Top-down Validation of European Halocarbon Emission Inventories

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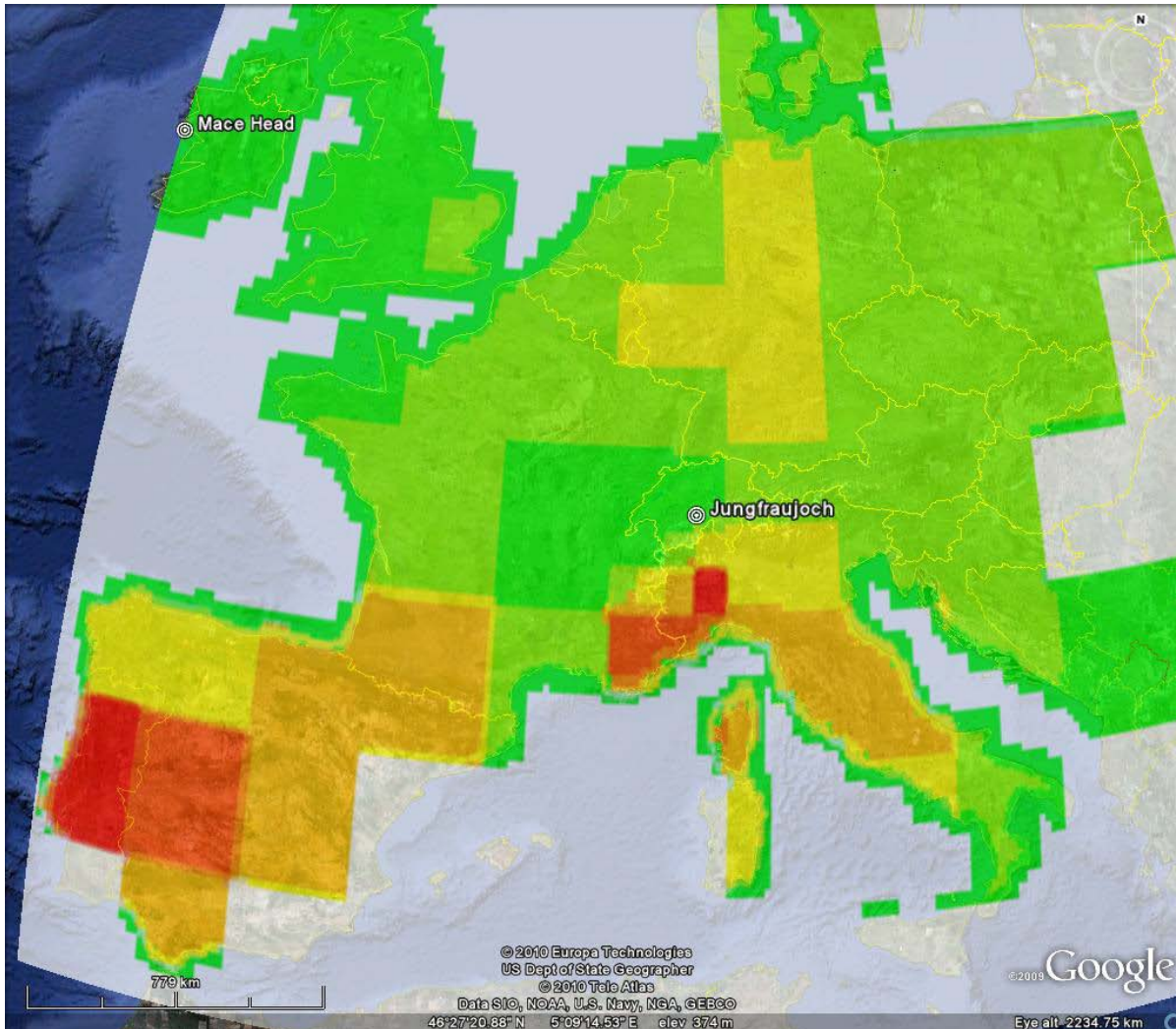
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[3] Climate Research, UK Met Office

EMPA

Materials Science & Technology
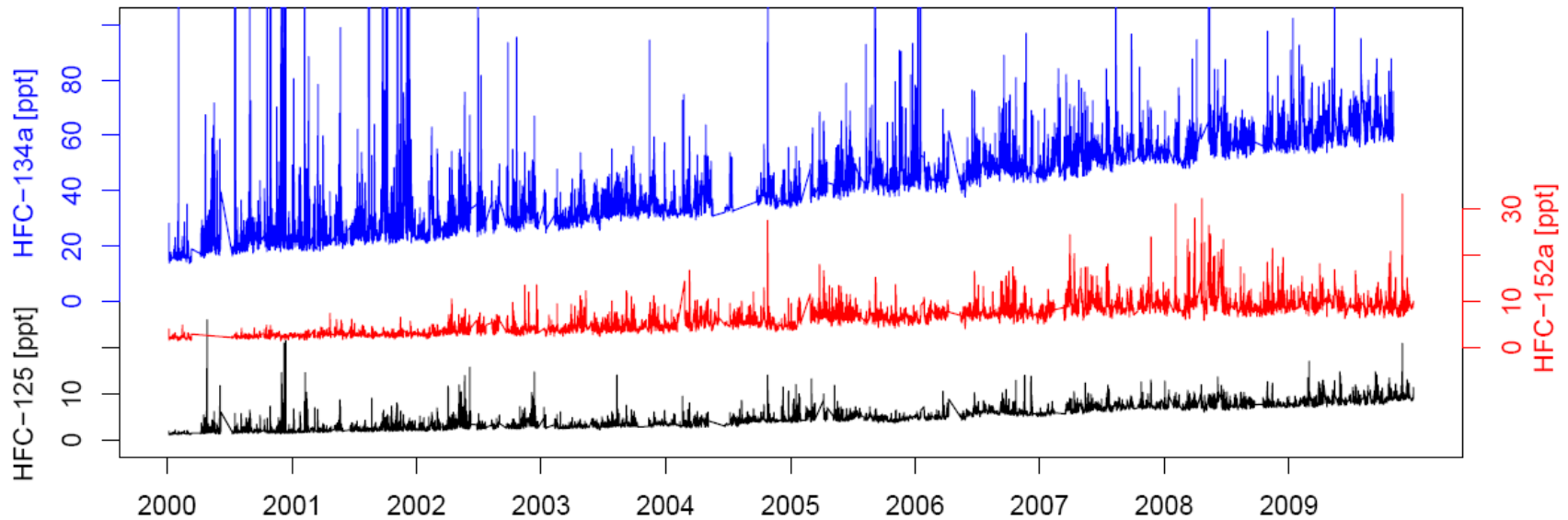
European HFC-23 emissions
## Mismatch of inventories and measurement-based estimates

