Calculating greenhouse gas emissions at global and regional scales using atmospheric measurements

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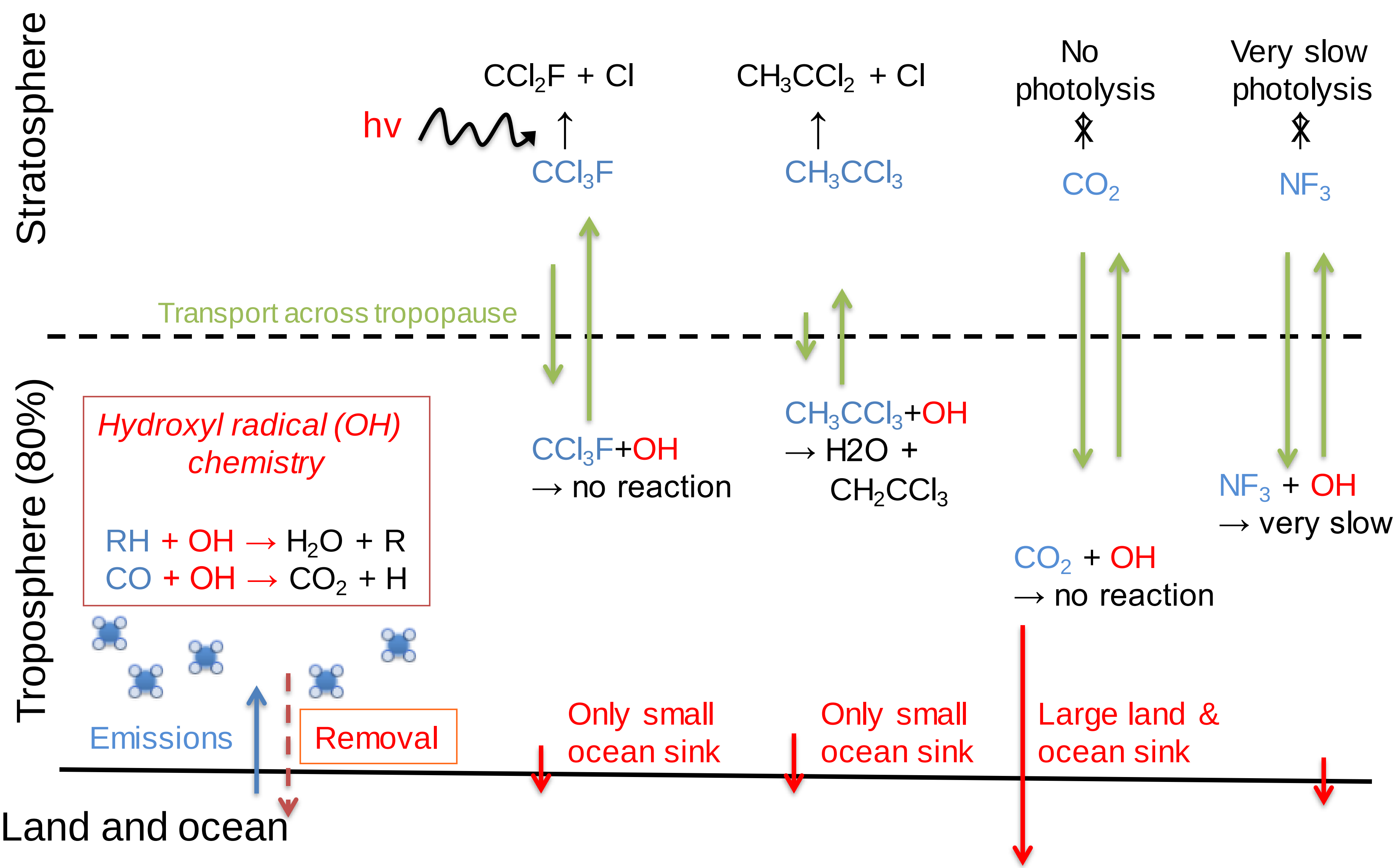
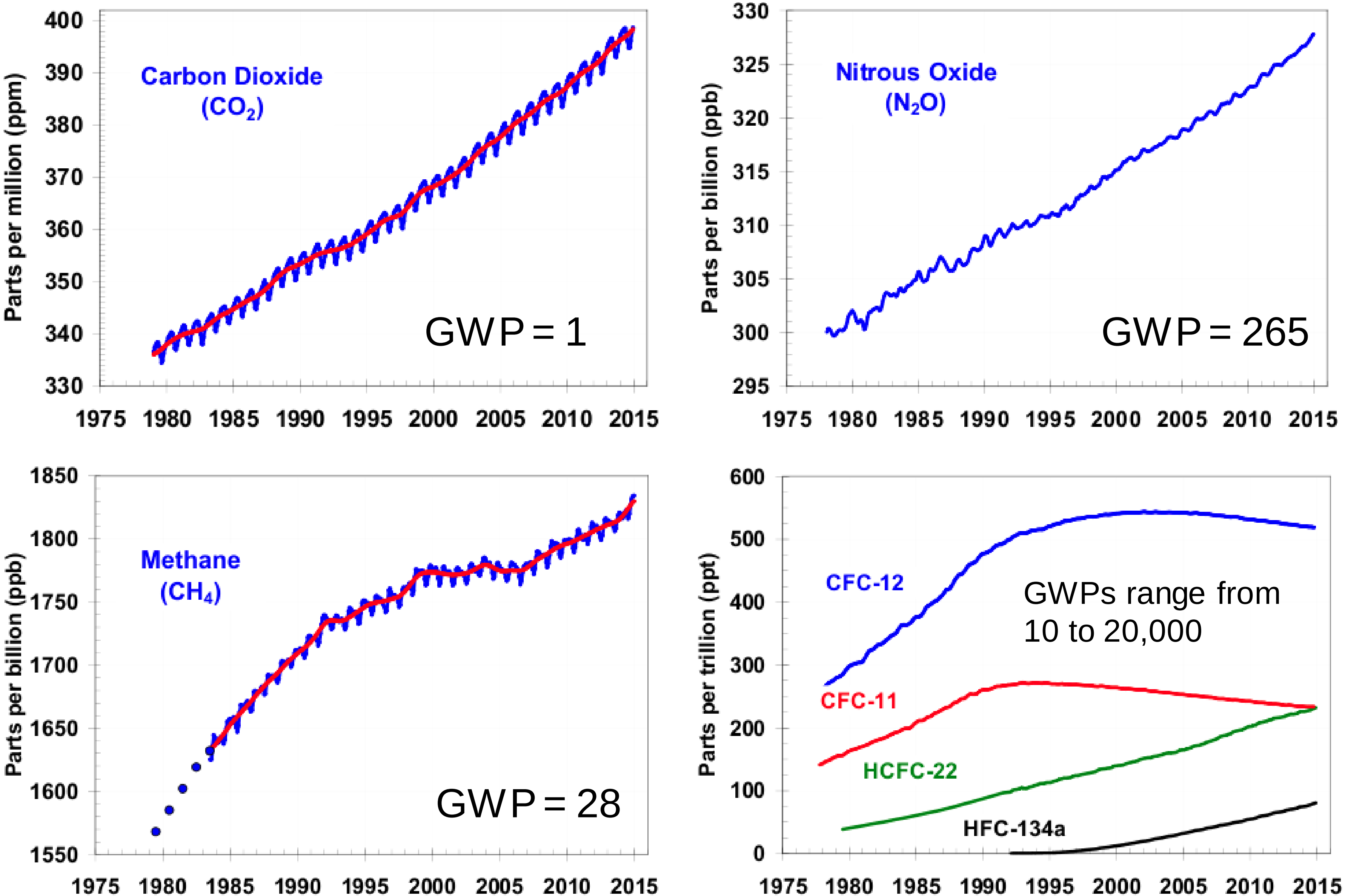
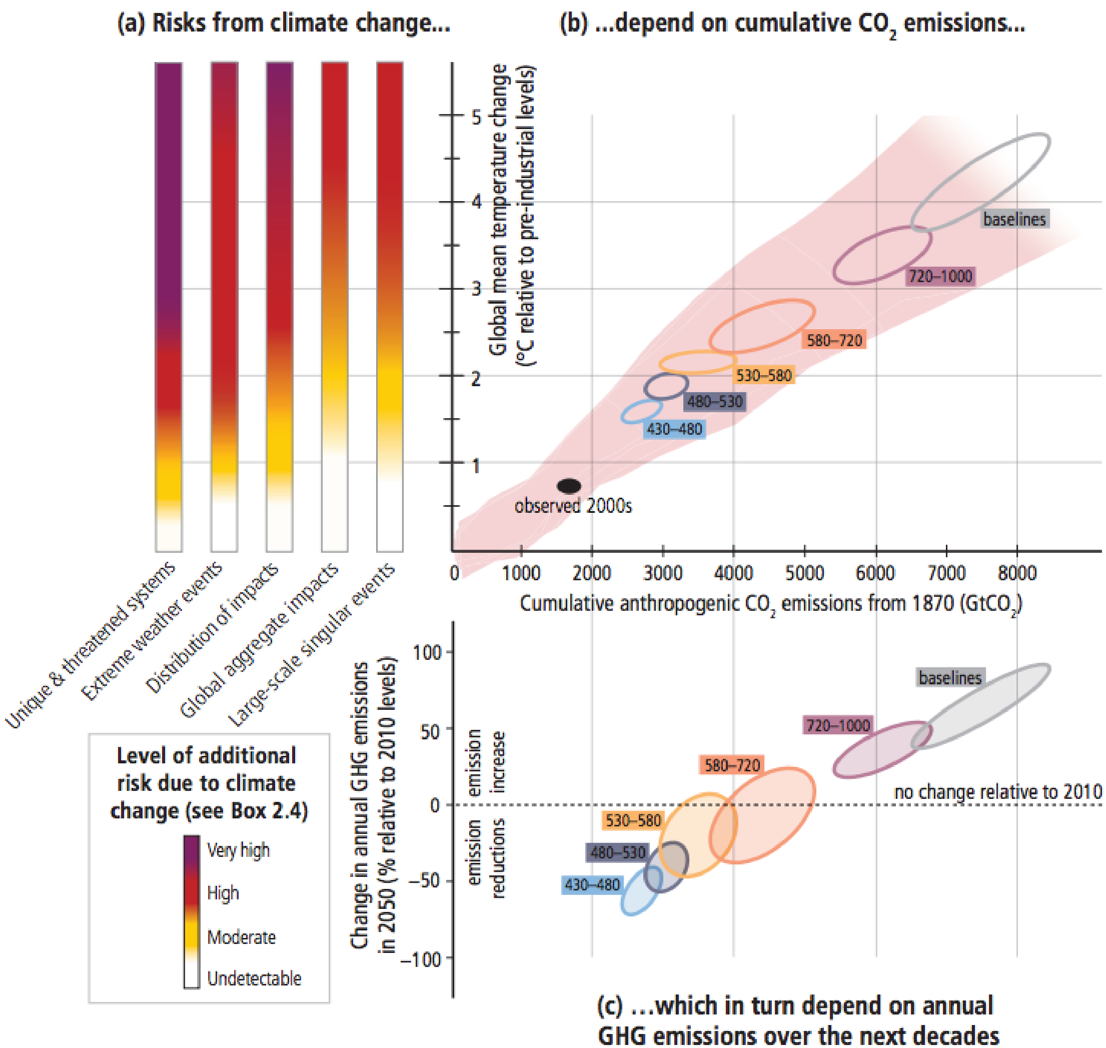
GLOBAL ATMOSPHERIC CHEMISTRY



