Western European emissions of CFC-11 (and CFC-12) inferred from atmospheric observations and inverse modelling

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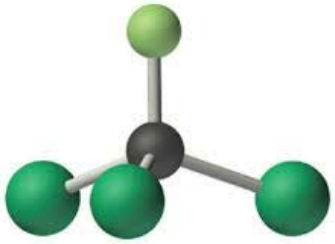
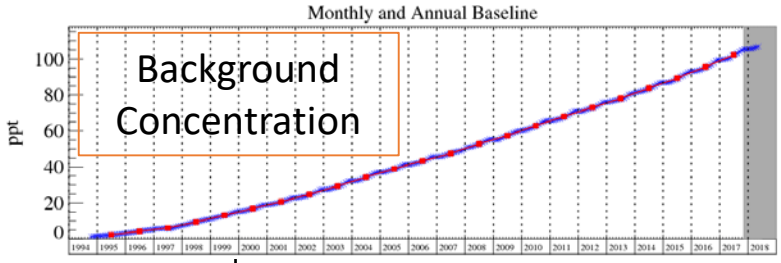
Why CFC-11?

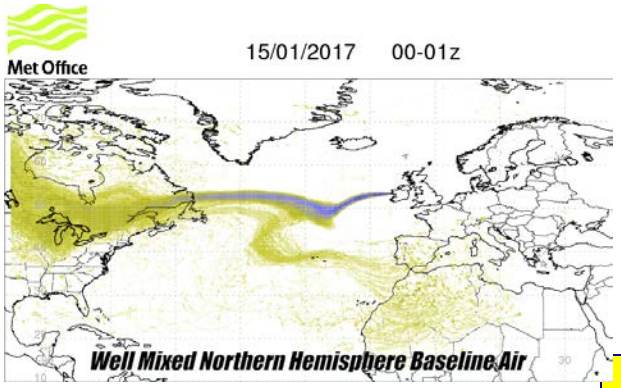


- Slow-down in the global decline of atmospheric CFC-11 from 2013 most likely caused by an increase in global emissions (Montzka et al, 2018);
- East Asia is a likely source of some, or all, of this increase (Montzka et al, 2018; Rigby et al, 2019);
- To close the global budget, we estimated emissions of CFC-11 and CFC-12 over Western Europe (IE, UK, FR, DE, BE, NE, LU, DK, IT, CH, AT, ES, PT) using atmospheric observations from 4 measurement sites of the AGAGE network;
- We compared results from 3 independent atmospheric inversion systems