Motivation

- Halocarbon emissions by country are estimated with bottom-up approach and reported to UNFCCC

- Top-down estimation provides an independent tool to verify these numbers
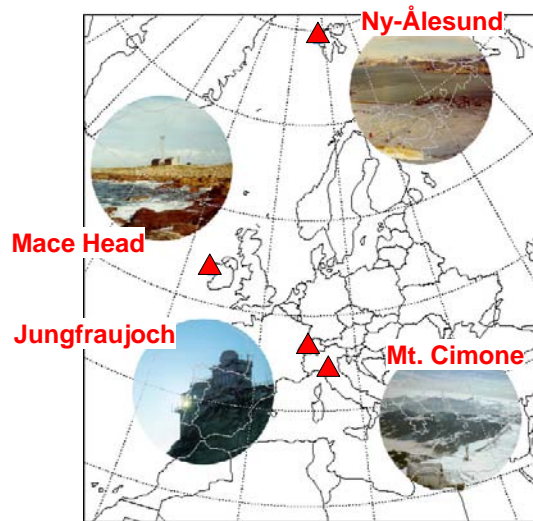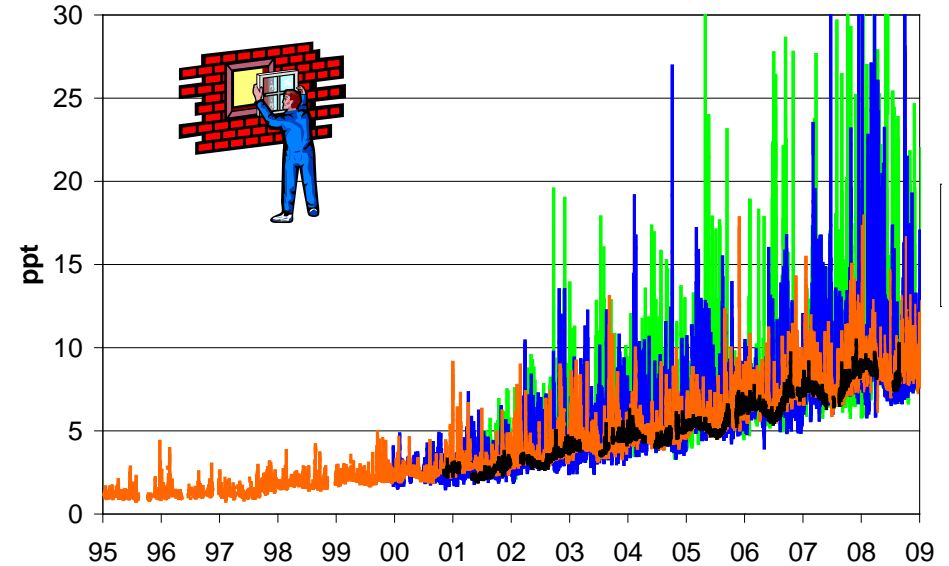
Jungfraujoch, Switzerland

Examples of European measurements: HFCs on the rise

HFC-134a

HFC-152a

- Ny-Ålesund
- Mace Head
- Jungfraujoch
- Mt. Cimone
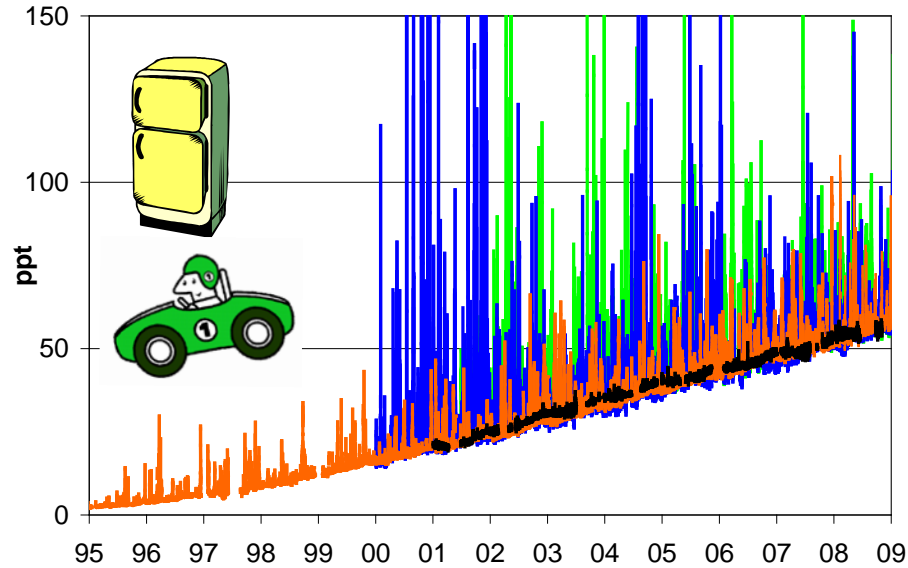
— Monte Cimone (I)
— Jungfraujoch (CH)
— Mace Head (IRE)
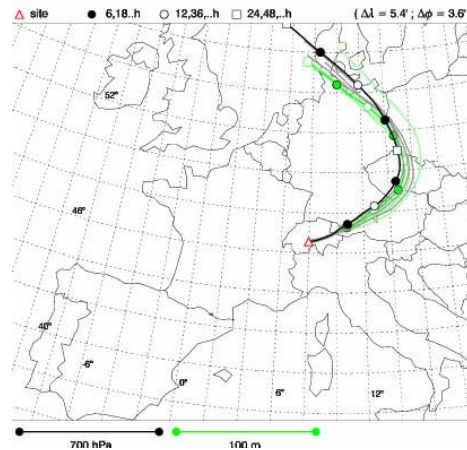— Ny-Alesund (N)

