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Resolving Discrepancies in High-GWP GHG Emissions Using Atmospheric Measurements: Recent AGAGE Results

"You can't manage what you can't measure."

-- business adage

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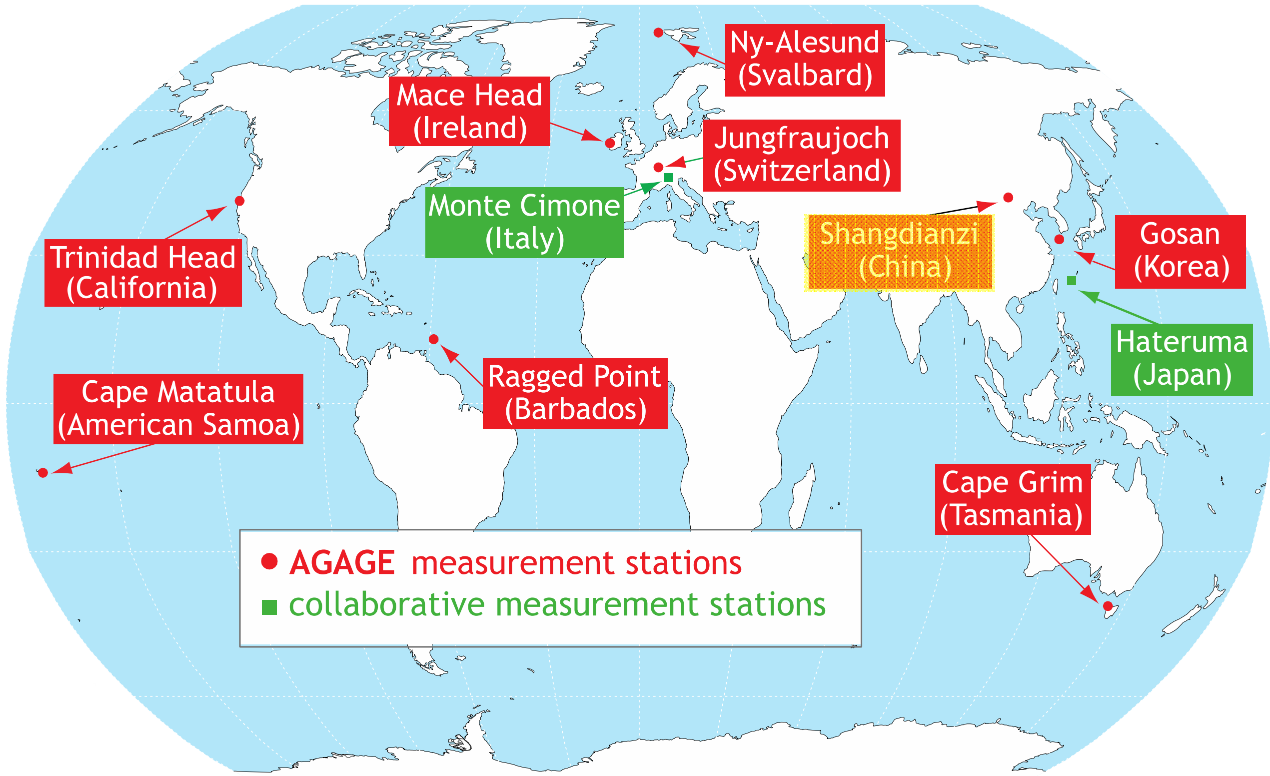
Advanced Atmospheric Gases Experiment (AGAGE)

AGAGE and its predecessors ALE and GAGE have been measuring changes in the composition of the global atmosphere since 1978.

AGAGE measures at high frequency over the globe almost all of the important species regulated by the Montreal Protocol to protect the stratospheric ozone layer, and almost all of the significant non-CO_2 gases in the Kyoto Protocol to mitigate climate change.



AGAGE stations occupy coastal & mountain sites around the world chosen to provide accurate measurements in clean and polluted air of trace gases whose lifetimes are long compared to atmospheric transport times.

AGAGE INSTRUMENTATION

1. Medusa GC-MS
2. GC-Multi-detector
3. Calibration

AGAGE currently measures over 50 environmentally active atmospheric trace gases in clean & polluted air using custom automated gas chromatographic and mass-spectrometric instrumentation.

AGAGE measured species. Medusa in Blue; GC-MD: Green; Both: Red.

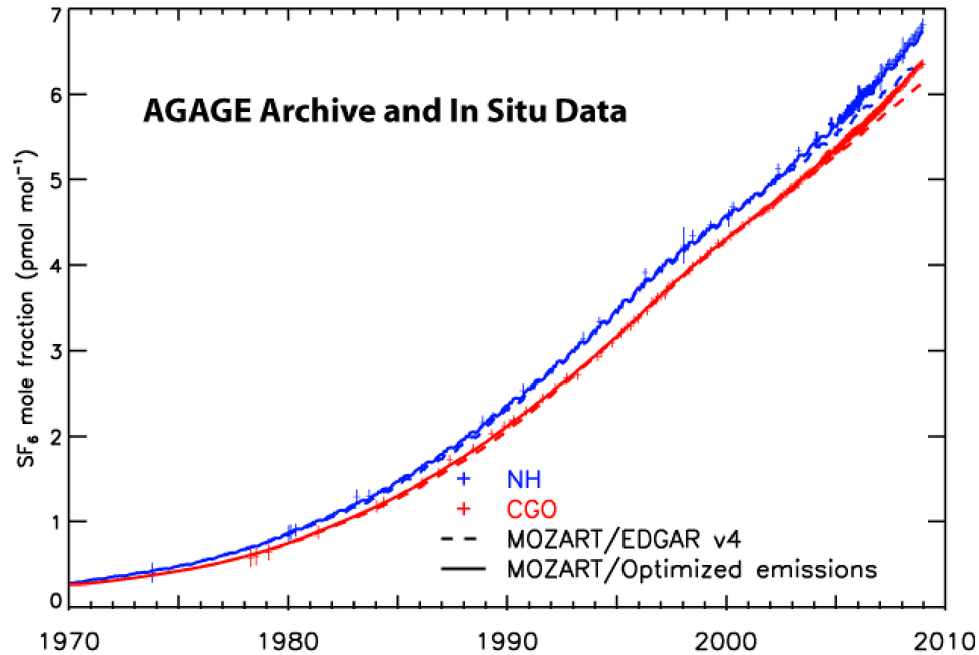
Compound	~NH (2014) (ppt)	Typical % precision	Compound	~NH (2014) (ppt)	Typical % precision
CF_4	81.7	0.15	CFC-114	16.35	0.3
C_2F_6	4.47	1	CFC-115	8.46	0.7
C_3F_8	0.62	3	H-1211	3.88	0.4
$c\text{-}C_4F_8$	1.47	1.5	H-1301	3.38	1.7
C_6F_{14}	0.30	3	H-2402	0.432	2
SF_6	8.46	0.6	CH_3Cl	555	0.2
SF_5CF_3	0.17	7	CH_3Br	7.2	0.6
SO_2F_2	2.12	2	CH_3I	1.0	2
NF_3	1.23	1	CH_2Cl_2	60	0.5
HFC-23	28.0	0.7	CH_2Br_2	1.4	1.5
HFC-32	10.9	3	$CHCl_3$	13	0.4
HFC-134a	83.7	0.5	$CHBr_3$	2	0.6
HFC-152a	9.8	1.4	CCl_4	83	1
HFC-125	17.6	0.7	CH_3CCl_3	3.8	0.7
HFC-143a	17.4	1	$CHCl{=}CCl_2$	0.1-1.0	3
HFC-227ea	1.10	2.2	$CCl_2{=}CCl_2$	1.4-3.5	0.5
HFC-236fa	0.135	10	COS	370-570	0.5
HFC-245fa	2.38	3	C_2H_6	500-2250	0.3
HFC-365mfc	1.00	5	C_3H_8	5-700	0.6
HFC-43-10mee	0.27	3	C_6H_6	10-100	0.3
HCFC-22	240	0.3	C_7H_8	2-40	0.6
HCFC-141b	25.5	0.5			
HCFC-142b	23.4	0.4			
HCFC-124	1.30	2	GC-MD Only	(ppb)	
CFC-11	235	0.2	CH_4	1835-1910	0.2
CFC-12	523	0.1	N_2O	327.5	0.05
CFC-13	3.02	2	* CO	80-140	0.2
CFC-113	73.1	0.2	* H_2	500-540	0.6