To understand the recent history of the air arriving at measurement stations

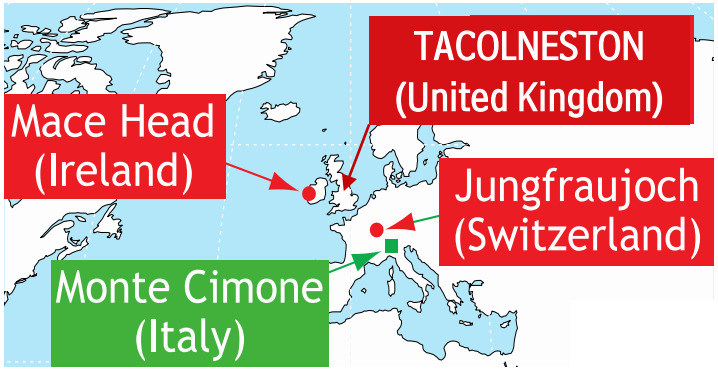
Atmospheric Transport Model



To estimate the surface emissions that best describe the observations

Prior Knowledge

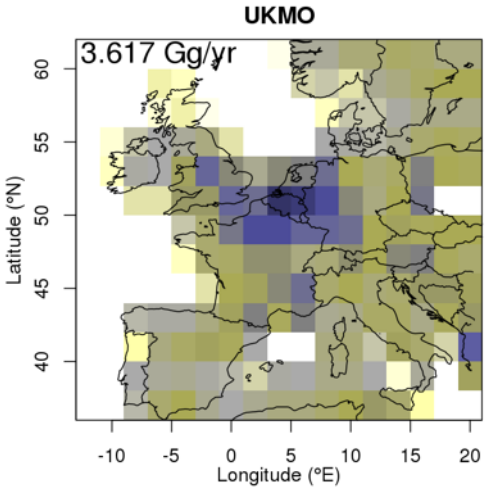
Atmospheric Observations of GHG



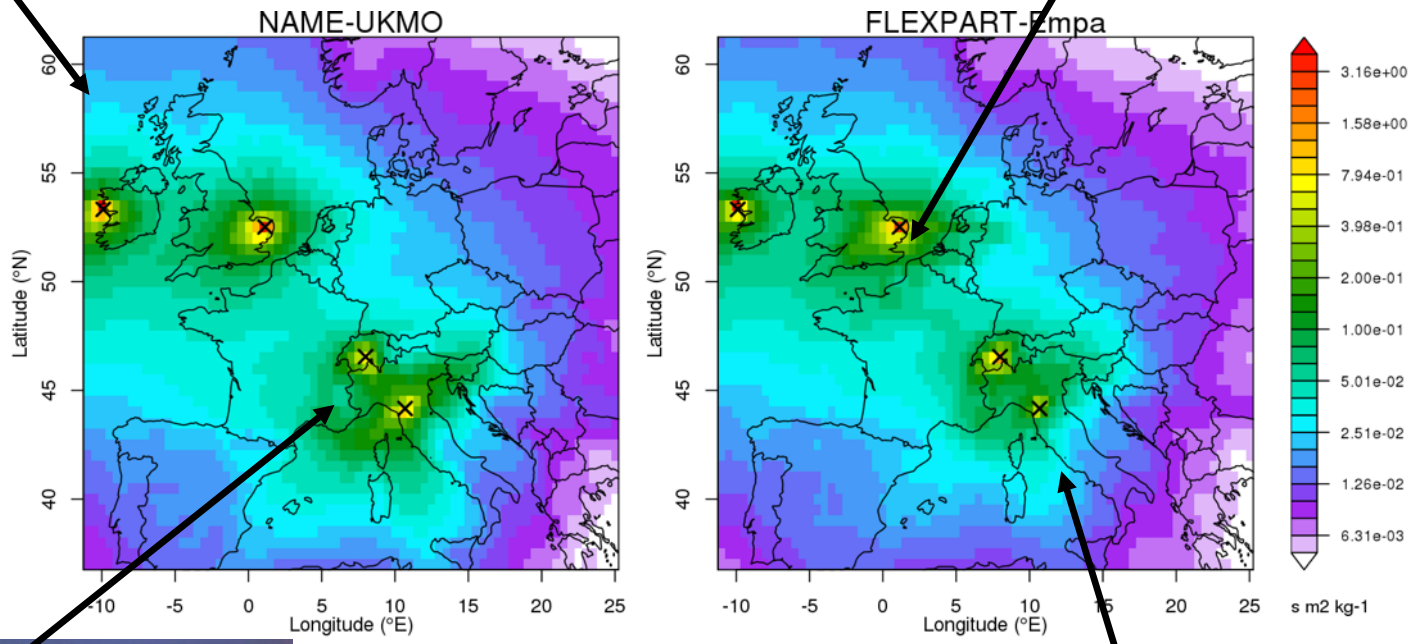
Inversion

Estimate of surface emissions

Uncertainties Estimated



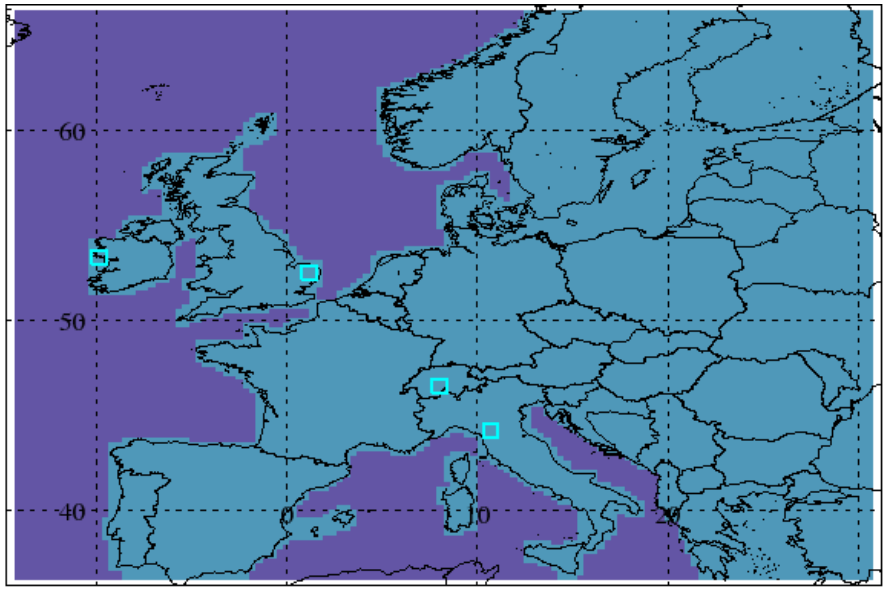
European atmospheric hi-frequency observations



Sensitivity Footprints from NAME and FLEXPART for the 4 atmospheric stations 2012-2017

3 Inverse Modelling Systems

- ECMWF-FLEXPART-Urbino
  - 20d back trajectories; 40,000 parcels; 3hrly
  - 1^{o}x1^{o} meteorology
  - Bayesian inversion
- ECMWF-FLEXPART-Empa
  - 10d back trajectories; 50,000 parcels; 3hrly
  - 0.2^{o}x0.2^{o} nested (Alps), 1^{o}x1^{o} global meteorology
  - Bayesian inversion
- UK-NAME-InTEM
  - 30d back trajectories; 40,000 parcels; 2hrly
  - 0.1^{o}x0.1^{o} nested (UK), 40-12km global meteorology
  - Bayesian inversion

