

# Variabilities in greenhouse gases over (South) Asia due to monsoon

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# Introduction

- Asia is one of the most intense source region of greenhouse gas emission
  - CO<sub>2</sub>: China, India, Japan are 1<sup>st</sup>, 3<sup>rd</sup> and 5<sup>th</sup> largest emitters (EDGAR/CDIAC/IEA)
  - CH<sub>4</sub>: about 40% of 535 Tg-CH<sub>4</sub>/yr is emitted from Asia (Patra et al., 2016)
  - Also a major source of N<sub>2</sub>O (N in general) due to nitrogen fertilizer use (Thompson et al., 2014; Saikawa et al., 2014)
- Long-lived tracers provide a different perspective of the transport pathways
- It is important to understand the source-receptor relationships before inverse modelling of regional sources and sinks

# Atmospheric Chemistry-Transport Model (ACTM): understanding and improving fundamentals

$$\frac{dCH_4(x, y, z, t)}{dt} = S_{CH_4}(x, y, t) - L_{CH_4}(x, y, z, t) - \nabla \cdot F_{CH_4}(x, y, z, t)$$

where,

$CH_4$  = concentration in the atmosphere

$S$  = Source/emission of constituent

$L$  = Loss/sink of constituent (mainly by OH reaction)

$\nabla \cdot F$  = transport by diffusion, advection and convection

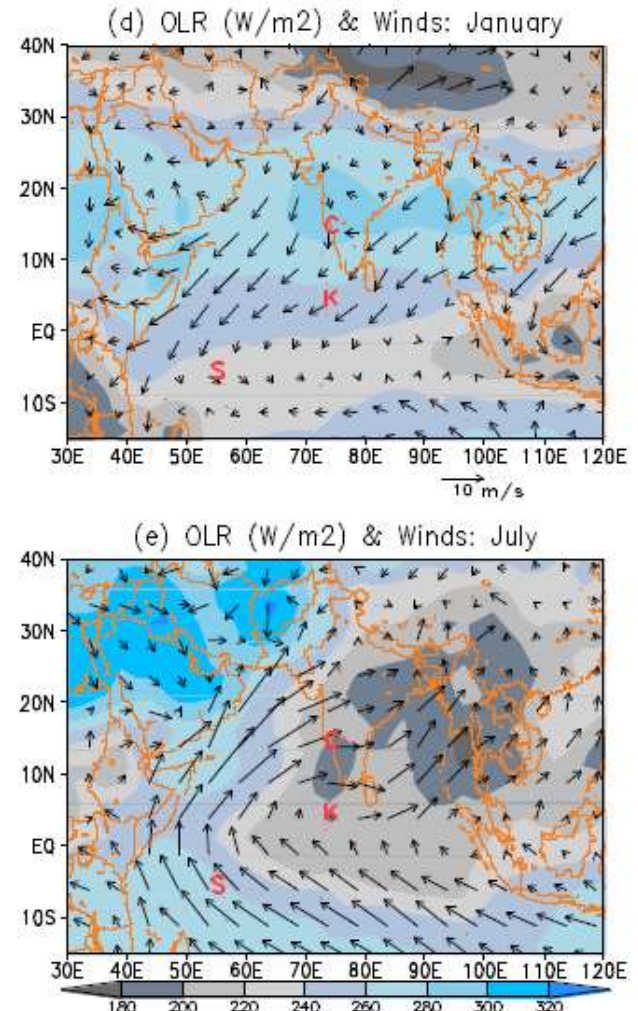
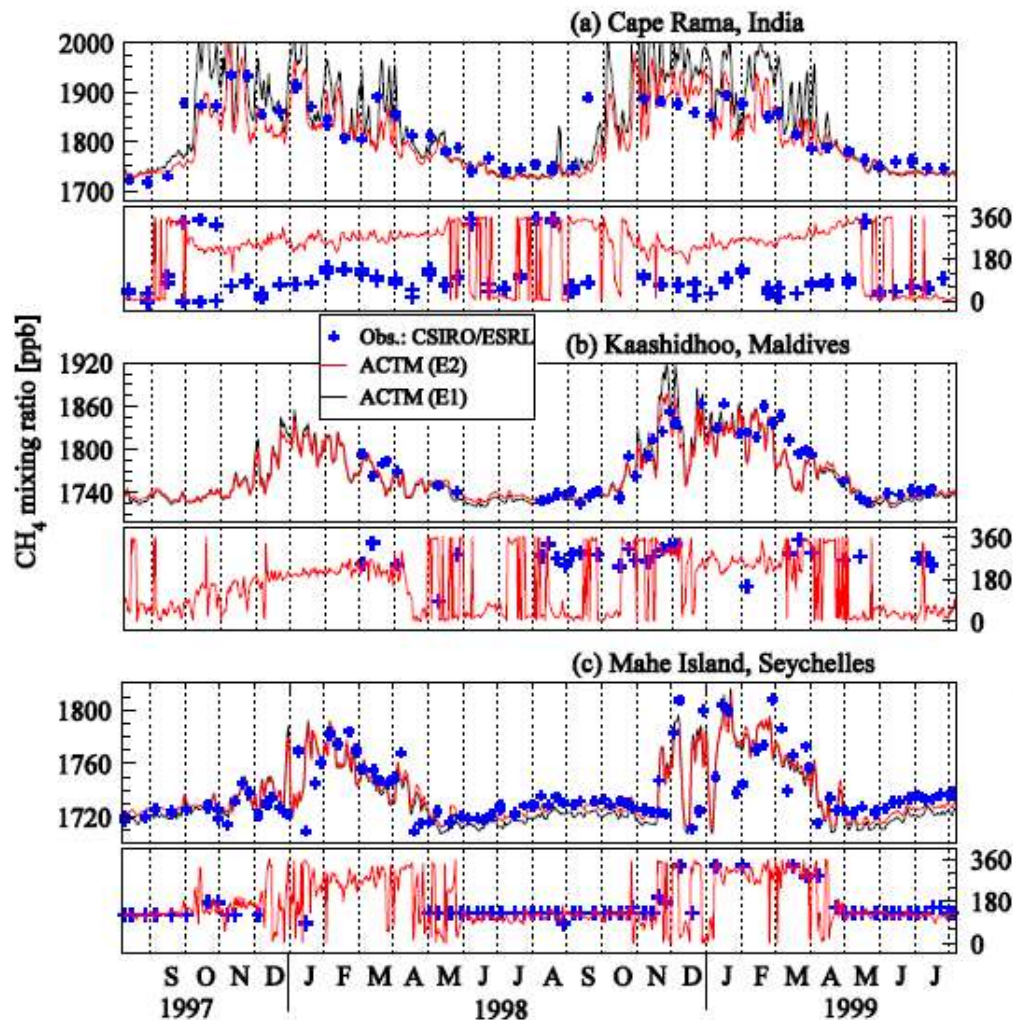
Species	Timescales/Lifetimes		
	Source	Loss~	Transp.
CO <sub>2</sub>	~Hour	100 yr	PBL/Dly
CH <sub>4</sub>	~Day	10 yr	Hem./Mn
N <sub>2</sub> O	~Day	120 yr	IH/Yr
SF <sub>6</sub>	~Year	>3000	IH/Yr