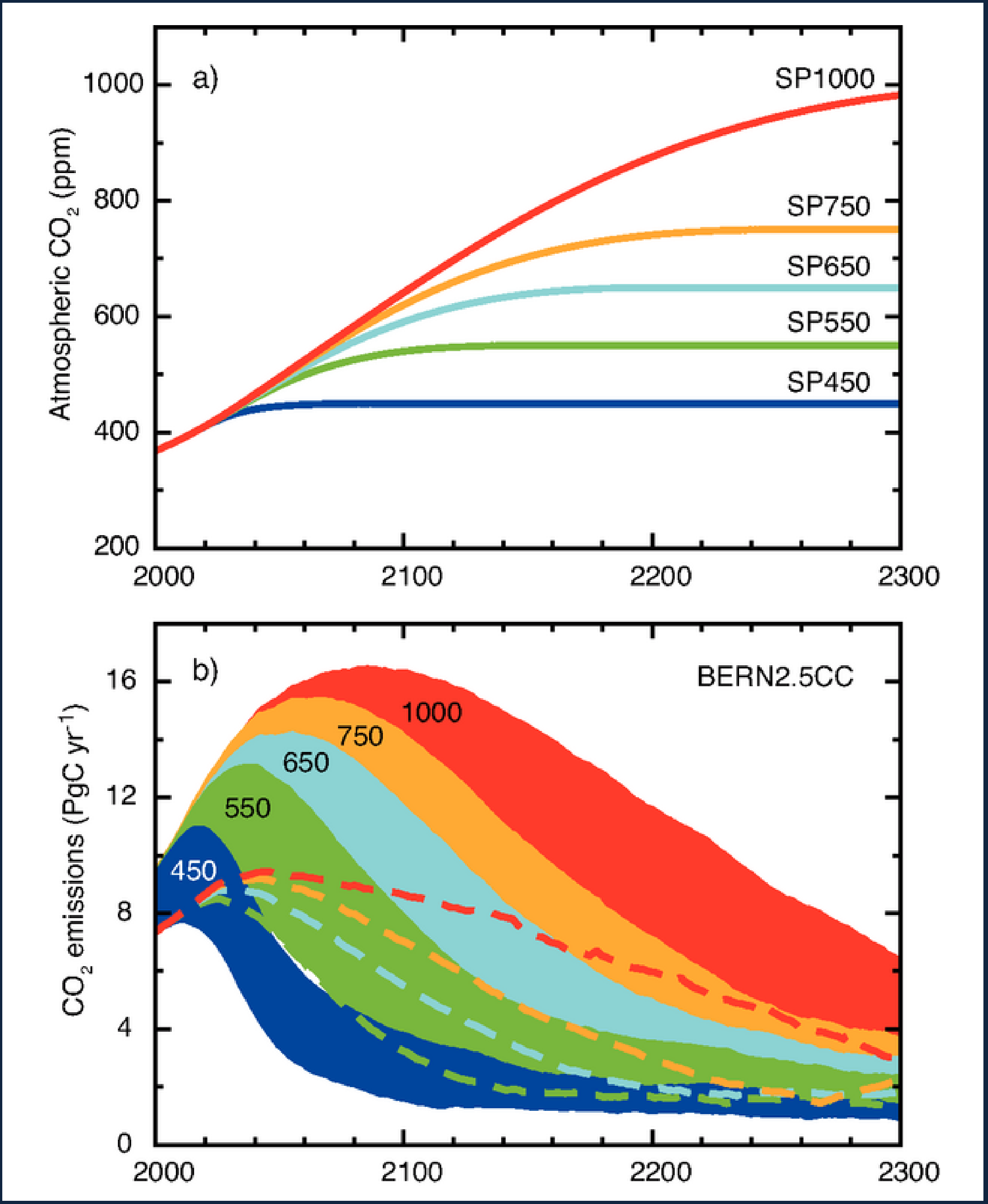


Carbon Dioxide
(CO_2)
GWP = 1
Parts per million (ppm)
Nitrous Oxide
(N_2O)
GWP = 265
Parts per billion (ppb)
Methane
(CH_4)
GWP = 28
Parts per billion (ppb)
CFC-12
CFC-11
HCFC-22
HFC-134a
GWPs range from
10 to 20,000
Parts per trillion (ppt)

Motivation for immediate and significant emissions reductions

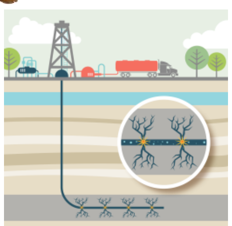


IPCC 5th Assessment Report (Executive Summary)



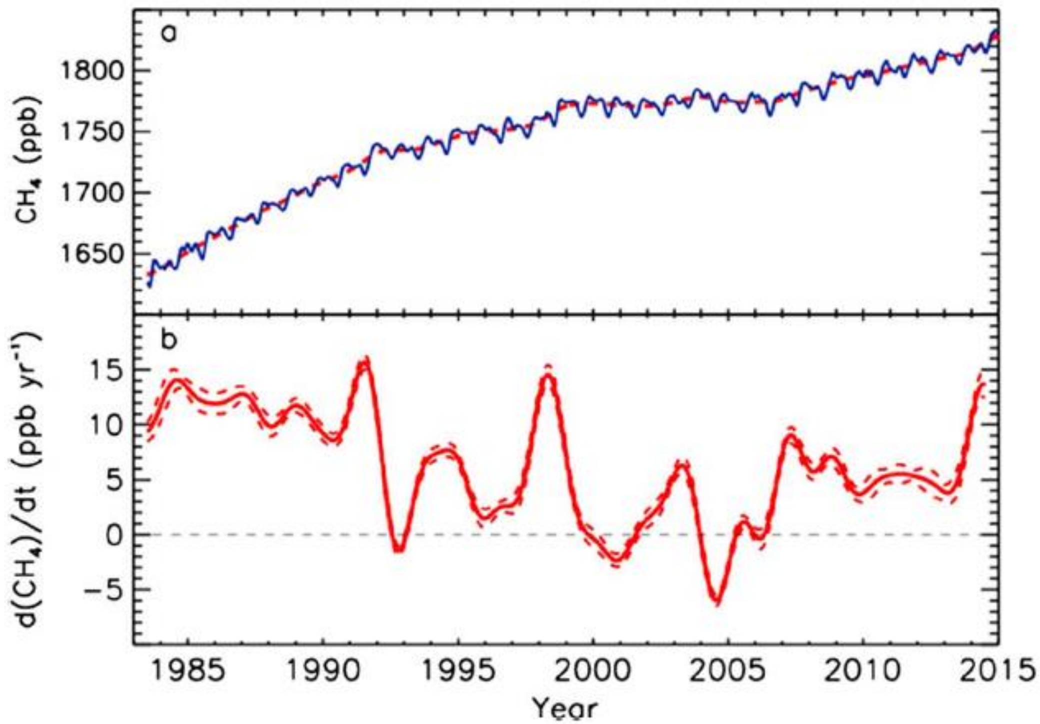
a)
Atmospheric CO_2 (ppm)
SP1000
SP750
SP650
SP550
SP450
1000
800
600
400
200
2000
2100
2200
2300
b)
BERN2.5CC
CO_2 emissions (PgC yr^{-1})
1000
750
650
550
450
16
12
8
4
0
2000
2100
2200
2300

Why atmospheric monitoring can play an increasingly important role



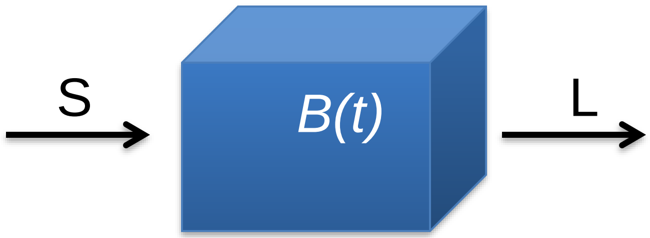
Uncertainty in scaling

- Provides a reality check – what's actually happening
- It is up to date – to direct timely policy
- Independent



A simple example of emissions verification from atmospheric measurements

1-box model of the atmosphere



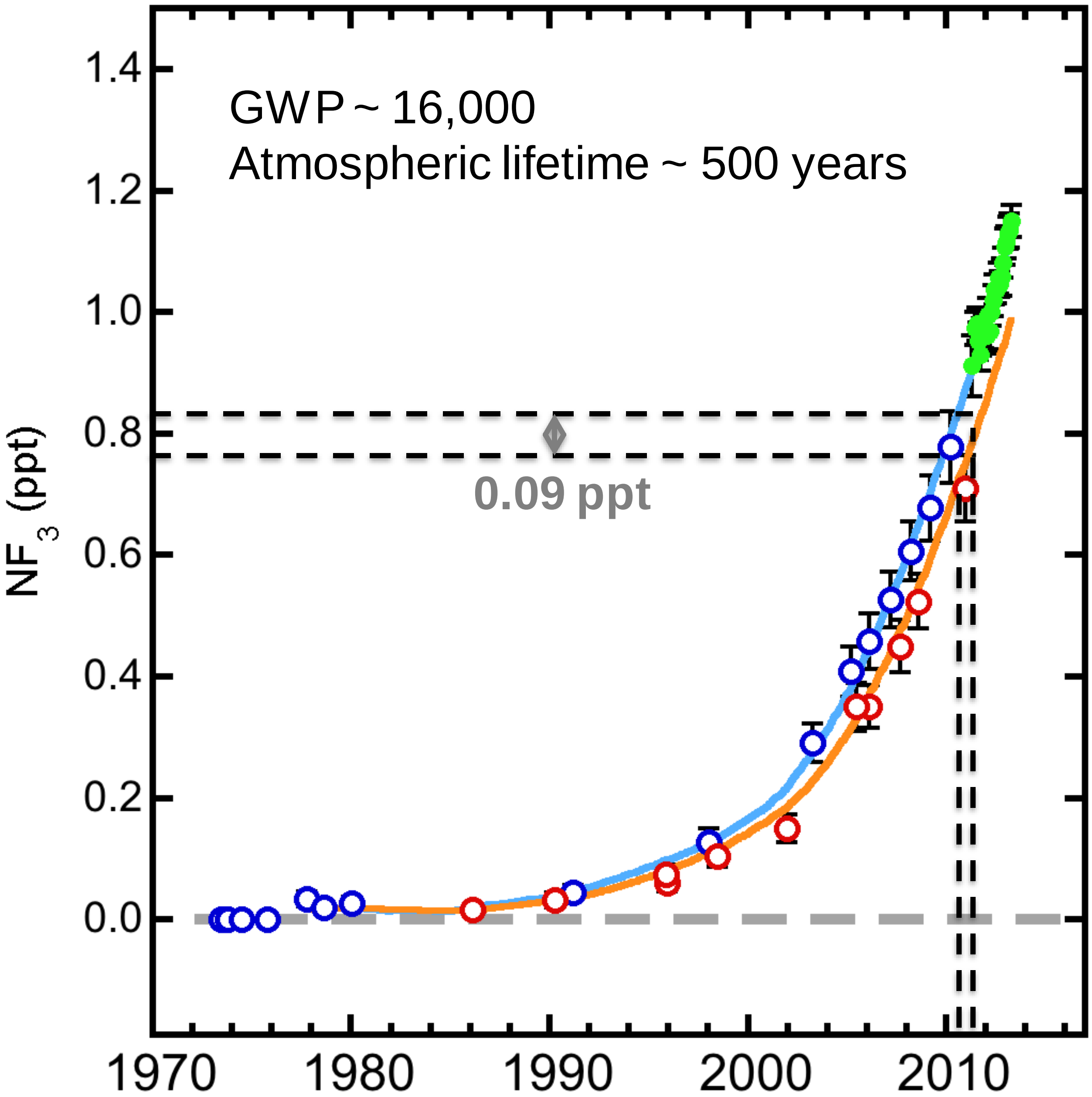
L is the removal rate e.g. Gg / year

S is the source rate

B(t) is the atmospheric burden at time *t*

$$\frac{\mathrm{d}B(t)}{\mathrm{d}t} = S - L \qquad L = \frac{B(t)}{t}$$

$$\frac{\mathrm{d}B(t)}{\mathrm{d}t} = S - \frac{B(t)}{t}$$

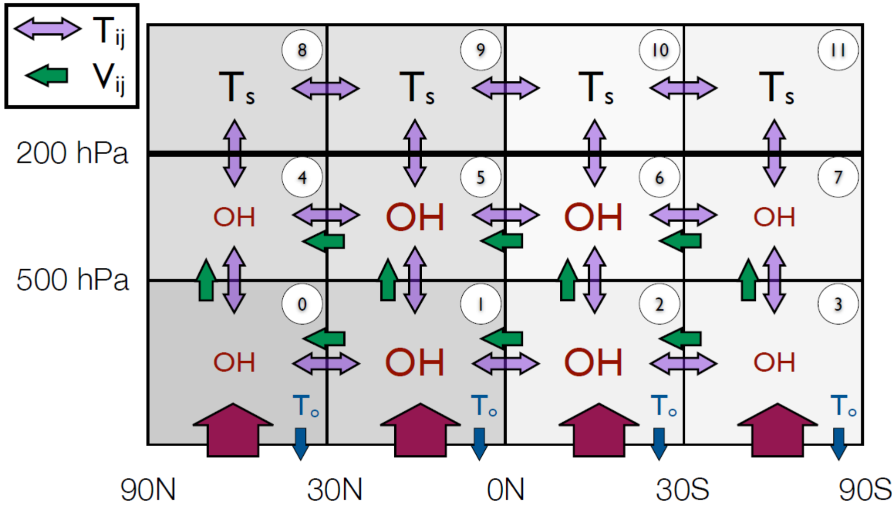


$$\frac{\mathrm{d}B(t)}{\mathrm{d}t} = S - \frac{B(t)}{t}$$

$$\frac{B(t)}{t} \to 0$$

$$\frac{\mathrm{d}B(t)}{\mathrm{d}t} = S$$

Bayesian inversions and more complex models



12-box model

$$\mathbf{y} = \mathbf{M}\mathbf{x}$$

$\mathbf{y} = (y_1 \quad \cdots \quad y_m)$ is the vector of simulated mole fractions