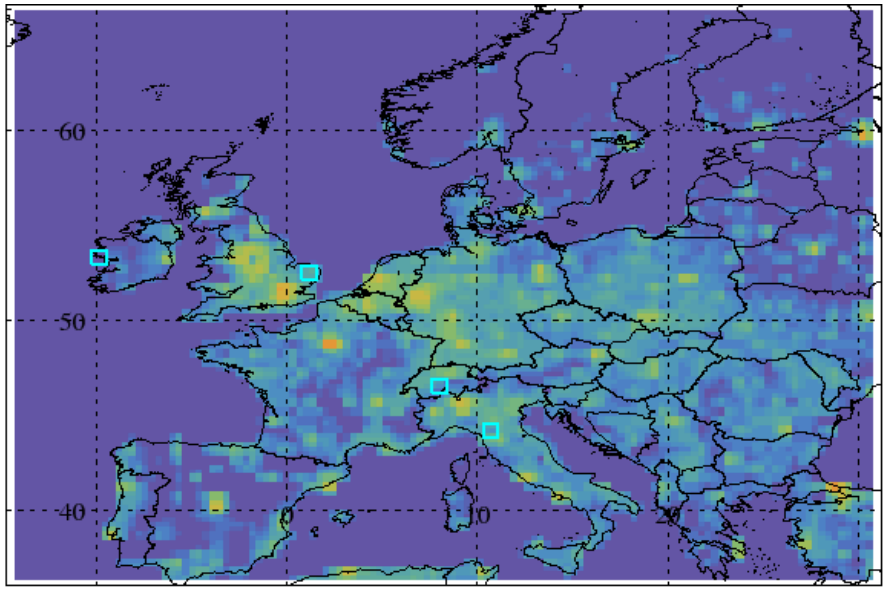


Maximum value = 2.62e-11 g/m²/s

0.00e+00 1.78e-11 5.63e-11 1.78e-10 5.63e-10

Uniform Land Prior

Two priors tested

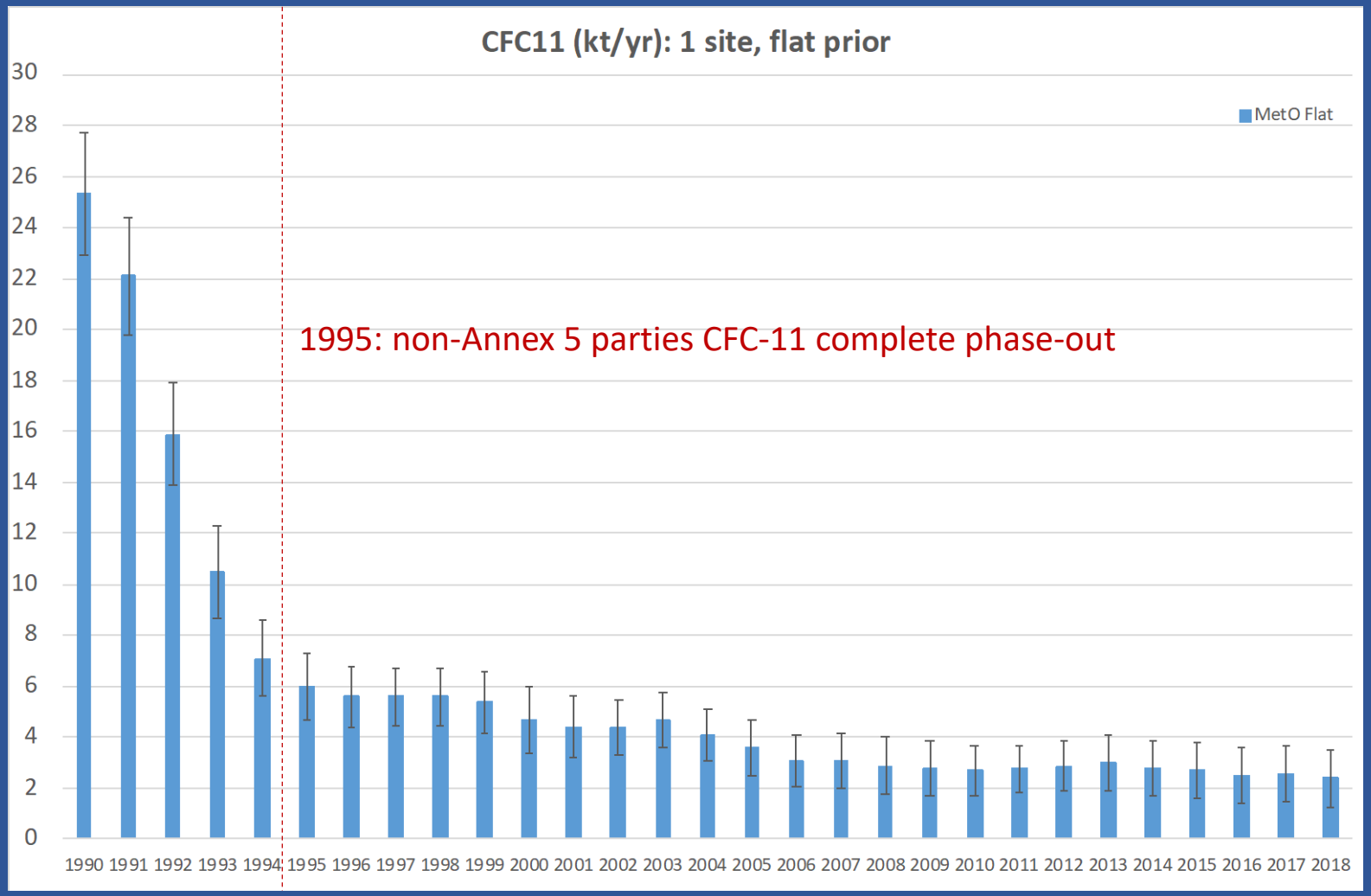


Maximum value = 9.45e-10 g/m²/s

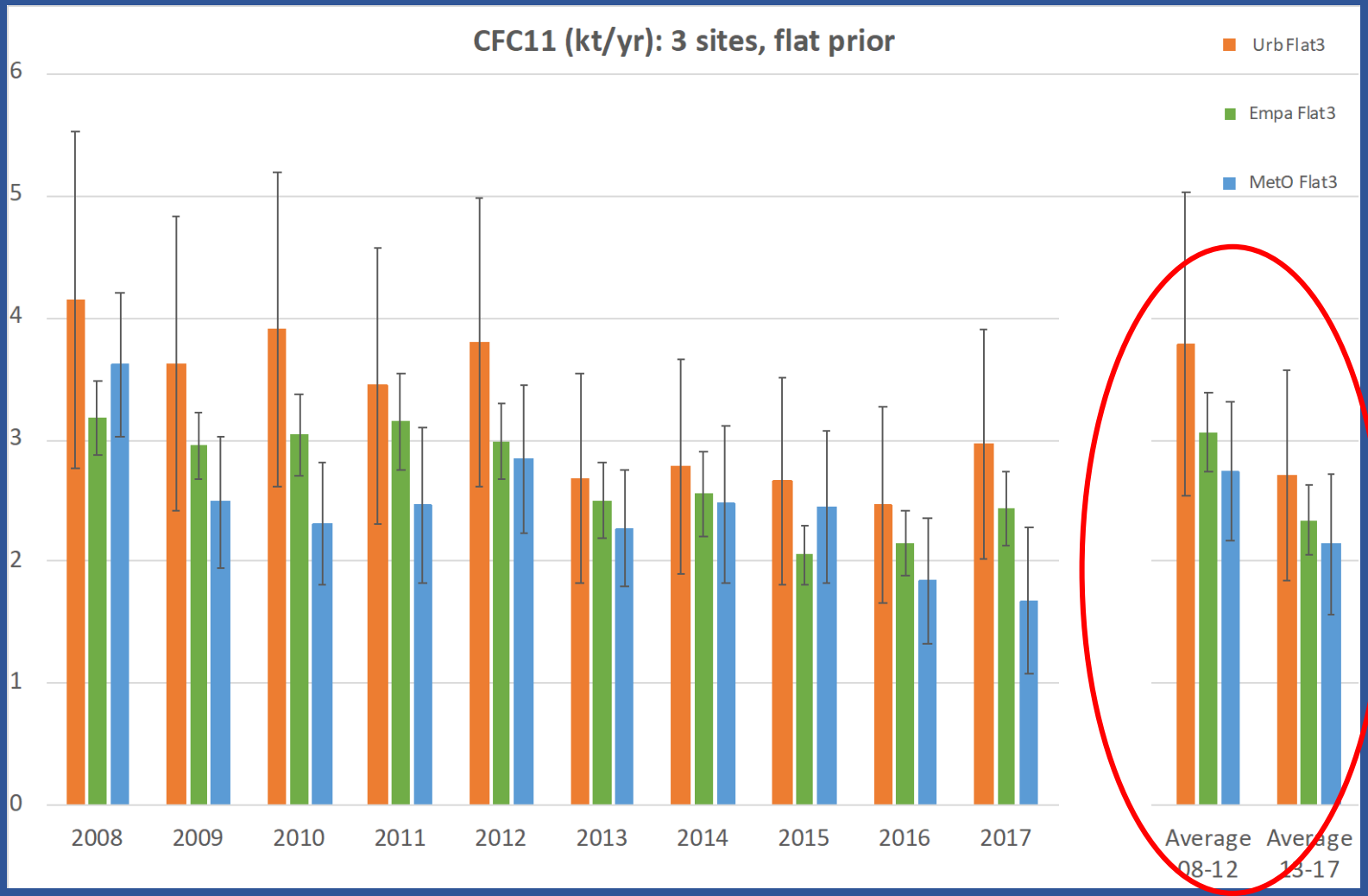
0.00e+00 1.78e-11 5.63e-11 1.78e-10 5.63e-10

