Validation of CH_4 surface emission using forward chemistry-transport model

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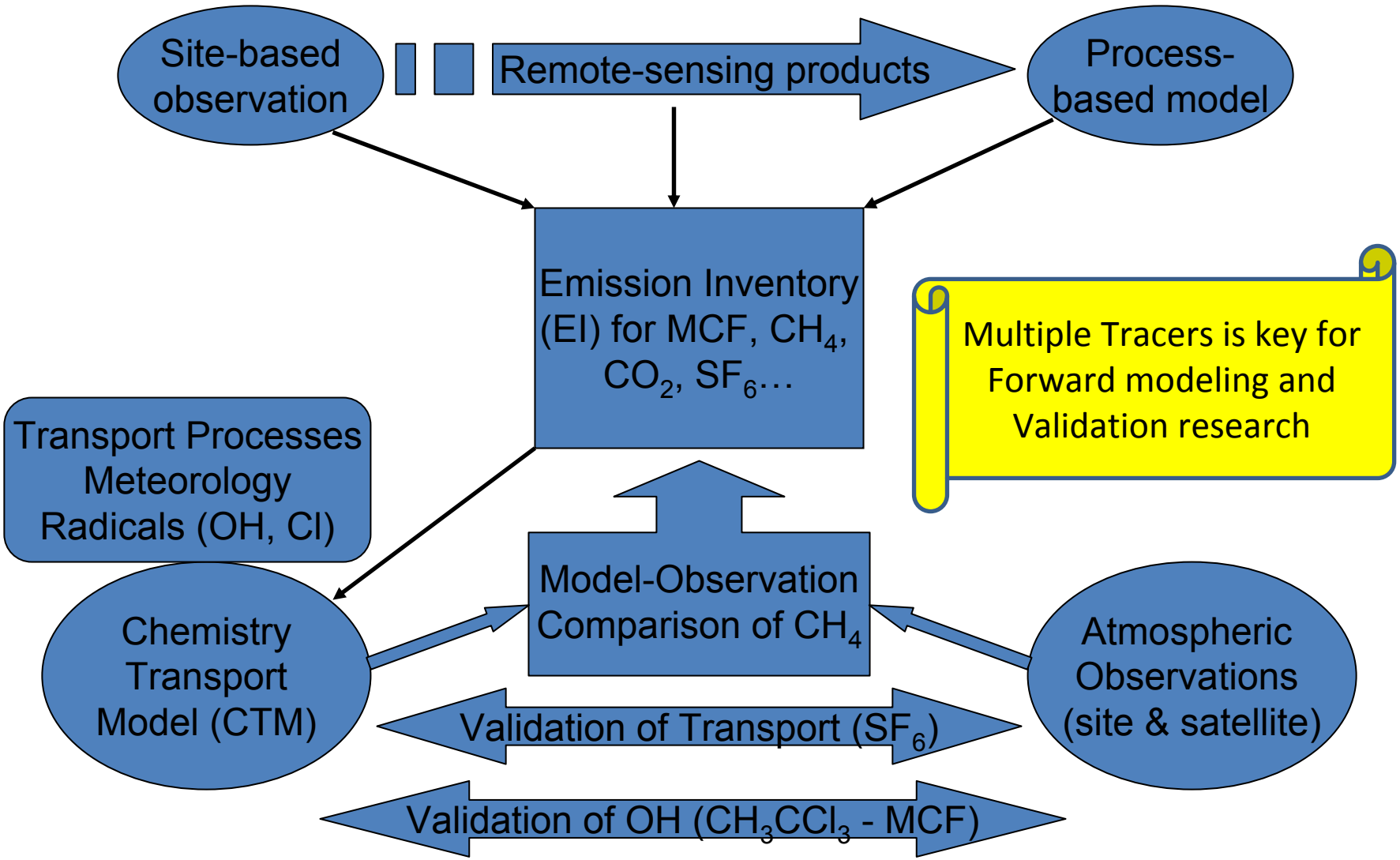
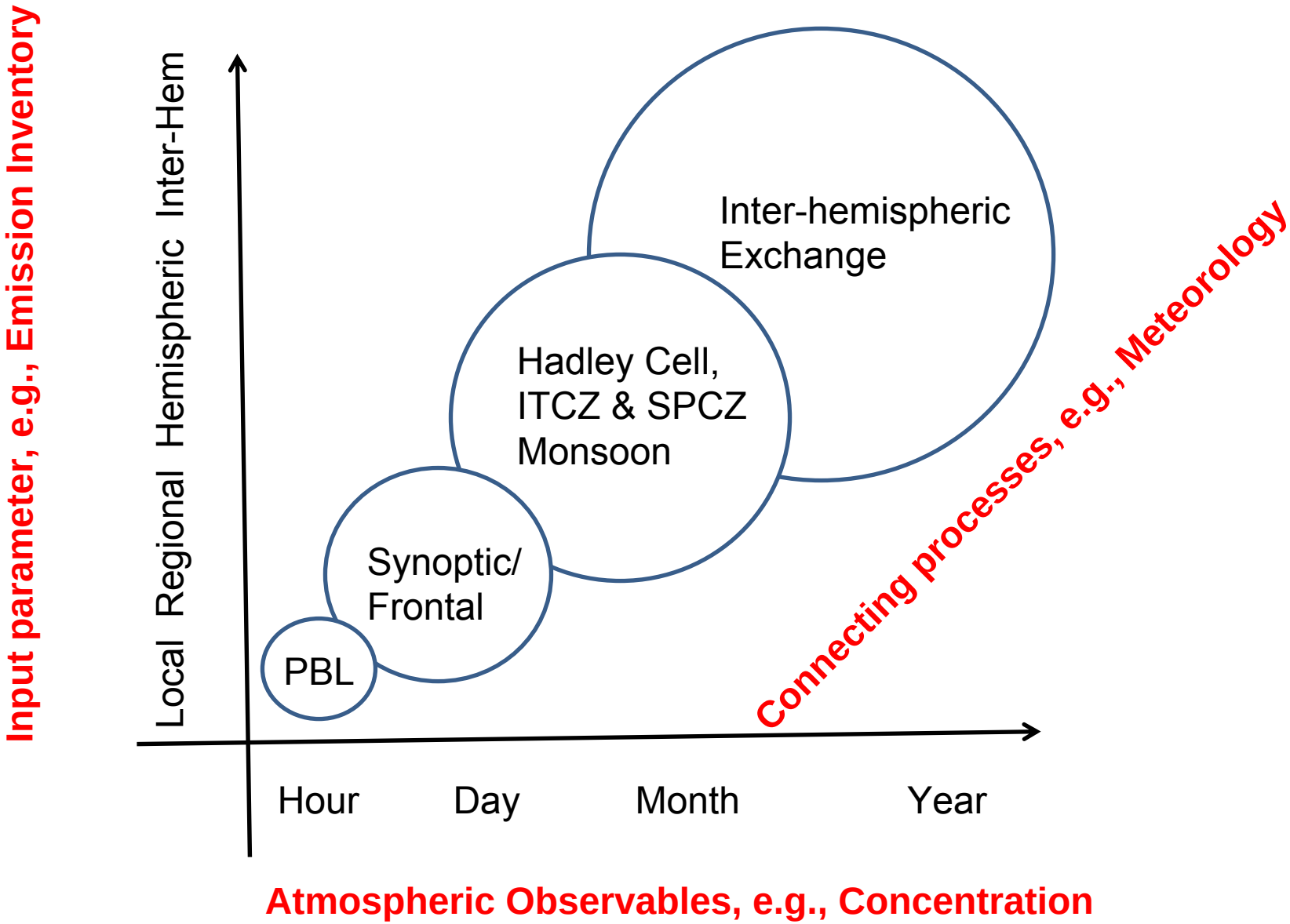
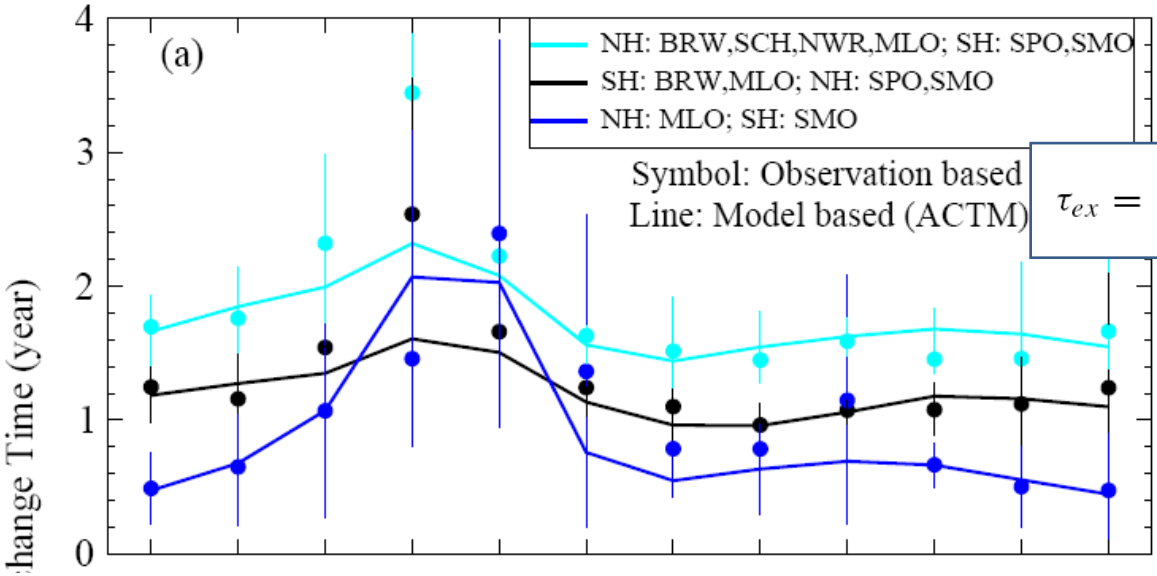
Introduction