We use JAMSTEC's **ACTM** in this study (transport is driven by the CCSR/NIES/FRCGC AGCM v5.7b, nudged with JMA reanalysis – JRA55)

We **characterised OH** for the global mean concentration (the main 'Loss' term) and NH/SH ratio (Patra et al., Nature, 2014)

09 June 2015; ACAM-Bangkok

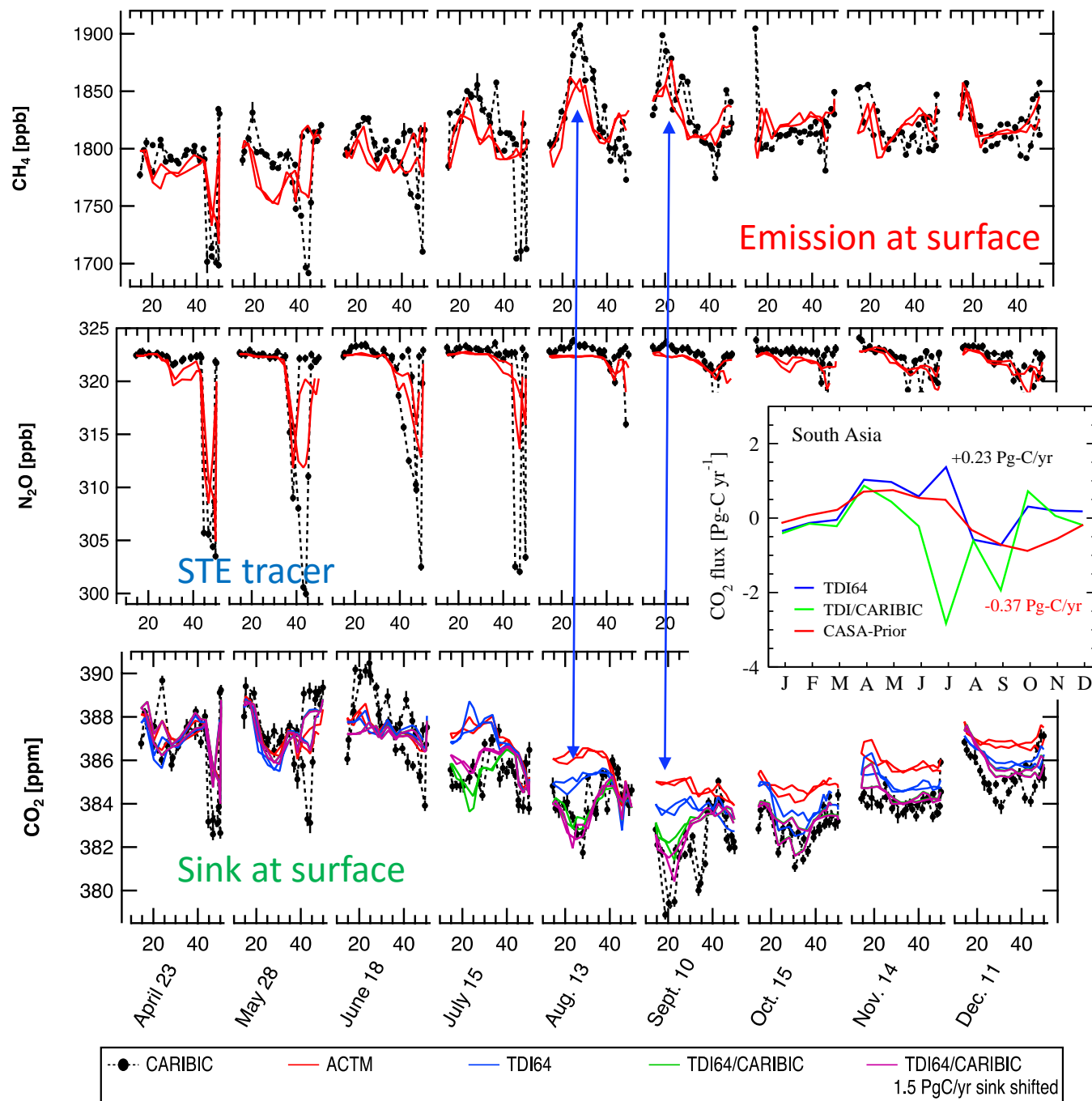
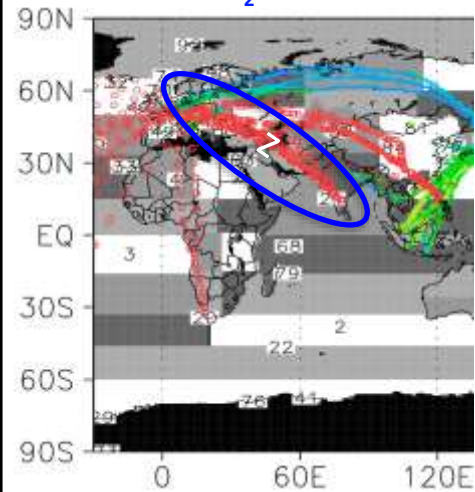
# CH<sub>4</sub> Seasonal Cycles at Surface – produced by chemical loss and South Asian monsoon