Population Weighted Prior

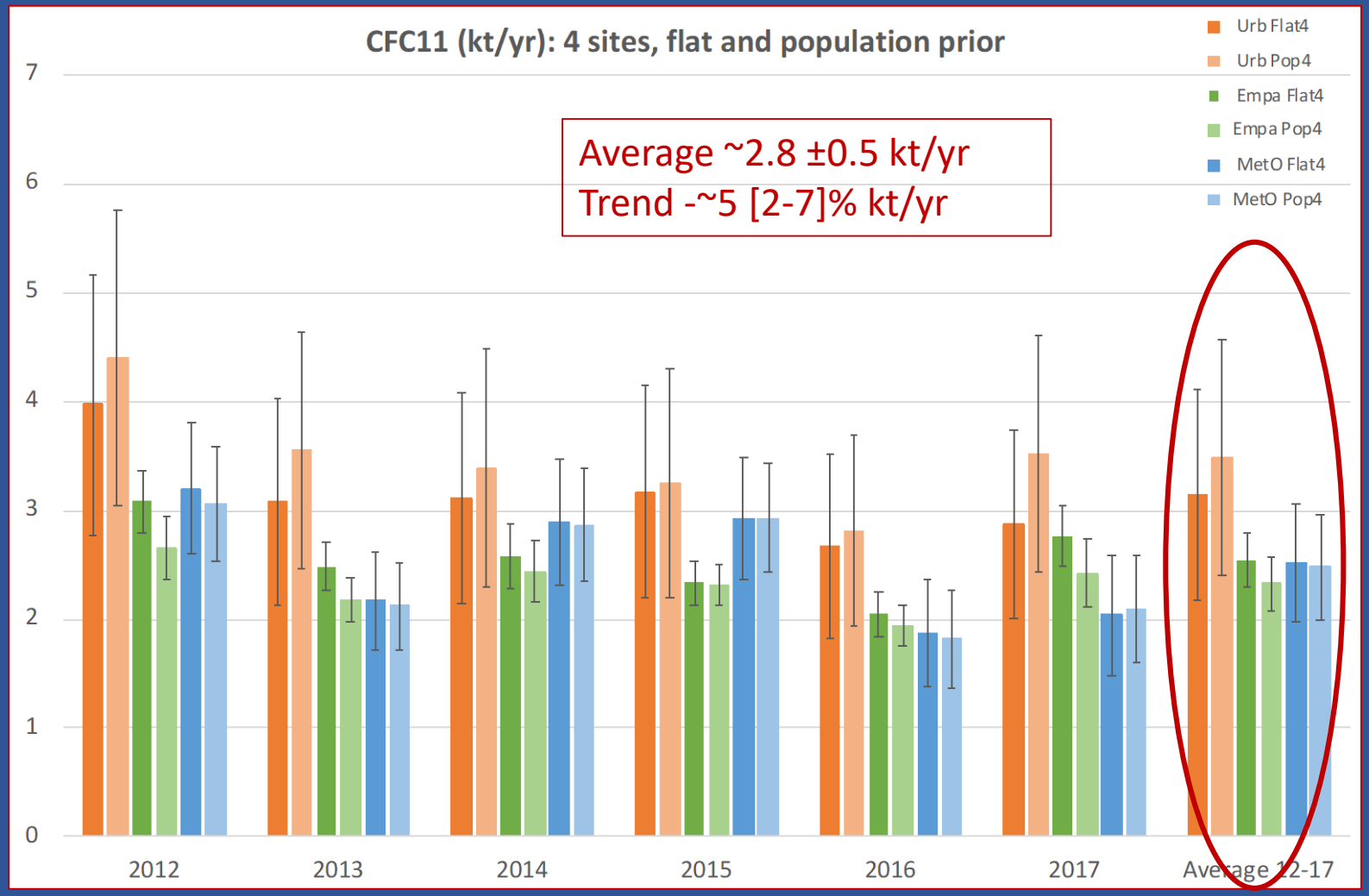
CFC-11 emissions from North Western Europe
1 observation site: MHD (UK-NAME-InTEM)



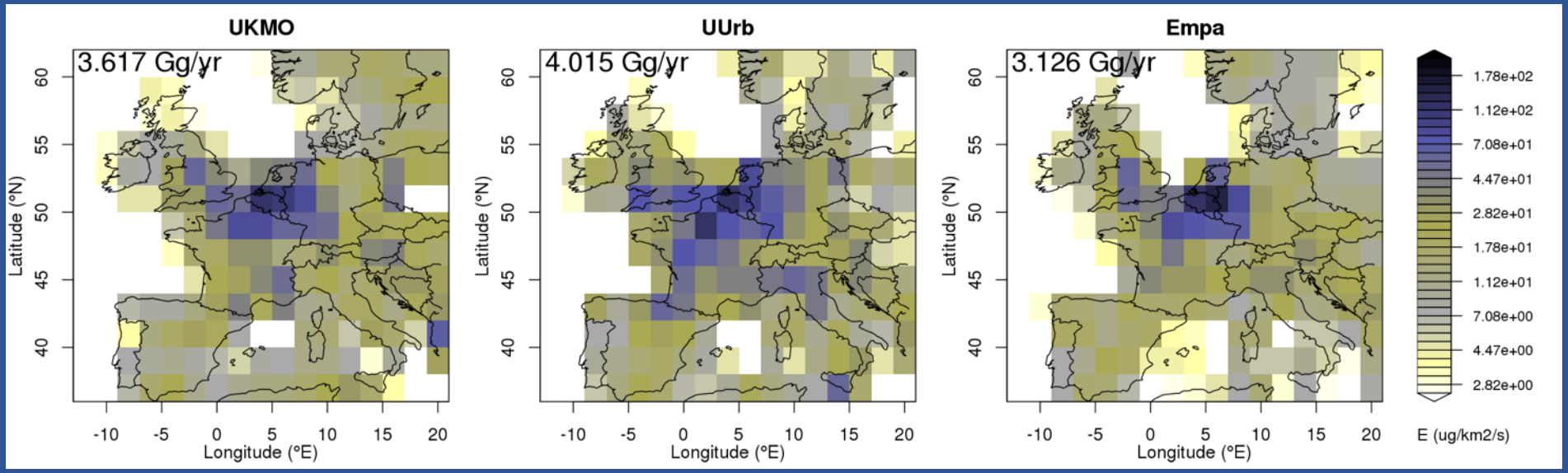
CFC-11 emissions from Western Europe
3 observation sites: MHD, JFJ, CMN (3 models)



