European Emissions of Chlorodifluoromethane (HCFC-22) Based on High Frequency Atmospheric Measurements and a Bayesian Inversion Method

Michela Maione

F. Graziosi, J. Arduini, F. Furlani, U. Giostra, S. Montzka, S. O'Doherty, A. Stohl, P. Bonasoni.

NOAA ESRL Global Monitoring Annual Conference, Boulder 20-21 May 2014

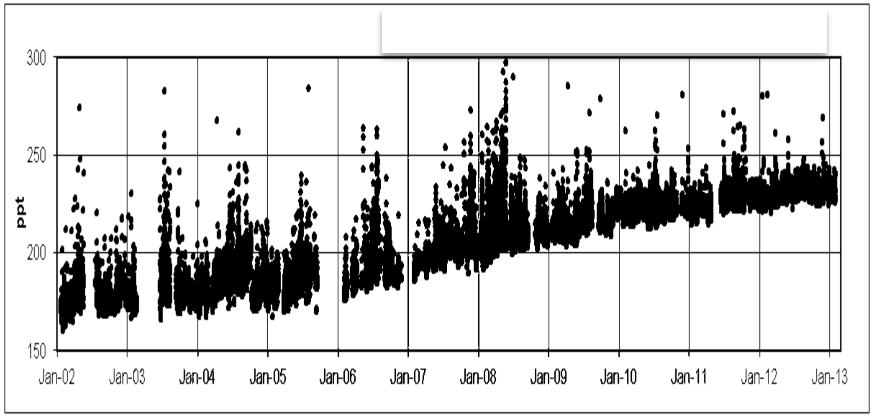
HCFC-22 Fact-sheet

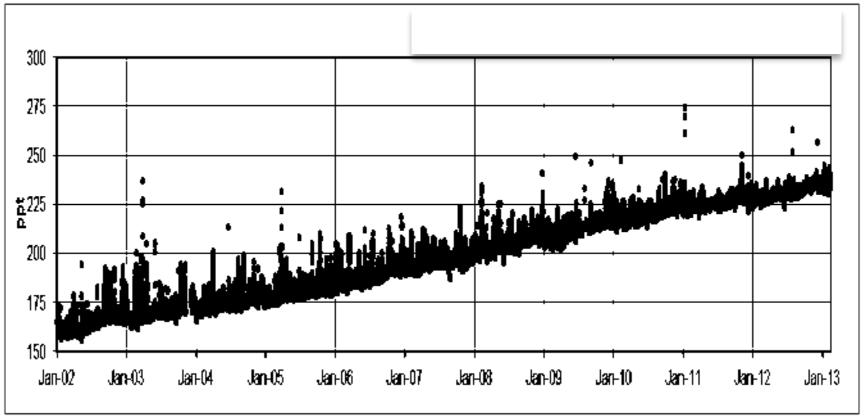
- ODP, *0.055*, GWP over 100 yr, *1790*, Lifetime, ca. *11.9 y*; third most abundant man made halocarbon;
- Interim replacement for many CFCs in refrigeration and air conditioning;
- Accelerated phase out schedule decided during the 19th Meeting of the Parties to the Montreal Protocol in 2007 but non-dispersive use (e.g., feedstock in fluoropolymer manufacture) not controlled;
- In Europe, starting from Jan 2010, only recycled HFCFs can be used for the service and maintenance of equipment;
- Currently, dominant emission for HCFC-22 is from banks contained in-use applications;
- Wide range of release times:
  - short (83% in the first year and the remainder in the second year)
  - medium (30% in the first year, the remainder in the next 10 years)
  - long (2% per year)

Aim of this study

- To provide estimates on the emissions of HCFC-22 on a European scale, down to the country level;
- To analyse the trend of such emissions over an 11 years period (2002 –2012), during which the accelerated restrictions on HCFCs production and consumption decided in 2007 at the 19th Meeting of the Parties to the Montreal Protocol have entered into force.