Top-down estimation of halocarbons
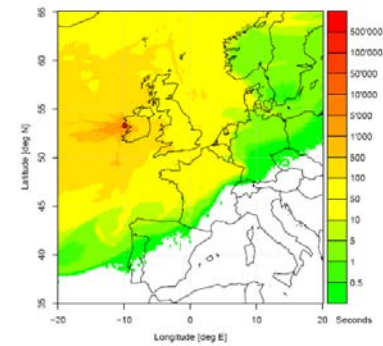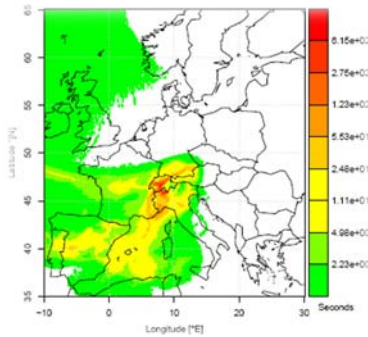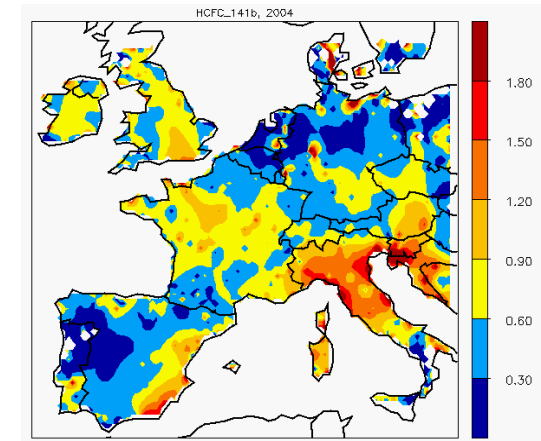
Modelling of European sources

1. Definition of pollution over background
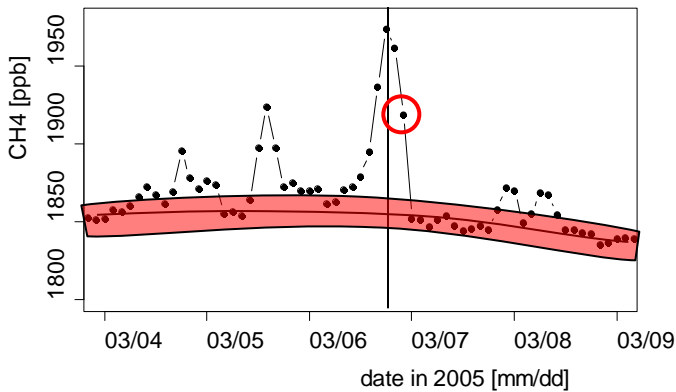
2. Calculate the trajectory for each measurement

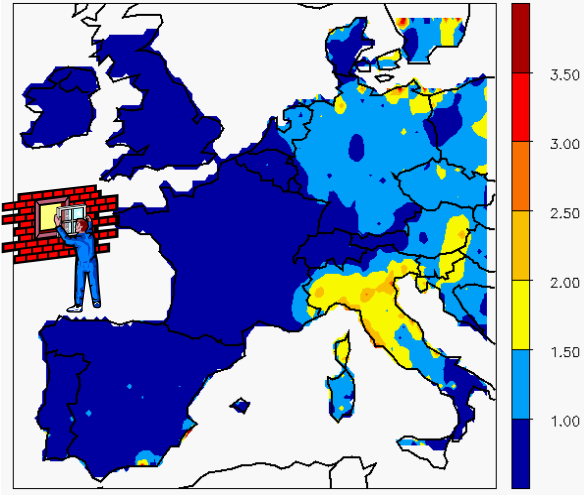**3.** Distribute the concentrations along the trajectory and average all points in the boundary layer

Jungfraujoch (Switzerland)
46.6°N, 8.0°W, 3580 m a.s.l
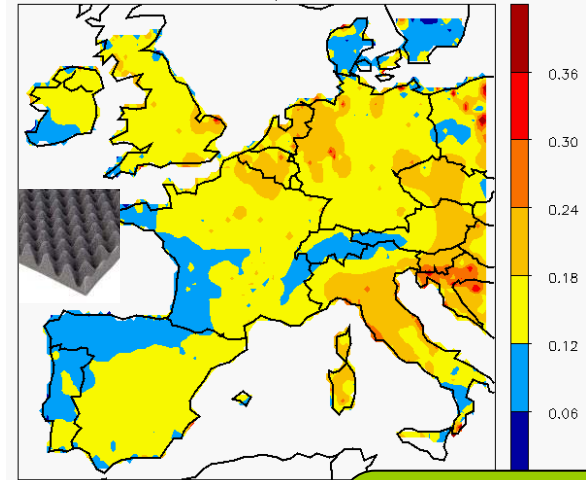free troposphere and polluted
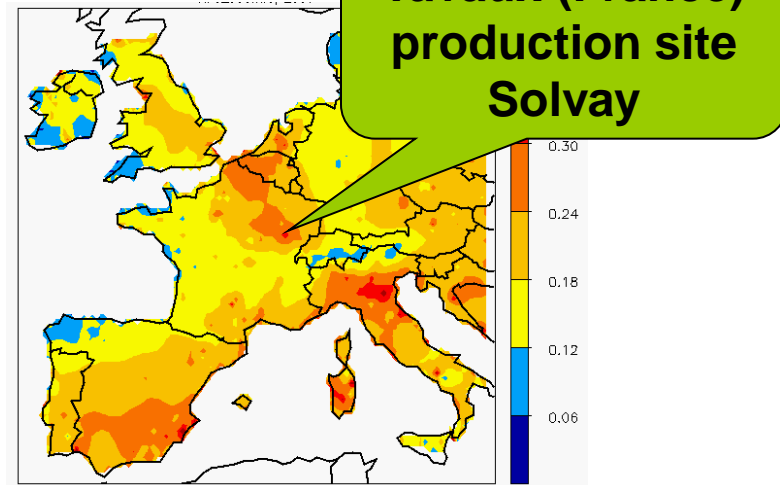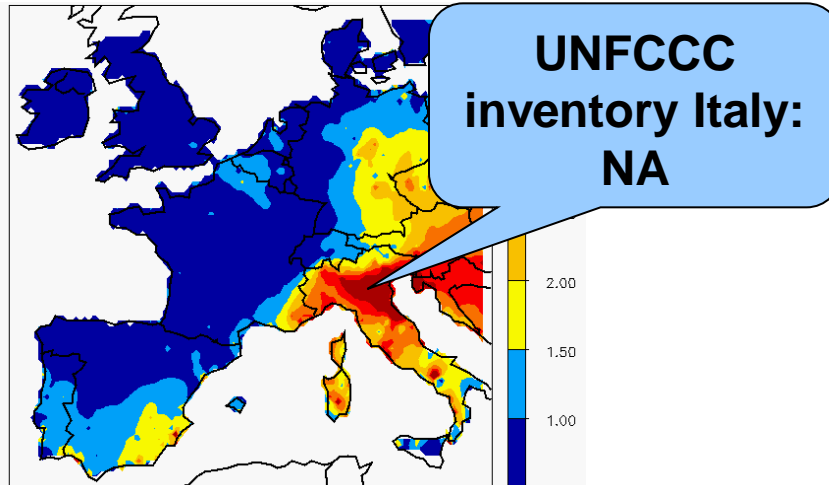boundary layer