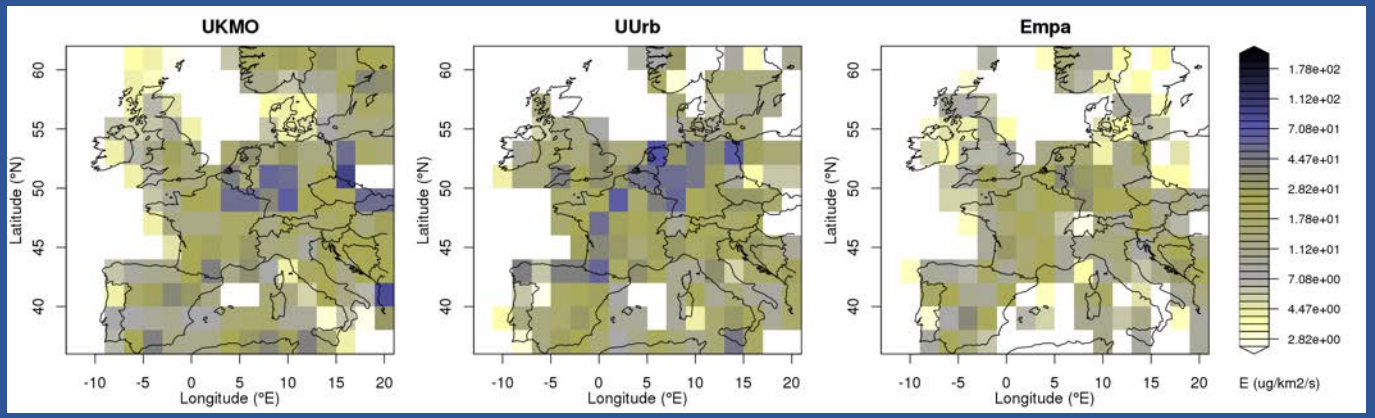
CFC-11 emissions from Western Europe
4 observation sites: MHD, JFJ, CMN, TAC (3 models)



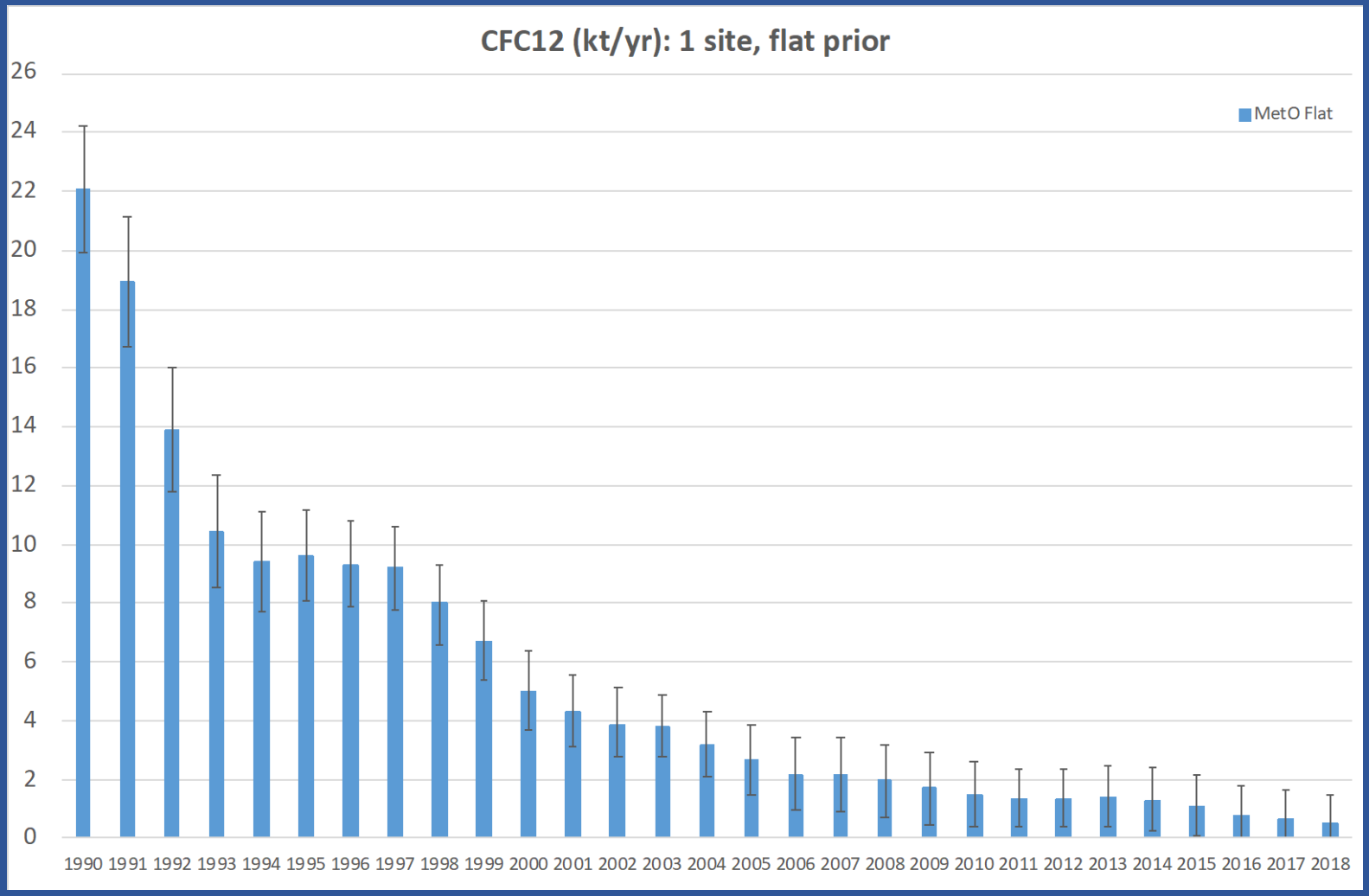
CFC-11 emissions over Western Europe
Geographical Distributions