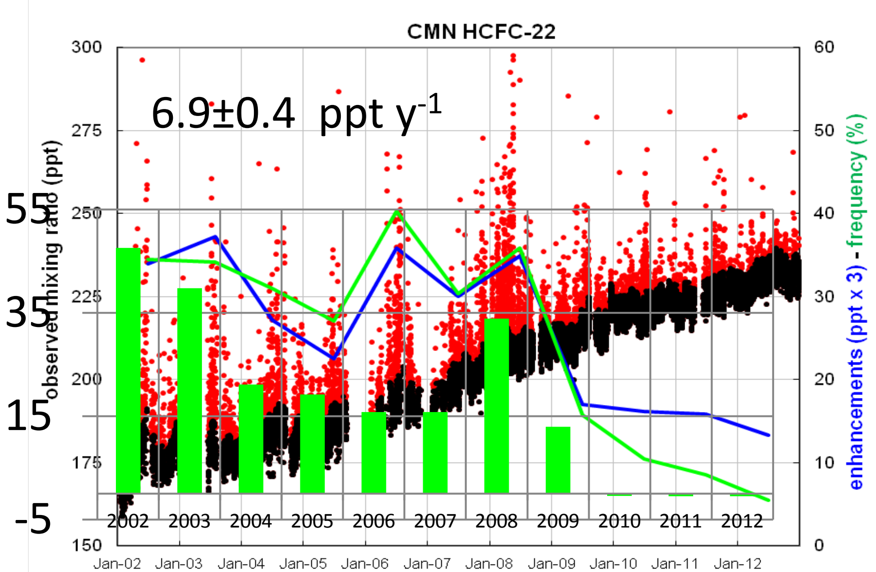
Time series

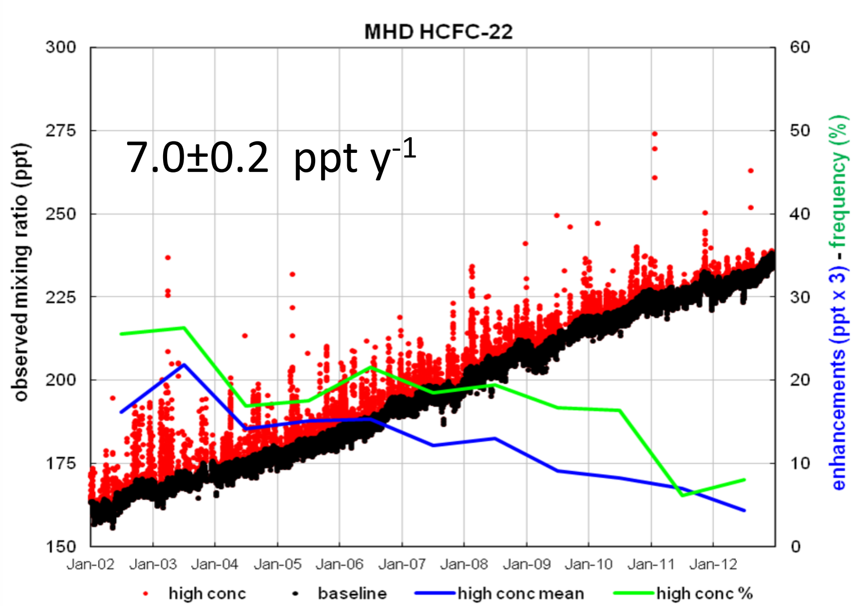




AGAGE

HCFC-22 trends





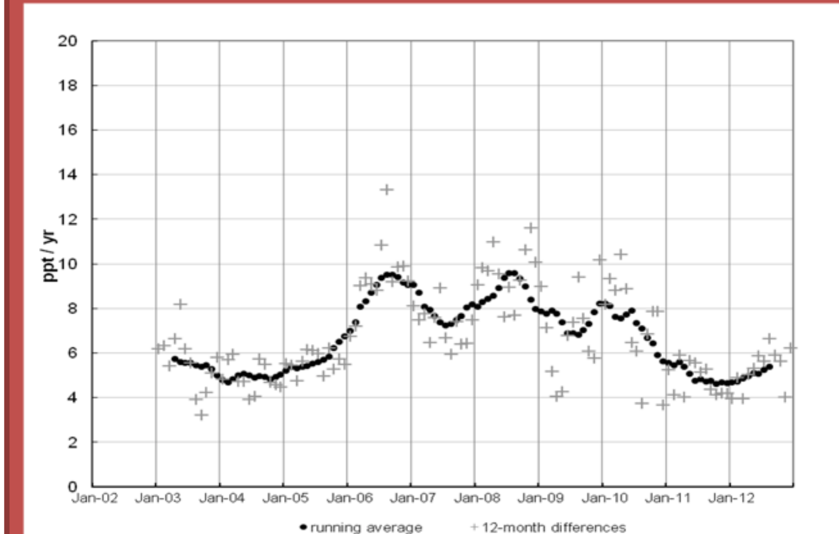
HCFC-22 time series at CMN and MHD.

Black dots: baseline, red dots: enhancements above the baseline;

Green line: Frequency (in %) of occurrence of enhanced values above the baseline;