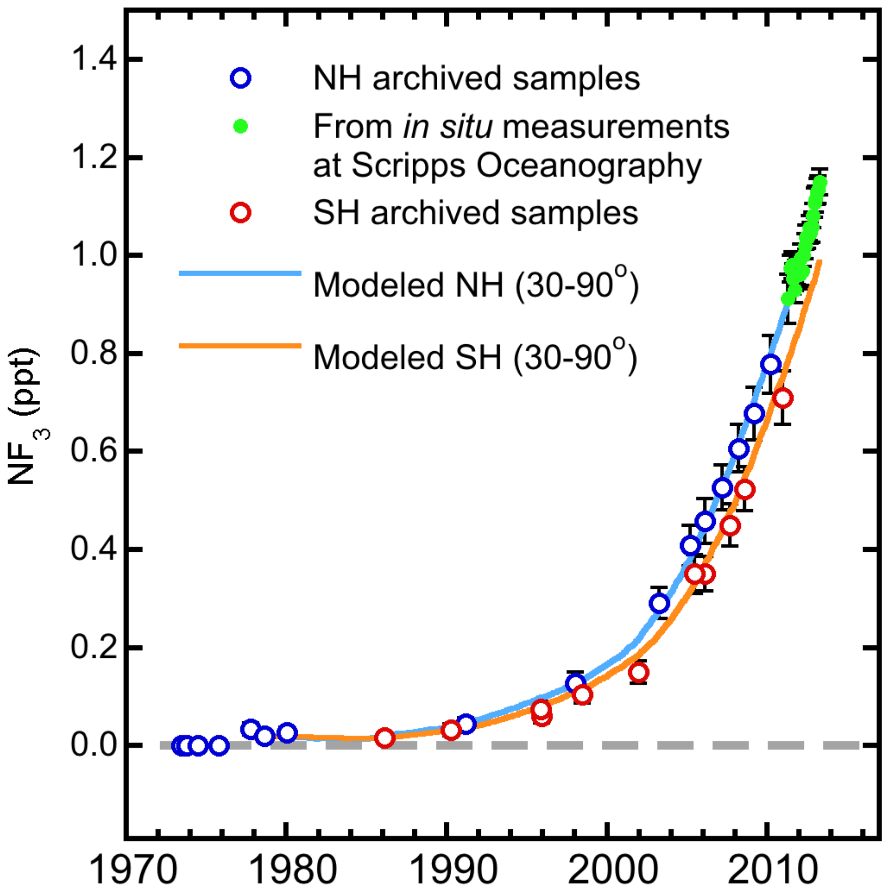
$\mathbf{x} = (x_1 \quad \cdots \quad x_n)$ is the state vector

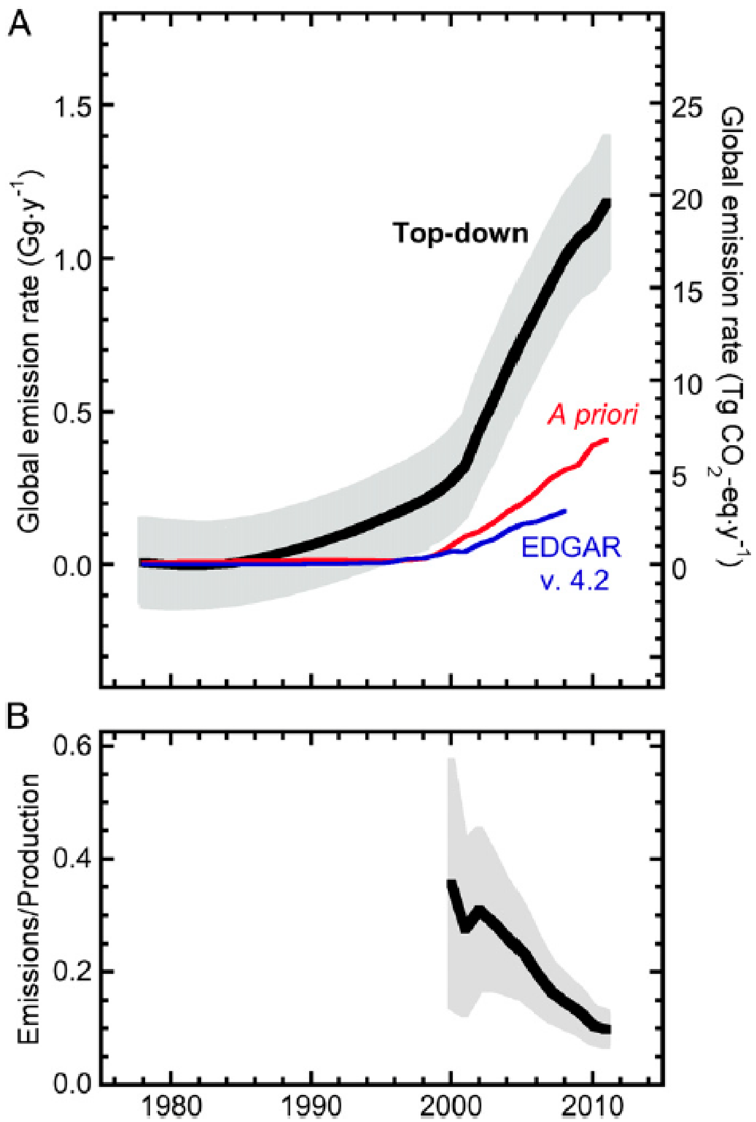
$\mathbf{M}$ is the sensitivity matrix (with dimension m x n)

$$\mathbf{M} = \begin{pmatrix} M_{1,1} & \cdots & M_{1,n} \\ \vdots & \ddots & \vdots \\ M_{m,1} & \cdots & M_{m,n} \end{pmatrix}$$

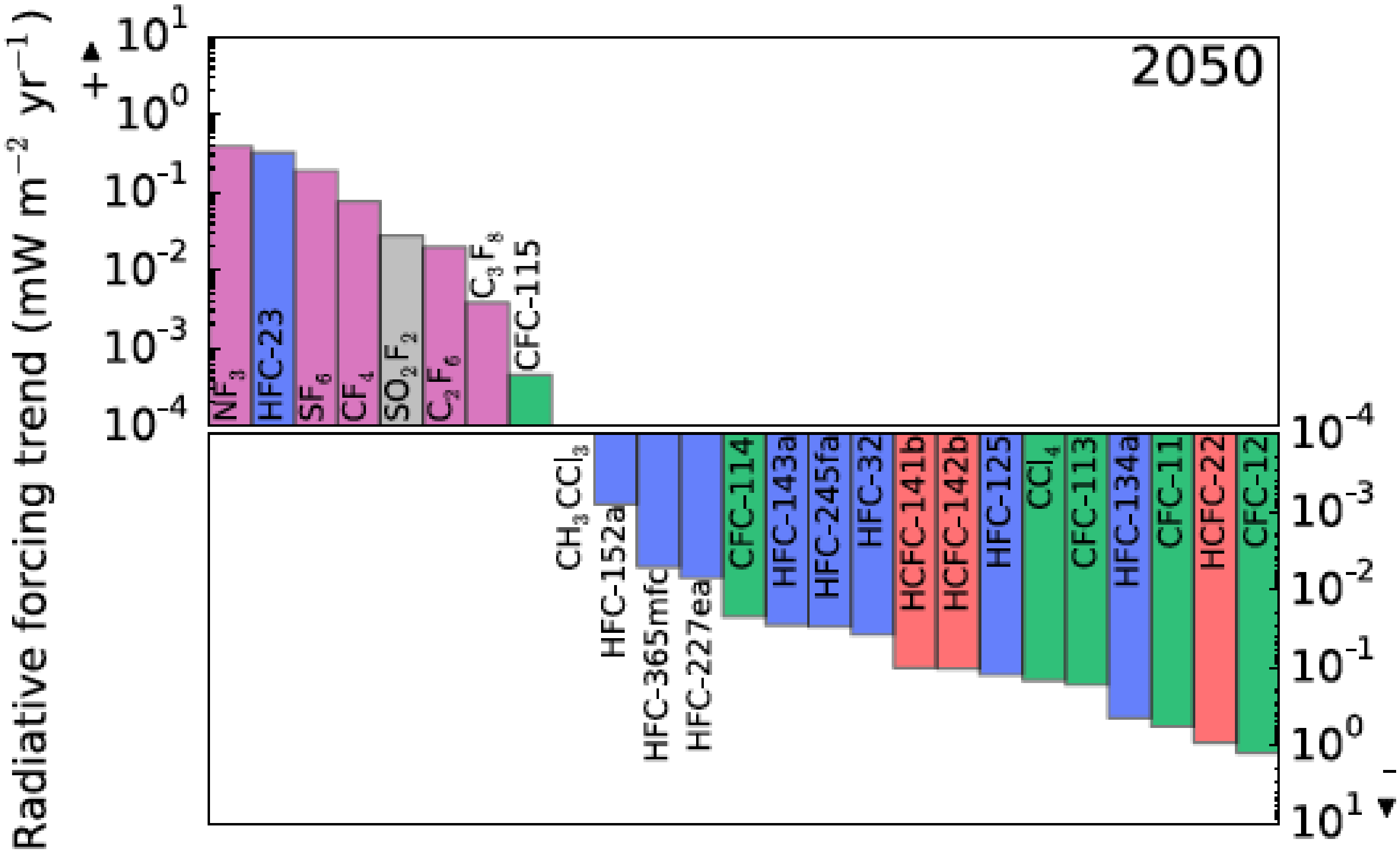
$$J = \frac{1}{2}(\mathbf{x} - \mathbf{x}_\mathrm{b})^\mathrm{T}\mathbf{B}^{-1}(\mathbf{x} - \mathbf{x}_\mathrm{b}) + \frac{1}{2}(\mathbf{M}\mathbf{x} - \mathbf{y}_\mathrm{o})^\mathrm{T}\mathbf{R}^{-1}(\mathbf{M}\mathbf{x} - \mathbf{y}_\mathrm{o})$$