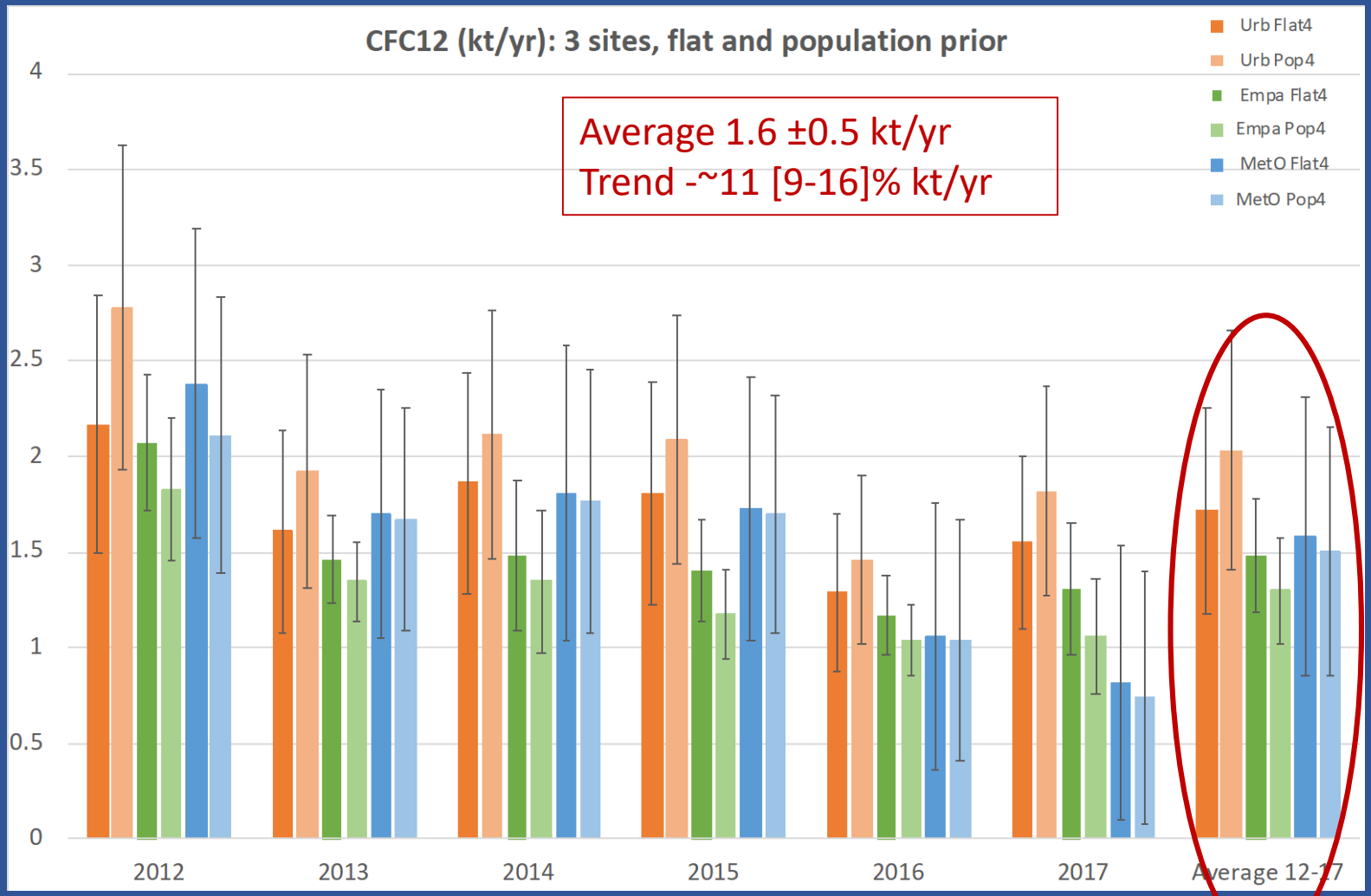
Uncertainties



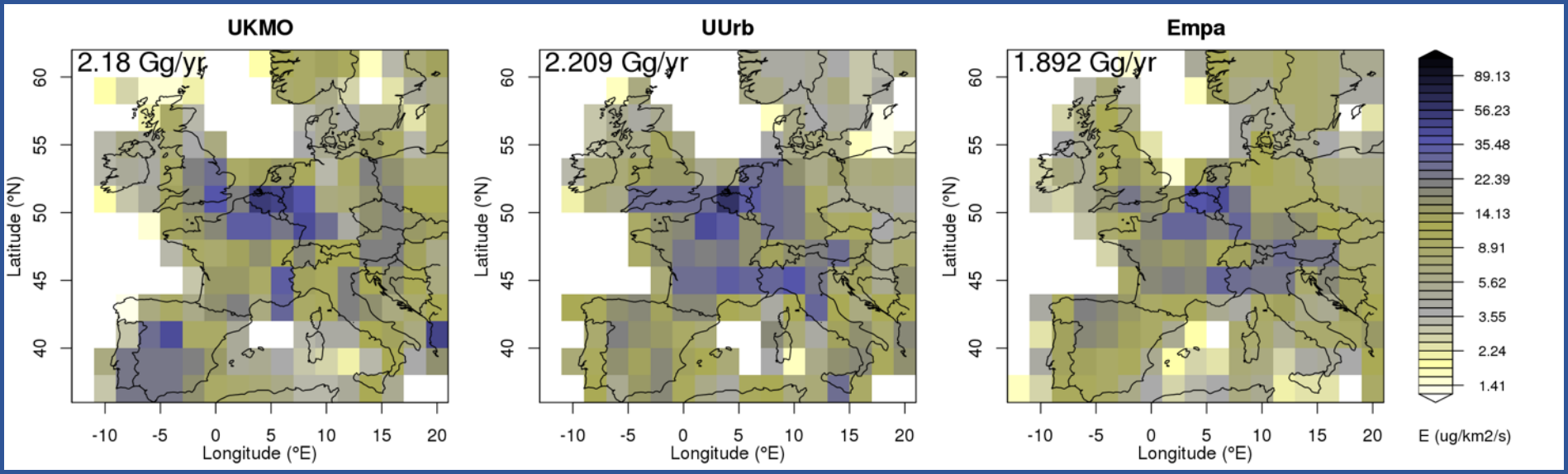
CFC-12 emissions from North-Western Europe
1 observation site: MHD (UK-NAME-InTEM)