Space-Time scales



ACTM transport validation: inter-hemispheric exchange time (τ_{ex})



Patra et al., ACP, 2009

$$\tau_{ex} = \left[\Delta c_{n-s}\left(\frac{E_n}{E_s}+1\right)\right] \Big/ \left[\frac{E_n}{E_s}\frac{dc_s}{dt} - \frac{dc_n}{dt}\right]$$

E_n = NH emission
E_s = SH emission
C_n = NH concentration
C_s = SH concentration

τ_{ex} Estimation Method	ACTM Based Mean±1σ	Observ. Based Mean±1σ	LH96 Mean	LG97 Mean	SD99 Range	BL04 Range	DR07 Range	AA08 Mean
Site based estimates								
Case 1	1.75±0.03	1.86±0.16	1.50	1.30				
Case 2	1.21±0.01	1.33±0.18						
Case 3	1.07±0.40	0.99±0.31						
Whole Hemisphere Based Estimates								
Case 4	1.37±0.04				0.76–1.97			
Case 5	0.70±0.01				0.55–1.26	0.80–1.2	0.78–1.02	0.70

ACTM framework for CH_4 simulation

(Patra et al., JMSJ, in acceptance)

- CCSR/NIES/FRCGC AGCM-based CTM (**ACTM**) run at resolution T42 L67 (top 90km)
- NCEP-2 reanalysis meteorology (U,V,T nudged)
- Hadley Center Sea-Surface Temperature & Sea-Ice Cover
- CH_4 chemistry (Sander et al., JPL Pub. 06-2, 2006) as:

$CH_4 + O^1D \rightarrow$ Products ($K_{O^1D} = 1.5 \times 10^{-10}$)

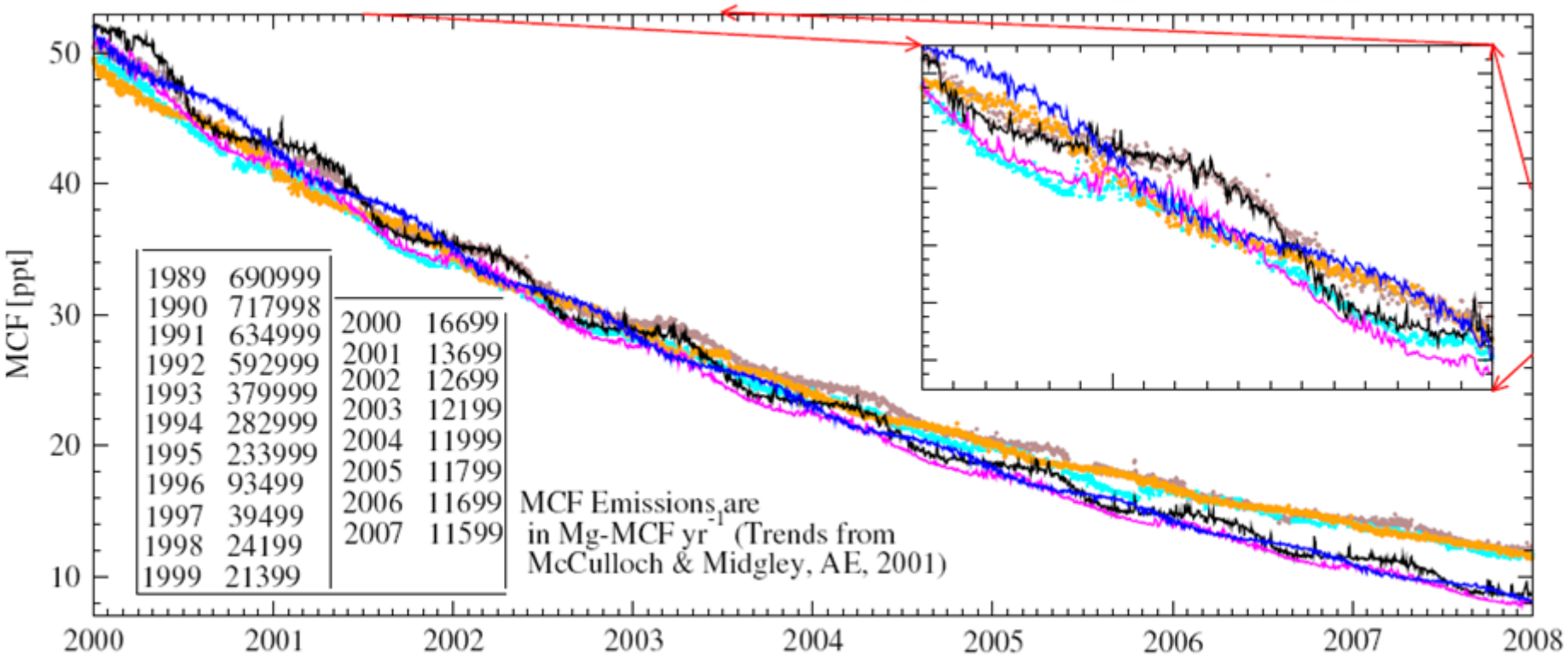
$CH_4 + OH \rightarrow CH_3 + H_2O$ ($K_{OH} = 2.45 \times 10^{-12} \exp(-1775/T)$)