Atmospheric measurements in verification





Arnold et al., 2012, *Analytical Chemistry*
Arnold et al., 2013, *PNAS*



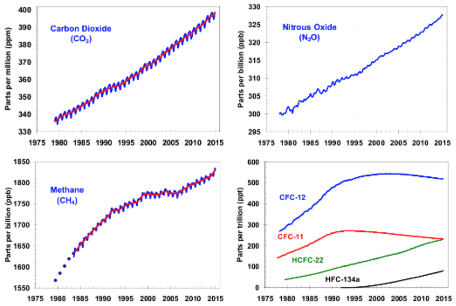
Rigby et al., 2014, *GRL*

Atmospheric monitoring between the source and the well-mixed atmosphere

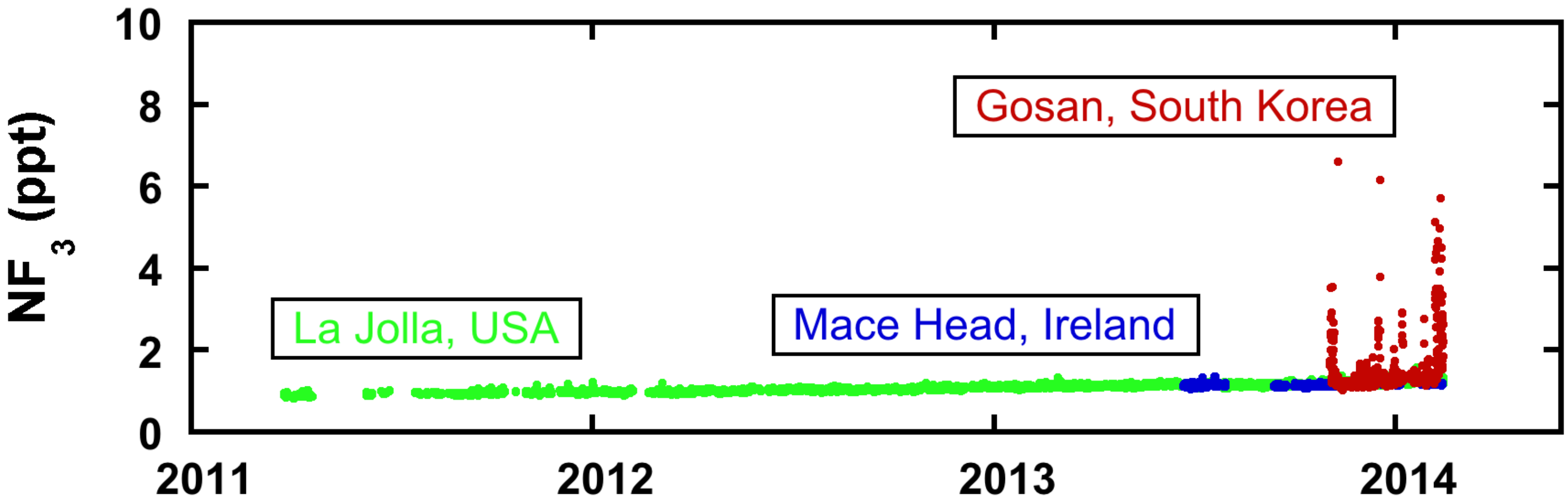
↔

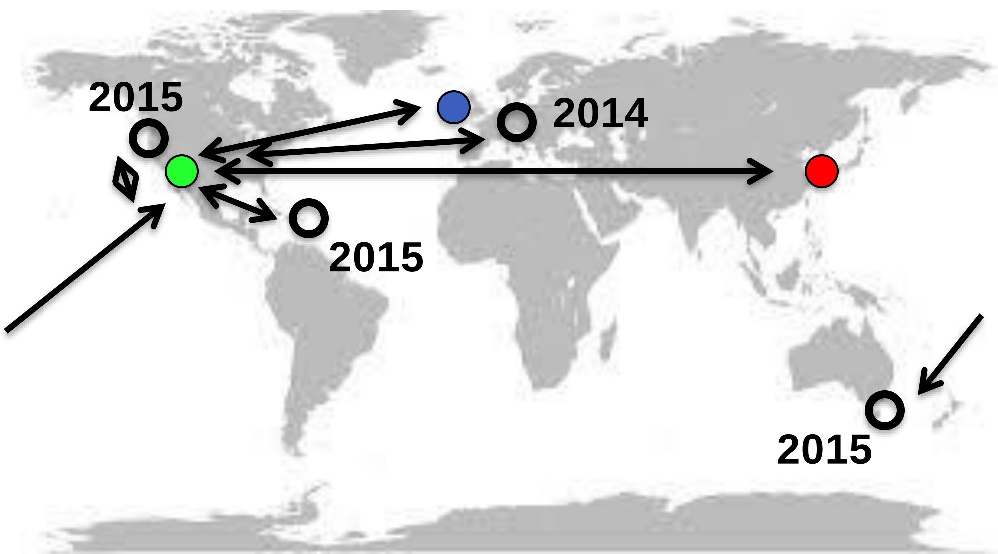
Understanding emission sources over 10-10^2-10^3 km **using continuous measurement methods**

↔



Higher spatial coverage



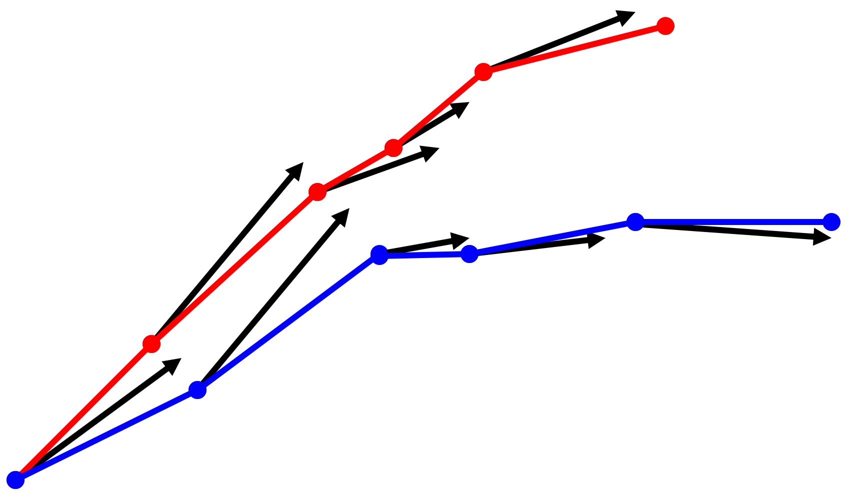


AGAGE NF_3 measurement network

Calibration and intercomparison – vital exercises in high-precision networks

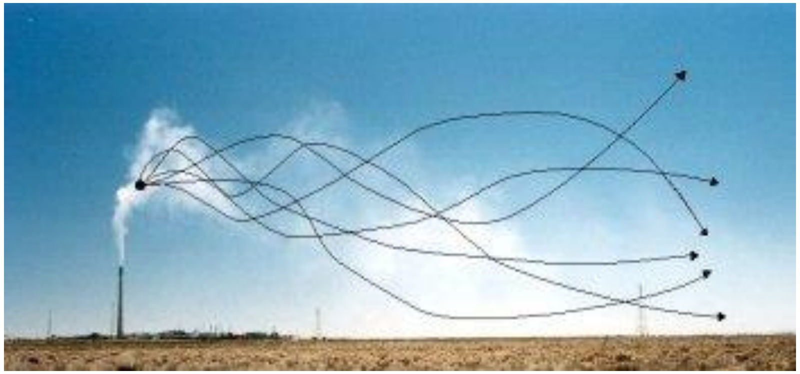
AGAGE now making 20,000 calibrated measurements per year globally

The tool of choice – Lagrangian particle dispersion models

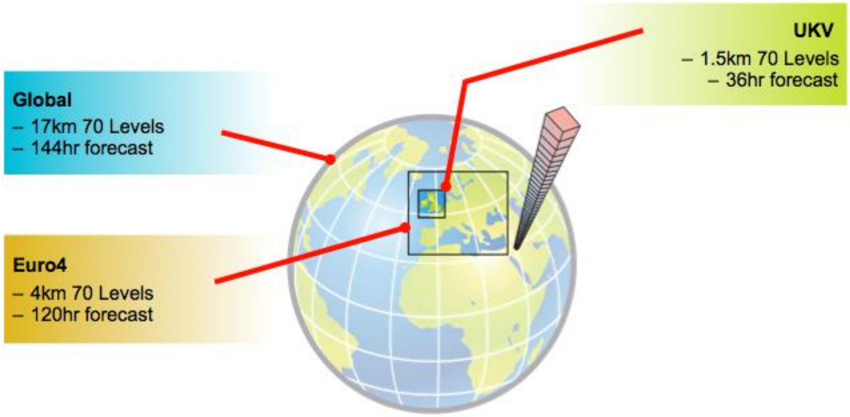


Black arrows show wind direction

Red and blue lines show movement of particles

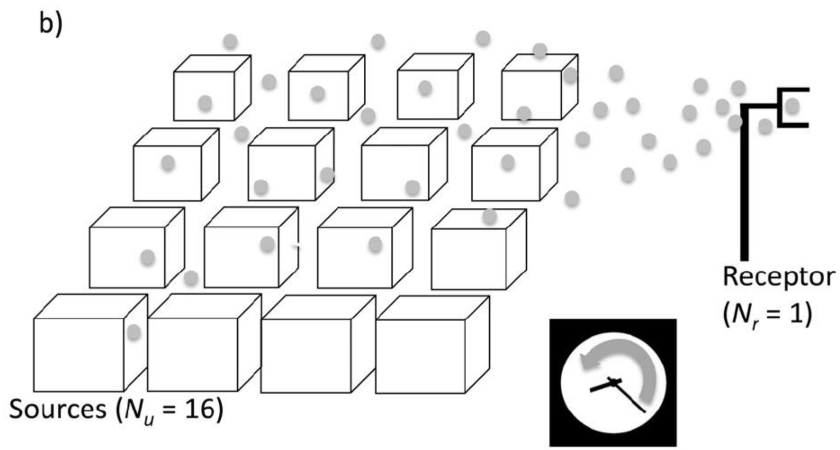


Met Office NAME model



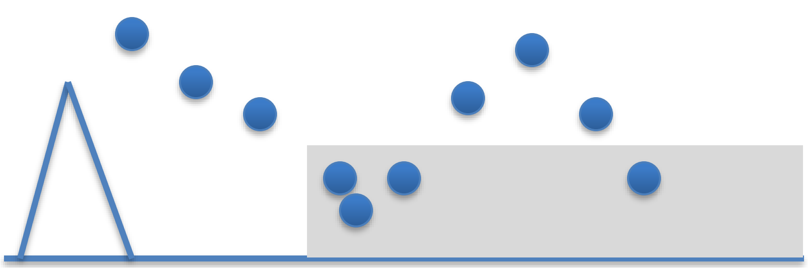
Met Office Unified Model (NWP)