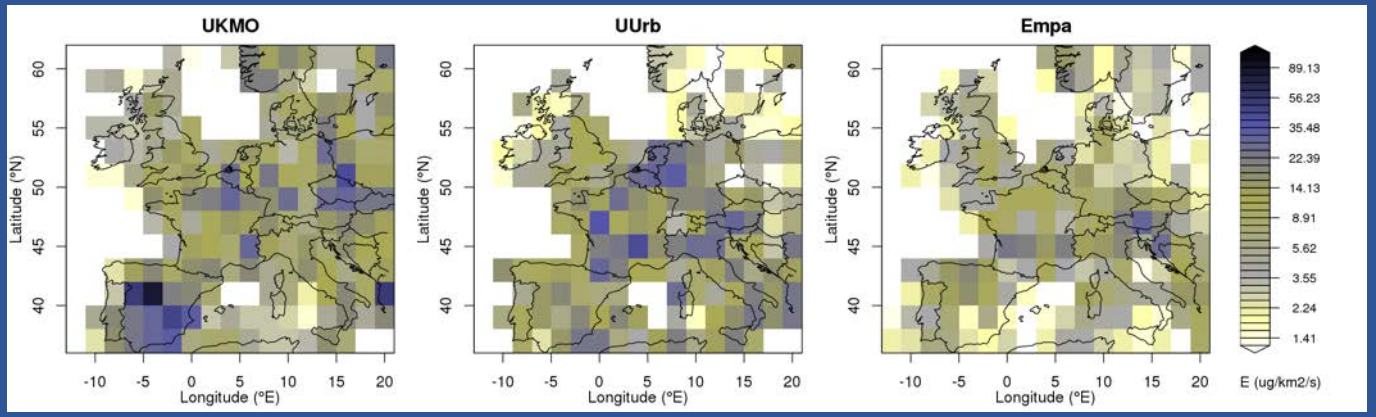
CFC-12 emissions from Western Europe
3 observation sites: MHD, JFJ, TAC (3 models)



CFC-12 emissions over Western Europe
Geographical Distributions



Uncertainties



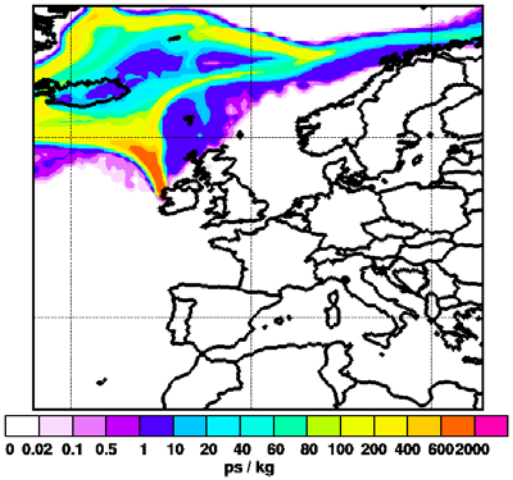
Summary

- 3 Inverse Modelling Systems used using two independent underpinning 3D meteorology (Met Office and ECMWF);
- Sharp decline in emissions from Western Europe in 1990s;
- CFC-11 emissions for Western Europe 2012-17 → 2.8 ± 0.5 kt/yr avg corresponding to less than 4% of global emissions;
- Avg decline 2012-17 → ~0.15 kt/yr (~5 [2-7]%/yr);
- Violation of the MP not likely, emission rates seem consistent with emissions from banks;
- In Europe the strongest CFC-11 source regions is BENELUX
  - By- product of HCFC-22 production?
  - Higher intensity of polyurethane (CFC-11) foam production and use in Benelux, vs higher use of extruded polystyrene (CFC-12) in Southern Europe?

Thank you!

Extra slides

FLEXINVERT (Uni Urbino)



- FLEXPART is a Lagrangian particle dispersion model (Stohl et al., 1998);

Model setting:

- SRR (Source Receptor Relationship) obtained from FLEXPART 20 d backward calculations;
- ECMWF data 1° x 1° resolution;
- 40.000 particles released every 3 h.

- The "SRR Source receptor relationship" value in a particular grid cell is proportional to the particle residence time in that cell and measures the simulated mixing ratio at the receptor that a source of unit strength in the cell would produce.
- Multiplying the SRR with an emission flux taken by an appropriate emission field gives the simulated mixing ratio at the receptors to be compared with the measurements
- The FLEXPART output is ingested by the inversion algorithm based on the analytical inversion method by Stohl et al. (2009);
- Minimization of cost function measure the misfit between model and observations and the measure the difference from a priori values.

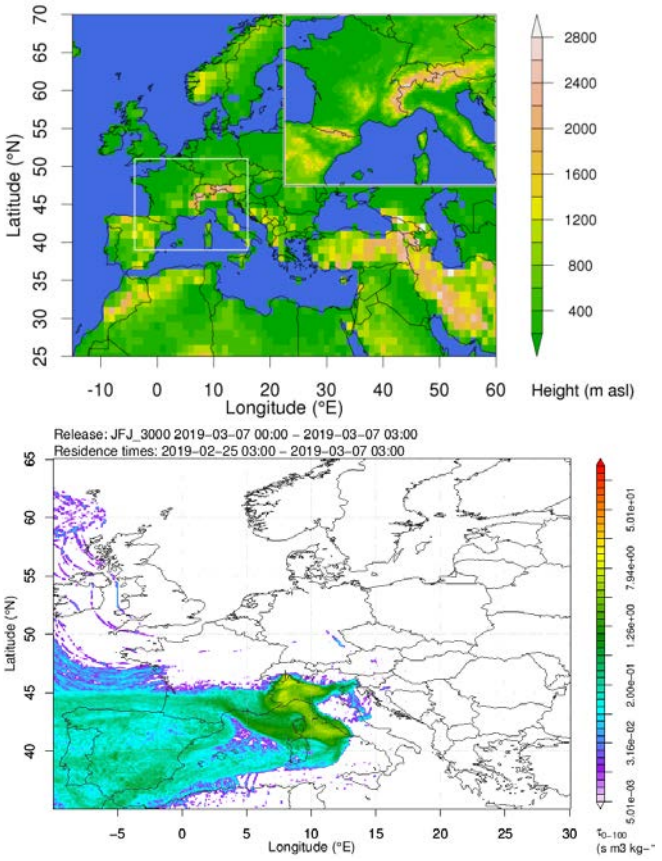
FLEXPART-Empa Inversion System

Transport

- FLEXPART-ECMWF (V9.2)
  - 0.2°x0.2° nest, 1°x1° global
  - Backward simulations for individual sites
  - 3-hourly releases of 50’000 particles per site
  - 10 day backward