$CH_4 + Cl \rightarrow CH_3 + HCl$ ($K_{Cl} = 7.3 \times 10^{-12} \exp(-1280/T)$)

$CH_4 + h\upsilon \rightarrow$ Products (wavelength dependent; not considered here)

- All the radicals are taken from CHASER/STRAT (Sudo et al., Takigawa et al.) models at monthly (or hourly) intervals

Validation of CHASER+STRAT OH using MCF



The comparison for the recent times raises a couple of questions:

1.Is MCF emission decreasing since 2000 or so. If yes, at what rate and what is present day emission magnitude

2.Is the OH production in CHASER is higher than the real values? If so, at which latitudes, longitudes or heights

Surface flux types and annual budget of CH_4

Sources	Range Estim. Reported by IPCC [2001]	A Priori Estimates, Tg CH_4/yr
Total wetlands	92-237	
Swamps		91[c]
Bogs and tundra		54[c]
Rice agriculture	25-100	60[d]
Ruminant animals	80-115	93[d]
Termites	20-20	20[e]
Biomass burning	23-55	52[f]
Energy	75-109	
Coal		38[d]
Natural gas and other industrial		57[d]
Landfills	35-73	50[g]
Ocean	10-15	10[h]
Hydrates	5-10	5[h]
Total source	**500-600**	**530**

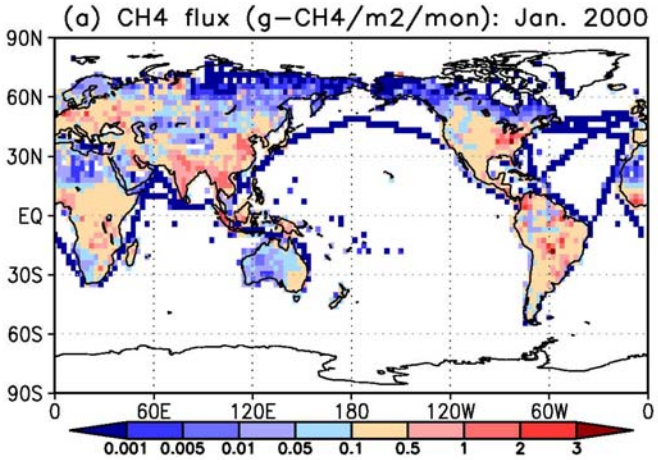
Sinks		A Priori Estimates, Tg CH_4/yr
Tropospheric OH	450-510	507[i]
Stratospheric loss	40-46	40[k]
Soils	10-30	30[k]
Total		**577**

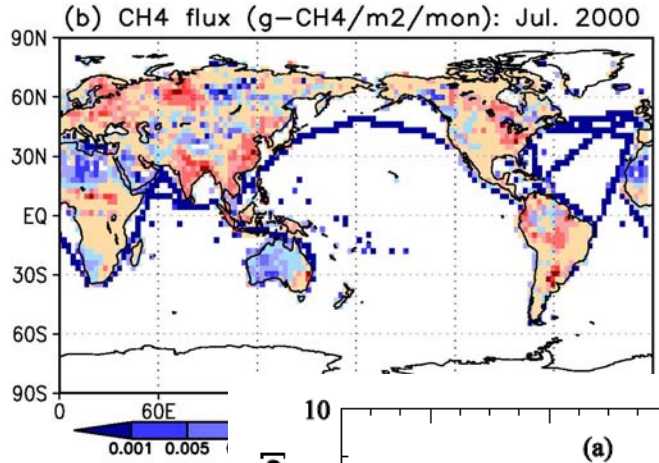
Year	Total emission (E2)	Tropospheric Budget (E2)	Year 2000 (E2)	Top emission country(E2)	Aggr. Emission (E2)
1988	569.4	**Industrial (EDGAR)***	**301.9**	Brazil	54.2
1989	570.6	Biofuel	16.0	USA	54.0
1990	571.1	Fossil fuel	102.9	Russia	51.3
1991	571.7	Industrial	0.9	China	47.4
1992	572.3	Agriculture	119.3	India	41.1
1993	572.9	Waste	62.7	Indonesia	30.1
1994	573.4	**Biogenic (GEIA)**	**273.0**	Canada	17.3
1995	574.0	Termites	20.5	Argentina	14.9
1996	574.3	Biomass Burning	59.8	Australia	11.7
1997	574.7	Rice	39.4	Thailand	10.7
1998	574.1	Swamps	104.4	Zaire	8.9
1999	574.5	Bogs	40.2	Nigeria	8.7
2000	575.0	Tundra	8.7	Sudan	8.6
2001	574.7	**Sinks**	**~580**	Mexico	8.1
2002	574.2	Trop. Loss	551	Venezuela	7.1
2003	574.9	Strat. Loss	29	Ukraine	6.6
2004	574.6	NH Loss	334	Vietnam	6.5
2005	574.8	SH Loss	246	Pakistan	6.4
2006	574.8	**Atmospheric Burden**	**4999**	Peru	6.3