Modelling of atmospheric mixing ratios at a regional scale



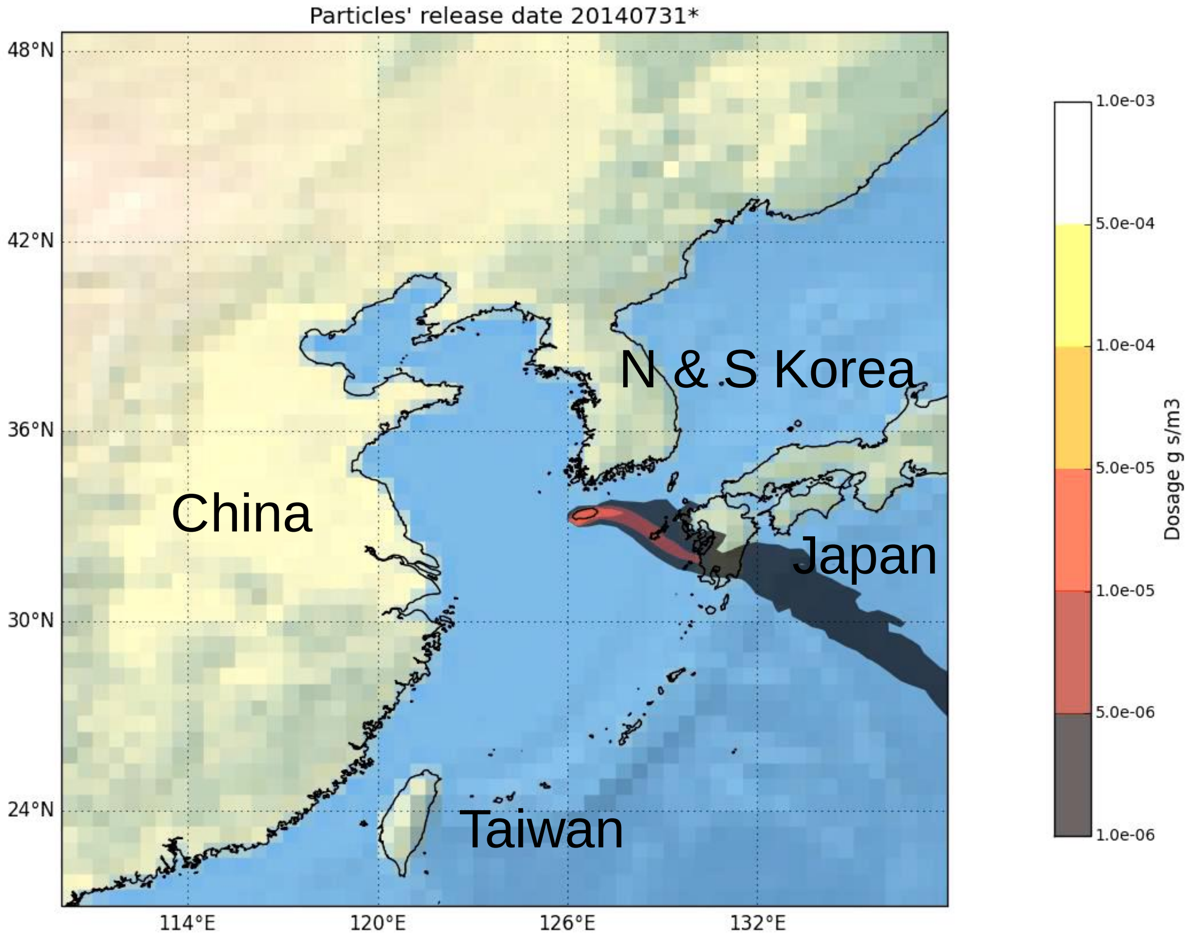
(Lin, *Lang. Mod. Atmos.*, 2012)

We release 20,000 **inert** particles over one hour from the measurement site and follow backwards for 30 days



→ Time integrated concentration in each grid box

Modelling of atmospheric mixing ratios at a regional scale

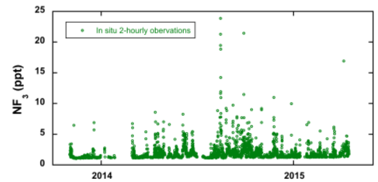


→ Sensitivity matrix

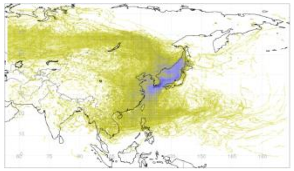
The effect of emissions from a specific grid to a concentration change at the receptor

Combining model and observation data

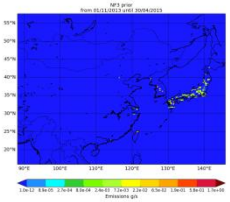
Measurements (*y*)

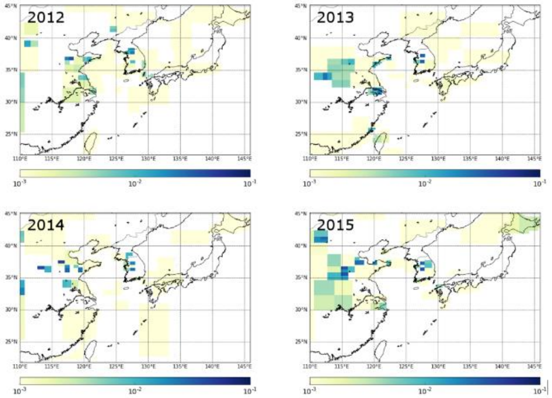


Sensitivity matrix (M)



Prior estimate (x_b)

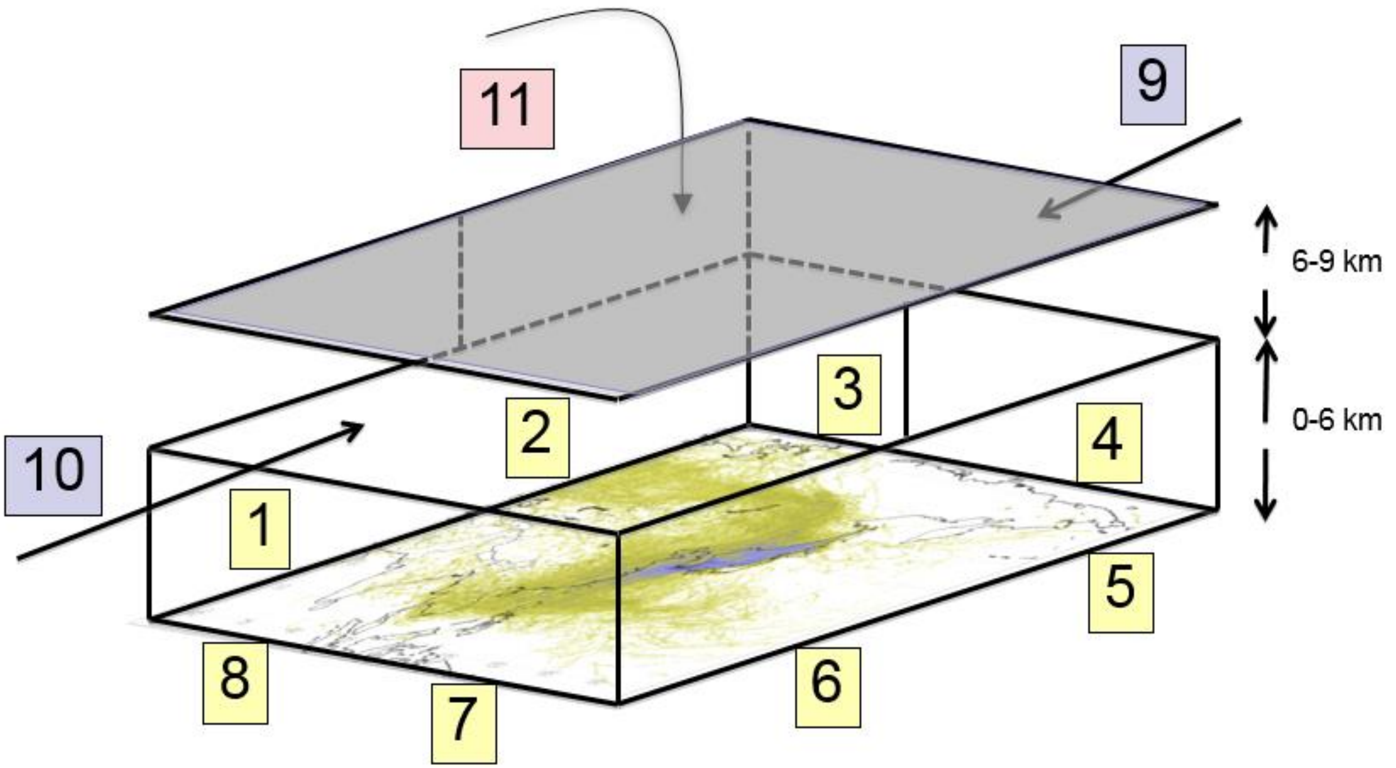




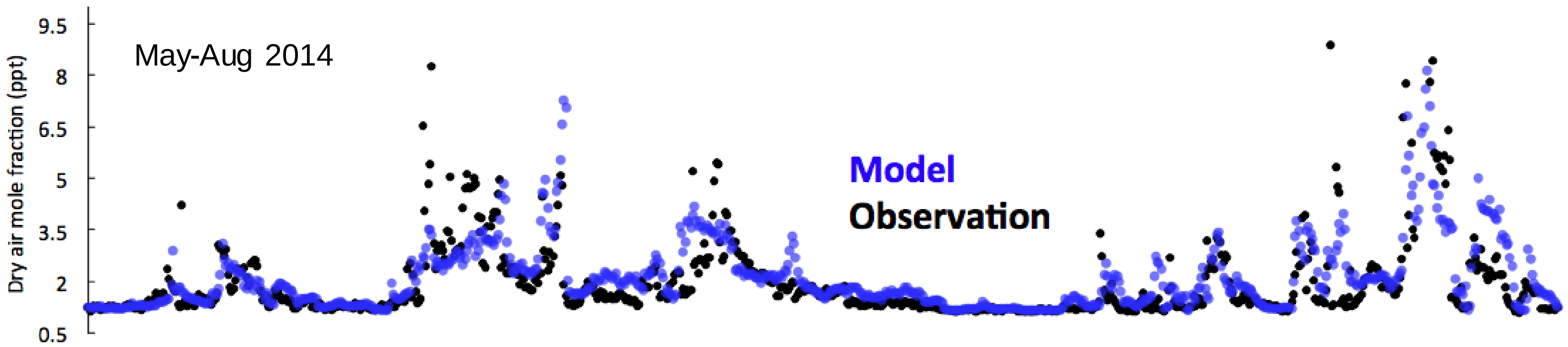
Posterior estimates (*x*)

$$J = \frac{1}{2}(\mathbf{x}-\mathbf{x}_\mathrm{b})^\mathrm{T}\mathbf{B}^{-1}(\mathbf{x}-\mathbf{x}_\mathrm{b}) + \frac{1}{2}(\mathbf{M}\mathbf{x}-\mathbf{y}_\mathrm{o})^\mathrm{T}\mathbf{R}^{-1}(\mathbf{M}\mathbf{x}-\mathbf{y}_\mathrm{o})$$

Accounting for the variable/uncertain background atmospheric contribution



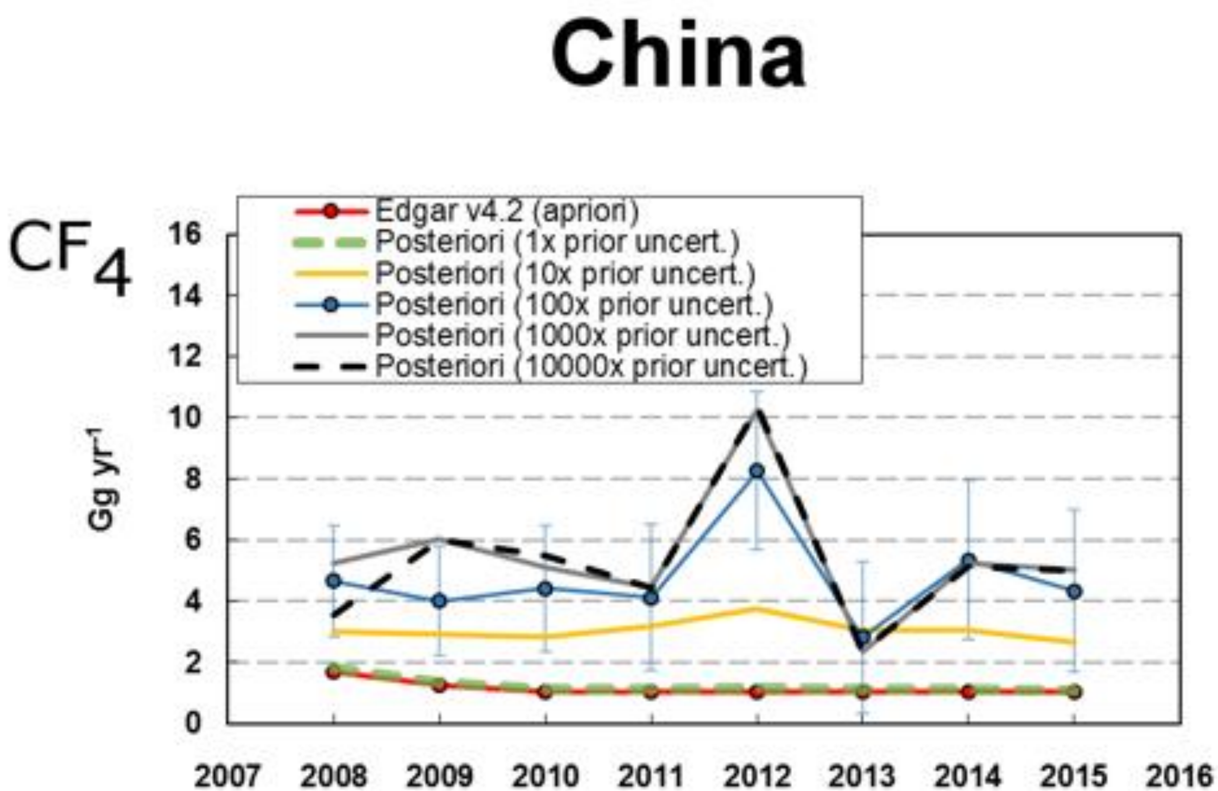
The biggest issue is in understanding how well the model behaves at each measurement time