*CO and H_2 are measured at Mace Head and Cape Grim Only.
(ppt = parts per trillion, ppb = parts per billion)

Public Data Archive at:
http://cdiac.ornl.gov/ndps/alegage.html

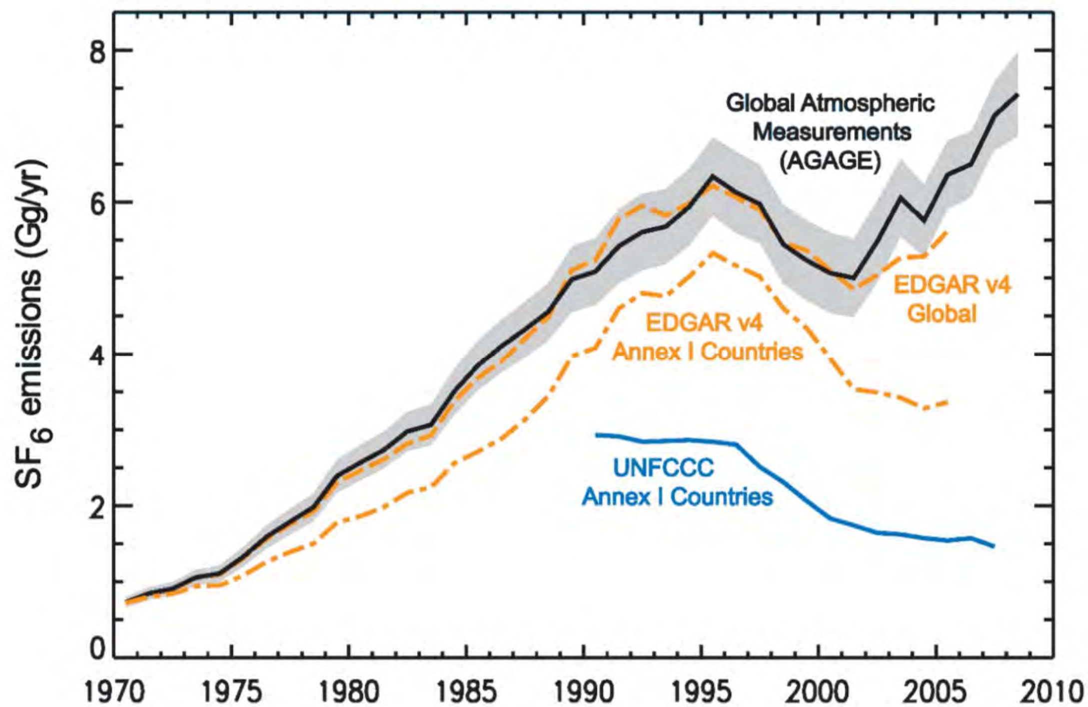
Electrical & electronics industries emit very powerful greenhouse gases

(e.g. SF_6 with GWP = 22,800. SF_6 is used in electrical distribution systems.)



Deduced changes in SF_6 emissions from 1971 onwards using AGAGE and NOAA *in situ* data, and AGAGE archive tank, data.

Rigby *et al.*, *Atmos. Chem. Phys.*, 2010



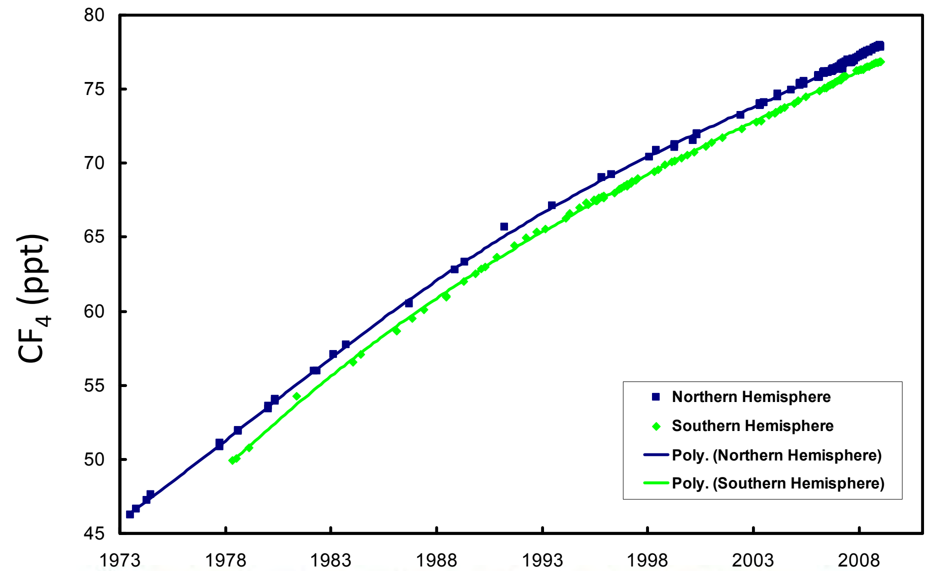
Aluminum and electronics industries also emit very powerful PFC greenhouse gases

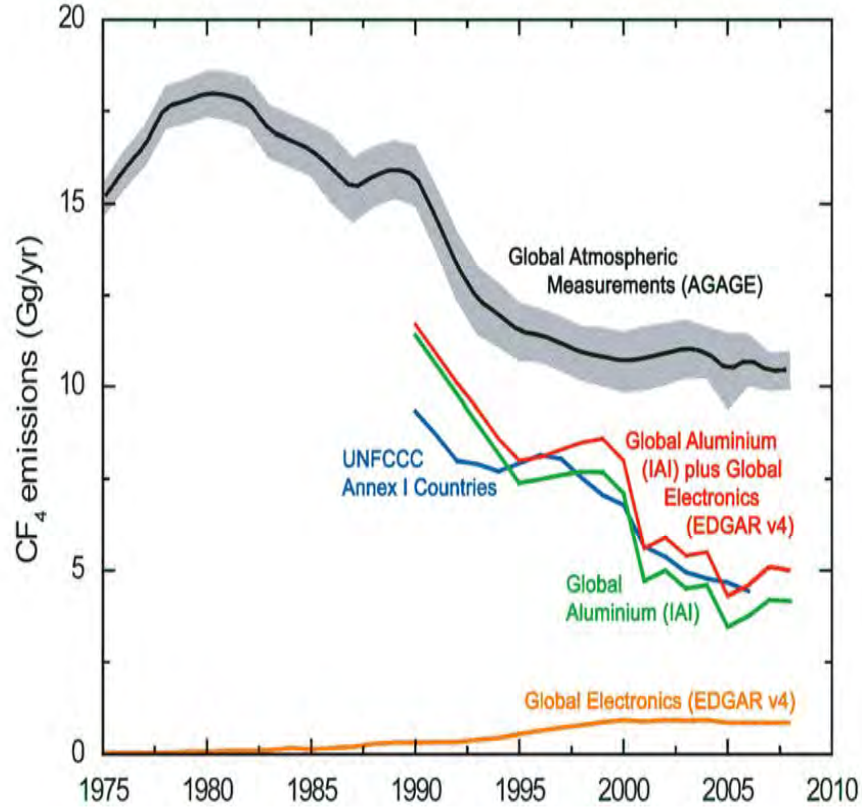
(e.g. CF_4 with GWP = 7400).