Modelling of European sources

HFC-152a

HFC-365mfc

2005

2008

UNFCCC inventory Italy: NA

Tavaux (France) production site Solvay

Modelling of European sources
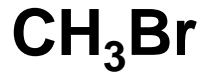
CH$_3$Br
regulated Montreal Protocol

Critical use exemptions by Montreal Protocol

2005
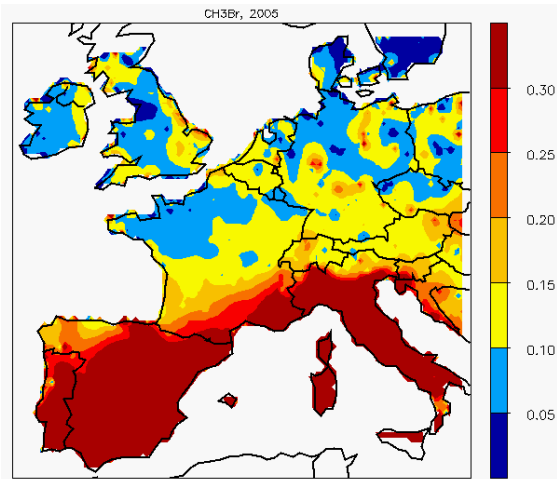
4.392 t
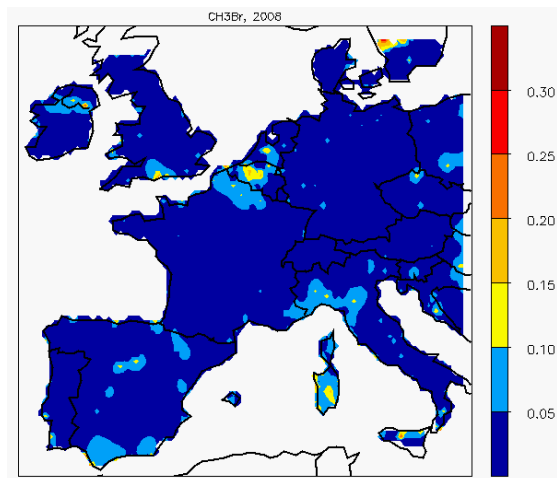
SO$_2$F$_2$
replacement (greenhouse gas)

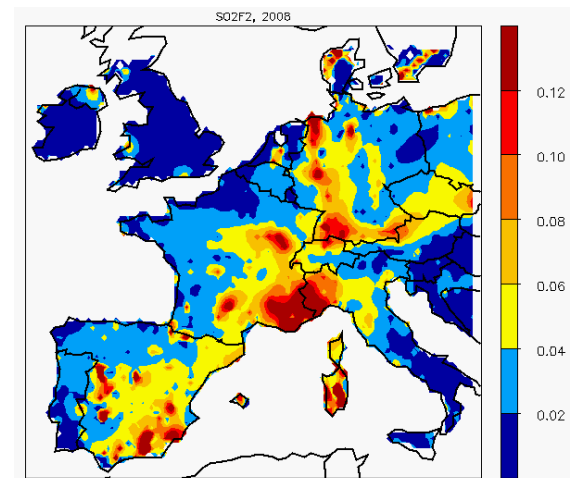**2008**

689 t

Modelling of European sources

2. Method:

- Dispersion model (FLEXPART)

- ^{222}Rn measurements for quality assurance of model

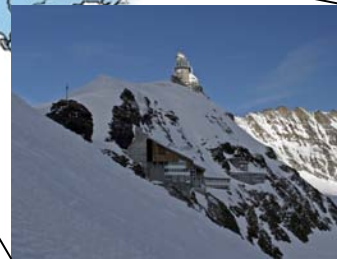- Kalman filter to fit emissions to measurements

Mace Head (Ireland)
53.3°N, 9.9°W, 25 m a.s.l
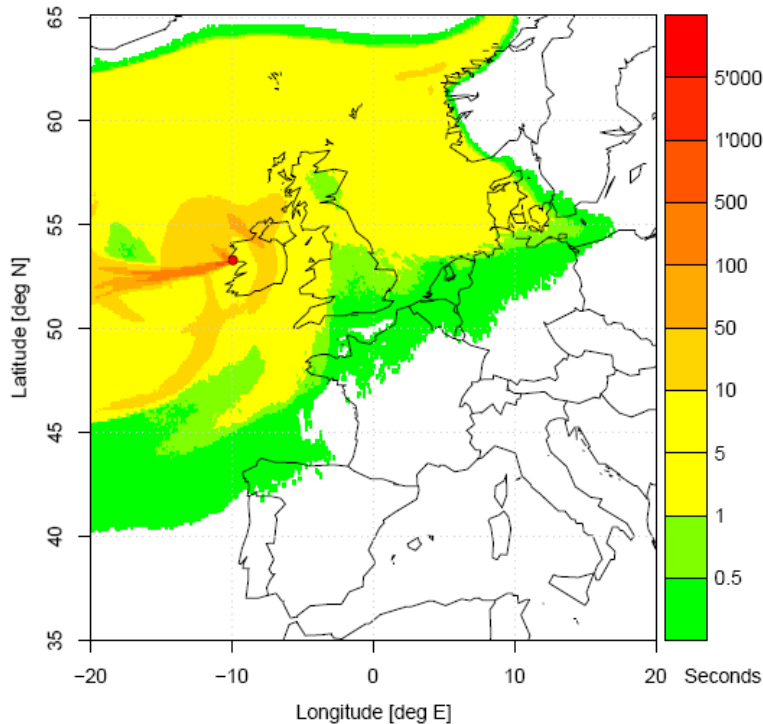Influenced by both marine
and continental air masses