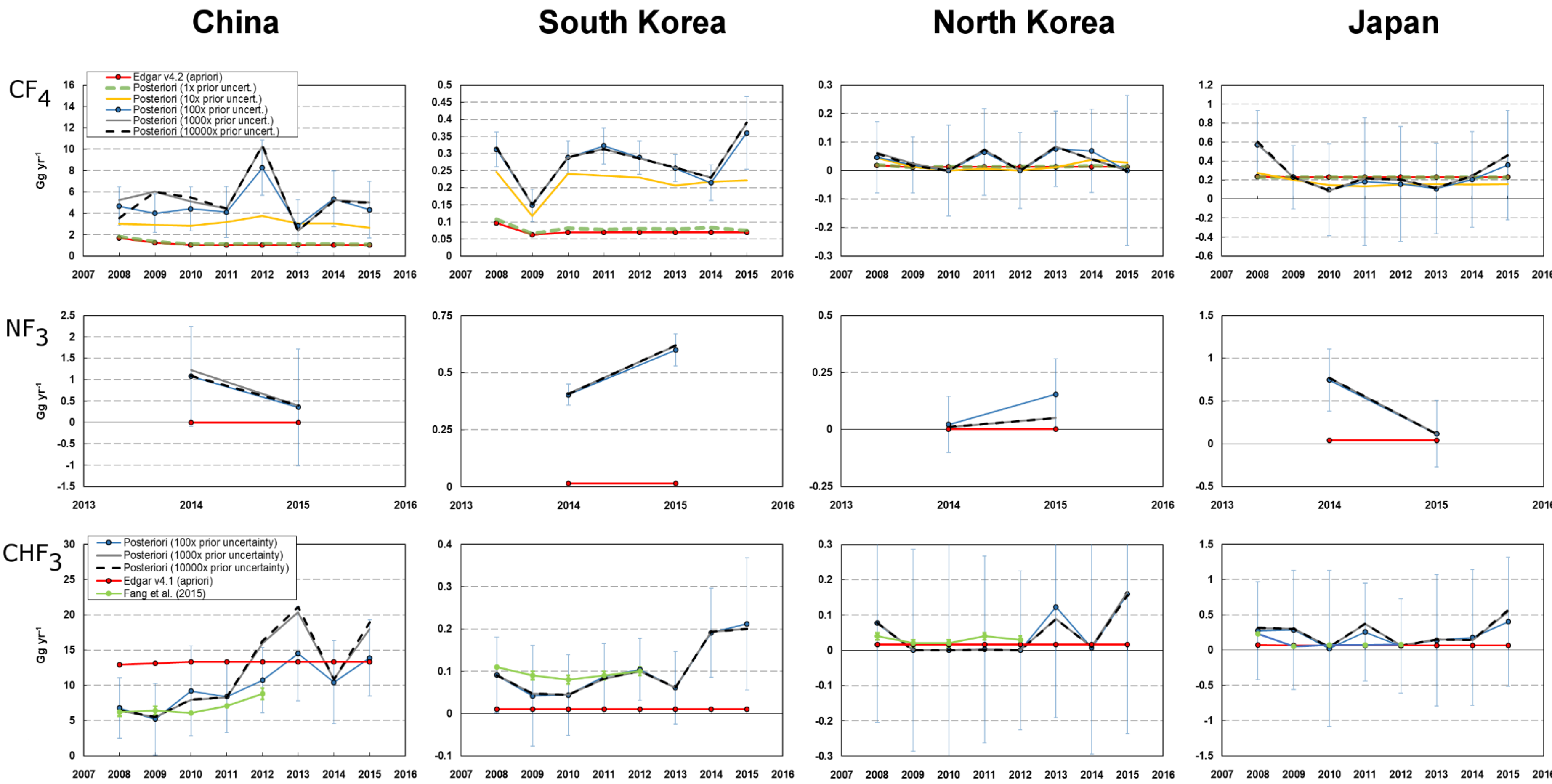
$$J = \frac{1}{2}(\mathbf{x} - \mathbf{x}_\mathrm{b})^\mathrm{T}\mathbf{B}^{-1}(\mathbf{x} - \mathbf{x}_\mathrm{b}) + \frac{1}{2}(\mathbf{Mx} - \mathbf{y}_\mathrm{o})^\mathrm{T}\mathbf{R}^{-1}(\mathbf{Mx} - \mathbf{y}_\mathrm{o})$$

$$\sigma_{model} = \sigma_{baseline} \times factor_{BLH} \times factor_{Topography} \times factor_{Inlet}$$

.. And how well is prior information understood?

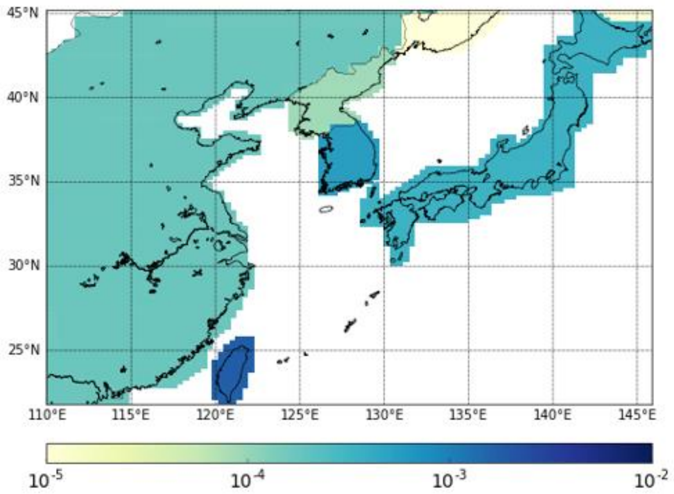


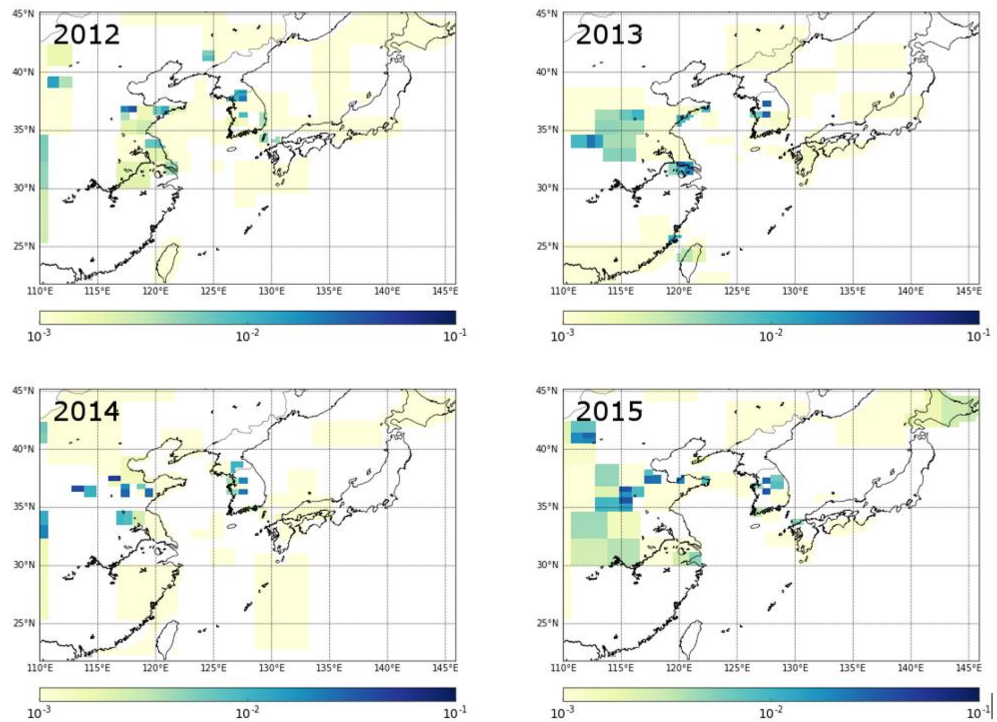
in prep

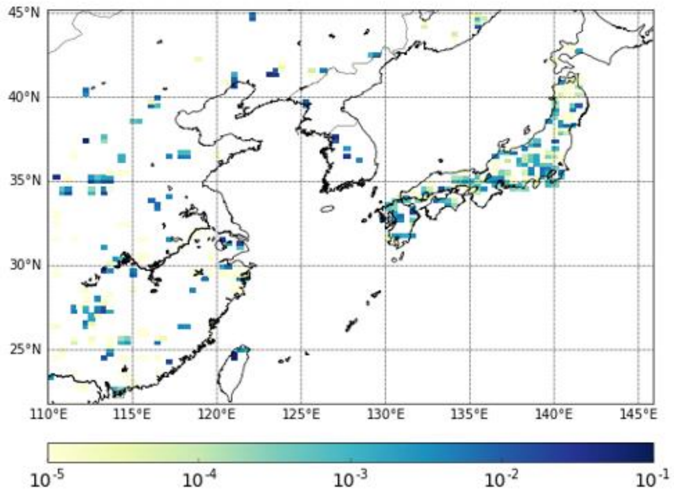

China
South Korea
North Korea
Japan
CF4
NF3
CHF3
Gg yr-1
Edgar v4.2 (apriori)
Posteriori (1x prior uncert.)
Posteriori (10x prior uncert.)
Posteriori (100x prior uncert.)
Posteriori (1000x prior uncert.)
Posteriori (10000x prior uncert.)
Posteriori (100x prior uncertainty)
Posteriori (1000x prior uncertainty)
Posteriori (10000x prior uncertainty)
Edgar v4.1 (apriori)
Fang et al. (2015)

In prep

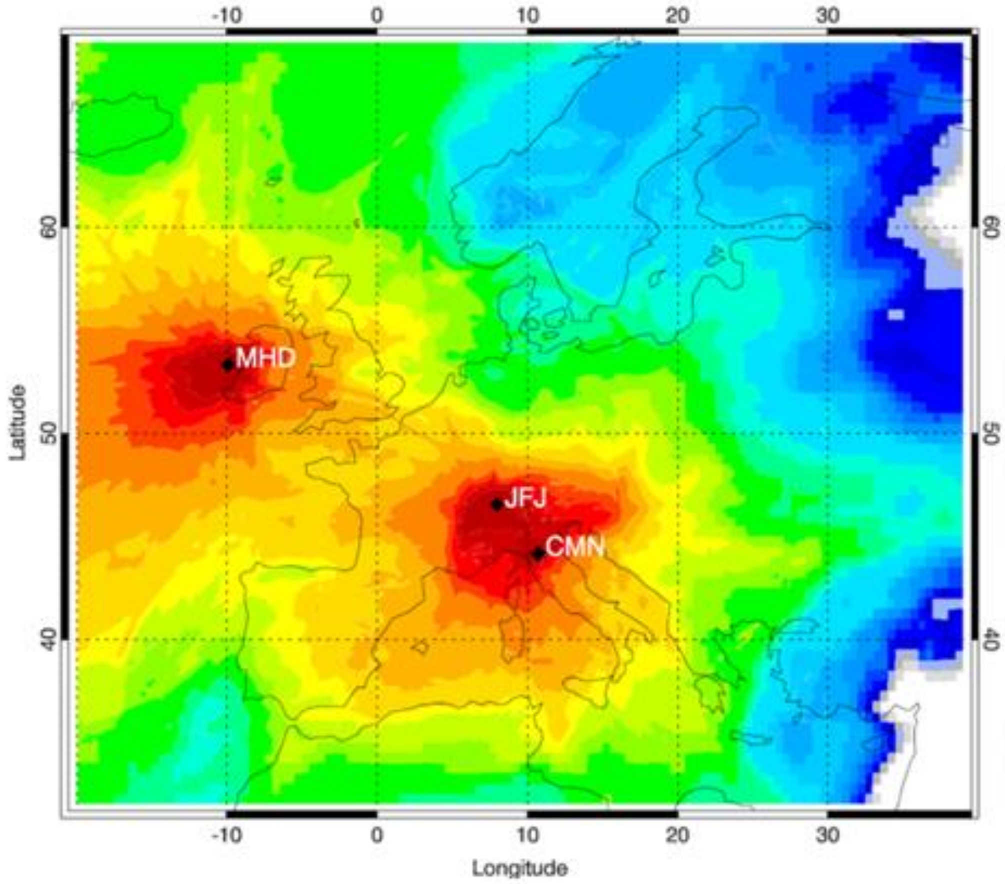
Uncertainty at higher spatial resolution