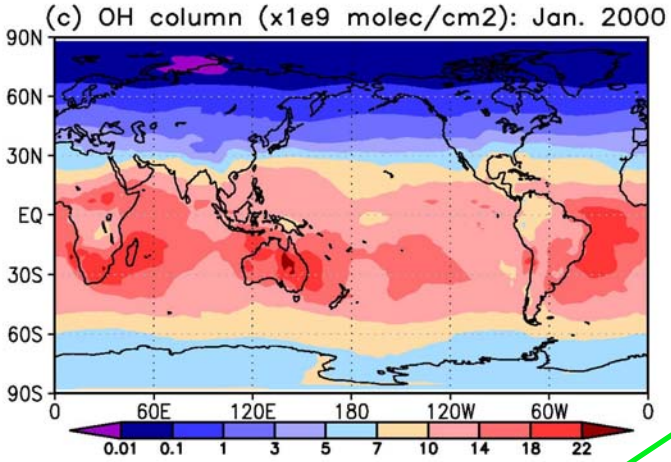
Mikaloff Fletcher et al., GBC, 2004

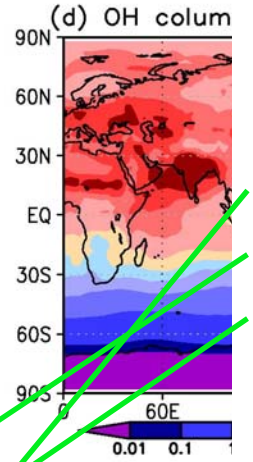
Patra et al., JMSJ, 2009, in acceptance

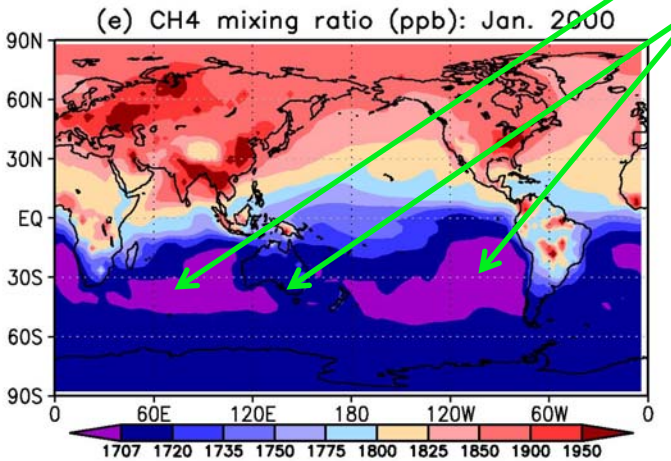
CH_4 emission, sinks, and concentration

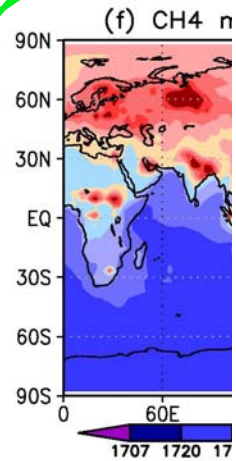


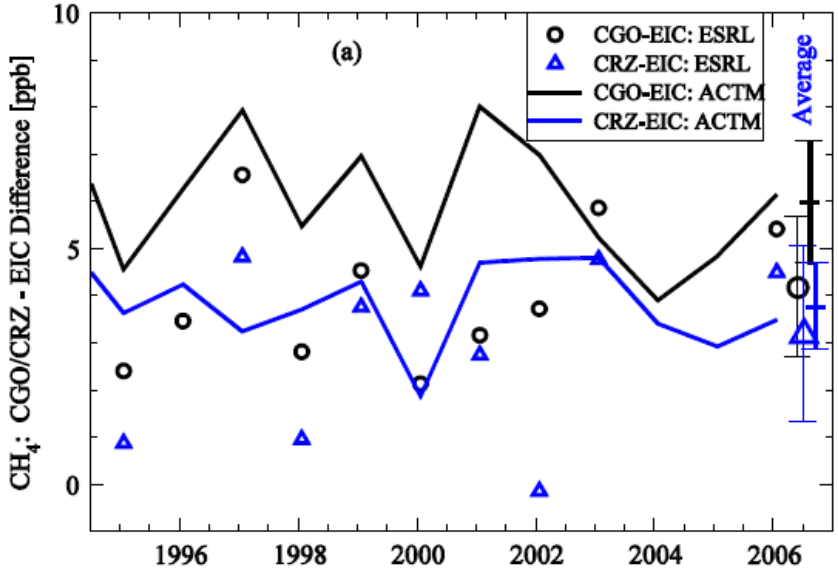


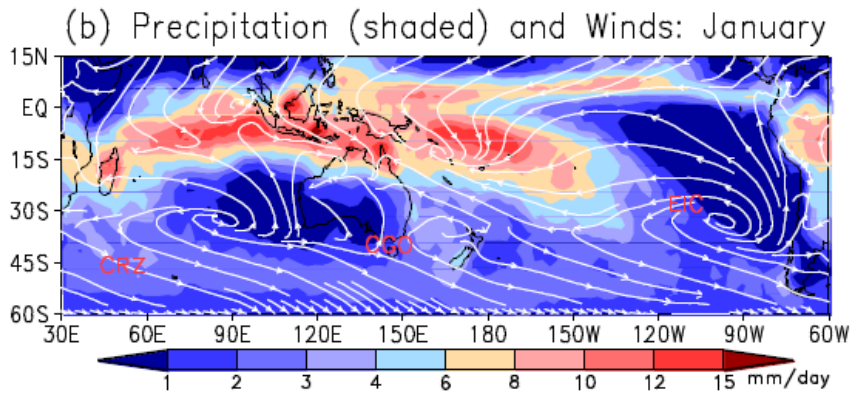




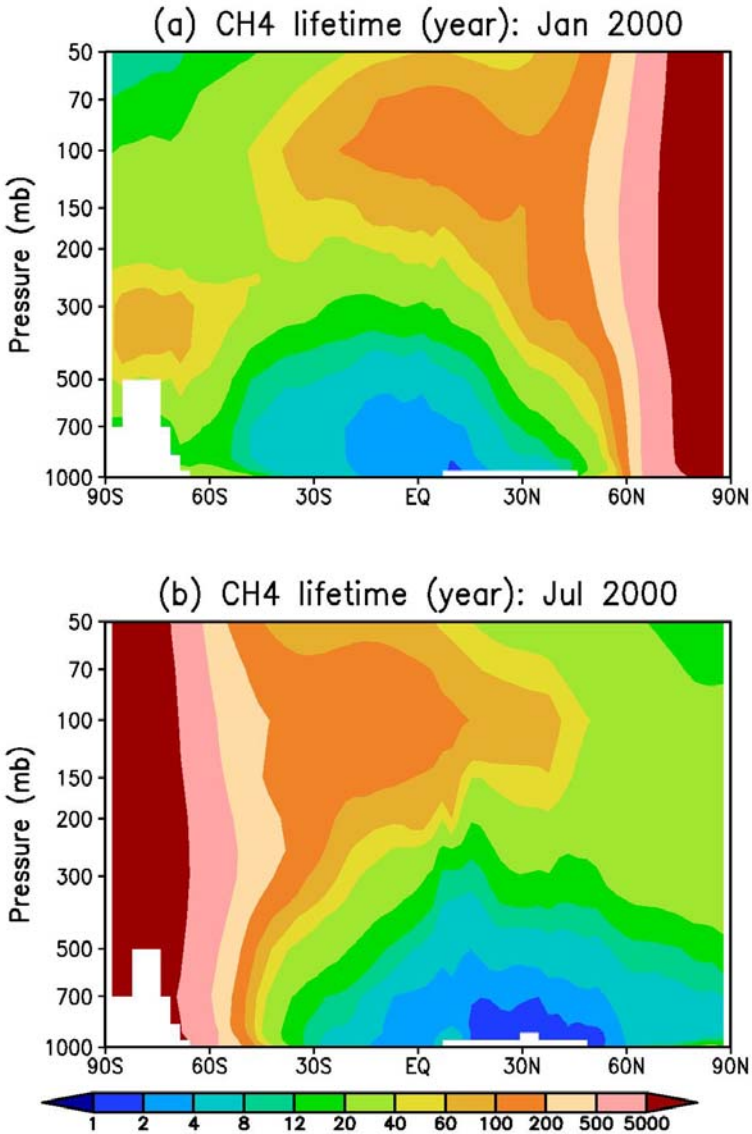








CH_4 lifetime and budgets



Instantaneous CH_4 Lifetime

$= 1/[K_{OH} \cdot OH + K_{O^1D} \cdot O^1D + K_{Cl} \cdot Cl]$

(useful for understanding the dominance of dynamics vs. chemistry on variability)