AGAGE In Situ & Archive measurements show rapid but decelerating rise.

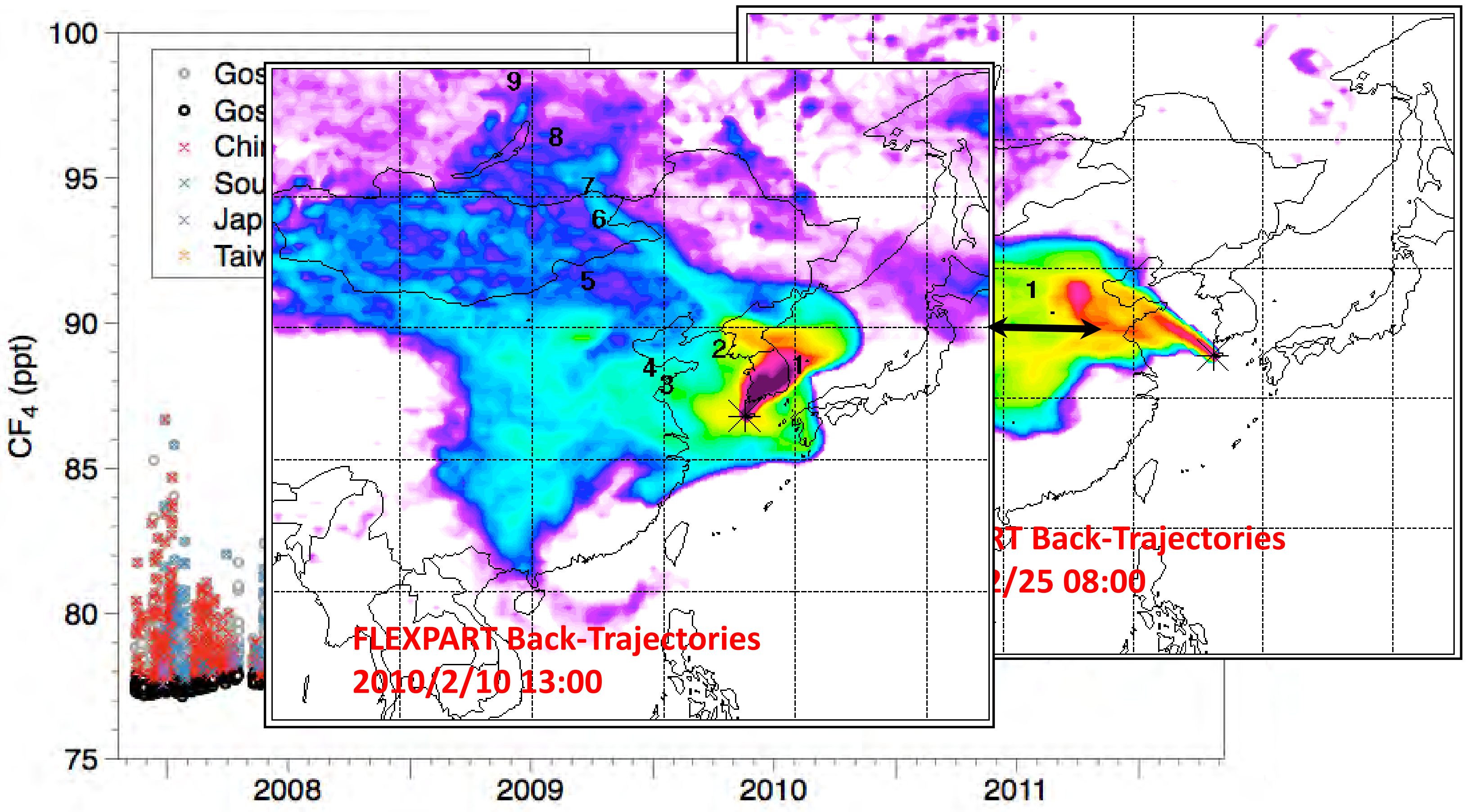
Deduced emissions for CF_4 between 1975 & 2009 from atmospheric measurements show post-1980 decrease driven by improved technology.