Model intercomparison project for SF_6, HFC-125 and HFC-134a



Brunner, Arnold et al., 2017, ACPD

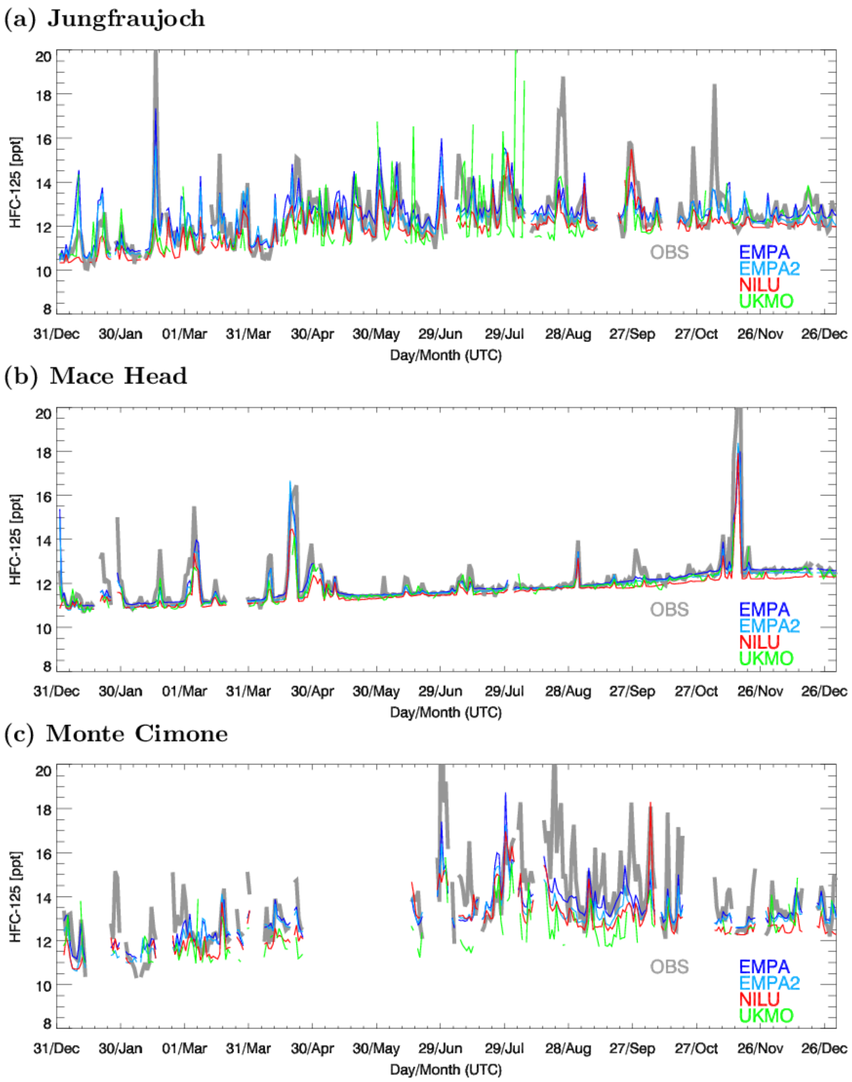
Model intercomparison project

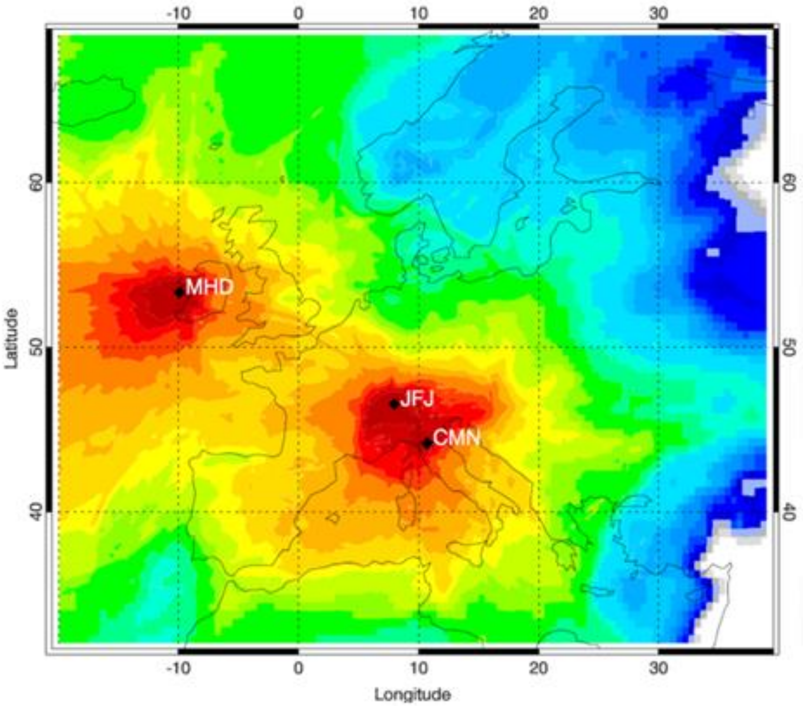
Table 1: Overview of inversion systems

Characteristic	EMPA	EMPA2	NILU	UKMO
Approach	Extended Kalman Filter (ExKF)	Bayesian	Bayesian	Bayesian
Transport model	FLEXPART	FLEXPART	FLEXPART	NAME
Meteorology	ECMWF analyses 0.2°x0.2°, 3hrly	ECMWF analyses 0.2°x0.2°, 3hrly	ECMWF analyses 0.2°x0.2°, 3hrly	UKMO analyses 0.352° x 0.234°, 3hrly
Computational domain	Nested, global	Nested, global	Nested, global	45°W - 40°E, 25°N - 80°N
Inversion grid	0.1°x0.1°min., reduced according to residence time	0.1°x0.1°min., reduced according to residence time	1°x1° over land, reduced over ocean and far eastern boundary	0.352° x 0.234° min., reduced acc. to residence time and within country boundaries
State vector length (e=emiss., b=background, o=other)	1092 (1083*e* + 3*b* + 6*o*)	M1: 522 e + 84 b M2: 522 e + 84 b M3: 405 e + 56 b FLAT: 522 e + 56 b	M1: 1140e M2: 1140e M3: 1140e FLAT: 1140e	~150e + 11 b
Assimilation time resolution	3-hourly means	3-hourly means	Daily means	3-hourly means once per day
Spatial correlation of prior	500 km	None	200 km over land 1000 km over sea	None
Backwards mode run time	5 days	5 days	10 days	19 days
Prior background mole factions	None, continuously estimated by ExKF	60-day REBS window, biweekly reference points	See Thompson and Stohl (2014) and description below.	Mace Head baseline for all sites, see Manning et al. (2011)
Temporal correlation of observation error	Red-noise Kalman filter	None	None, assumed negligible for daily means	None, assumed negligible with one value per day
Key references	Brunner et al., 2012	Stohl et al., 2009, Vollmer et al., 2009	Thompson and Stohl, 2014	Manning et al., 2011