Jungfraujoch (Switzerland)
46.6°N, 8.0°W, 3580 m a.s.l
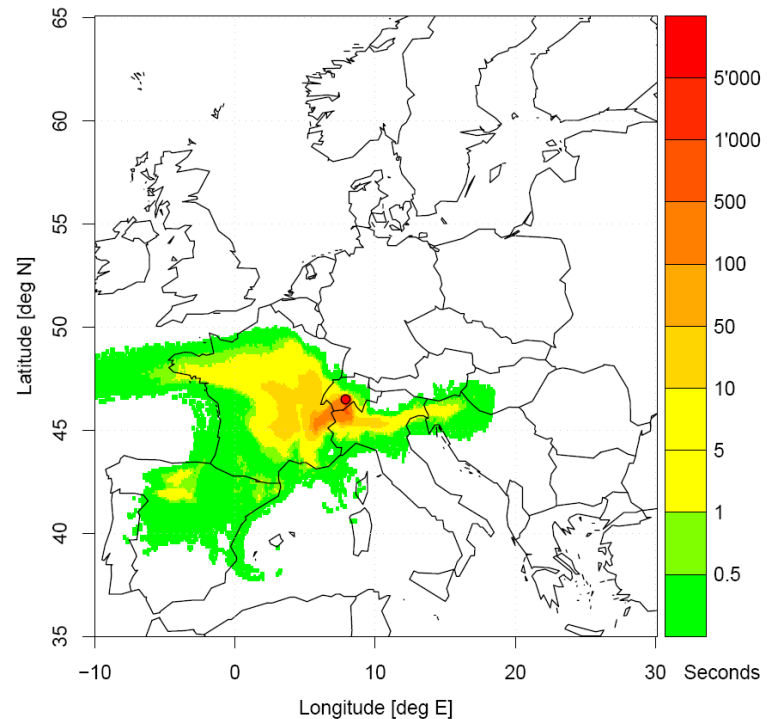free troposphere and polluted
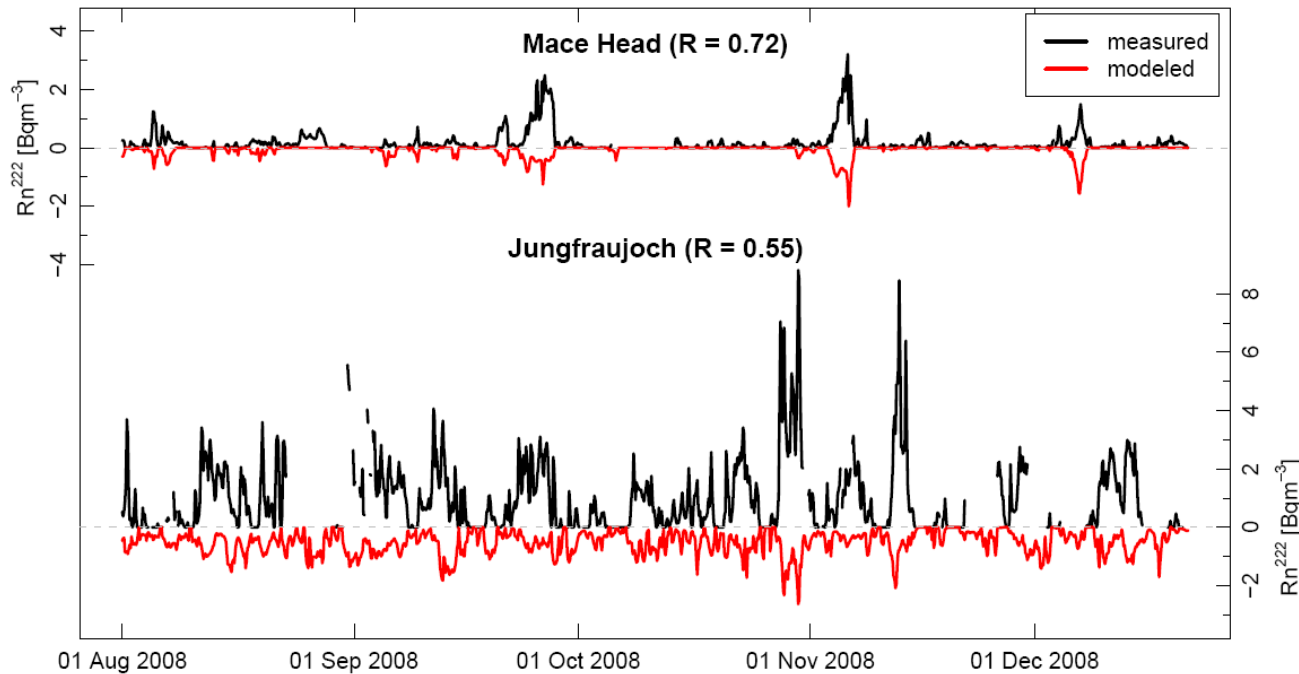boundary layer

Dispersion Modeling

- LPDM FLEXPART used to determine origin of air masses
- Driven by ECMWF windfields
- Footprint: map of residence times of particles in lowest 100m above model ground
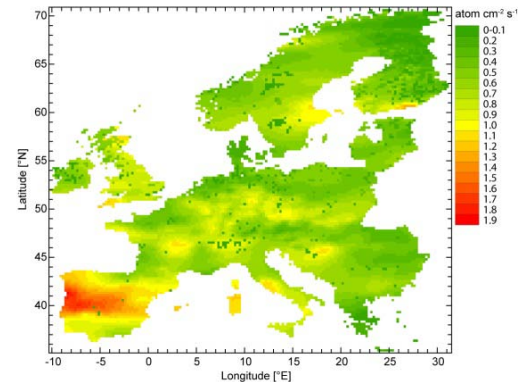
Radon-222 as atmospheric tracer
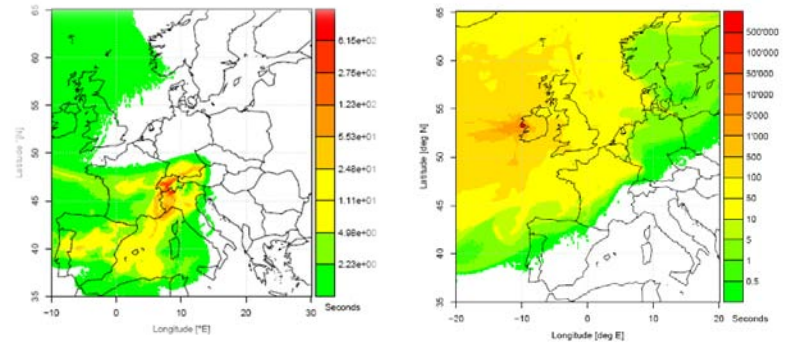