Mühle *et al.*, *Atmos. Chem. Phys.*, 2010





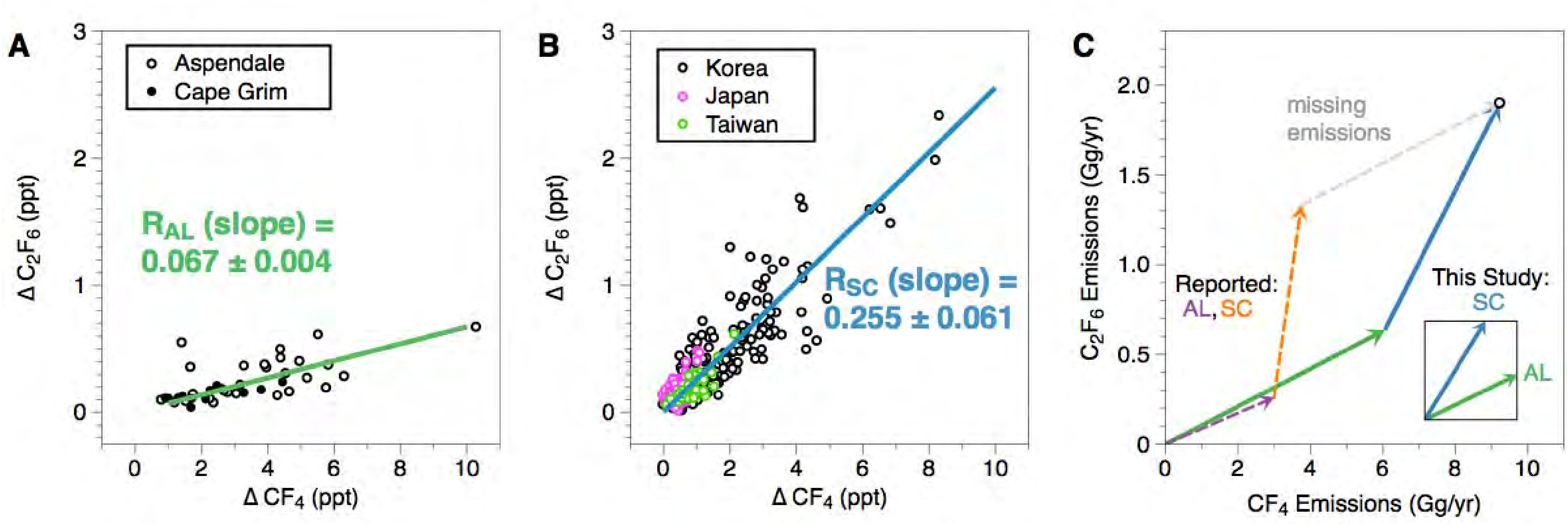
Analyzing Pollution Events



Japan, Taiwan events occur in spring, fall seasons

J. Kim *et al.,* Geophys. Res. Lett., 2014

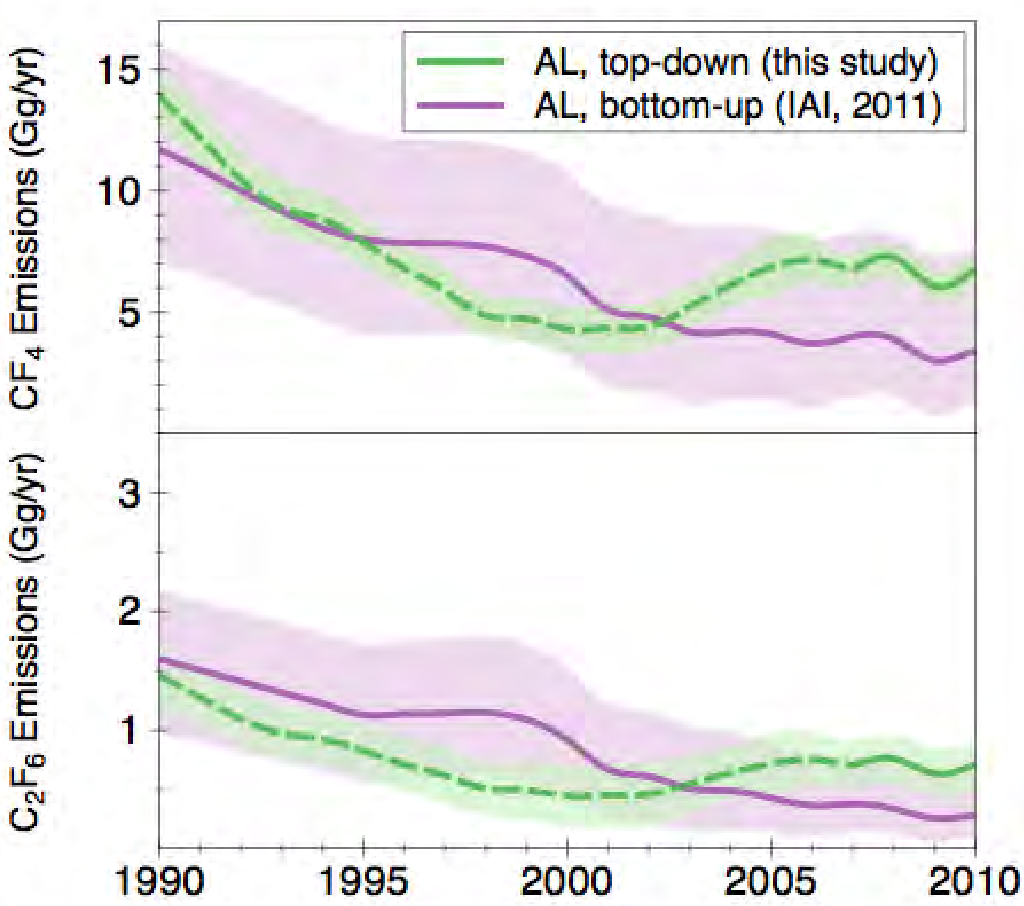
PFC Emission Ratios by Industry



If these examples are typical, industry-specific emission ratios can be used to decompose the global total PFC emissions to separate industry-specific totals.

J. Kim *et al.,* Geophys. Res. Lett., 2014

Aluminum Industry (AL) Top-Down vs. Bottom-Up

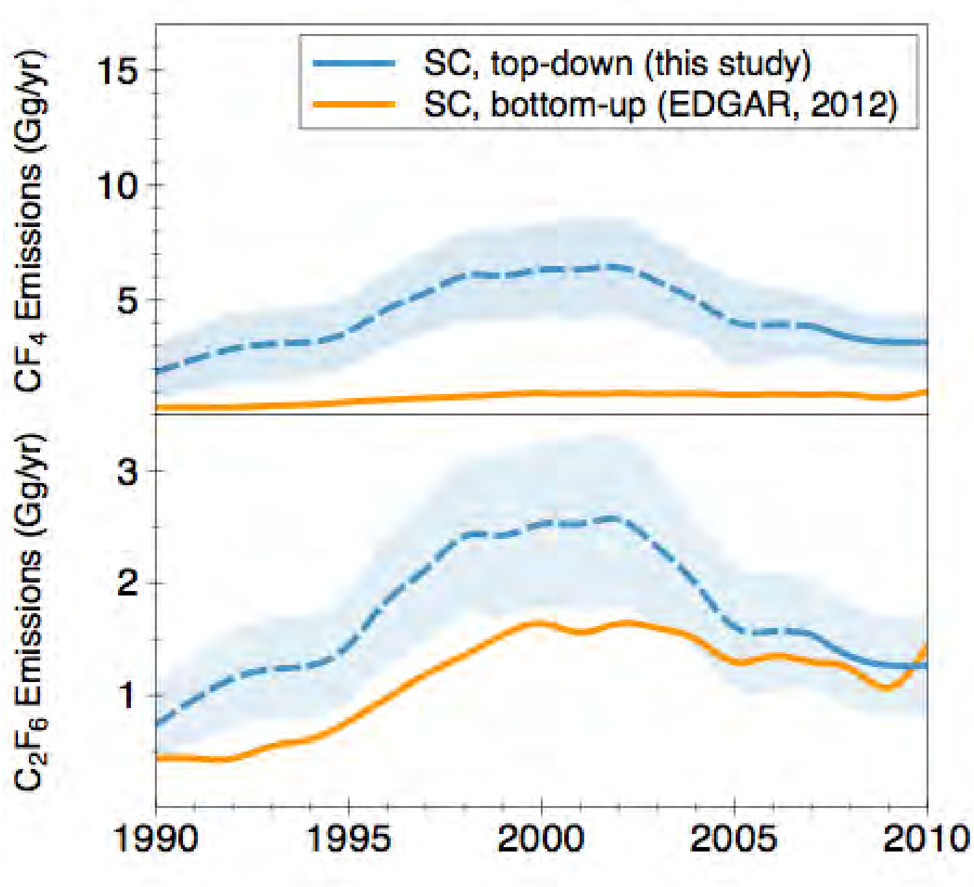


Aluminum industry global emissions agree within estimated uncertainties, but this masks important differences and trends.

J. Kim *et al.*, Geophys. Res. Lett., 2014

AGAGE

Semiconductor Industry (SC) Top-Down vs. Bottom-Up



Semiconductor industry global emissions discrepancies are significant, especially during the 1995-2005 decade and especially for CF_4.

PFC emissions from this industry are complicated and difficult to quantify.