Atmospheric Lifetime = burden/loss

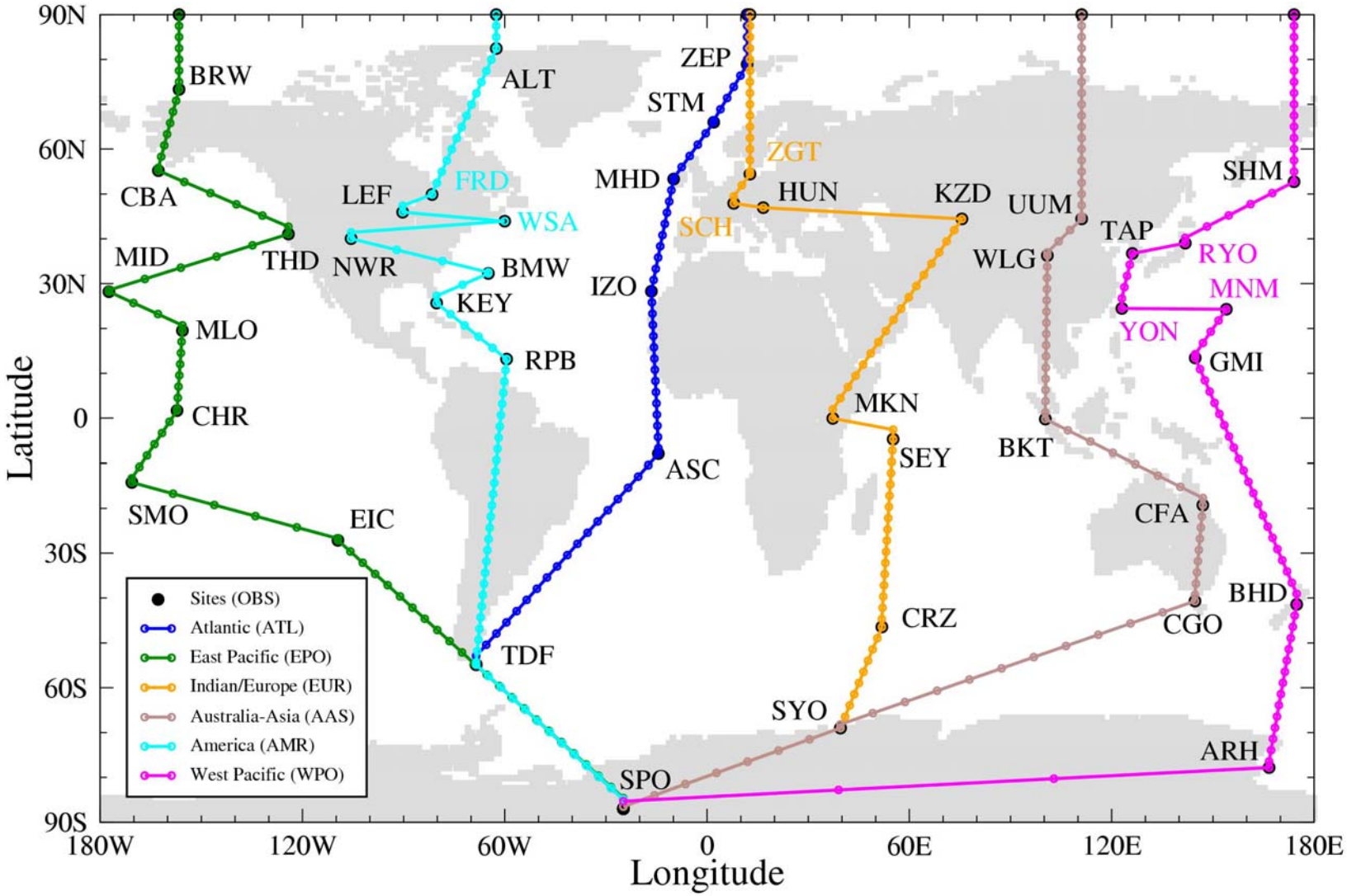
(Prather et al., IPCC, 2001)

CH_4 L. T. = 4999 Tg/580 Tg yr^{-1} = 8.62 years

Estimates	Atmospheric Lifetime	References
IPCC TAR	8.4	Prather et al.
IPCC FAR (prescribed)	8.67±1.32(m#26) 8.45±0.38 (m#12)	Stevenson et al., JGR, 2006
This work (full model)	8.62	Patra et al., JMSJ, 2009

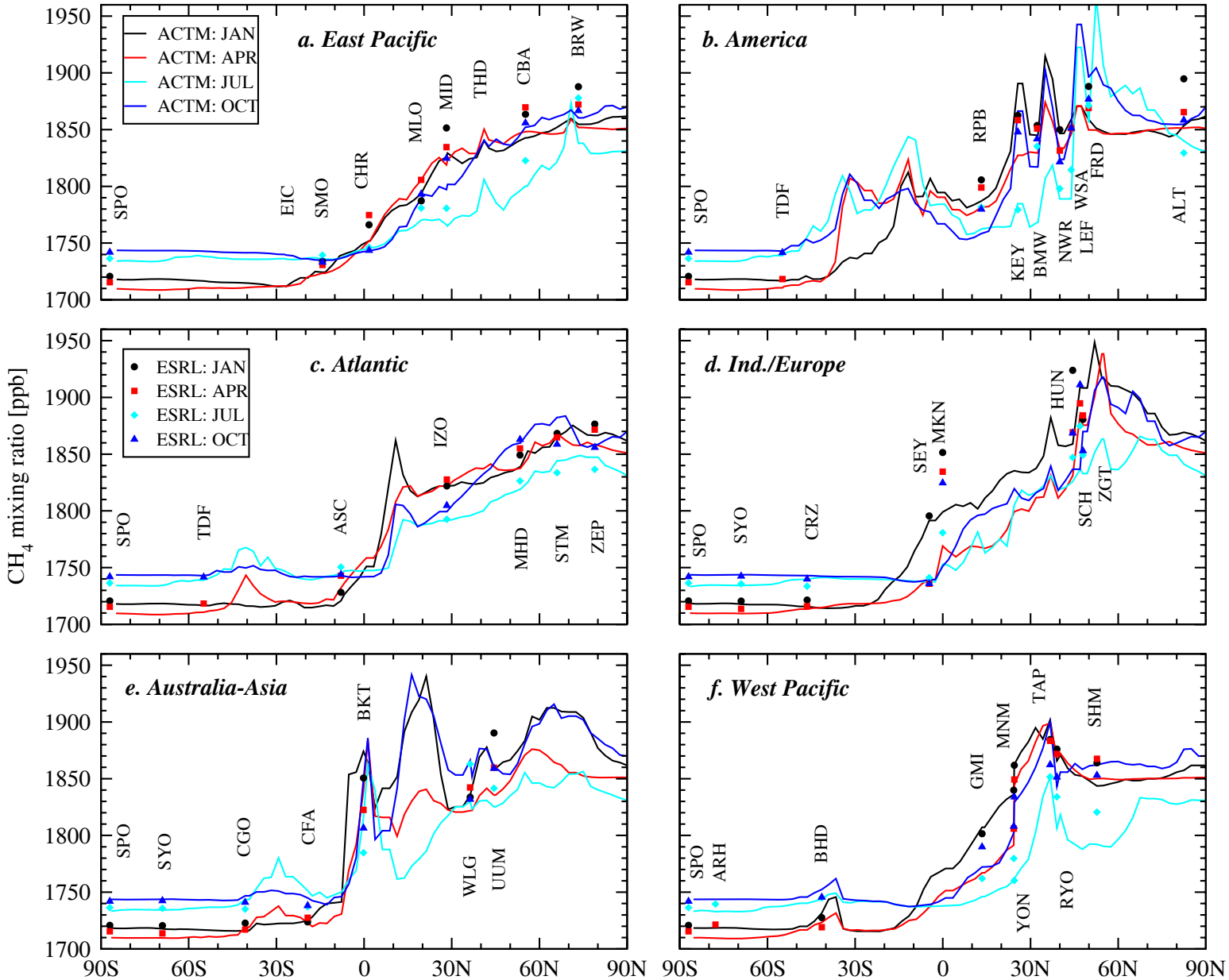
CH_4 Measurement Sites – can we track emissions?