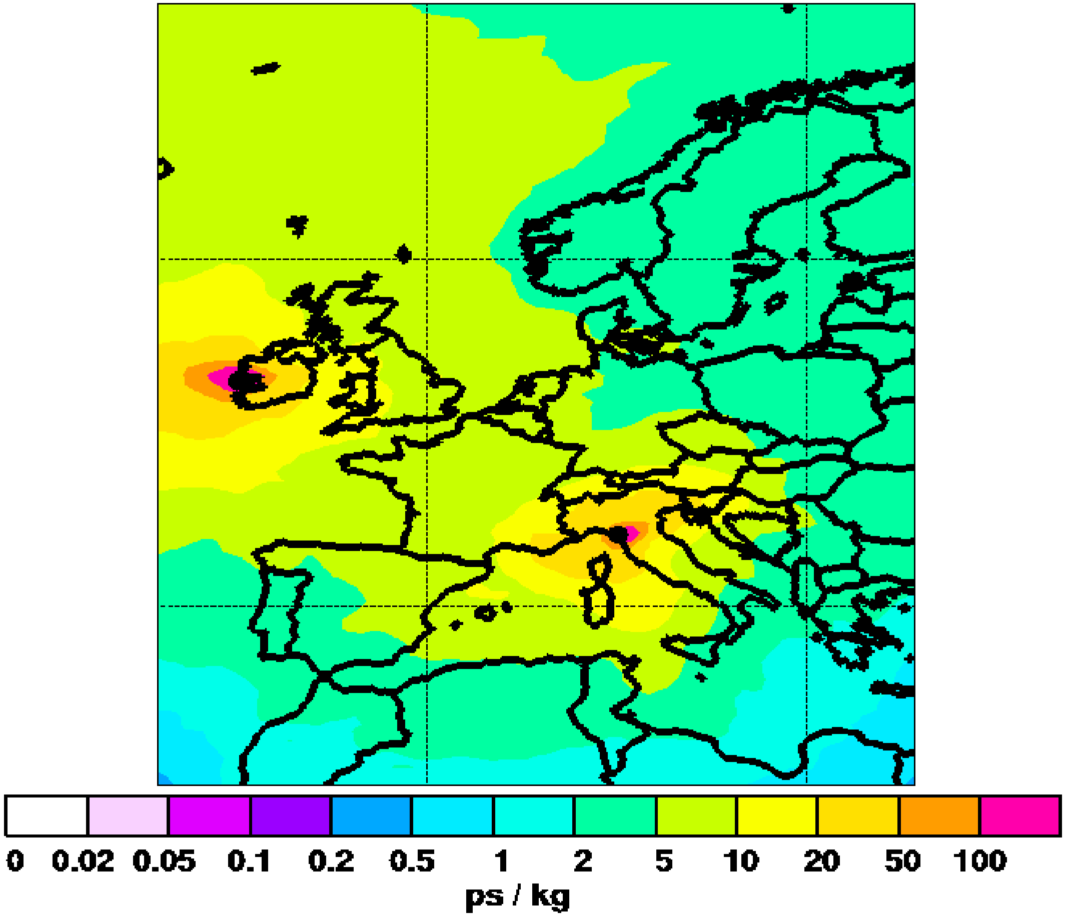
Blue line: enhancements' average annual intensity, in ppt*3

UNEP EU consumption in Gg y^{-1}



Average monthly growth rates of HCFC-22 from measurements at CMN and MHD, derived from smoothed 12-month differences in the mixing ratio means over the previous 12 months.

SRR - Source Receptor Relationship



SRR (Source Receptor Relationship) obtained from FLEXPART 20 d backward calculations averaged over two years (2008-2009)

ECMWF nested data 0.25° x 0.25° resolution

40.000 particles released every 3 h

The FLEXPART output can be ingested directly by the inversion algorithm based on the analytical inversion method by Stohl et al. (2009)