J. Kim *et al.,* Geophys. Res. Lett., 2014

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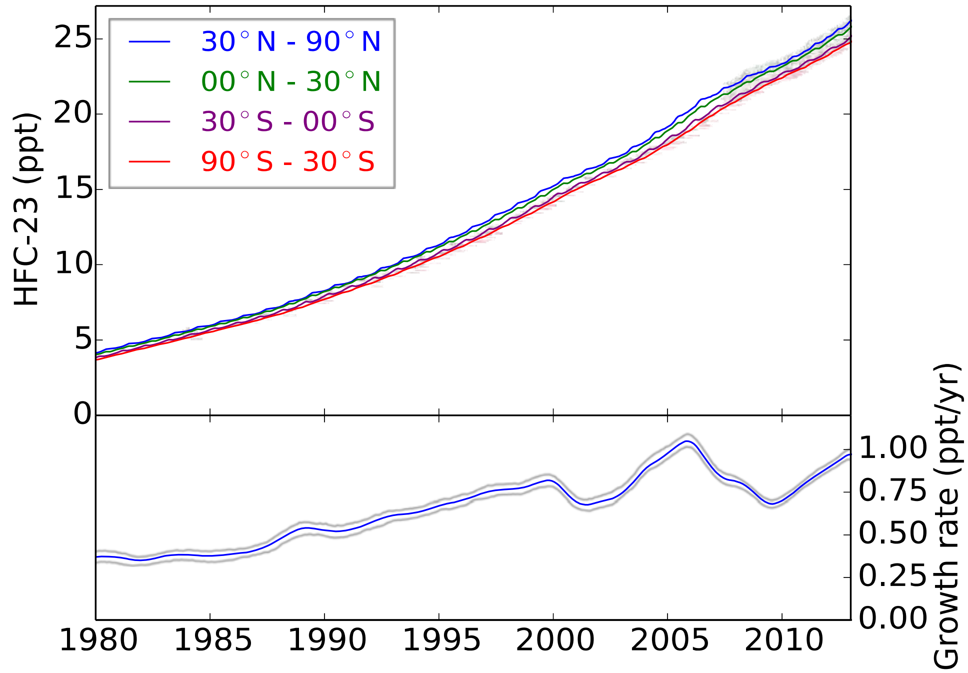
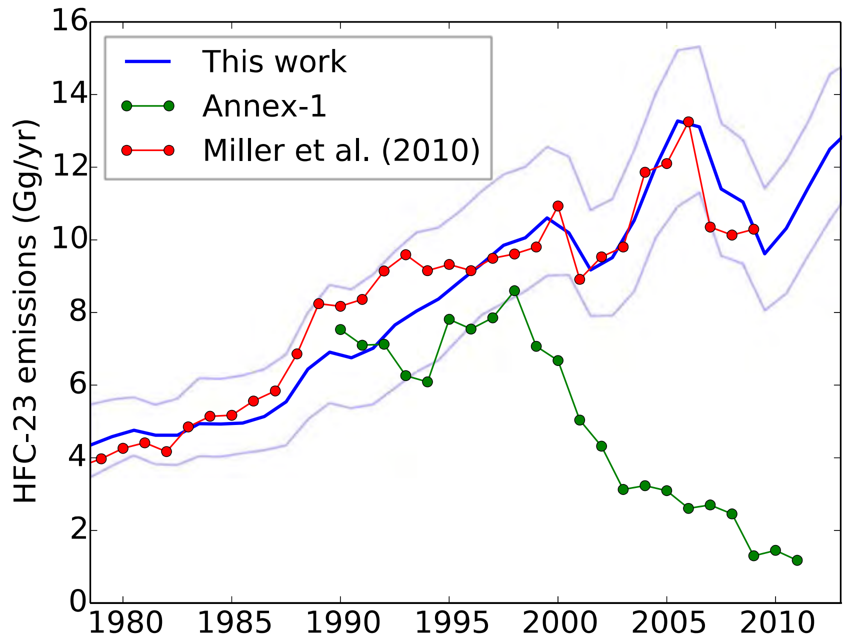
HFC-23 (CHF_3) is a powerful greenhouse gas (GWP = 14,800) produced as a by-product of HCFC-22 production.

AGAGE *in situ* & archive tank measurements show historic rise, recent slowdown & more recent increase.

Global HFC-23 emissions show temporary drop in emissions consistent with incineration of this gas under the Kyoto Protocol CDM program. Most recent increase is possibly related to increased HCFC-22 production without CDM.

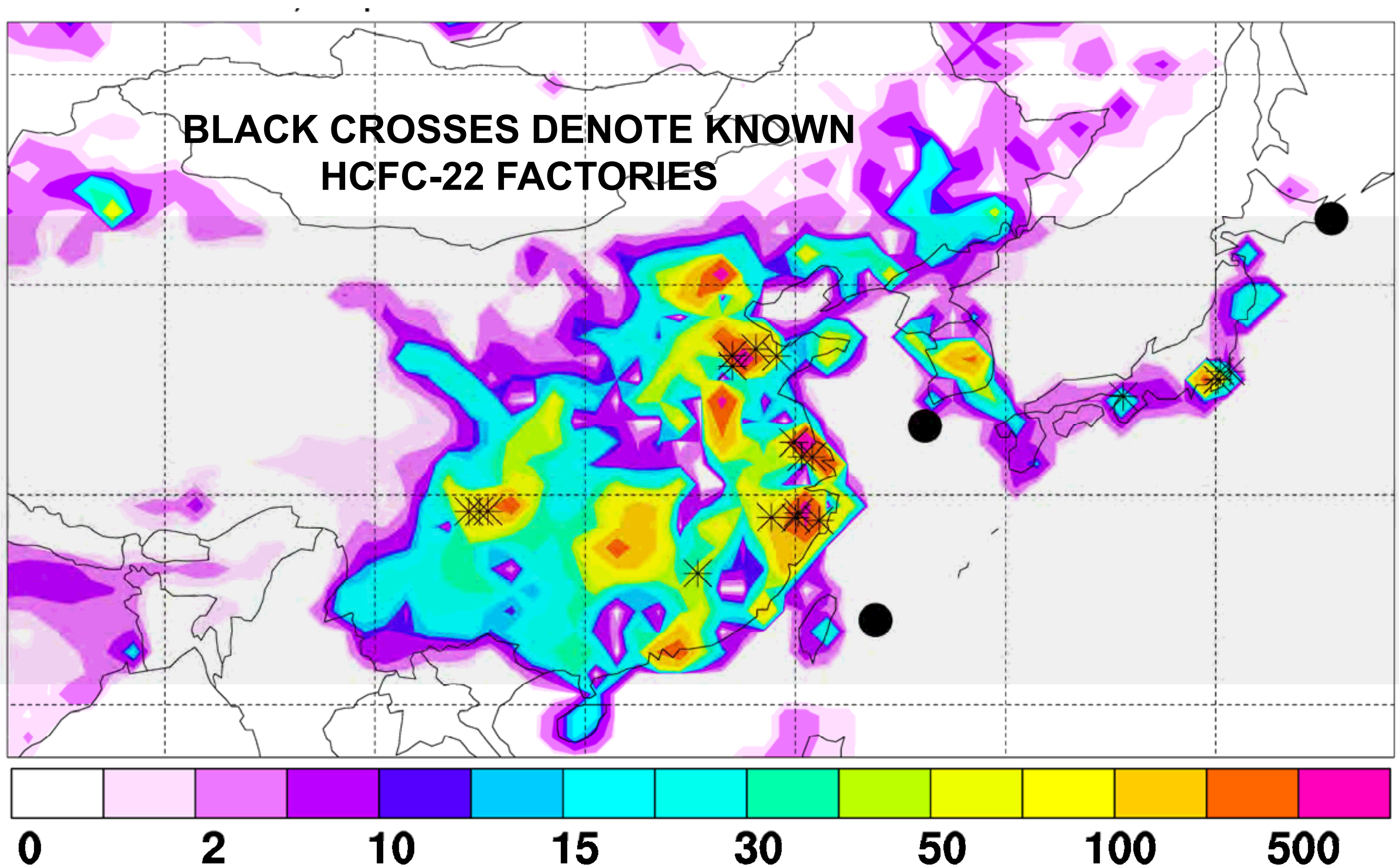
Miller *et al.*, *Atmos. Chem. Phys.*, 2010:
Rigby et al. unpublished update