# CH<sub>4</sub>, N<sub>2</sub>O, CO<sub>2</sub> Variability over India, between Chennai- Frankfurt

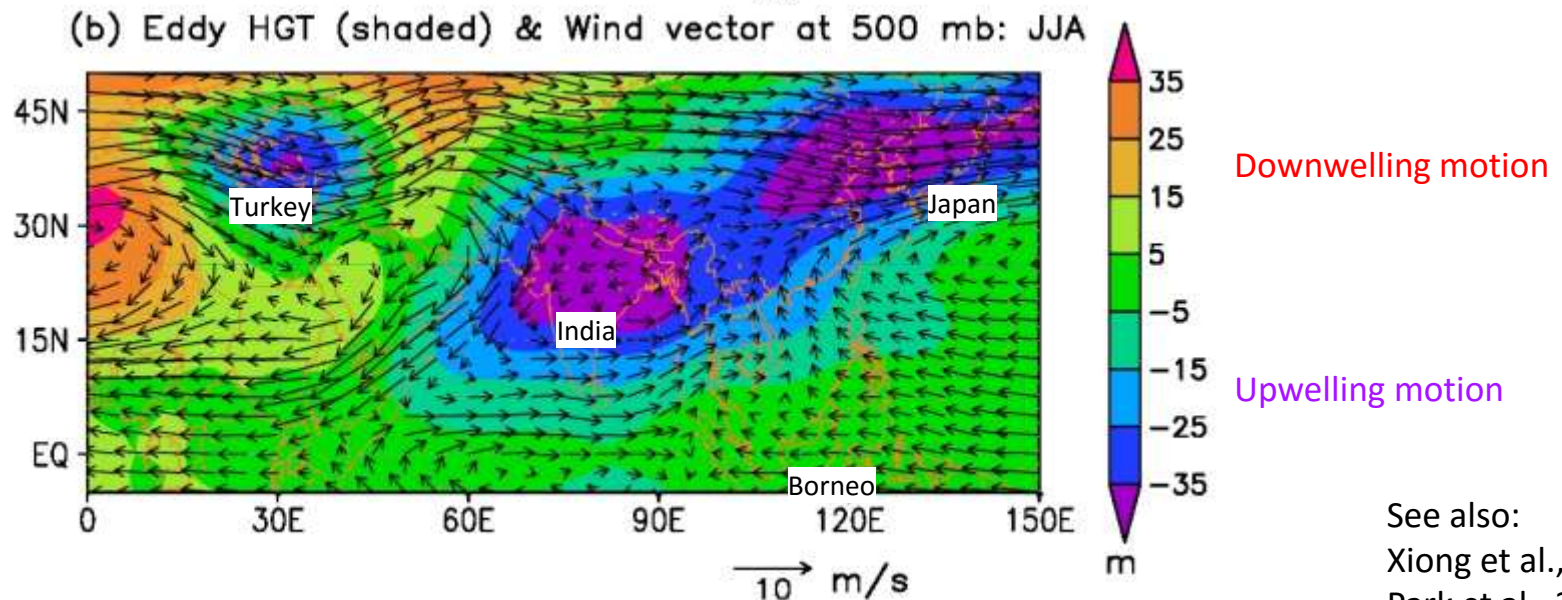
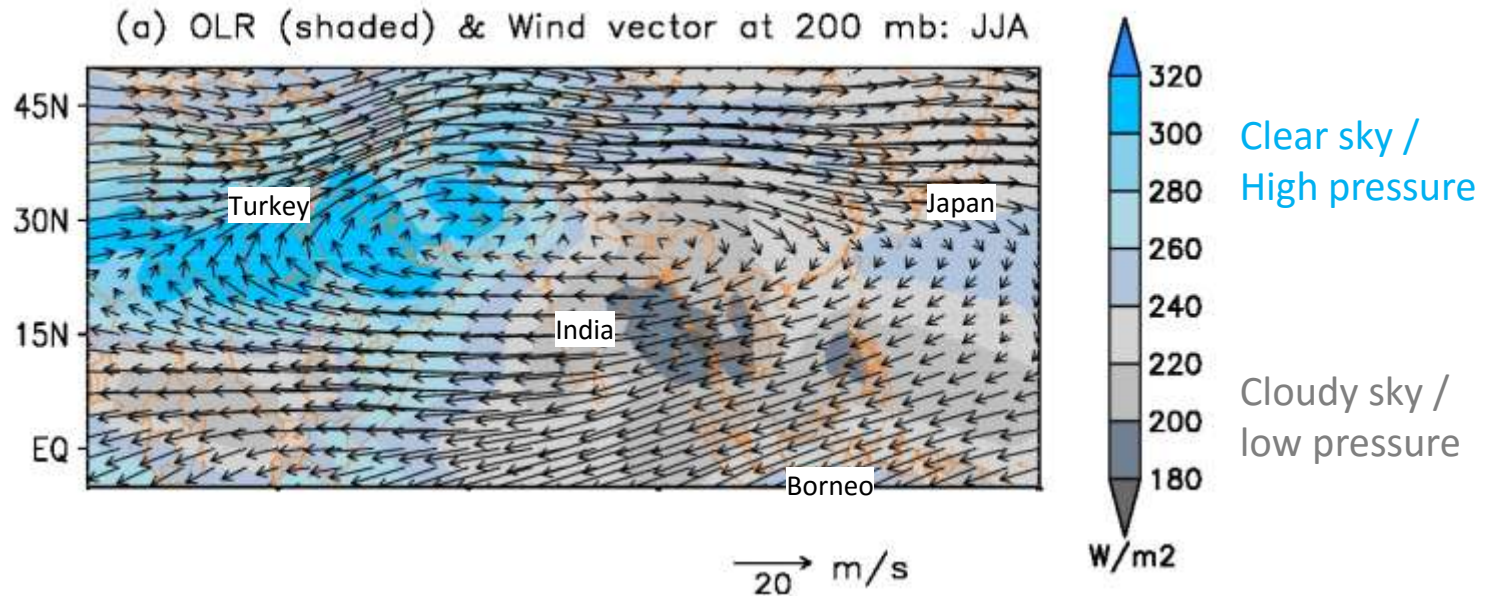


