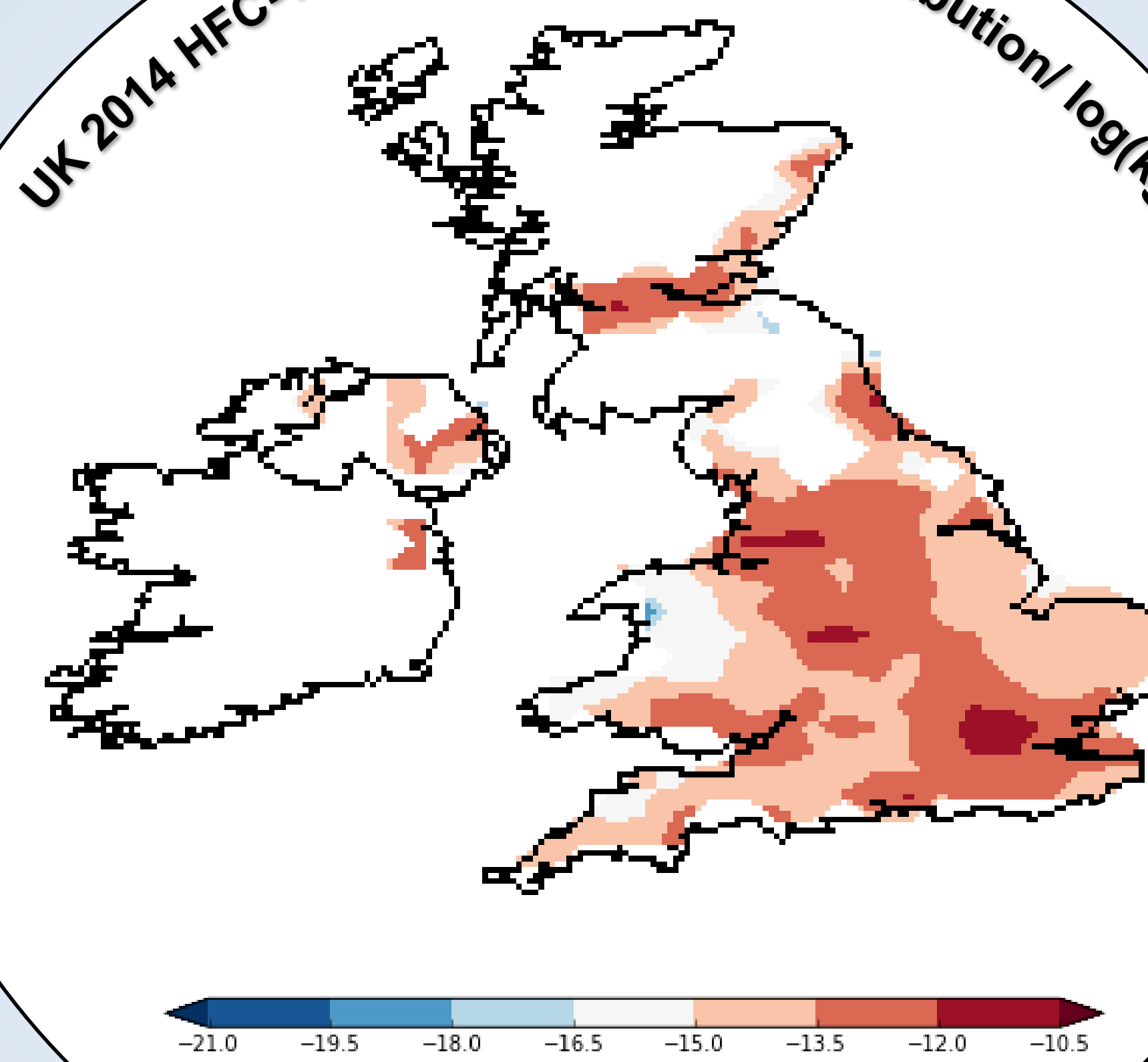
- Westerly/South Westerly wind direction; minimal continental influence
- >10 knot wind speed, providing efficient transport of pollutants
- Minimal cloud cover

Using the NAME (Numerical Atmospheric Modelling Environment) particle dispersion model, we select samples representative of clean inflow and take an average to obtain background values for each HFC in each measurement window.

Significant mole fraction enhancements, above background values, are observed for all 6 HFCs:

- 85% of HFC-134a samples above the inflow average
- Comparable fractions for HFC-125 (88%) and HFC-152a (86%)
- Smaller fractions for HFC-143a (59%), HFC-32 (65%) and HFC-227ea (63%)

UK 2014 HFC-134a scaled emissions distribution/ log(kg m⁻² s)



We infer UK HFC emissions for 2014 using the Model – Observation Correlation (MOC) method. Each HFC observation (mol mol⁻¹) is compared to model output, combining NAME with the spatial emissions inventory EDGAR (v42, 2010) to generate a modelled HFC enhancement at each sample point.