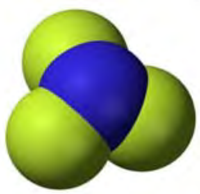


HFC-23 (CHF_3) East Asian Emissions Estimation

AGAGE and NIES Atmospheric Measurements at 3 Stations for 2008
(pg/m^2/sec, Stohl et al., *Atmos. Chem. Phys.*, 2010)



Global Hemispheric Trends and Growth Rates of Nitrogen Trifluoride (NF_3)

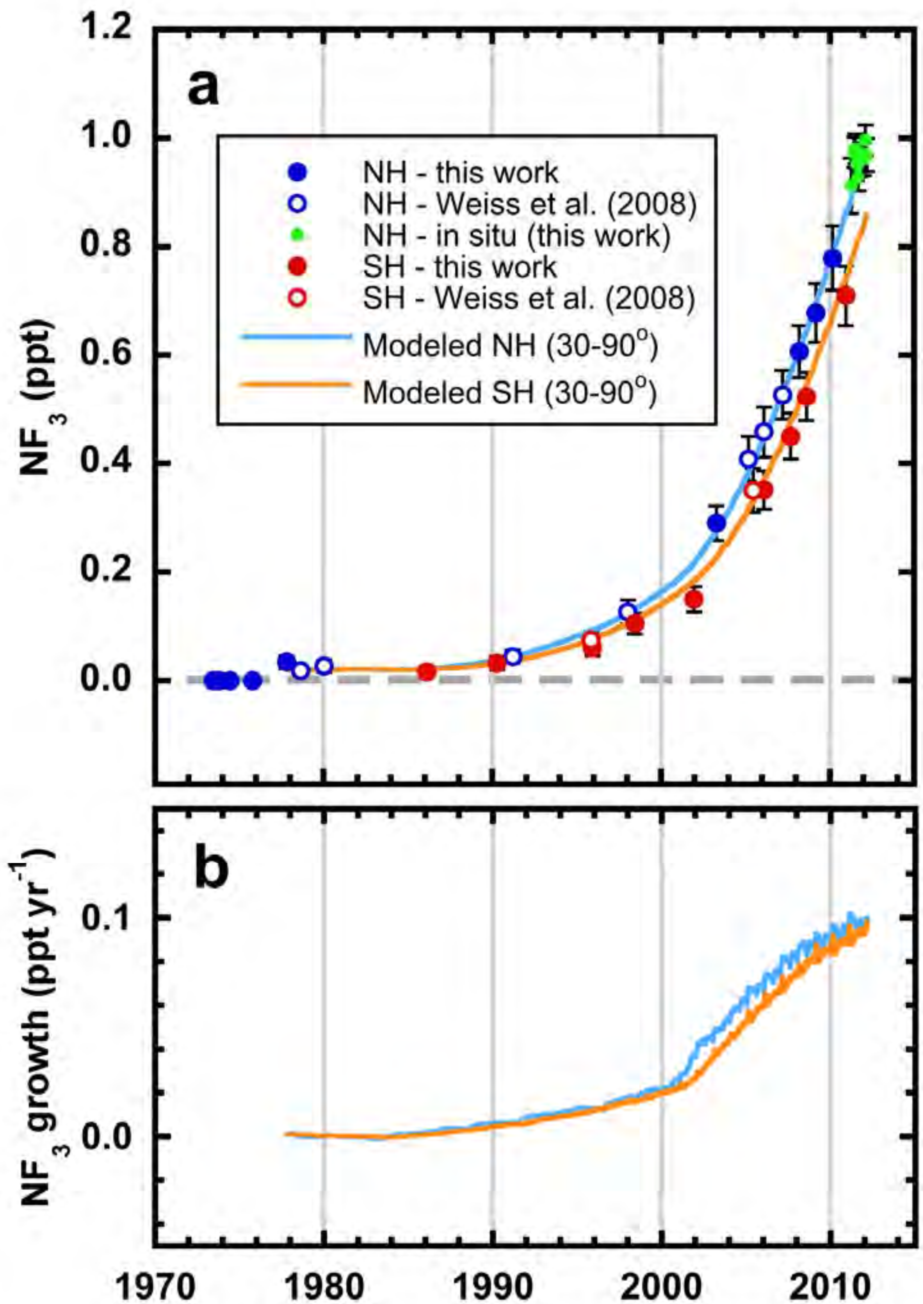


Lifetime ~ 490 years
GWP ~ 16,600

Used as a fluorine plasma source in electronics manufacturing (replacing C_2F_6)

Trends Fitted with AGAGE 2-D 12-Box Inverse Model

Weiss et al. (*GRL*, 2008)
Arnold et al. (*PNAS*, 2013)