CARIBIC CO<sub>2</sub> data for 2008



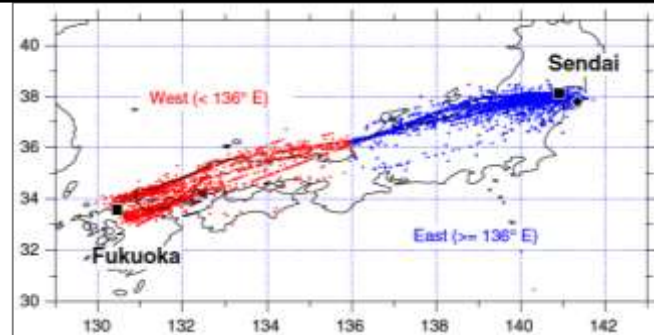


# Dynamical conditions: South to East Asian transport corridor

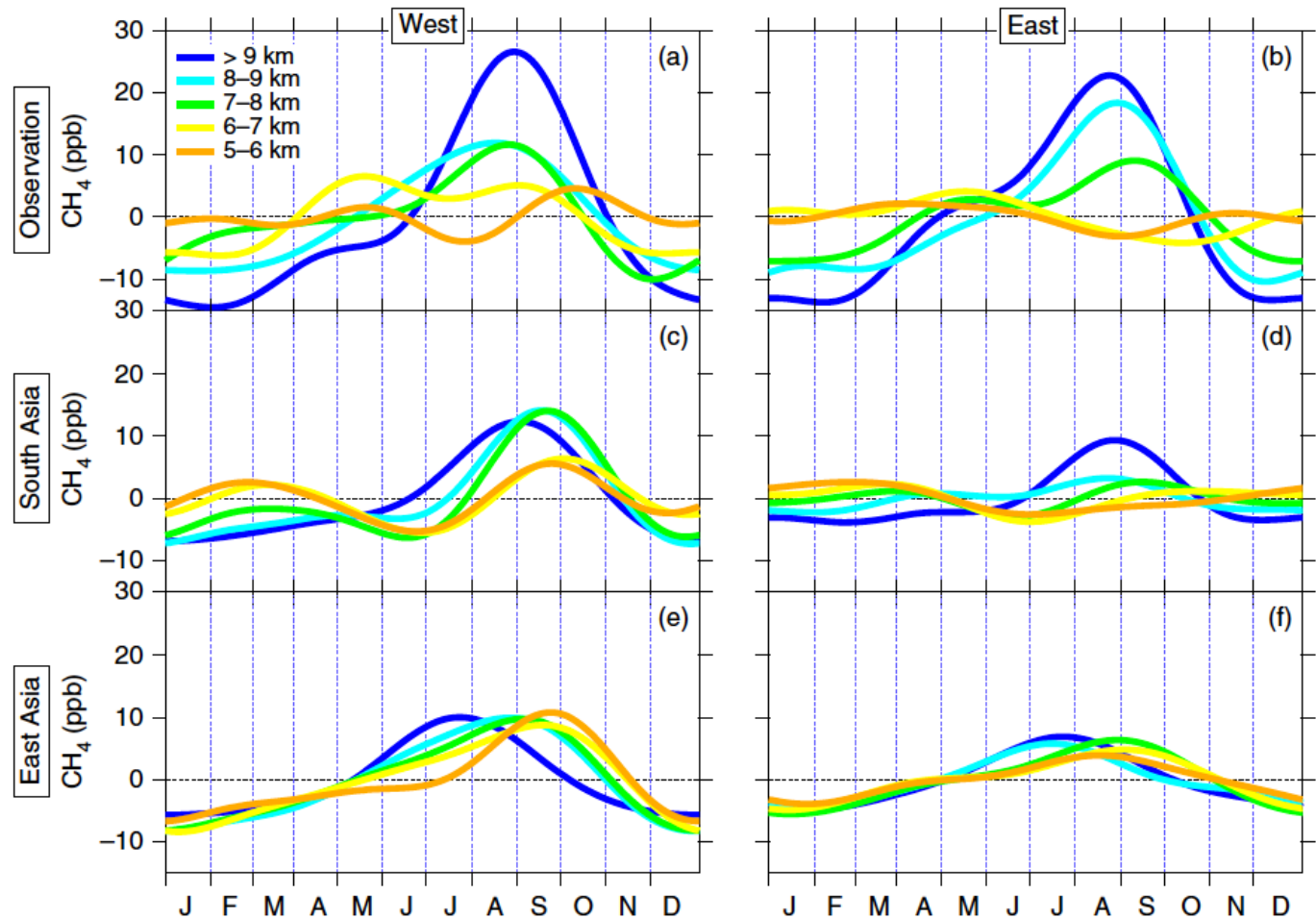


See also:  
Xiong et al., 2009  
Park et al., 2007

# Transport of CH<sub>4</sub> emission signal from India to Japan



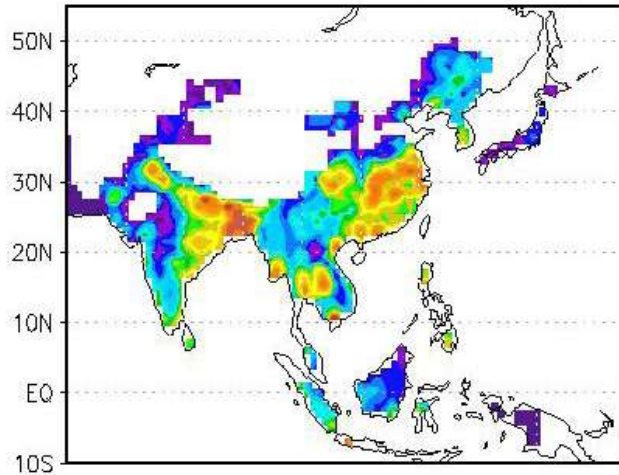
ACCLIP



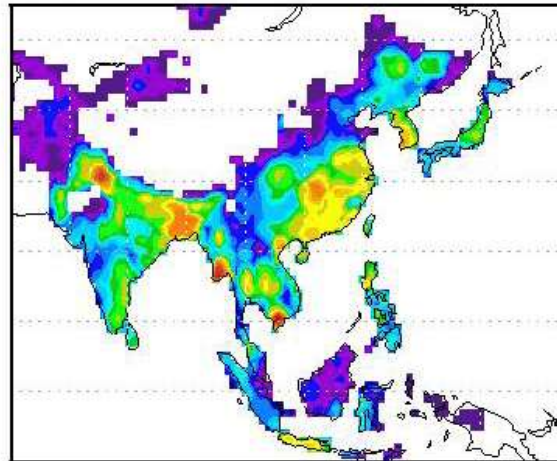


# CH<sub>4</sub> flux from rice fields: uncertainties in emissions

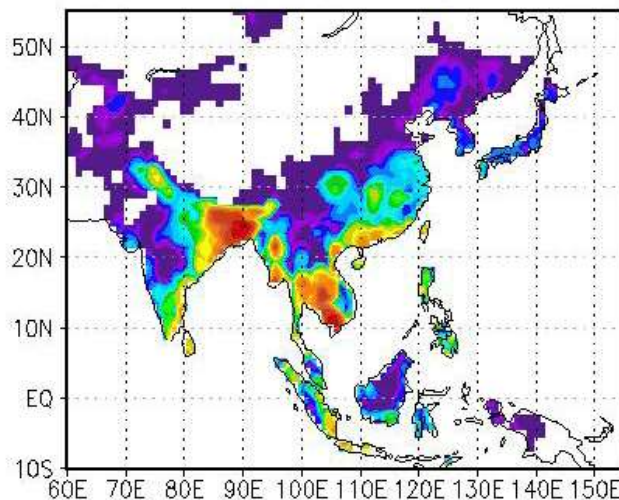
(b) EDGAR3.2 flux (39.3 Tg/yr)



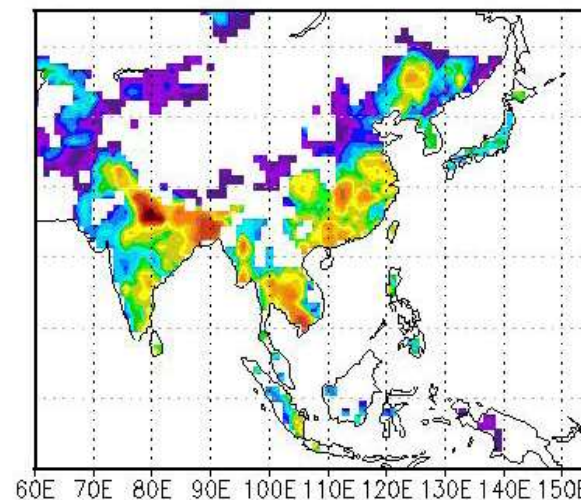
(c) REAS flux (26.7 Tg/yr)