²²²Rn flux map:

Szegvary et al. (ACP, 2007)

- Radon-222: uniformly emitted from soils → excellent tracer for vertical transport and mixing
- ²²²Rn used to evaluate how accurately transport is simulated
- Good agreement between observed and modeled concentrations

Kalman filter to estimate surface emissions

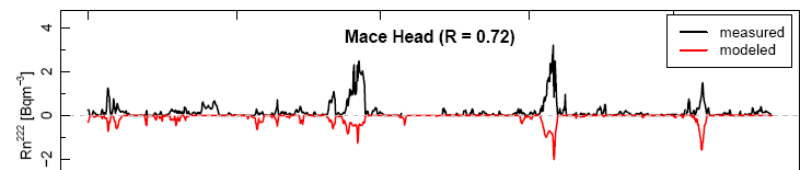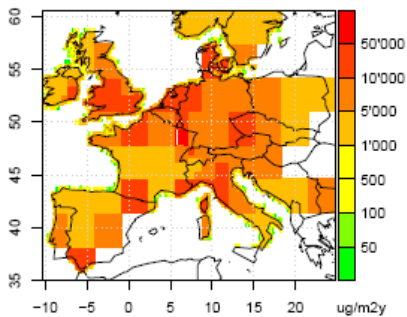Measurements z_k
(- background bg_k)

Footprints H_k

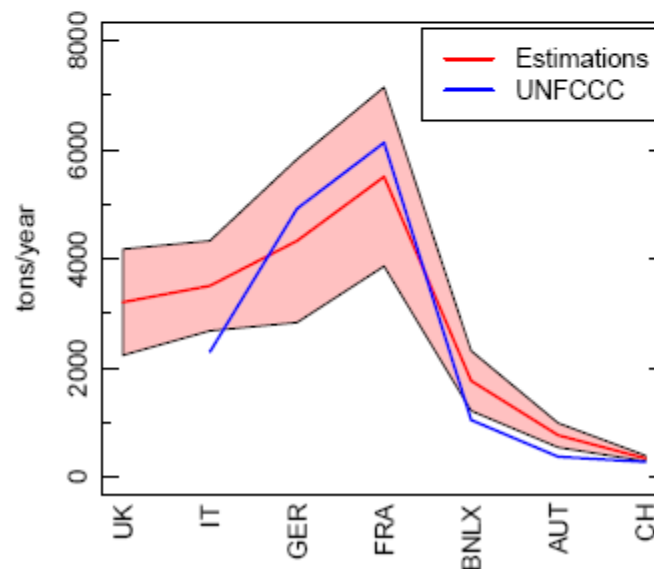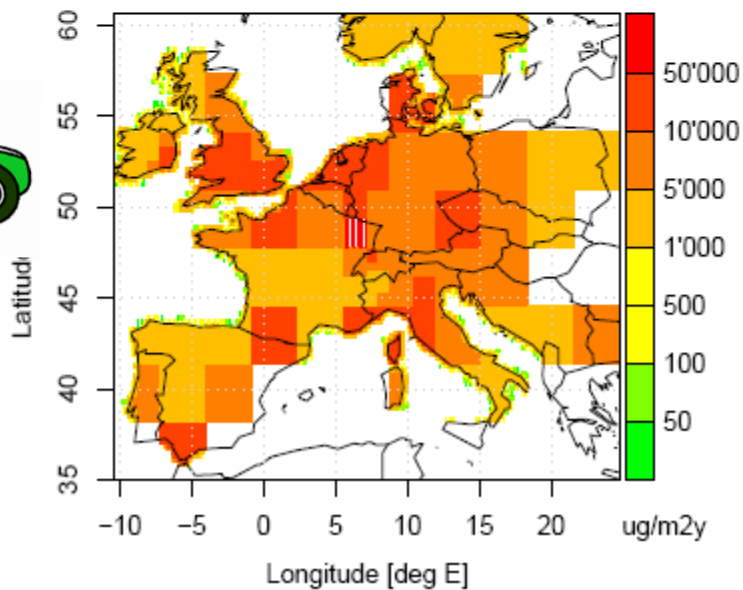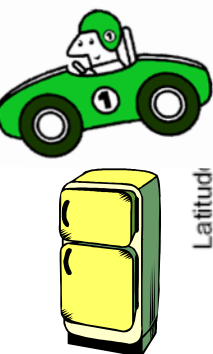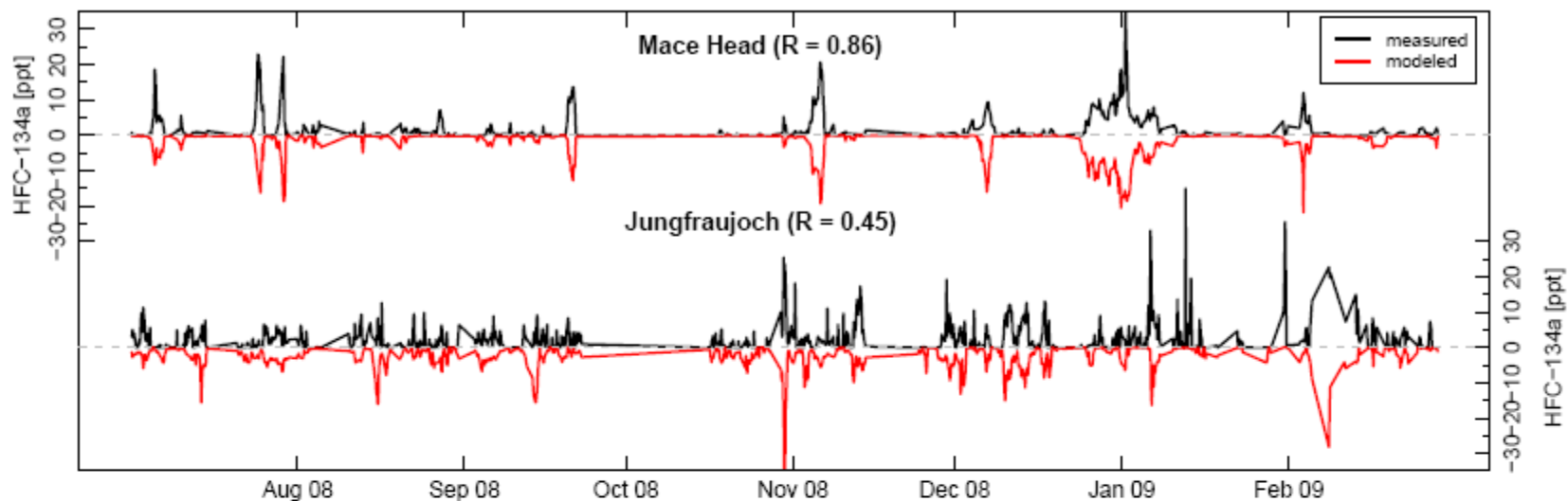$$x_k = 1 \cdot x_{k-1} + K_k (z_k - bg_k - H_k x_{k-1})$$
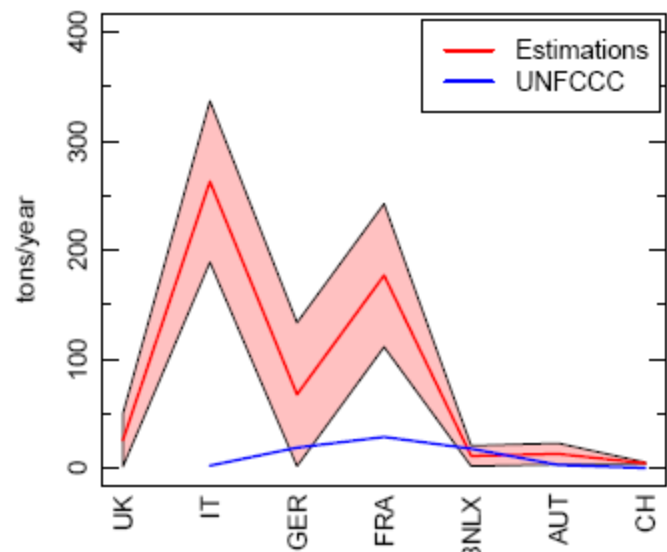
Estimated emission
field at time k