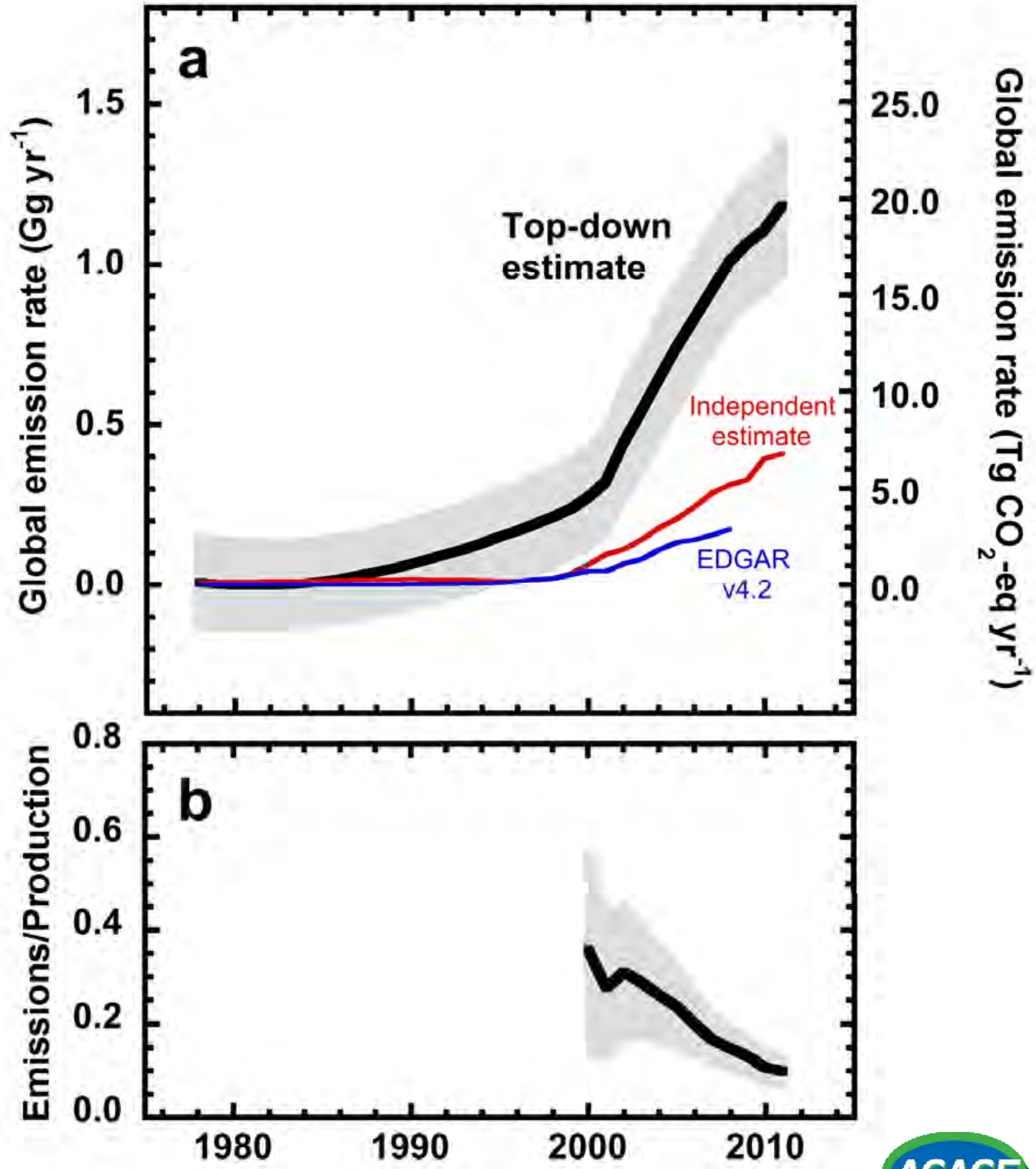


Global NF_3 Emissions

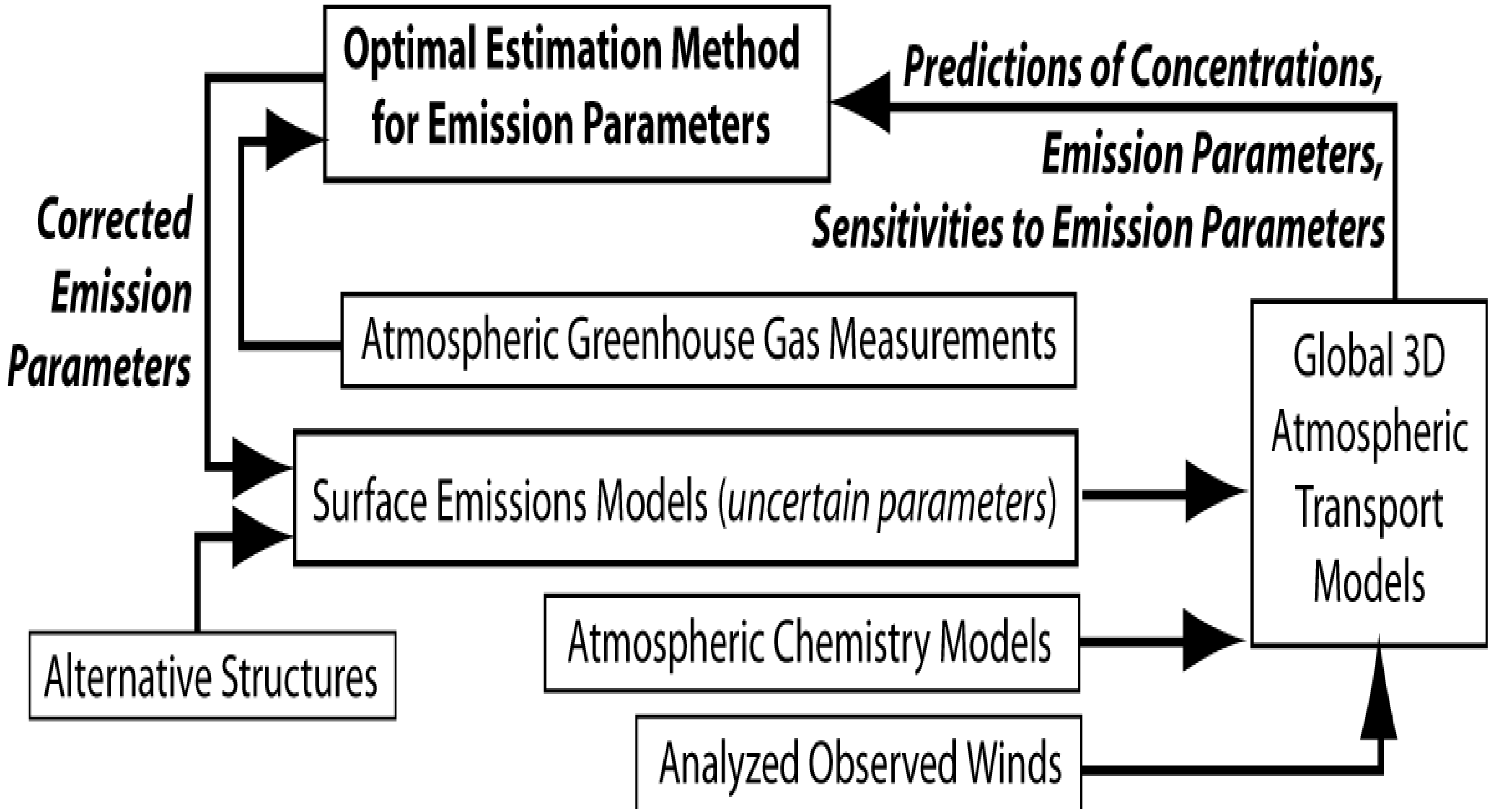
a) NF_3 global top-down emission rates compared to industry and EDGAR v4.2 bottom-up estimates.

b) NF3 global emission/production ratio (emission factor) based on top-down emissions divided by global industry production data. Note that while emissions are rising steeply, emission factors are falling steeply, from about 30% a decade ago to about 9% today, but they are still short of the 2% industry target.

Arnold *et al., PNAS*, 2013



The Way Forward -- The Estimation Challenge



“Regulating emissions without verification against actual accumulation in the atmosphere is like going on a diet without weighing oneself.”