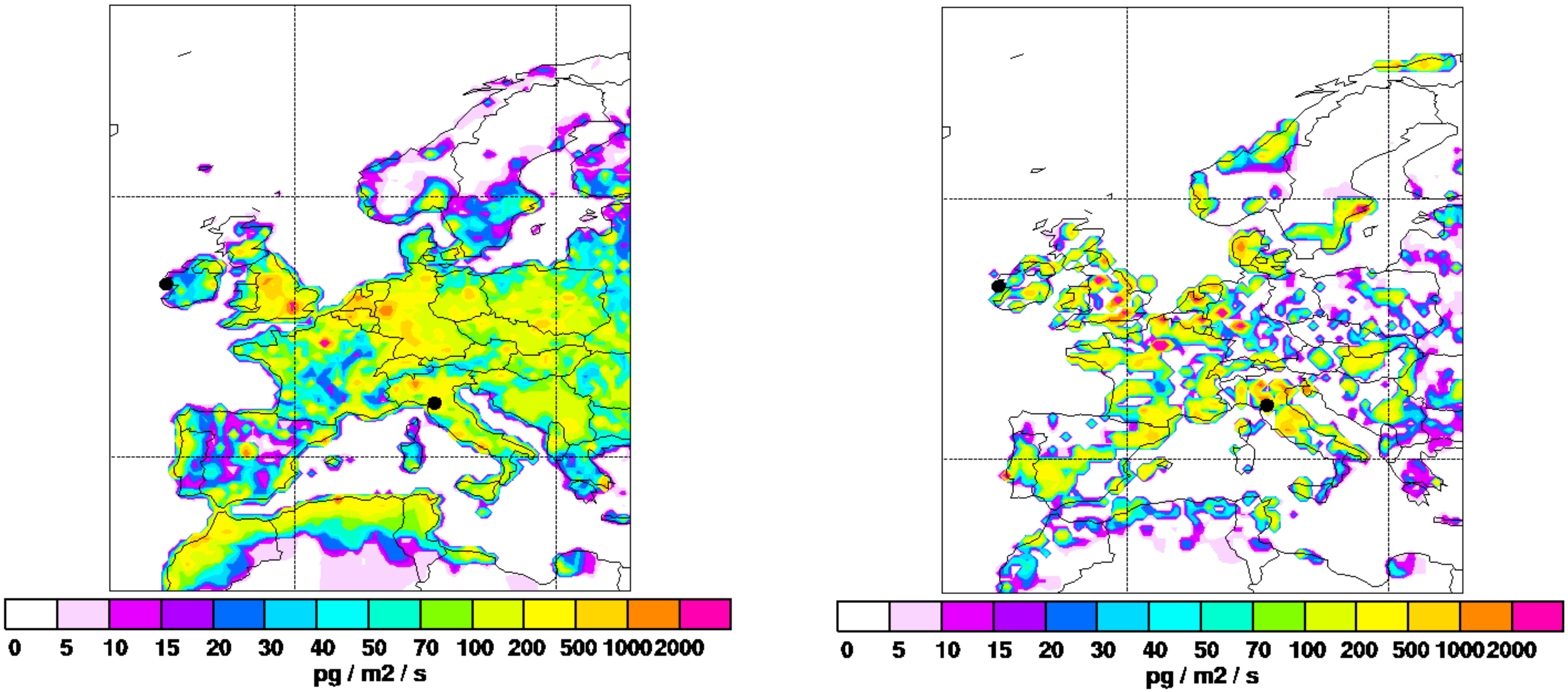
A priori emission field

- 57% of the UNEP aggregate total HCFC consumption (1989 – 2012);
- Consumption from 1982 to 1989 (20-y release) extrapolated from the UNEP 1989-1994 consumption data, assuming an exponential trend fitting data with R^2=0.94;
- Consumption data divided by categories (short, medium and long release) and emissions simulated using the EF as in McCulloch et al. (2006);
- Emissions disaggregated within each country's borders according to a gridded population density data set.

+

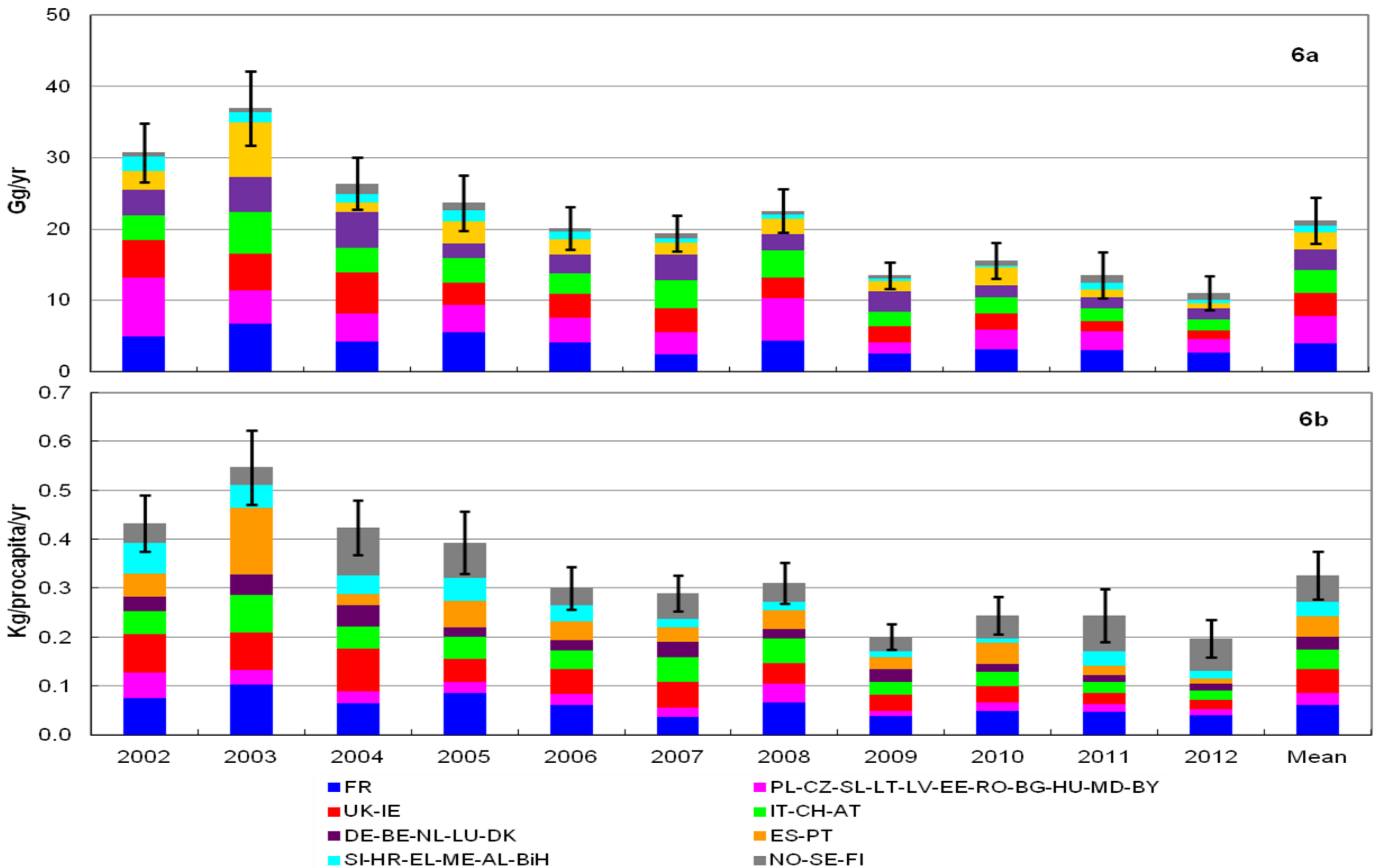
- Emissions reported by the E-PRTR inventory (2007 – 2011), declared by industries during normal operations and accidents (does not consider the emissions from banks);
- 2002-2006 and 2012 emissions extrapolated;
- E-PRTR emissions are only about 4 % of the emissions calculated through UNEP consumption data combined with the McCulloch EF.