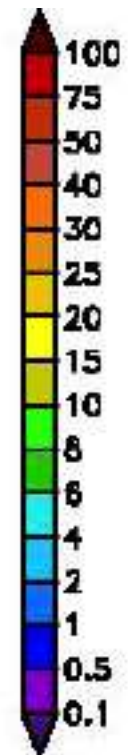
(d) VISIT (Cao) flux (39.2 Tg/yr)



(e) VISIT (WH) flux (43.0 Tg/yr)

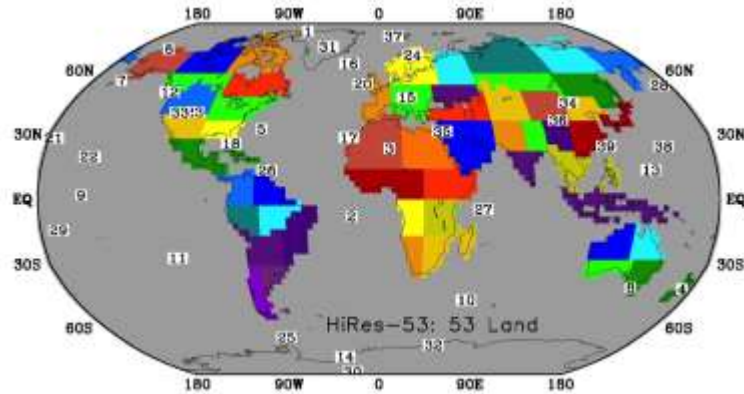


mg/m<sup>2</sup>/day





# CH<sub>4</sub> emission trends – East Asia vs. the Tropics



$$C_S = (G^T C_D^{-1} G + C_{S_0}^{-1})^{-1}$$

$$S = S_0 + (G^T C_D^{-1} G + C_{S_0}^{-1})^{-1} G^T C_D^{-1} (D - D_{ACTM})$$

$S_0$  = regional prior sources

$C_{S_0}$  = Prior source covariance = 50% of region-total emission for each month

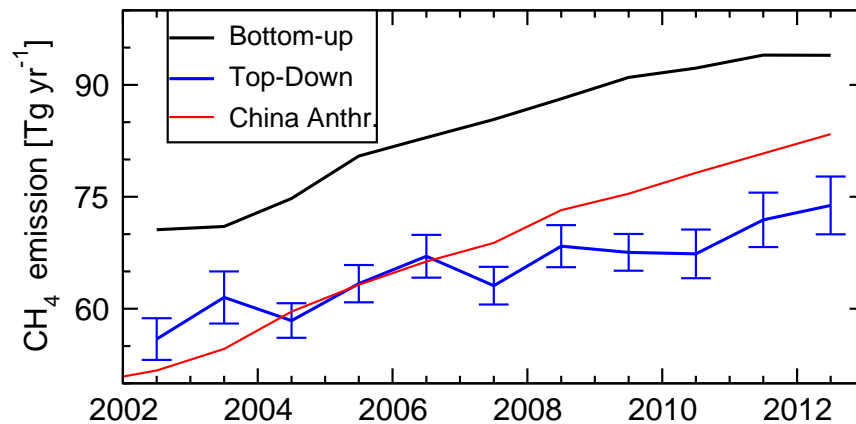
$D$  = atmospheric concentration data

$C_D$  = Data covariance = 10 ppb; 5 ppb for measurements + 5 ppb for model

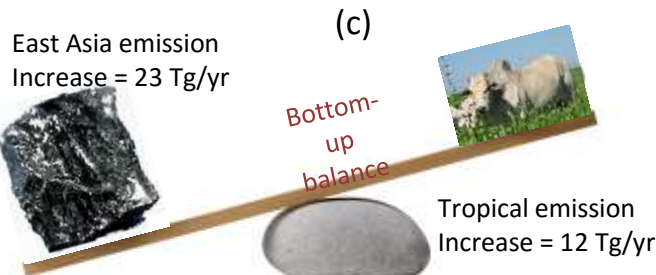
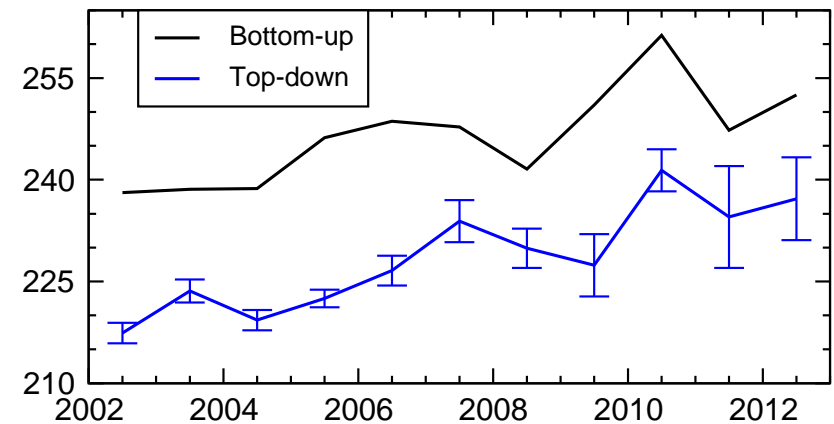
$D_{ACTM}$  = ACTM simulation using  $S_0$