Kalman gain: $K_k = f(Q_k, R_k)$

Error covariance R_k
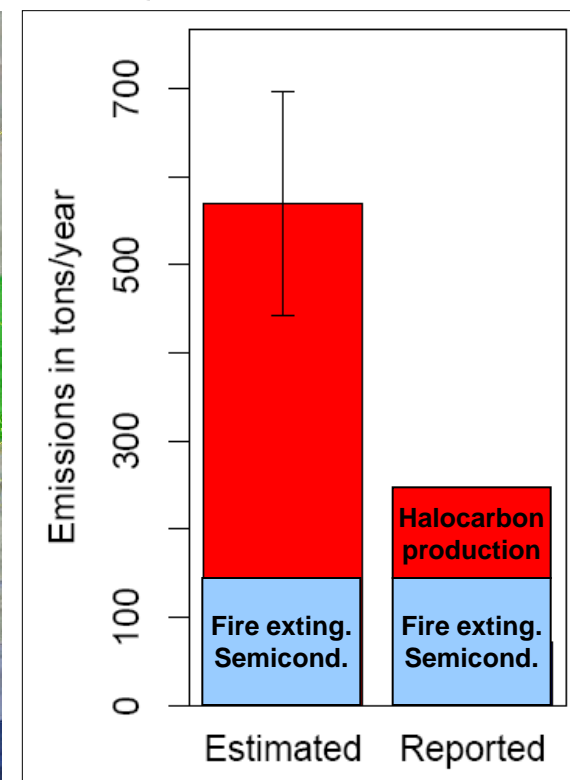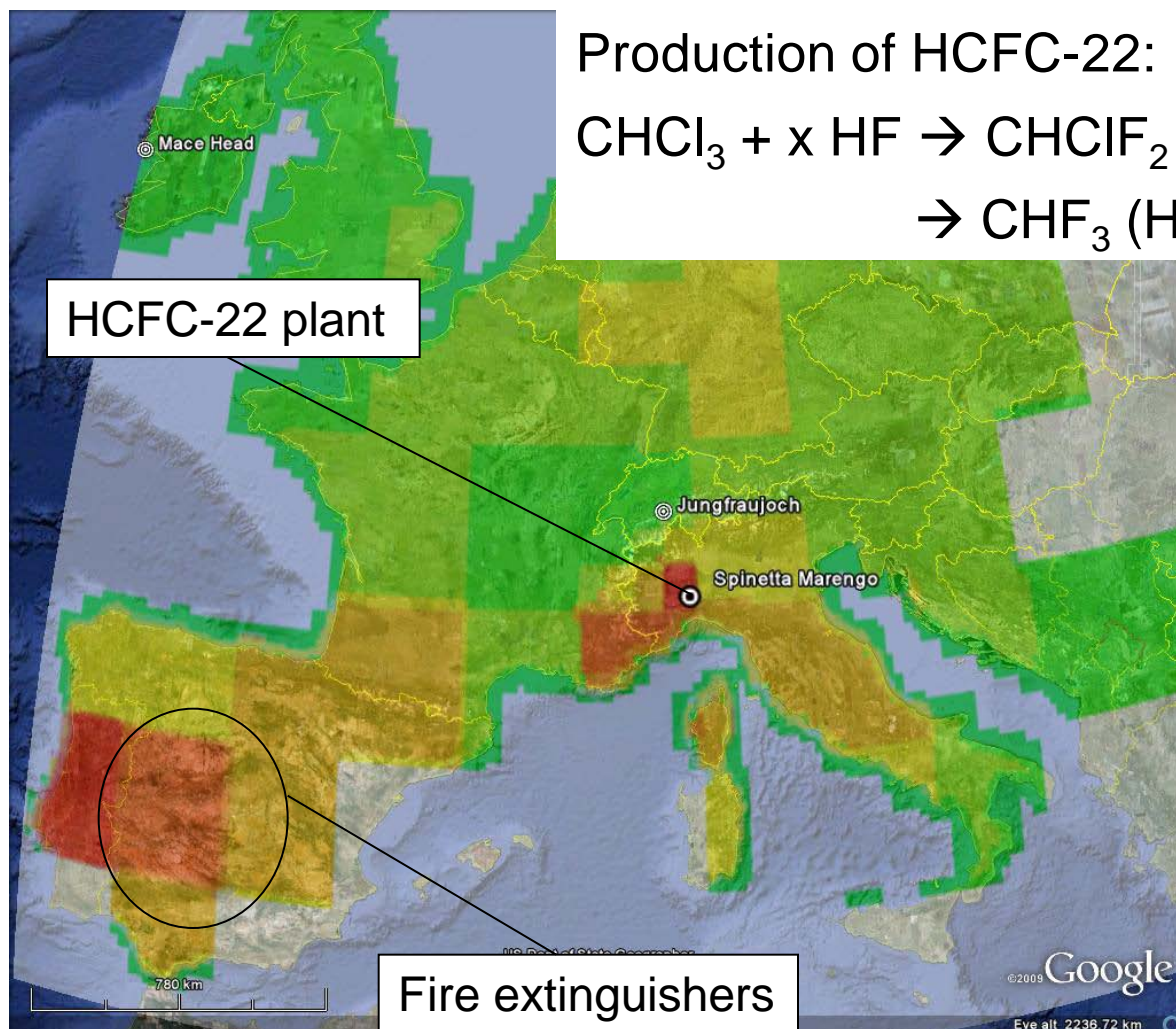derived from ^{222}Rn