Emission estimates

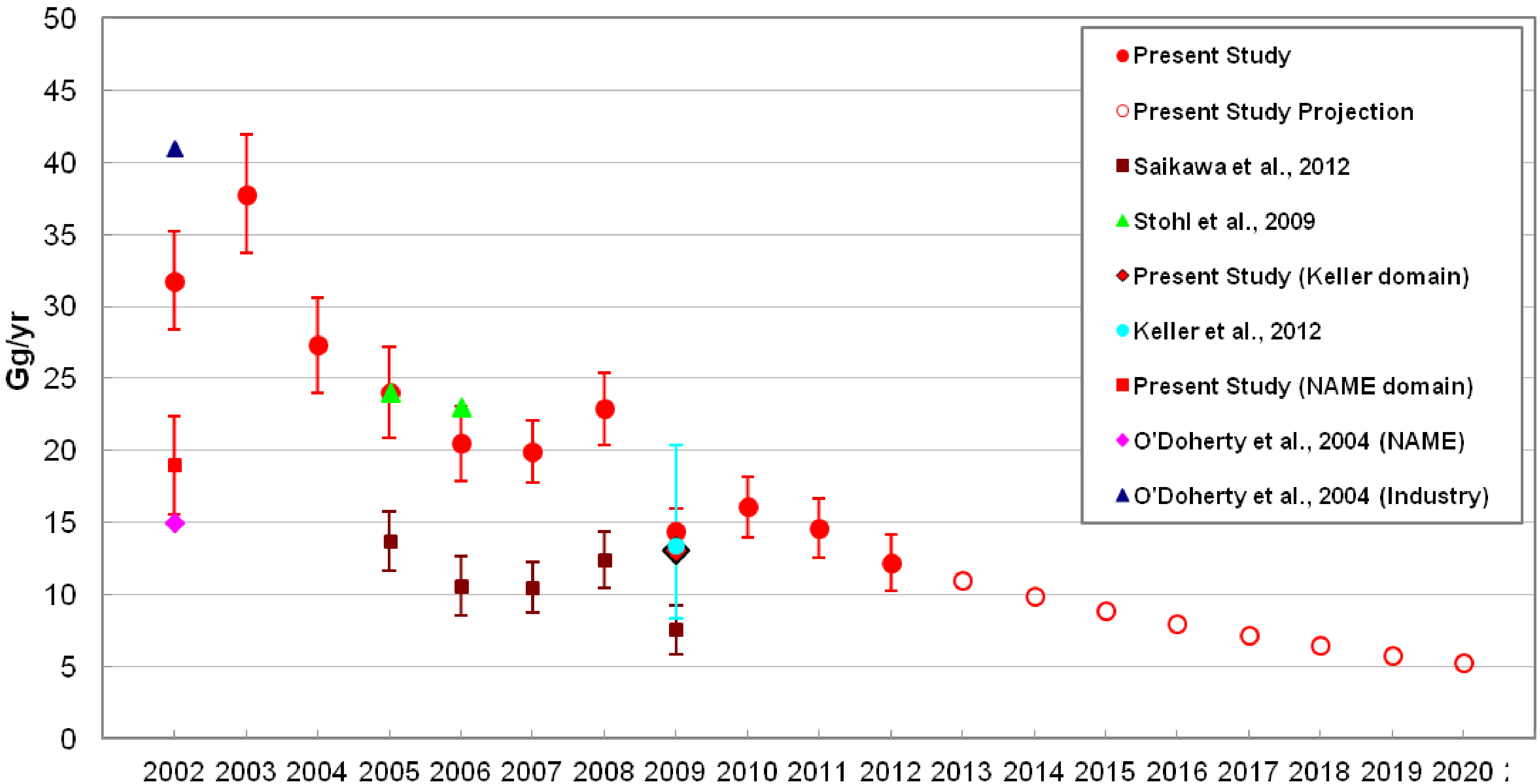


a) *A priori* distribution of HCFC-22 emissions from the European Geographic Domain in 2007; b) *A posteriori* distribution for the same domain and year. Number of grid cells 3400, emission uncertainty 120 %.