$G$  = Green's functions for regional source-receptor relationships

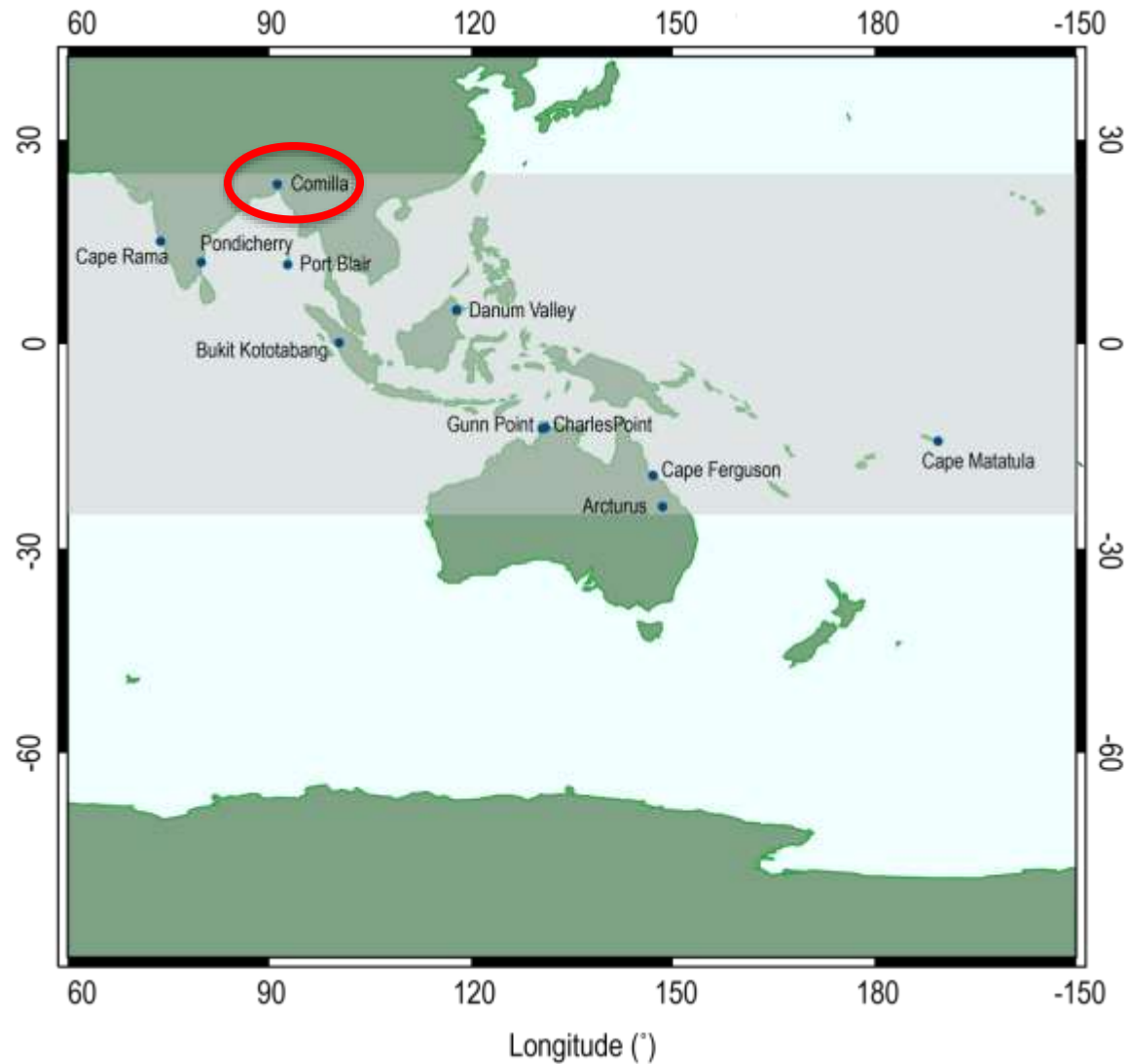
(a) East Asia: China, Japan, Korea



(b) Tropical Land



# Global and Asia-Australian Regional GHGs Network



Courtesy of: Marcel van der Schoot, Pep Canadell

# JAMSTEC established site in collaboration with Dhaka Univ/NIES

Observatory of Bangladesh Meteorol. Dept.  
Comilla (23.45°N, 91.20°E)

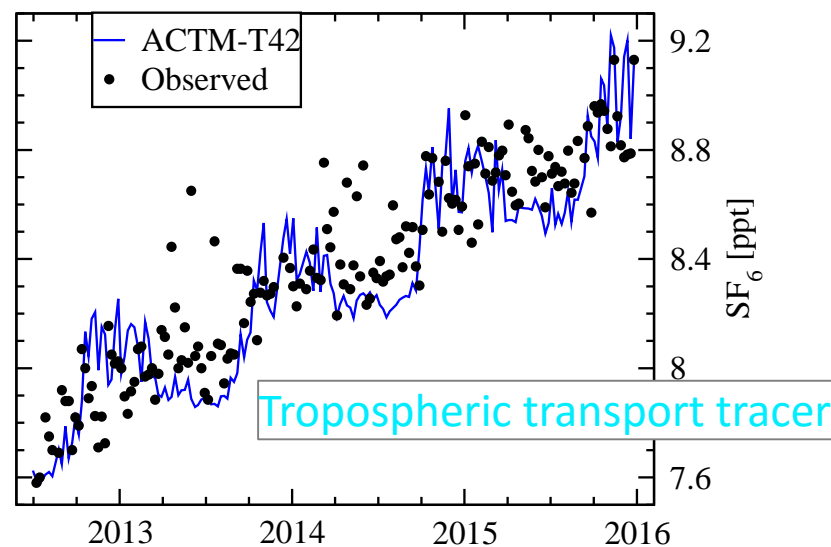
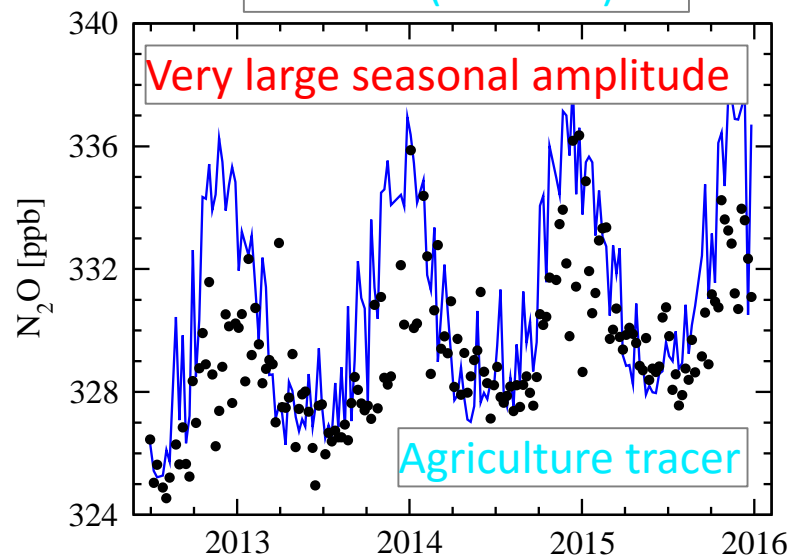
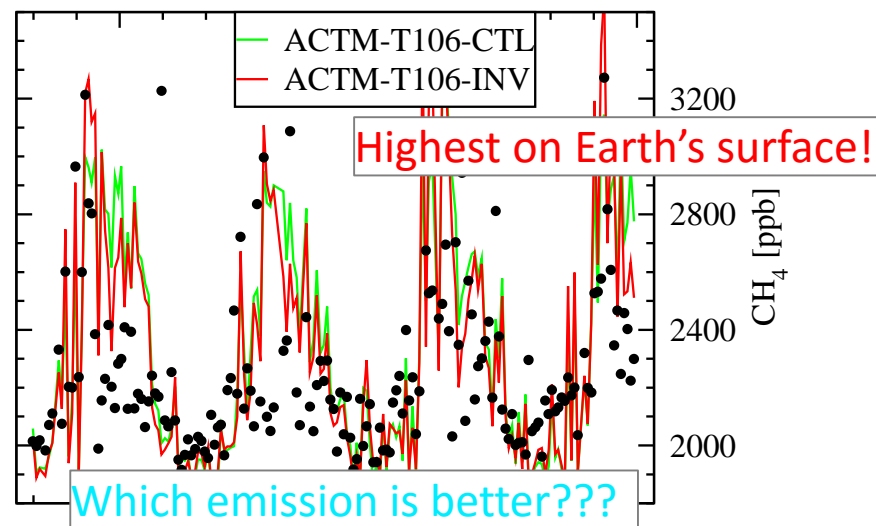
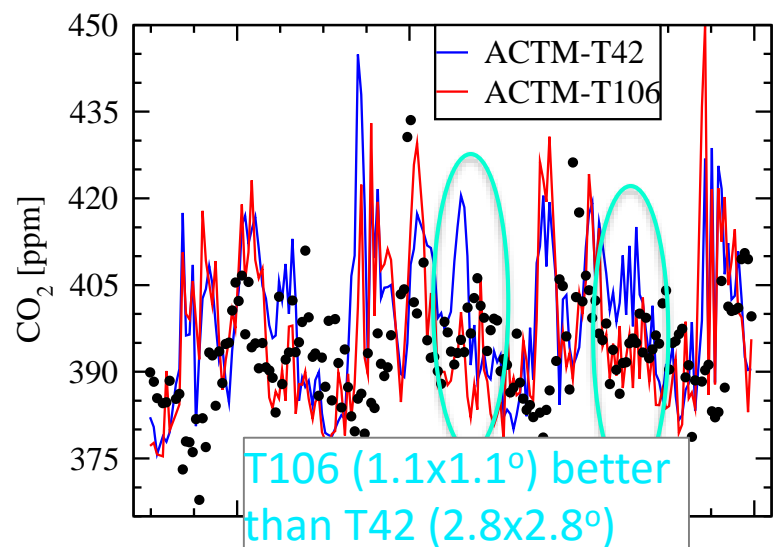