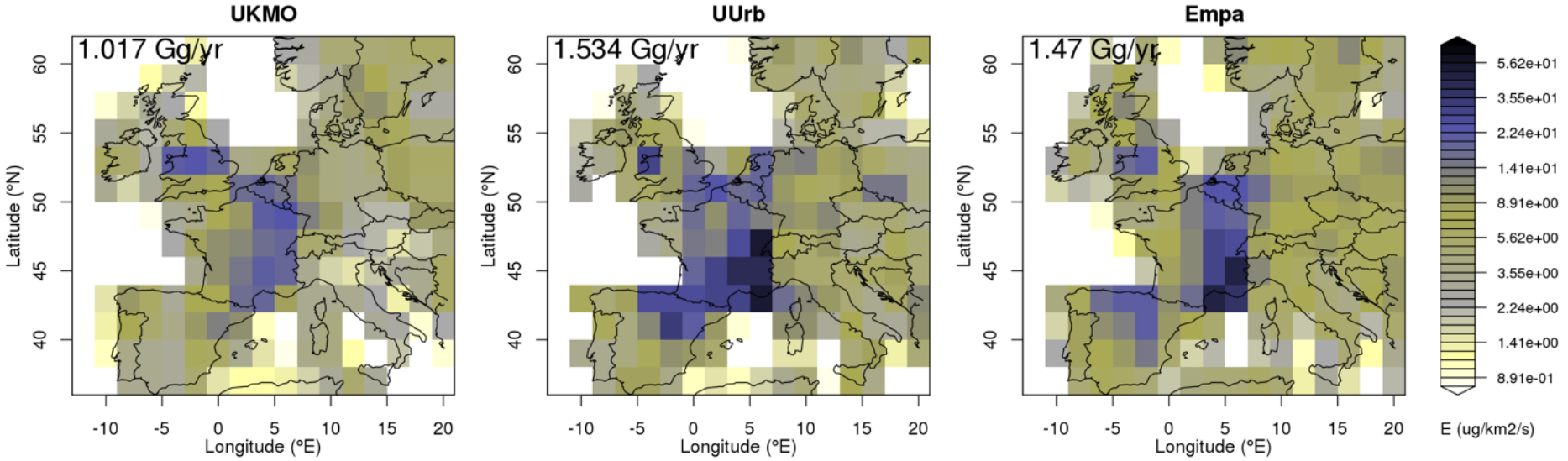
Inversion

- Bayesian inversion (Stohl et al. 2009, Henne et al. 2016)
  - Reduced inversion grid
  - Baseline for each site part of state vector
  - Positive solution enforced by iterative adjustment of a priori uncertainty
  - Spatio-temporal correlations considered in covariance matrices

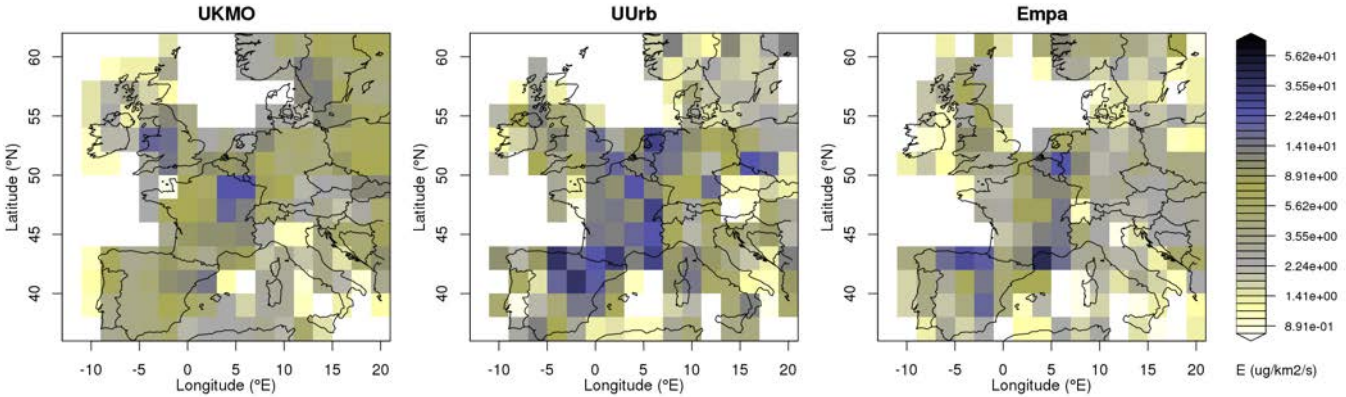


source sensitivity: 1 site, 1 time

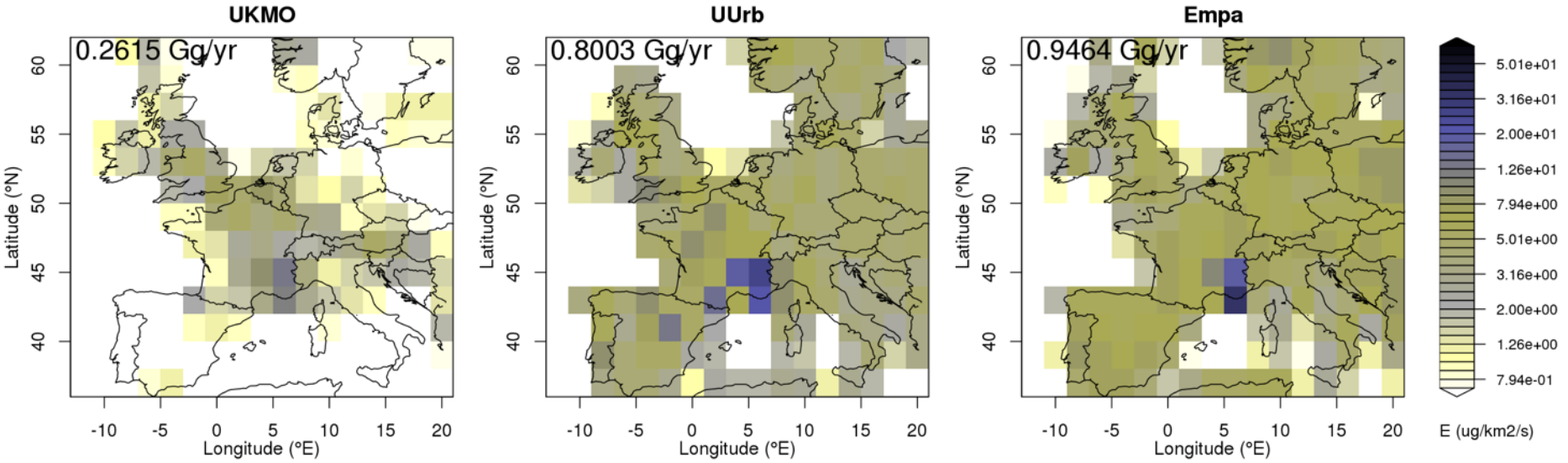
CCl_4 emissions over Western Europe
Geographical Distributions



Uncertainties



CH_3CCl_3 emissions over Western Europe
Geographical Distributions



Uncertainties