(~50 used here; >100s are in operation in 2007)



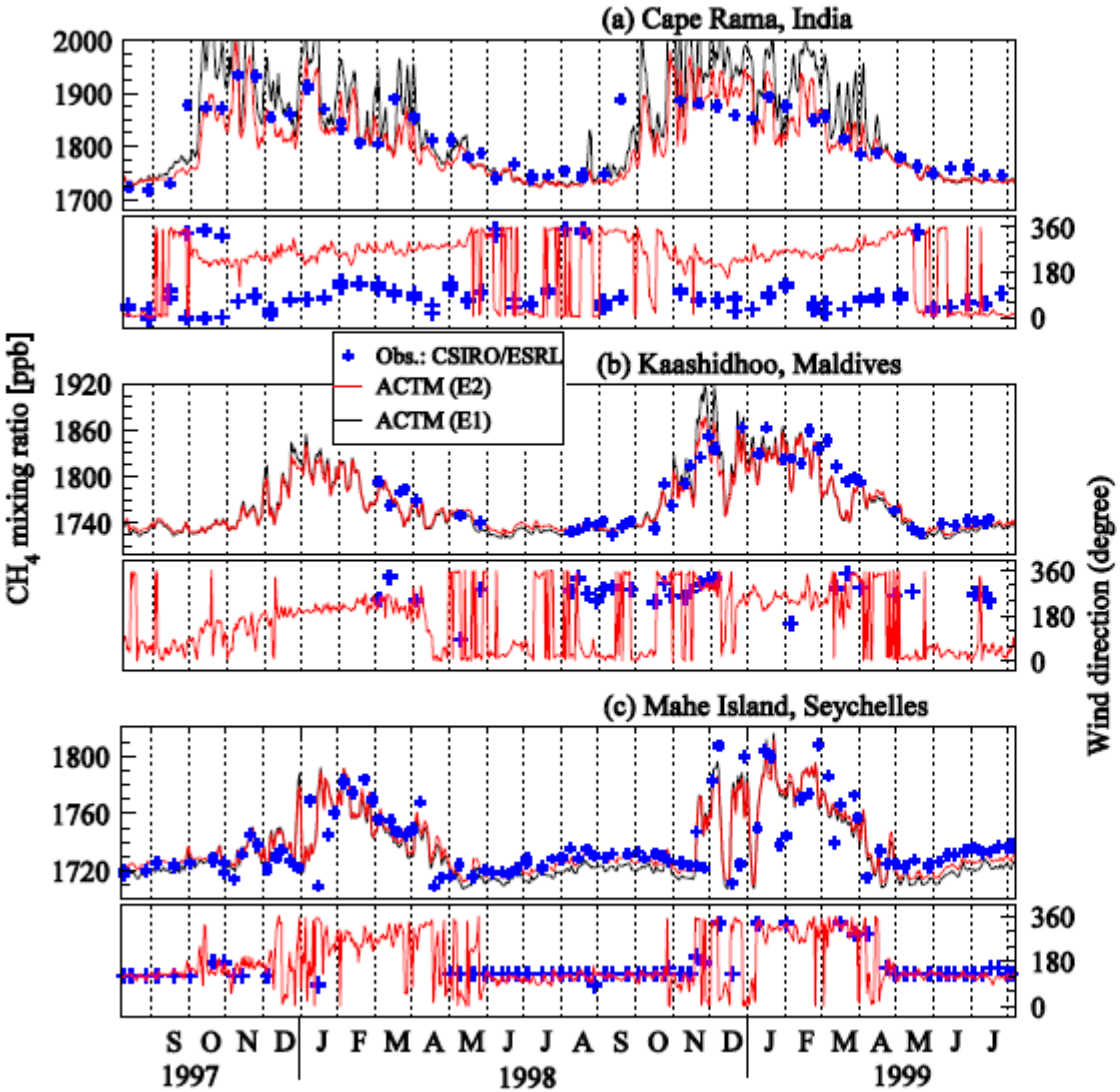
Contributing Institutes: 1. NOAA/ESRL, 2. FEA, Germany, 3. JMA, Japan, 4. EC, Canada, 5. NIWA

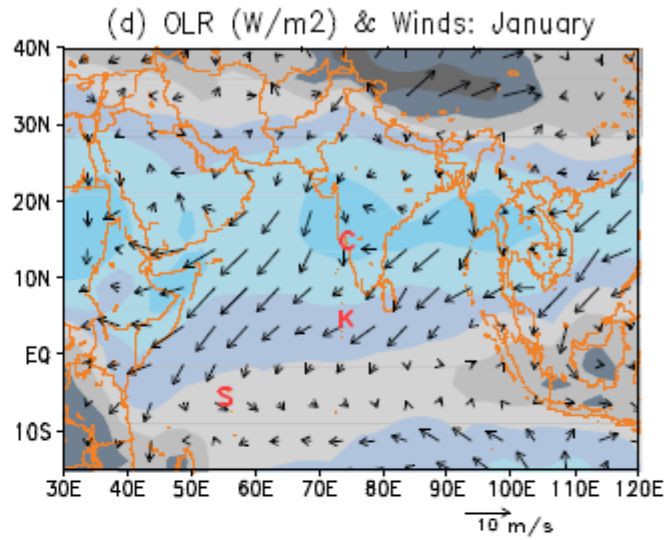
CH_4 Latitudinal gradients: seasonal and longitudinal variations

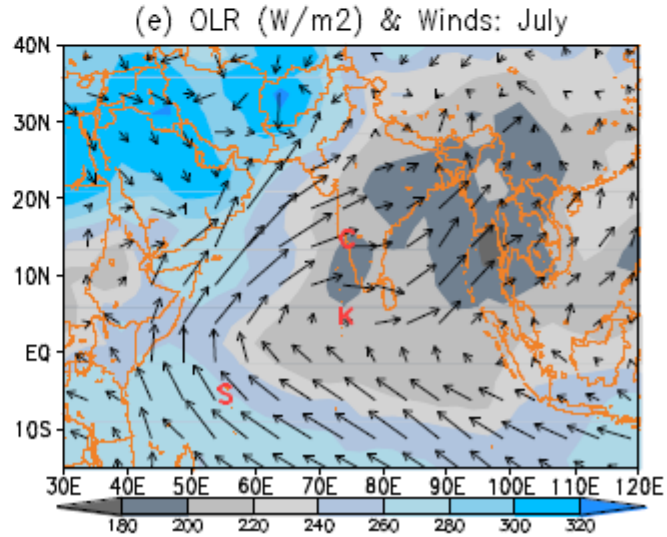


Seasonal cycle in the extra tropics/high latitudes is controlled by chemical loss and surface emission

CH_4 Seasonal Cycles in the tropics – produced by chemical loss and dynamics (monsoon)