- We geographically aggregate the a priori emissions, and apply a scaling factor obtained from the line of best fit (gradient) to infer the UK flux (Gg yr⁻¹)
- The intercept of this line can be used to estimate the background atmospheric abundance of the HFC in question

Figure 3: The MOC method. HFC-134a observations (mol mol⁻¹) are compared to model output to generate a scatter plot. The red line indicates the line of best fit (R = 0.65, Intercept = 8.58 E-11), the blue line indicates a gradient of 1

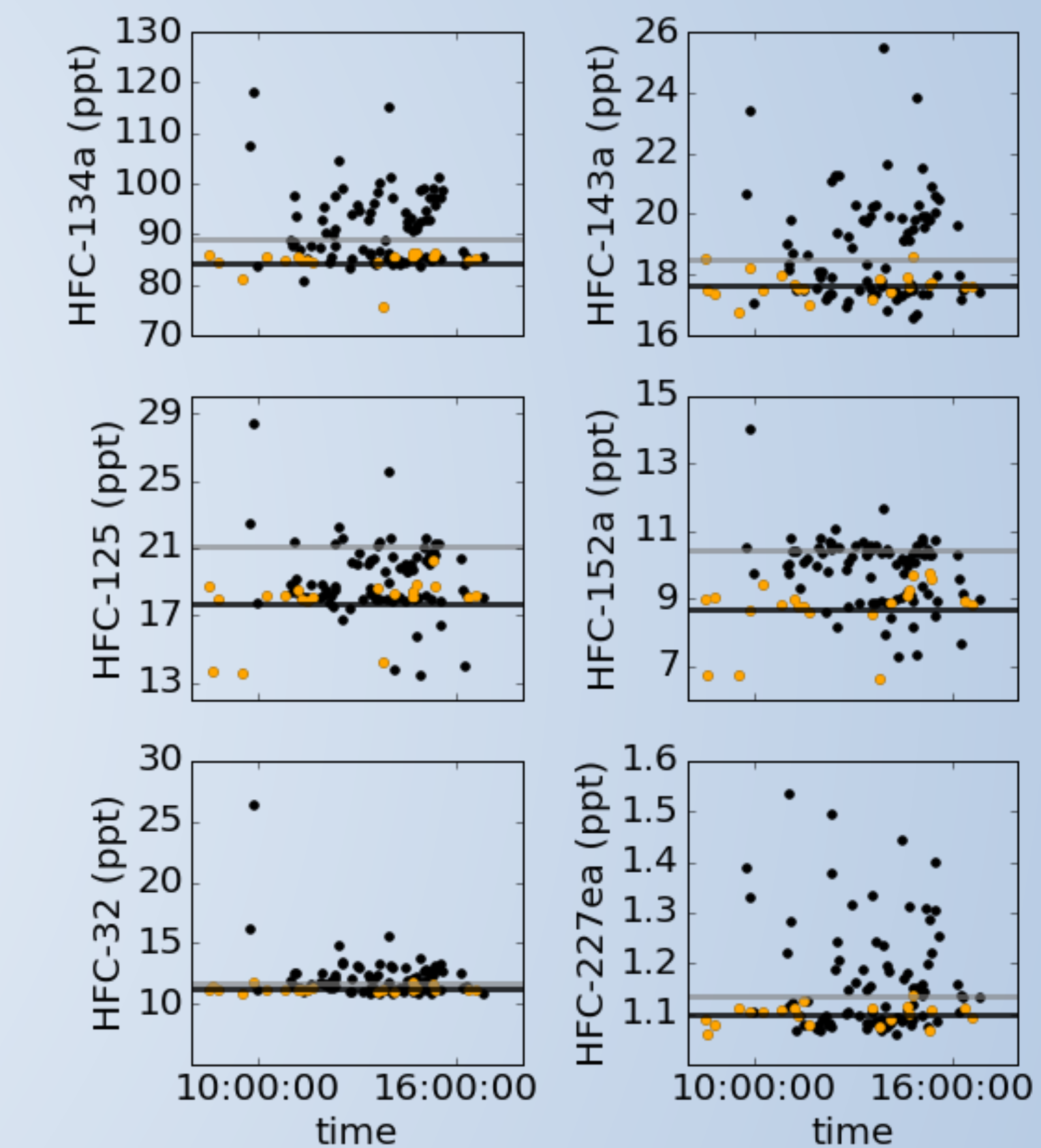
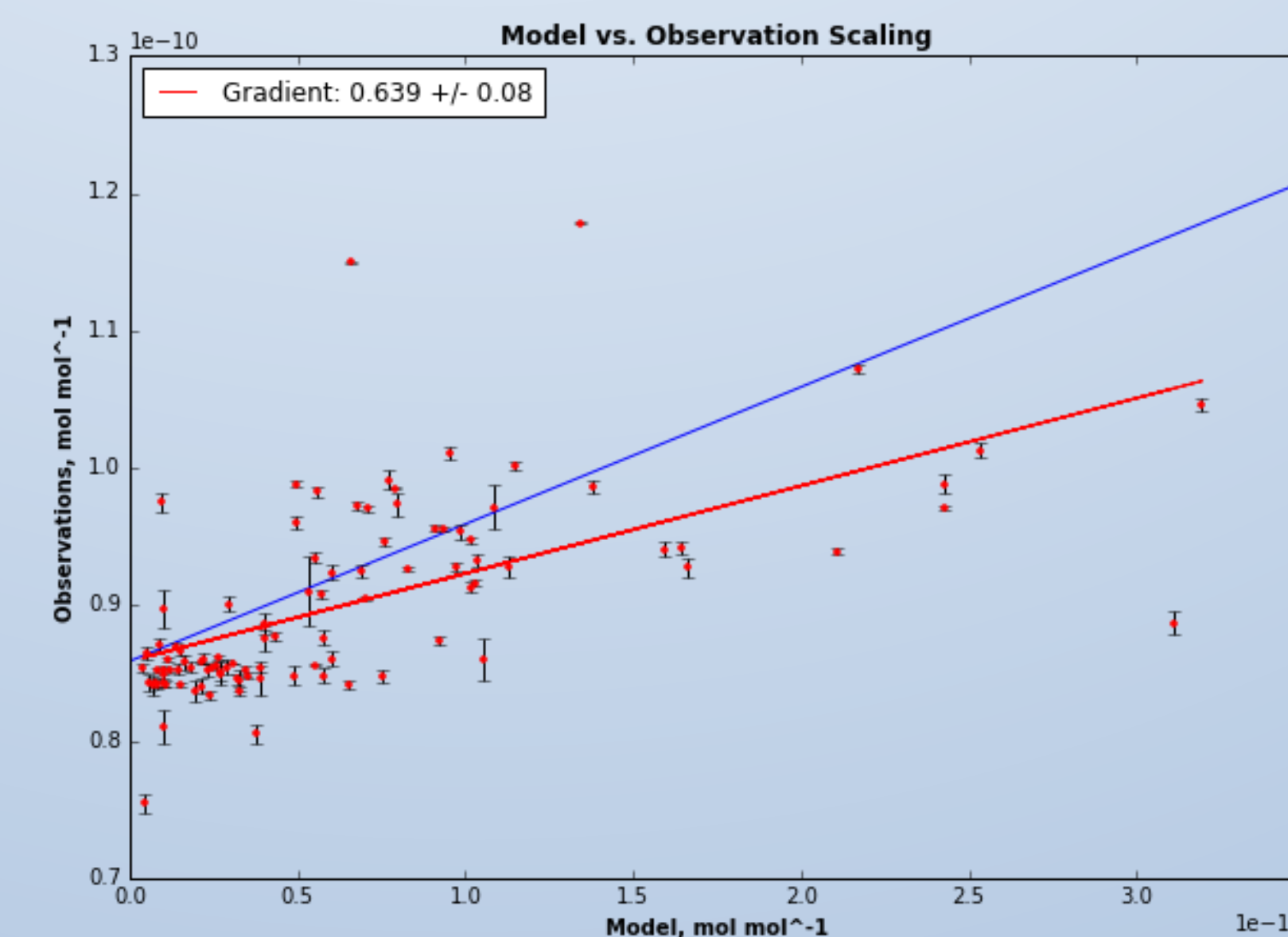


Figure 2: Mixing ratios of six HFCs collected as whole air samples above the UK during 4 consecutive days in Sept 2014. Yellow circles represent unpolluted inflow. Black circles indicate the remaining samples. The black line is the average of the inflow and the grey line is the average plus two standard deviations.

3) UK HFC emission estimates

2) General trends in HFCs observed above the UK

References.

- 1) Millet, D. B., Atlas, E. L., Blake, D. R., Blake, N. J., Diskin, G. S., Holloway, J. S., Hudman, R. C., Meinardi, S., Ryerson, T. B., and Sachse, G. W.: Halocarbon emissions from the United States and Mexico and their global warming potential, *Environ. Sci. Technol.*, 43, 1055–1060, 2009
- 2) Ganesan, A. L., et al. "Characterization of uncertainties in atmospheric trace gas inversions using hierarchical Bayesian methods." *Atmospheric Chemistry and Physics* 14.8 (2014): 3855-3864.

Acknowledgements:

We thank all staff, ground crew and pilots at FAAM for their continued assistance and advice during flight campaigns, and the Met Office ADAQ group for the provision of the NAME model and UM data. Daniel Say is funded by a NERC studentship.