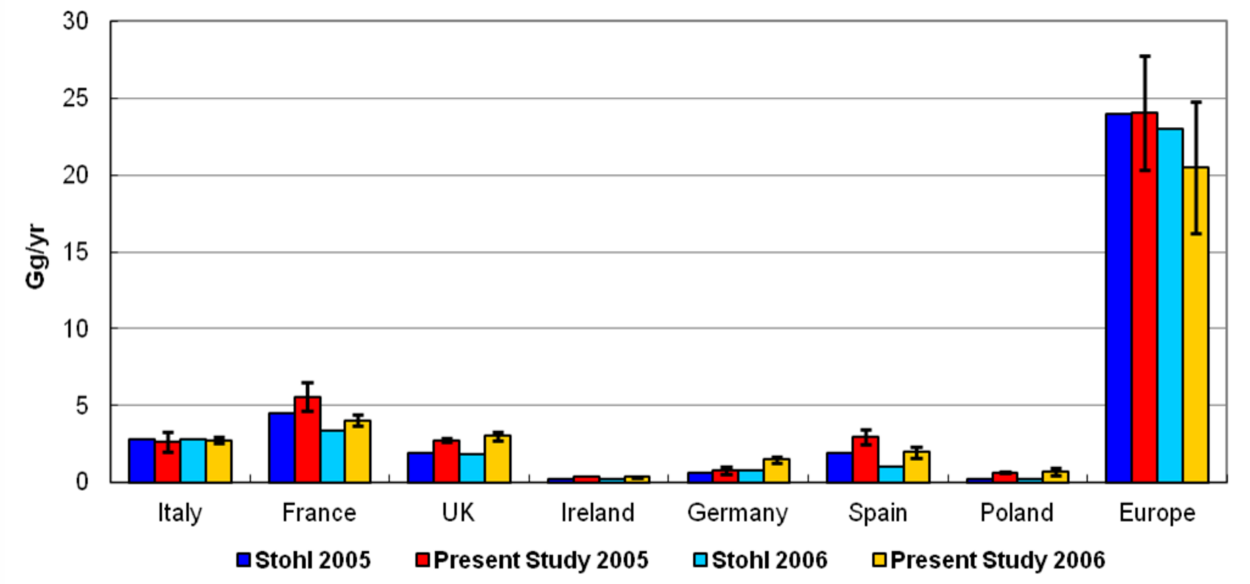
European Country Emissions



Comparison with other top down estimates



Comparison with other studies

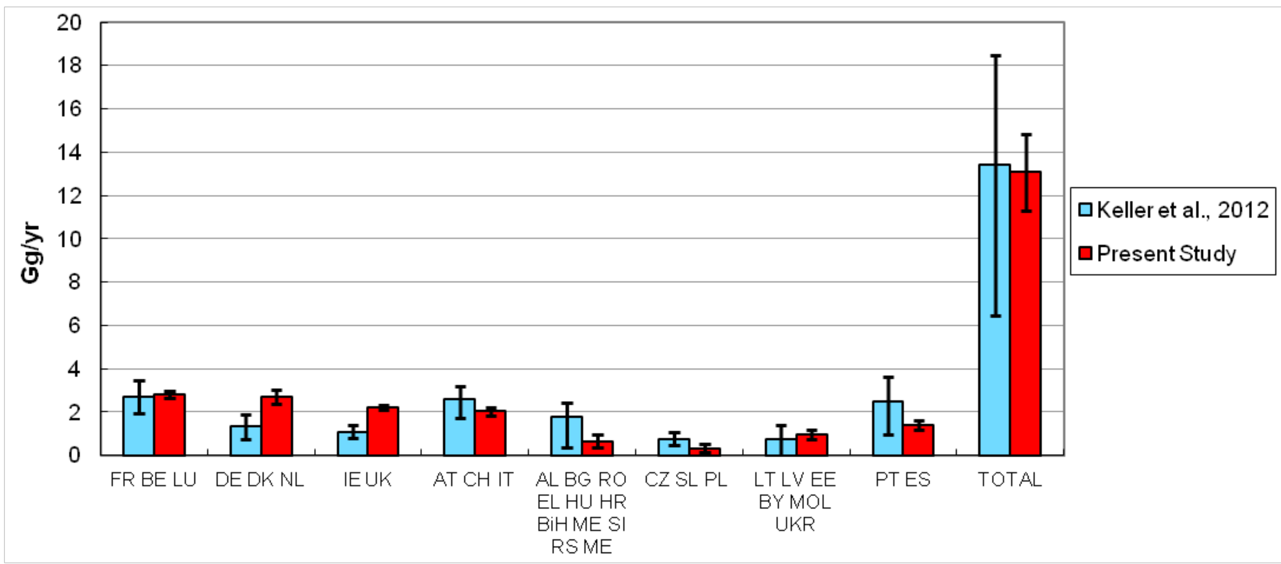


Stohl et al., 2009

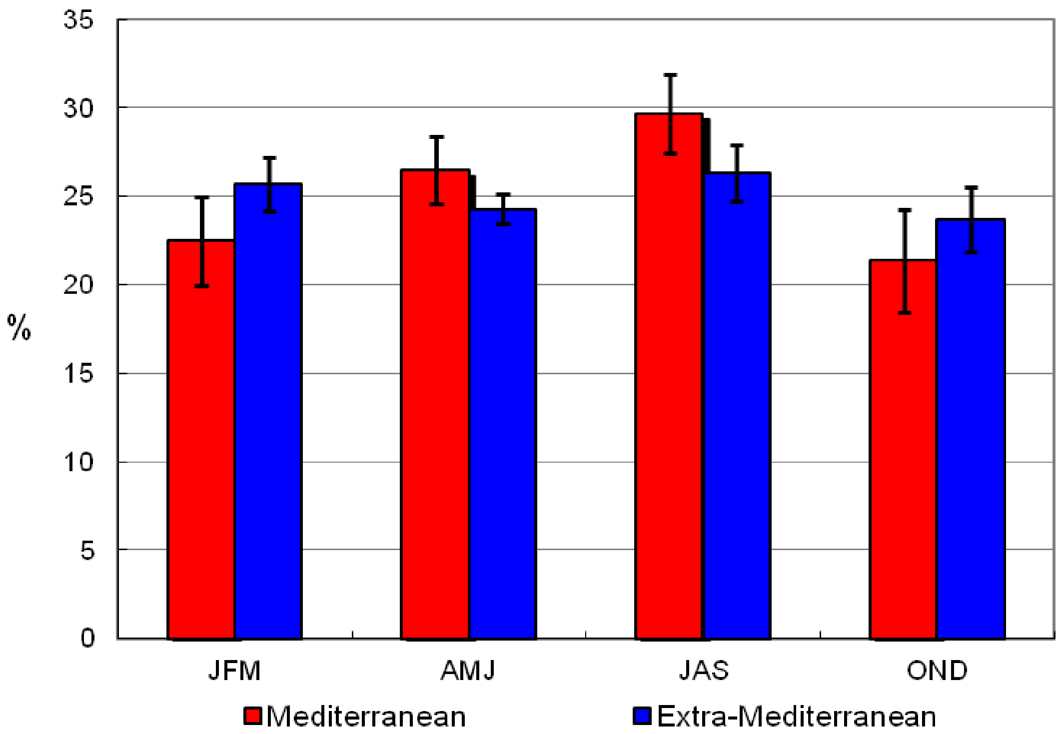
- ✓ 2005&2006 emissions
- ✓ Same modelling approach
- ✓ Different *a priori*
- ✓ Different observation data

Keller et al., 2011

- ✓ 2009 emissions
- ✓ Similar modelling approach
- ✓ Different *a priori*
- ✓ Different observation data



Emission Seasonal Cycles



Three-monthly average HCFC-22 emission estimates. Red bars, Mediterranean regions; blue bars, extra Mediterranean regions. (JFM: January-February-March; AMJ: April-May-June; JAS: July-August-September; OND: October-November-December).

Summary and next steps

- Background values are still increasing as a consequence of global emissions, especially from developing countries;
- A decrease in the atmospheric growth rate of HCFC-22 has been recorded since 2008;
- Annual European emissions show a constant decrease with two slight deviations from this trend in 2003 and 2008;
- Emissions have decreased from 56.0±6.0 tCO_{2}-eq in 2002 to 21.5±3.4 in 2012;
- The largest emissions are from France and a large macro area including 11 eastern European countries. Highest per-capita emissions are from France and UK-Ireland;
- In the countries facing the Mediterranean basin the emissions in summer are ca 25% higher than those in the colder months.;
- Once identified the correct EF, we will be able to estimate the current and future contribution of banks to the total emissions.