## Funded by:

Asia-Pacific Network (APN): Patra and Canadell  
Ministry of Environment, Japan (PI: S. Hayashida)



# Analysis of Comilla measurements using ACTM



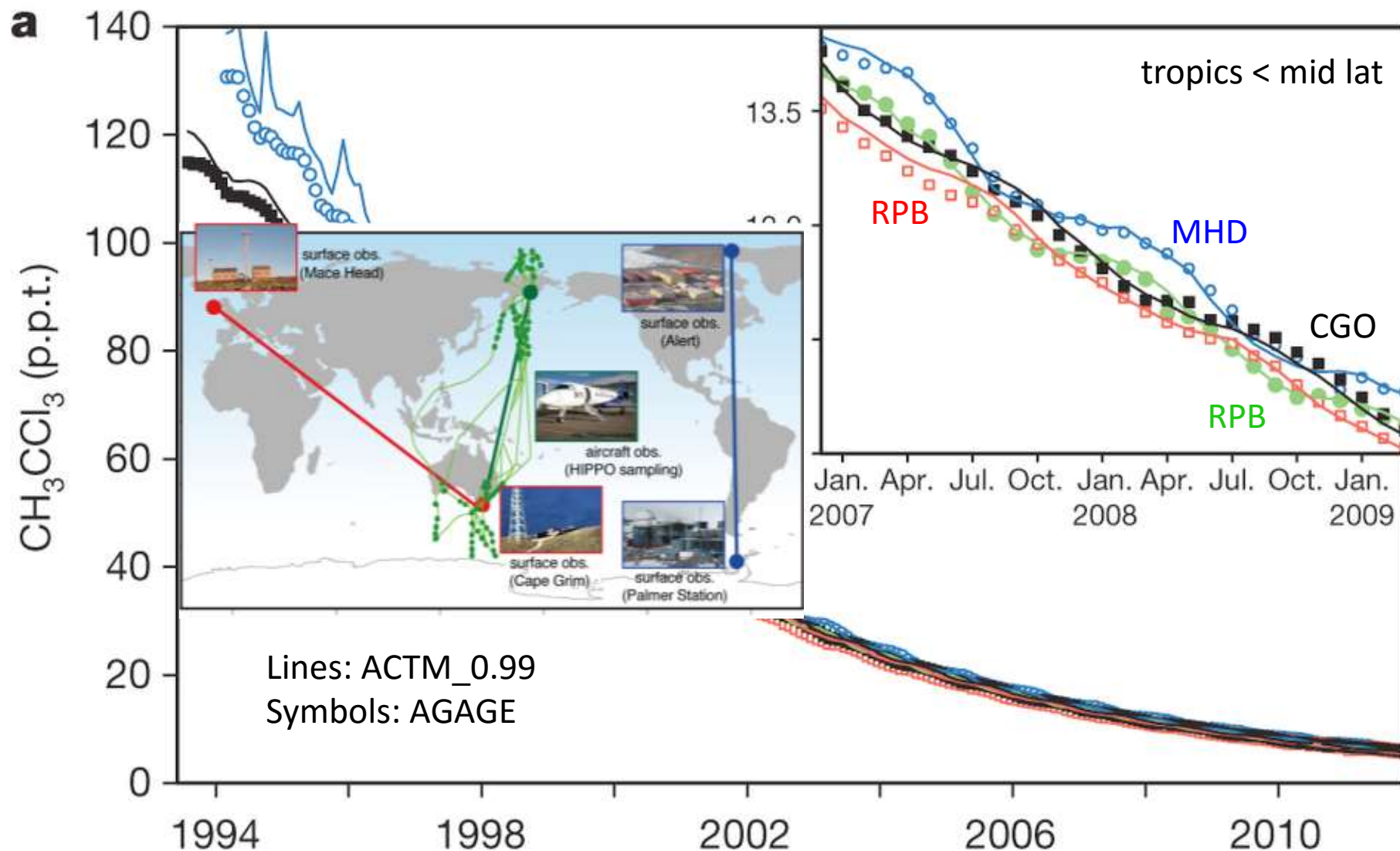


# Conclusions

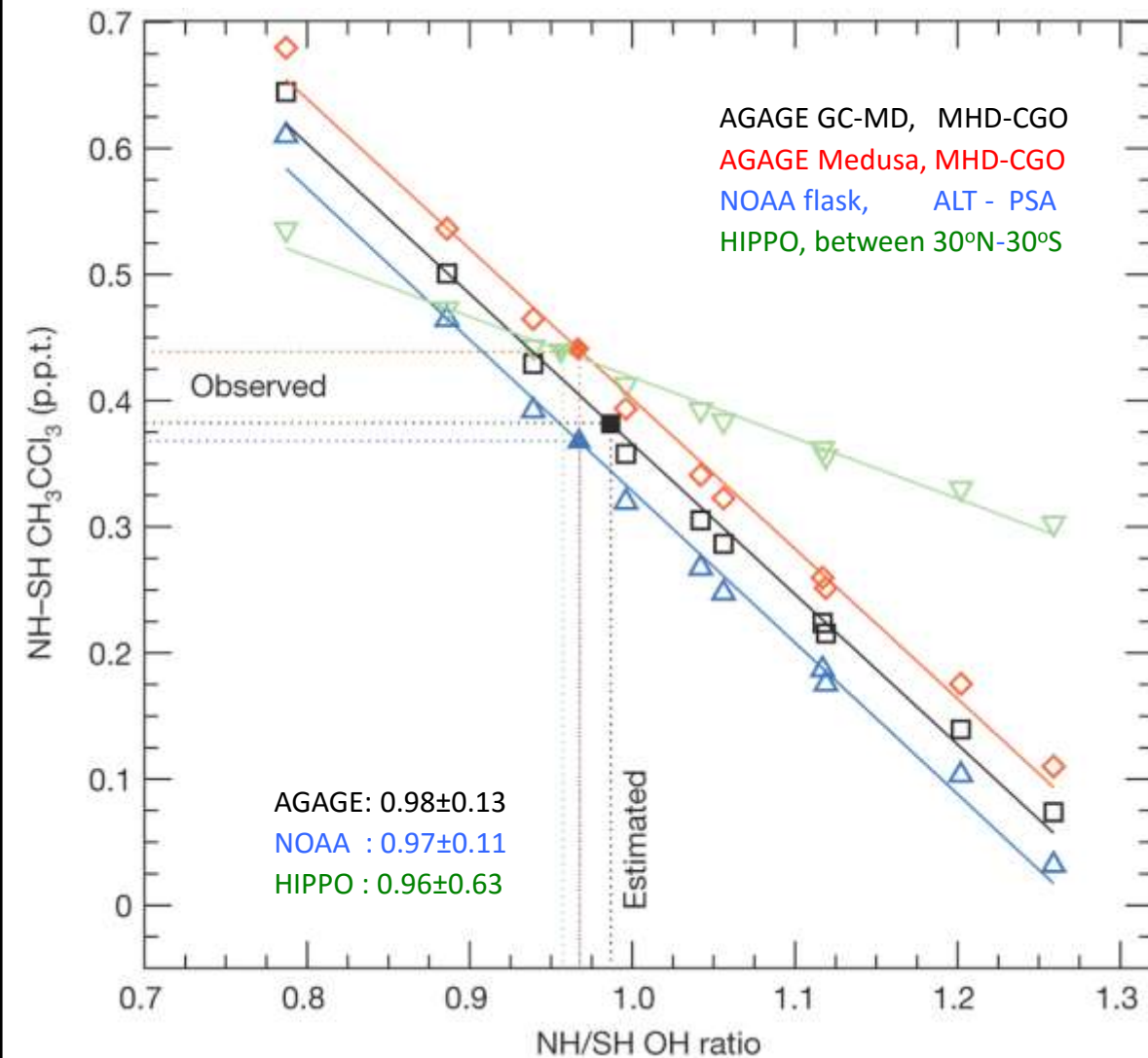
- Variations of GHGs concentrations near the Earth's surface and in the upper troposphere are strongly affected by monsoon
- Transport of CH<sub>4</sub> emission signals from South Asia can be observed by aircraft measurements over Japan (South and East Asian Transport Corridor - SEATC)
- Emissions of CH<sub>4</sub> by inventories are overestimated for China (East Asia) and the tropics – a conclusion following characterisation of OH
- New measurements from Comilla, Bangladesh are promising for separating contributions from emission vs transport patterns on high CH<sub>4</sub> over the eastern IGP

Thank you

# Temporal evolutions of $\text{CH}_3\text{CCl}_3$ in the atmosphere



# Various OH fields with different NH/SH OH ratios



Method	NH/SH ratio	Analysis of period	Reference
$^{14}\text{C}$ -CO, box model	Significantly less than 1	1989-1990	Breninkmeijer et al., 1992
$\text{CH}_3\text{CCl}_3$ , box model	$0.87^* \pm 0.15\%$	1998-1999	Montzka et al., 2000
$\text{CH}_3\text{CCl}_3$ , 3-D model	$\sim 0.98$	1991-2000	Krol and Lelieveld, 2003
ACCMIP – IPCC AR5, full Atmos. Chem. model	$1.28 \pm 0.10$ (range: 1.13 to 1.42)	1980-2010	Naik et al., 2013
$\text{CH}_3\text{CCl}_3$ , 3-D model; transport validated	$0.97 \pm 0.12$	2004-2011	Patra et al., 2014



# ACAM-2017 notes

- **Monday Morning -2**
- **Geywui Wang : PNAS 2016**