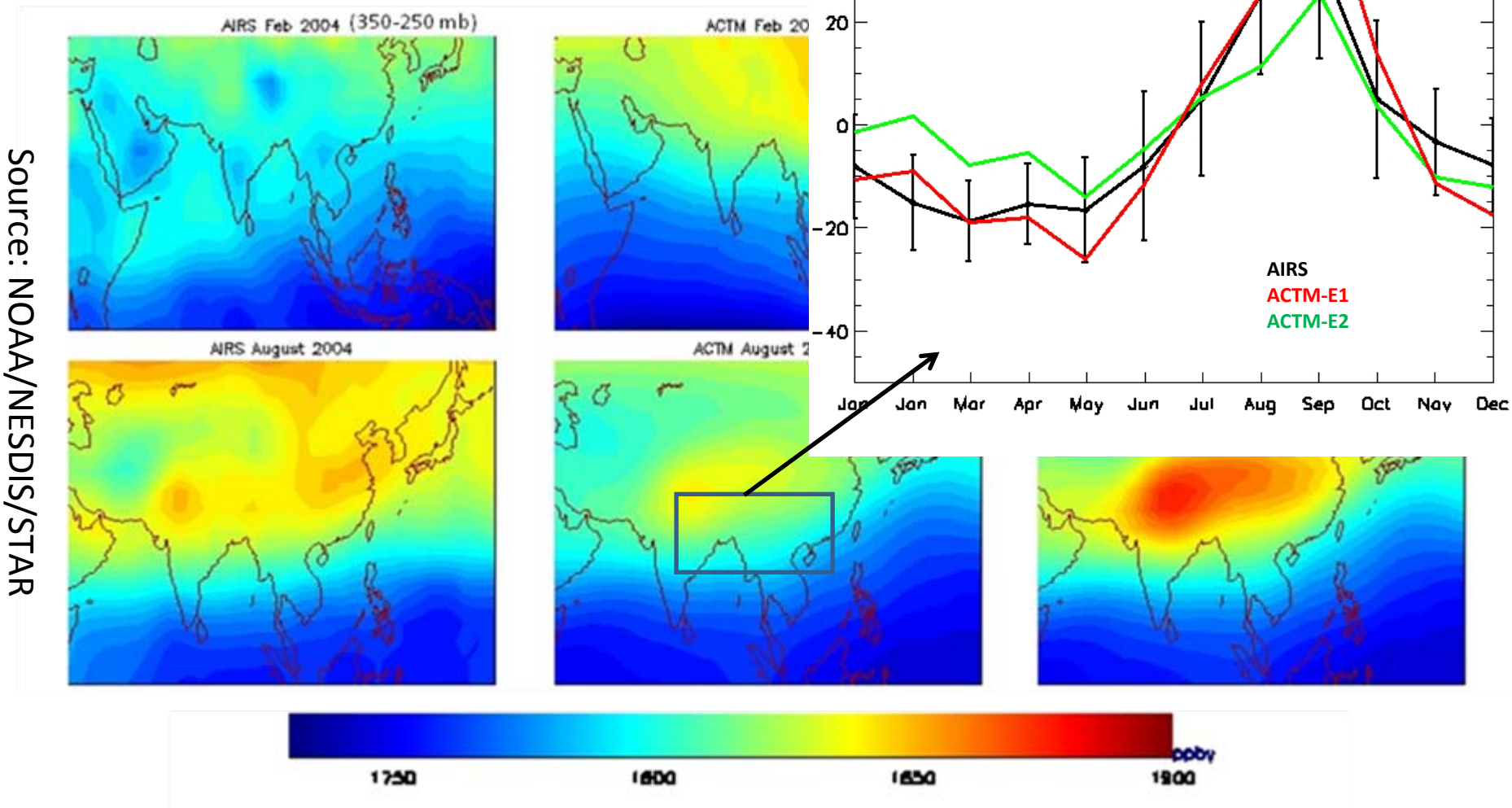




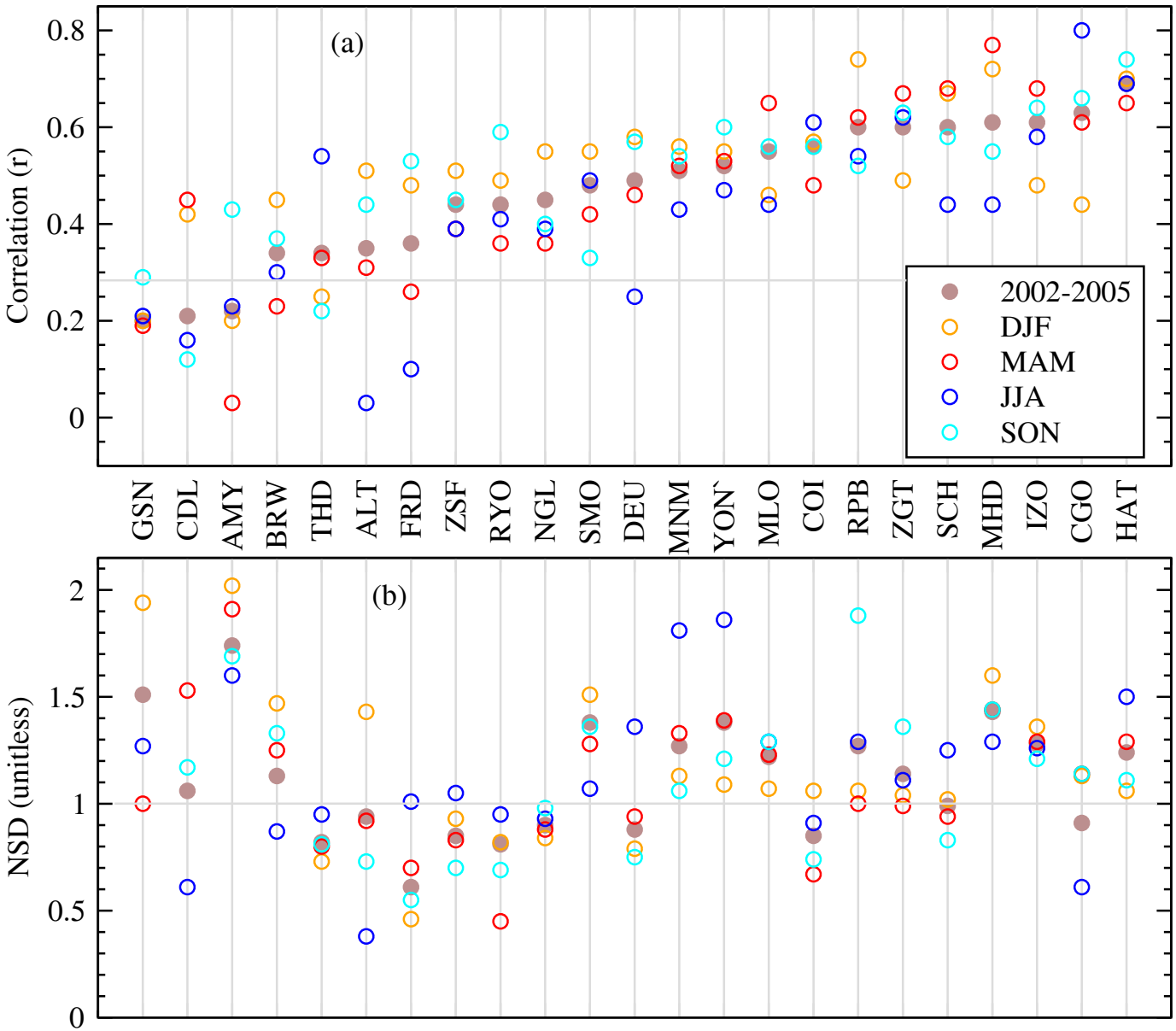
Preliminary comparison with AIRS/Aqua satellite retrievals