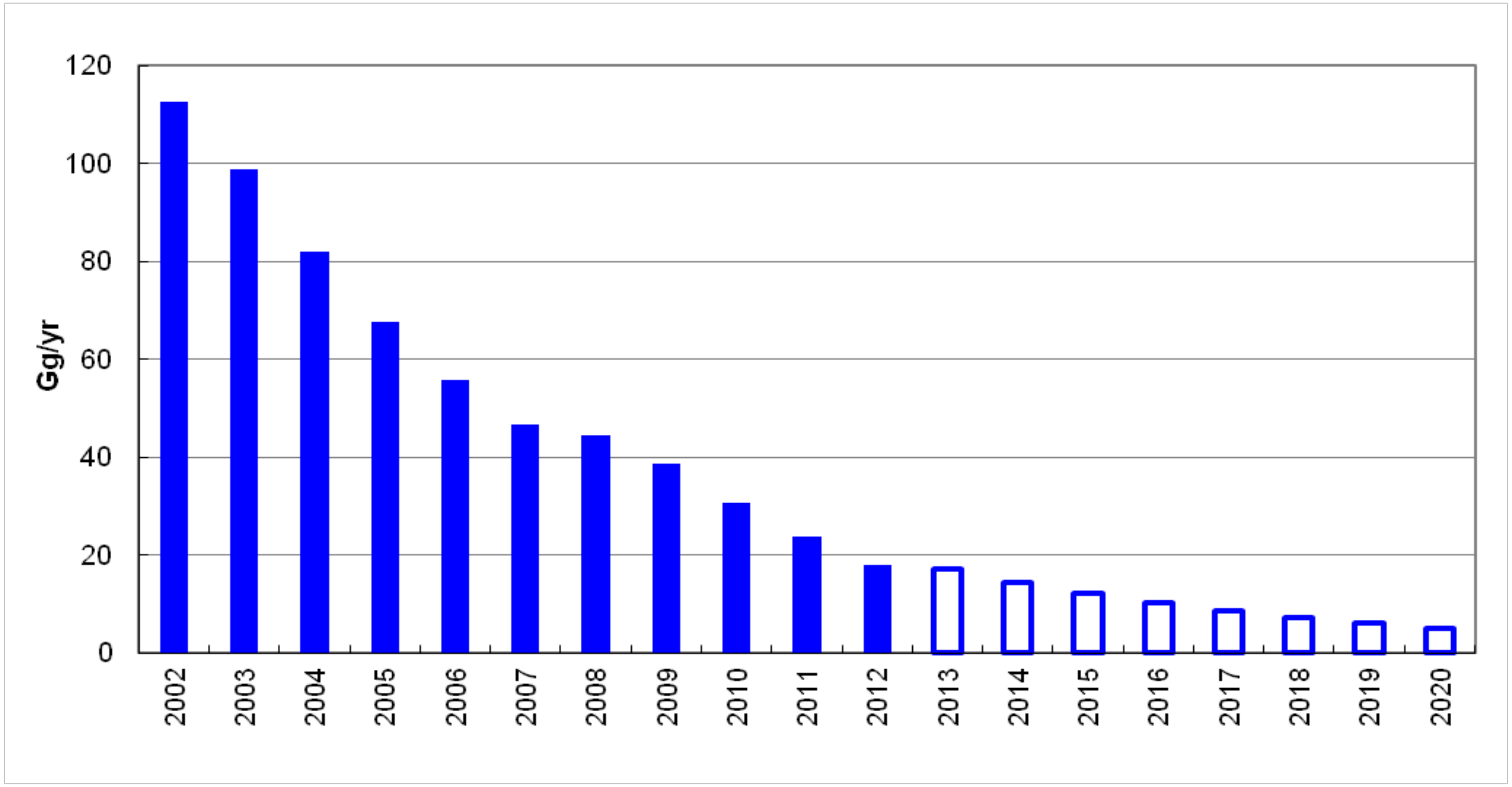
Acknowledgements

- The AGAGE scientific team and calibration scale;
- National Research Council of Italy;
- CINFAI University Consortium;
- NOAA GMD


Thank you!

Results

Banks



Estimated (solid columns) and predicted up to 2020 (open columns) emissions from Banks (EM_BANKS) present in the European Geographic Domain.

- Statistical parameters characterizing the model performance at the stations CMN and MHD. *sd*, standard deviation; N number of observation; E_a, RMS error a priori; E_b, RMS error a posteriori; $1 - E_a/E_b$, relative error reduction; E^n_b, a posteriori error normalized with the standard deviation of the observed concentration minus baseline; r^2_a, squared Pearson correlation coefficients between the observations and the total a priori; r^2_b, squared Pearson correlation coefficients between the observations and the total a posteriori; r^2_{ba}, the squared Pearson correlation coefficients between the observations and the a priori baseline; r^2_{bb}, the squared Pearson correlation coefficients between the observations and the a posteriori baseline; r^2_{ea}, squared Pearson correlation coefficients between the observation minus the a priori baseline and the modelled a priori; r^2_{eb}, squared Pearson correlation coefficients between the observation minus the a posteriori baseline and the modelled a posteriori.

2009	Station	Mean	sd	N	Mean	sd	N	E_a	E_b	$1-E_a/E_b$	E^n_b	r^2_a	r^2_b	r^2_{ba}	r^2_{bb}	r^2_{ea}	r^2_{eb}
ECMWF_N	CMN	215,33	6,42	1709	5,76	4,44	0,23	5,76	4,44	0,23	0,69	0,29	0,50	0,26	0,32	0,10	0,16
	MHD	212,15	4,25	2063	2,49	1,80	0,28	2,49	1,80	0,28	0,42	0,72	0,82	0,59	0,60	0,39	0,55
ECMWF	CMN	215,33	6,42	1709	5,80	4,38	0,24	5,80	4,38	0,24	0,69	0,30	0,53	0,28	0,32	0,08	0,31
	MHD	212,15	4,25	2063	2,53	1,89	0,25	2,53	1,89	0,25	0,45	0,71	0,81	0,59	0,61	0,36	0,51

Table I. HCFC-22 phase out schedule as decided during the 19th Meeting of the Parties to the Montreal Protocol, Montreal, Canada, 2007.

Non A-5 consumption		A-5 consumption		Non A-5 production		A-5 production	
Base Level: 1989[a]		Base Level: Average 2009–10		Base Level: 1989[b]		Base Level: Average 2009–10	
Freeze: 1996		Freeze: Jan 2013		Freeze: Jan 2004		Freeze: Jan 2013	
% reduction	**Year**	**% reduction**	**Year**	**% reduction**	**Year**	**% reduction**	**Year**
35	Jan 2004	10	Jan 2015			10	Jan 2015
75	Jan 2010	35	Jan 2020	75	Jan 2010	35	Jan 2020
90	Jan 2015	67.5	Jan 2025	90	Jan 2015	67.5	Jan 2025
99.5	Jan 2020[c]	97.5	Jan 2030[c]	99.5	Jan 2020[c]	97.5	Jan 2030[c]
100	Jan 2030	100	Jan 2040	100	Jan 2030	100	Jan 2040

[a] Average HCFC consumption + 2.8% of 1989 CFC consumption

[b] Average HCFC production + 2.8 per cent of 1989 CFC production

- From the AFEAS data set the percentage of sales for different categories were taken (on average: short release times 10% of total, medium release 85% of total, long release 5% of total), and we applied such percentages to the total European consumption data. Having obtained the consumption data divided by categories (short, medium and long release) from the whole period of interest (1982 - 2012) we simulated the emissions using the appropriate emission function (EF) described by McCulloch et al. (2006). The EUMSs emissions values obtained through this EF and the emissions from countries that are not EUMSs but included in the European Geographic Domain (EGD), are disaggregated within each country's borders according to a gridded population density data set (CENSIS 2005-2010).