Brunner, Arnold et al., 2017, ACPD

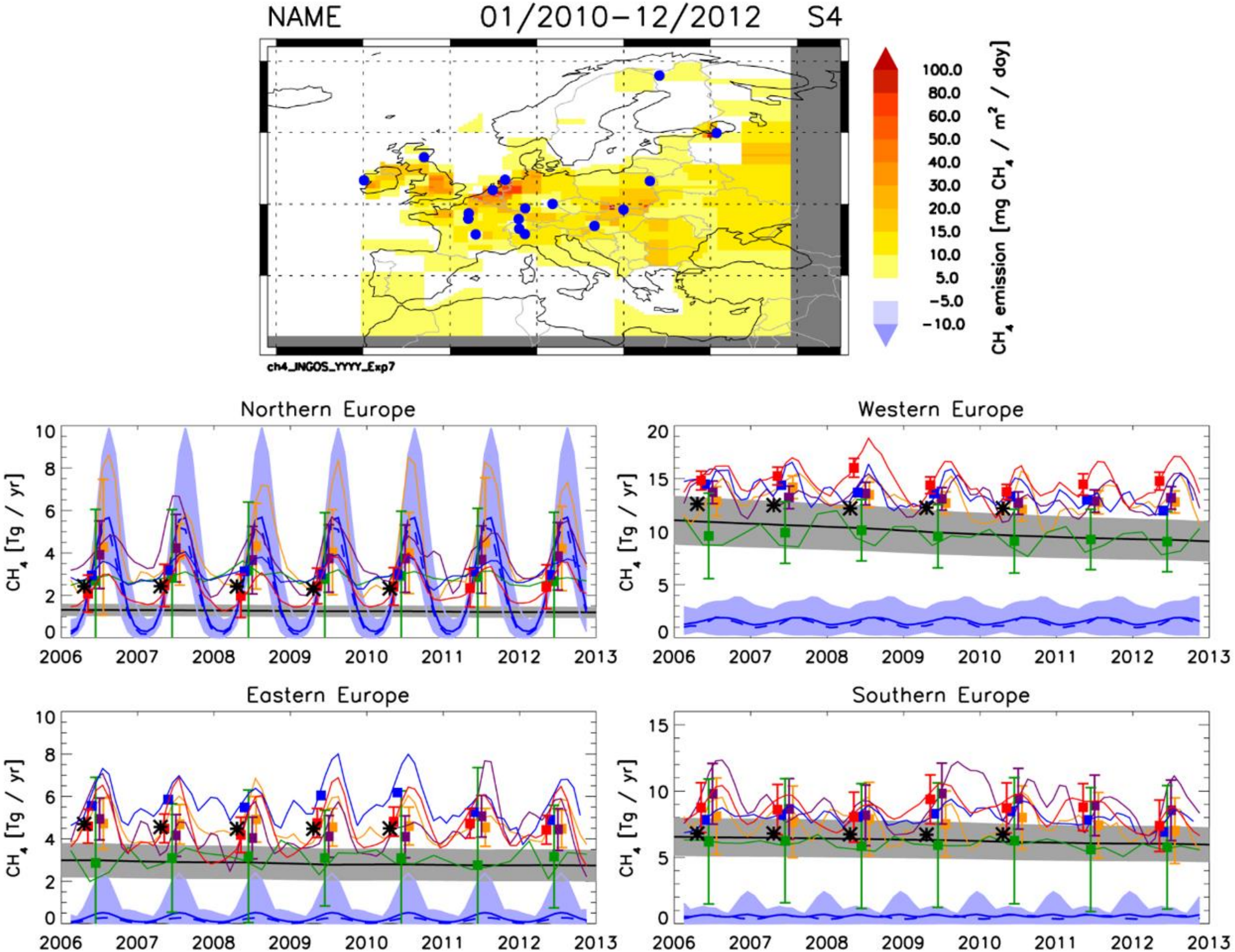
Model intercomparison project





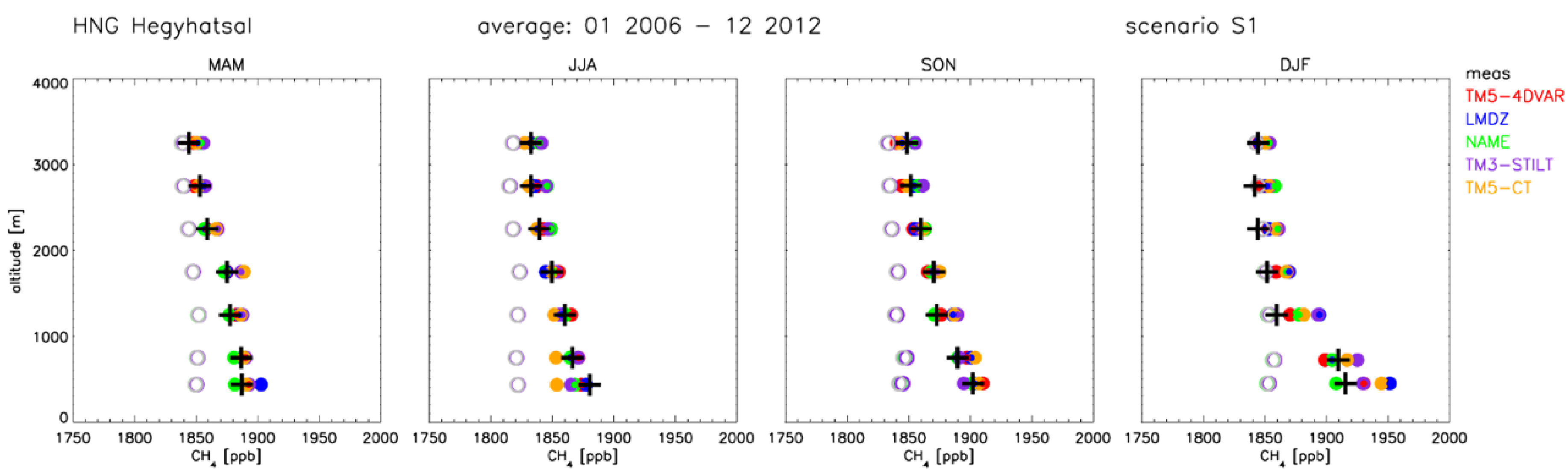
Brunner, Arnold et al., 2017, ACPD

European methane model intercomparison project



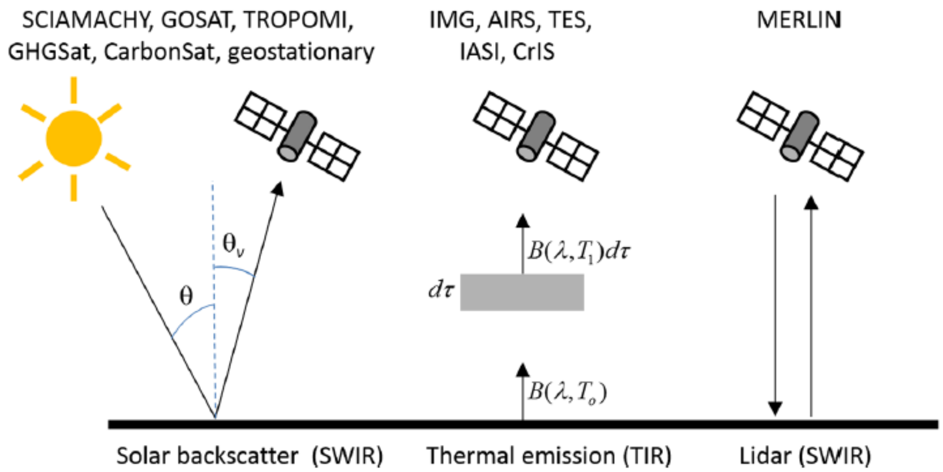
Bergamaschi et al., submitted

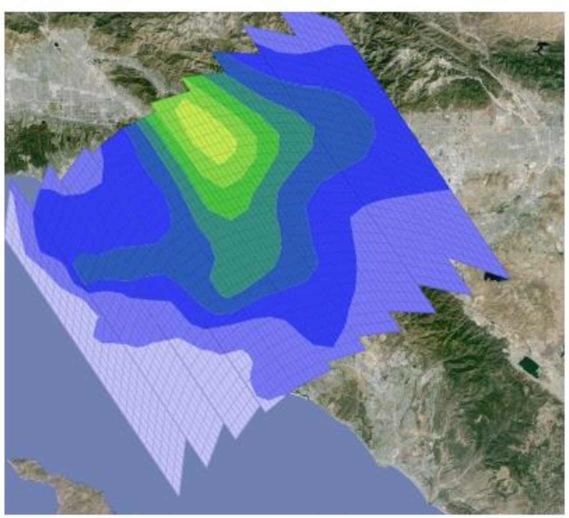
Model validation using aircraft measurements



Bergamaschi et al., submitted

Need to prepare for satellite observations





Launch ~2018
Column CO_2 measurements over cities

http://www.jpl.nasa.gov/missions/orbiting-carbon-observatory-3-oco-3

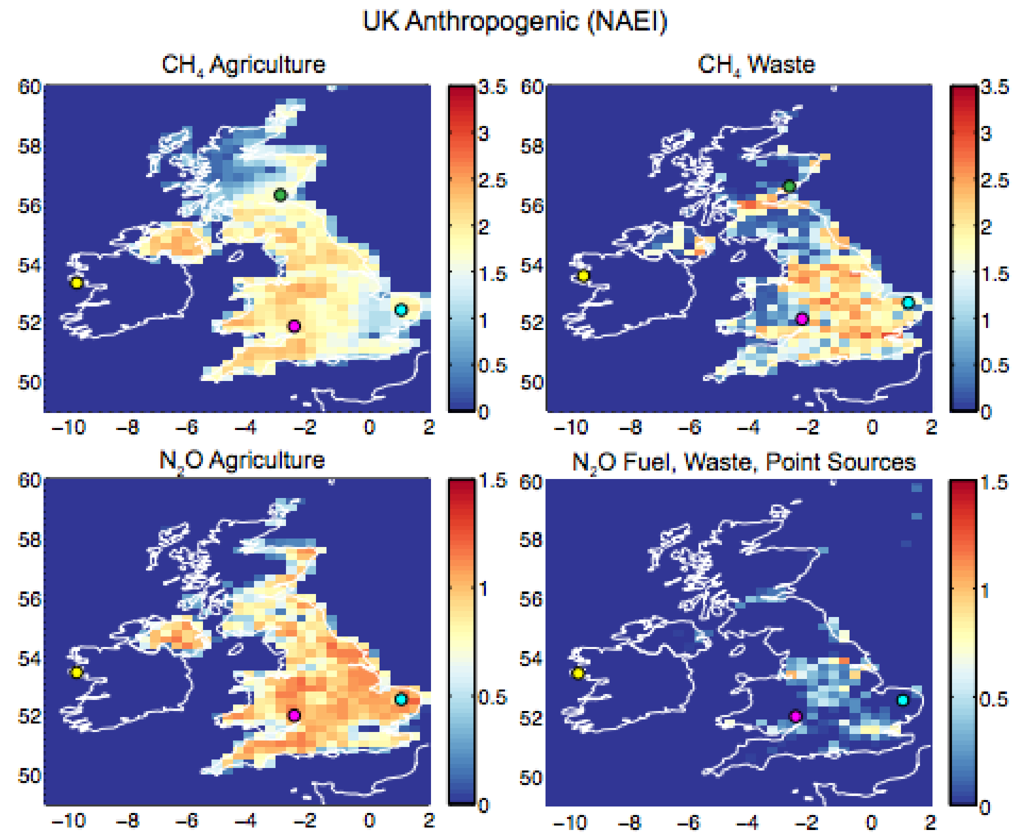
Nitrous oxide and methane have complex mixed source emissions

<u>CH_4</u>

- Wetlands (microbial)
- Fossil fuels
- Biomass burning
- Landfills
- Ruminants
- Rice cultivation

<u>N_2O</u>

- Soils
- Ocean
- Biomass burning
- Sewage
- Fossil fuel burning



Ganesan et al., *ACP*, 2015

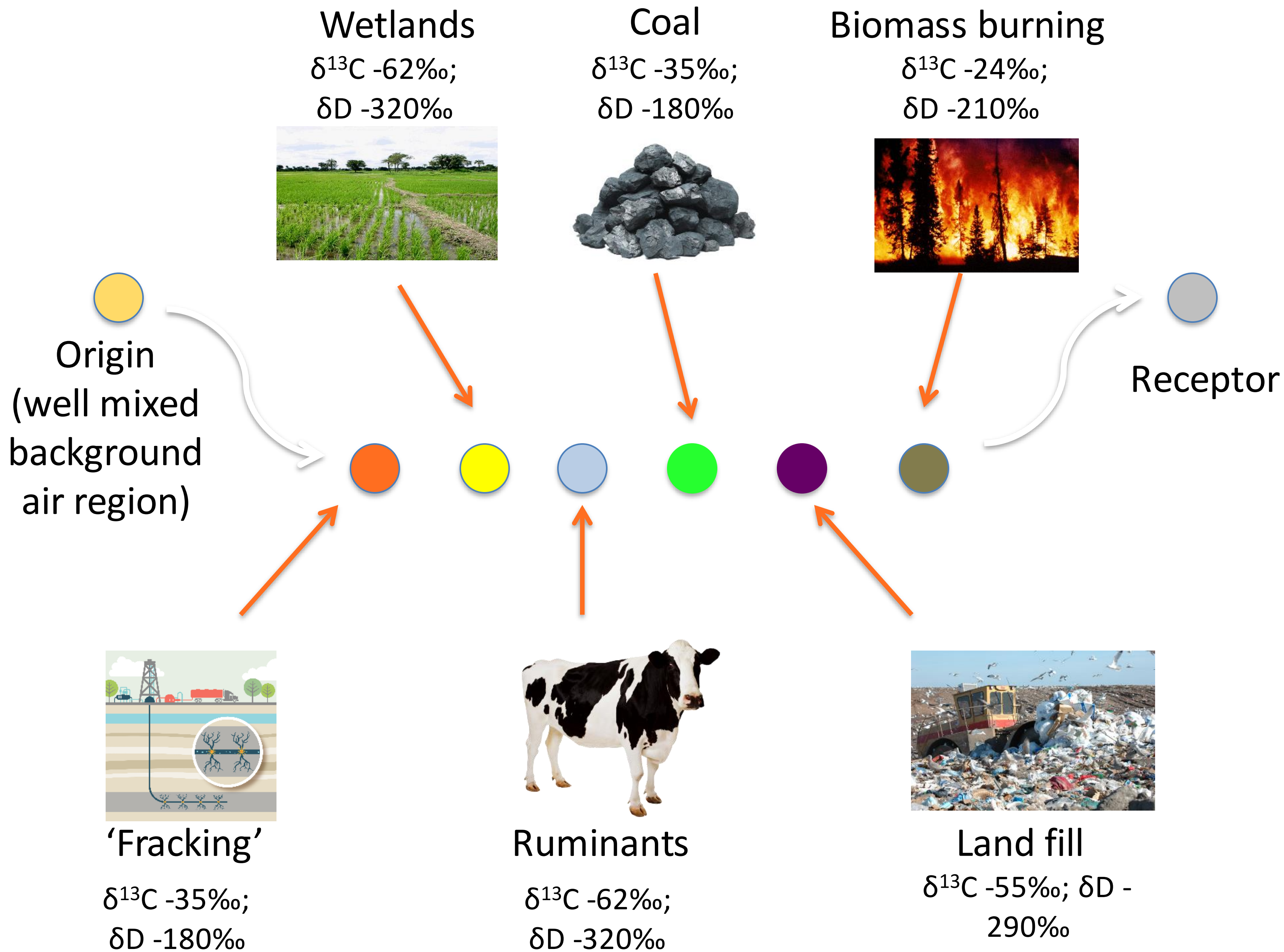
A large range of CH_4 emissions:

1.65–2.67 Tg yr−1 (Ganesan et al., 2015)

1.7–3.0 Tg yr−1 (MO for DECC)

1.8–3.0 Tg yr−1 (Bergamaschi et al., *in prep*)

Methane sources have different isotopic signatures



Laboratory development of isotopologue measurements

CH_4 made of $^{12}CH_4$, $^{13}CH_4$, $^{12}CH_3D$.. and 7 more!

Preconcentration Technique

Laser spectroscopy