Mace Head (R = 0.72)

measured
modeled

Emissions of HFC-134a in 2008 (GWP = 1'430)

Emissions of HFC-23 in 2008 (GWP = 14'800)

Emissions of HFC-23 in 2008 (GWP = 14'800)

Production of HCFC-22:

$$CHCl_3 + x\ HF \rightarrow CHClF_2\ (HCFC\text{-}22) + x\ HCl$$
$$\rightarrow CHF_3\ (HFC\text{-}23) \quad\quad + x\ HCl$$

HCFC-22 plant

Fire extinguishers

Emissions in tons/year

Halocarbon production

Fire exting. Semicond.

Fire exting. Semicond.

Estimated   Reported
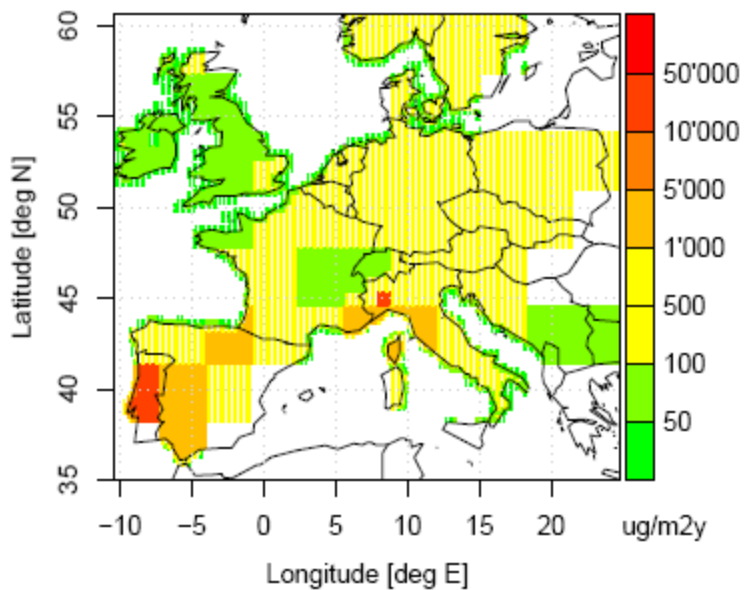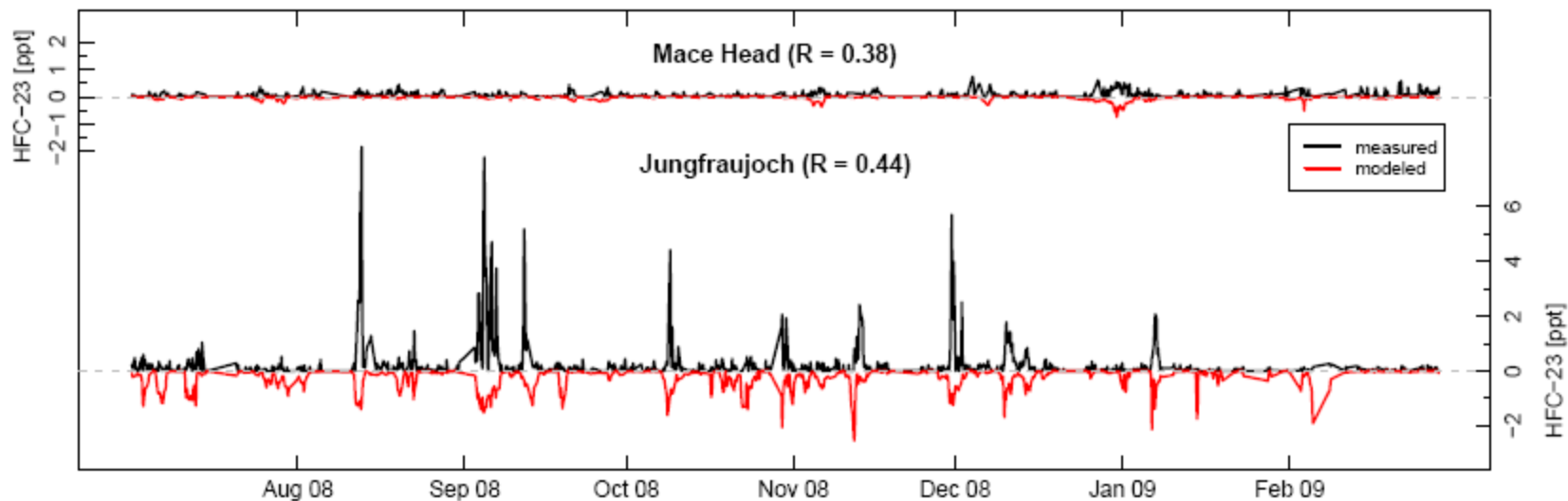
Italy: 250 t x 11700 =
2.93 Mt CO$_2$ eq.
0.5% of Italy (or CO$_2$ from 300.000 inhabitants)

Conclusions / Outlook

Conclusions

- Trajectory models useful for location of sources

- Particle dispersion model + ^{222}Rn +Kalman filter ➔

  validation of regional emissions for UNFCCC inventories

- European emissions of HFC-23 are likely underestimated

Outlook

- System of regional medium-polluted sites could be used world-wide for verification of emission inventories

Thank you for your attention

Halocarbon emissions in 2008: reported vs. estimated