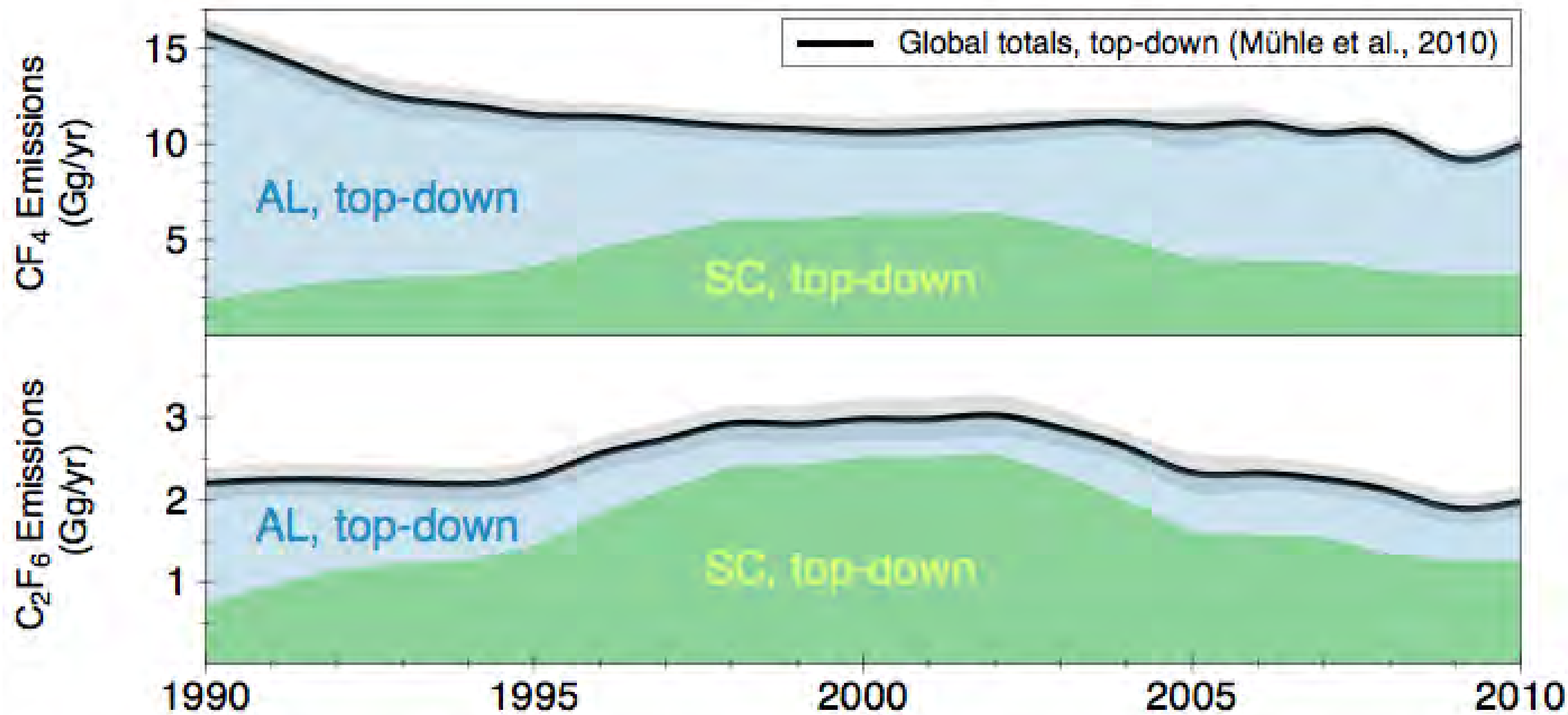
Thank You

Shangdianzi Station

Chinese Academy of Meteorological Sciences (CAMS)
China Meteorological Administration (CMA)

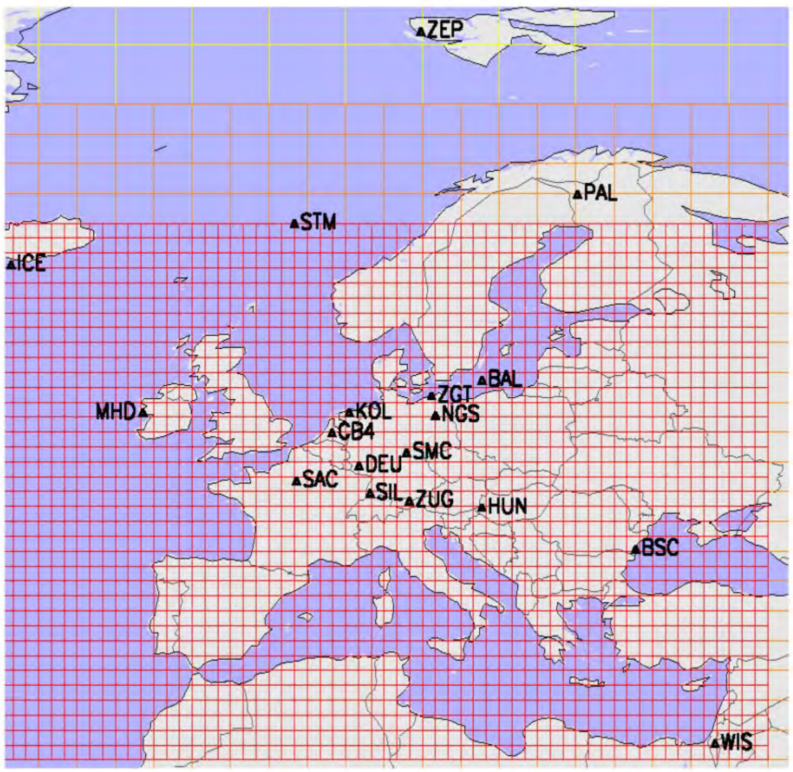
Operational: 2009-2012

Top-Down CF_4 and C_2F_6 Emissions by Industry vs. Time

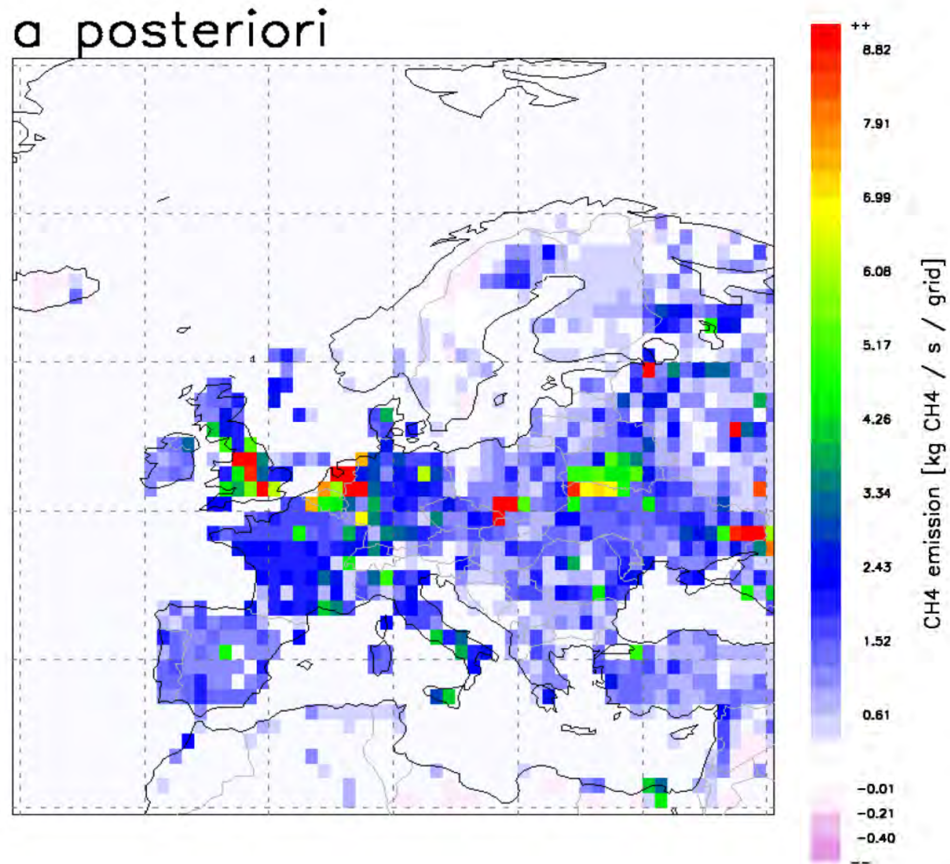


AGAGE

2001 European Methane Emissions (CH_4 , GWP = 25) from a 1° Nested Atmospheric Model *



16 European Stations
Within a 56-Station Global Network



2001 “Top-Down” European
Emissions Map

*Bergamaschi et al., *Atmos. Chem. Phys.*,5, 2431-2460, 2005