AIRS Precision : ~20 ppb
AIRS – ACTM = 20 ppb

E1 : 80 Tg-CH_4 Rice emis
E2 : 40 Tg-CH_4 Rice emis



Source: NOAA/NESDIS/STAR



Measure of

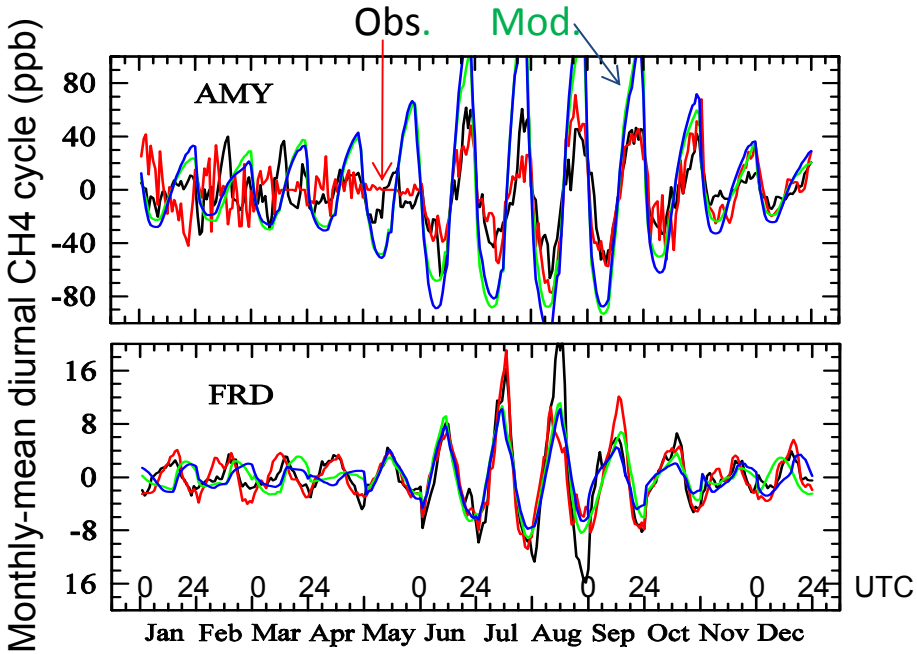
Synoptic peak shape / phase

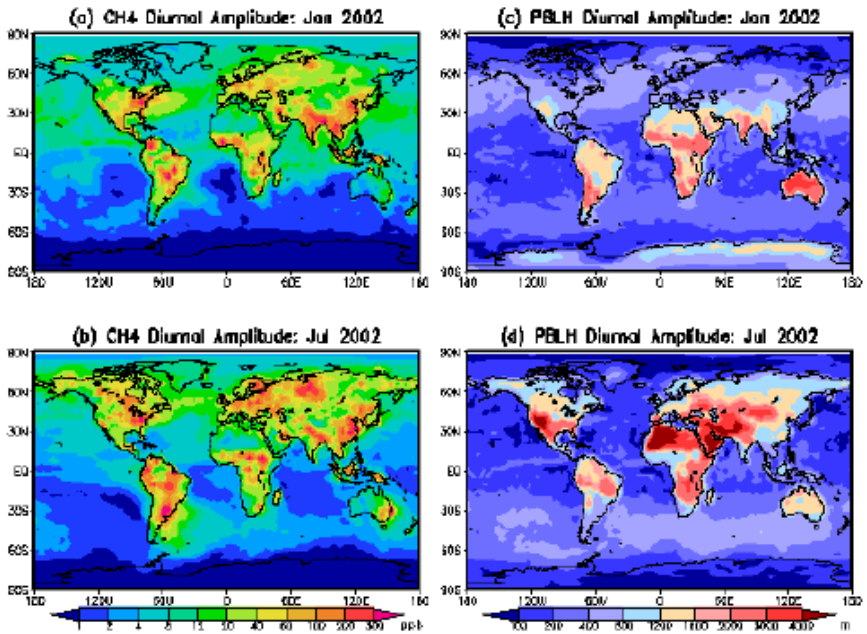
Measure of

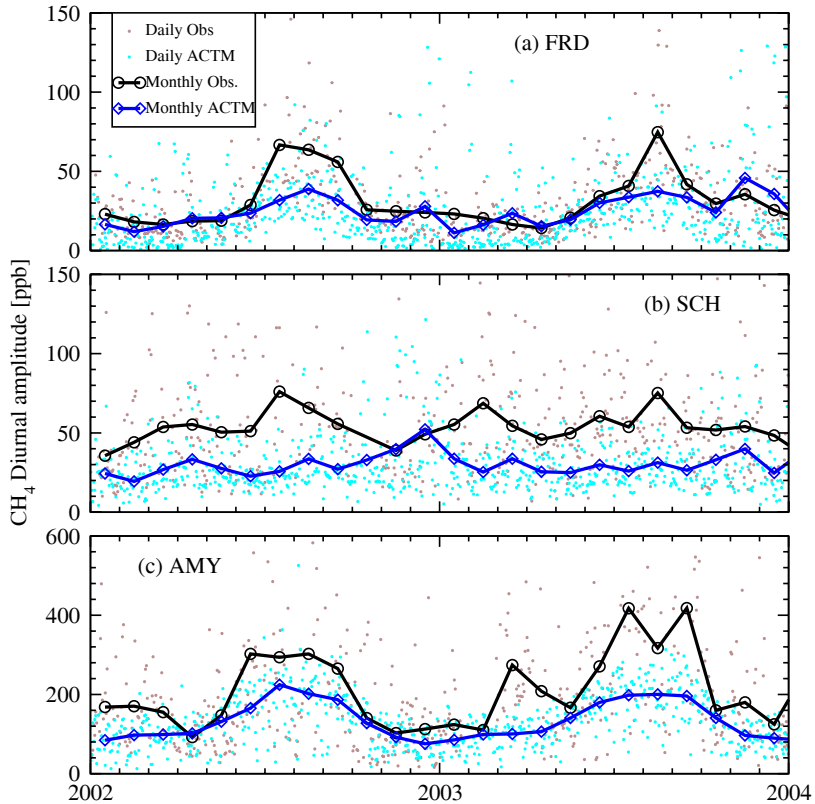
Synoptic peak height

$(\sigma_{obs}/\sigma_{mod})$
Ideal~1.0

CH_4 diurnal cycles: caused by local emission and PBL cycle







Conclusions

- ACTM CH_4 simulations have been optimized for a combinations of fluxes, radicals and transport
  - Examining seasonal cycles of IHGs at a 'set' of stations helped to prepare flux-types combinations
  - Very low (~5ppb) CH_4 longitudinal gradients between CRZ/CGO/EIC can be tracked
  - Seasonal cycle, synoptic variability and diurnal cycle; all needed to be checked for testing bottom-up fluxes

- The ACTM-AIRS comparison in the upper troposphere region reveal
  - A reversal of CH_4 seasonal cycle at about 400-300 mb height
  - Role of CH_4 transport by Asian monsoon during high surface emission period

- Both transport and chemistry components in ACTM have been validated independently using two different chemical tracers, SF_6 and MCF, respectively

Outlook

- Reduction in radical (e.g., OH) uncertainty by independent methodology
  - Can the MCF emission be available at more confidence? e.g., global/country total emission inventory for the users?
  - Time dependent gridded distribution of emissions

- Updating chemical (e.g., CH_4) emission distribution and strength
  - A concerted effort?, e.g., EDGAR gives annual mean emission due to rice cultivation
  - IAVs in wetland or other natural emissions? Validation strategy

- Increased use of remote sensing data for long-lived gases
  - Some issues persist with the product quality; more validation and comparison would help probing the causes
  - More dedicated satellite sensors, e.g., GOSAT, new-OCO?