  - $\text{SO}_2 + \text{OH} + \text{M}$  (gas phase reaction rate is slow  $\tau \sim 1\text{week}$ ),  $\text{SO}_2 + \text{H}_2\text{O}_2$  in aqueous phase reaction is very fast
  - $\text{SO}_2$  oxidation by  $\text{NO}_x$  is neglected in text books, but in China  $\text{NO}_2$  and  $\text{NH}_3$  are high (compared to say USA)
- **Puerto: CO, O<sub>3</sub> from Nepal**
  - Daily PBL height (ECMWF) is used for time series analysis – doesn't make sense (Q.)
  - Modelling work? (Crawford) It seems inventories are 4-5 times lower than that is needed for matching model with obs (Mark)
- **Abdus Salam**
  - Air pollution cost 1% GDP growth
- **Gong: Isoprene and its oxidation**
  - Nearly 40%  $\text{O}_3$  is produced by  $\text{OH} + \text{Isoprene}$  reaction at the Nangling site
- **Pratima Gupta: Emission and depositio of Black Carbon in the IGP**
  - How representative?
- **Fahim Khokar: Lohore smog(ke)**
  - Delhi smog formed within 30 min in one day of Nov (pictures between 3:30pm – 4pm)
  - 80% crop residus are cleared through open burning,
- **Monday Afternoon 1**
- **Helen Worden – MOPITT 17 years**
  - The trends from MOPITT (decreasing) & MLO (increasing) CO are not consistent.
- **Papori Dahutia : good analysis covering the East India/Bangladesh/Bhutan – aerosol vertical profiles (WRF-Chem run for her and Abdus Salam, Dhaka univ)???**
- **Crawford : HAM-AQ research opportunity ; installation of Pandora instrument, upwad looking  $\text{O}_3$ ,  $\text{NO}_2$  instrument**
- **Monday afternoon 2**
- **Harder (MPI) : ONO experiment using HALO (see Ojha et al., 2017)**
  - Injection of  $\text{NO}_x$  maintains recycling efficiency for OH
  - $\text{RO}_2$  inside the anticyclone elevated;  $\text{NO}_x$  increased due to lightning by about 30%
- **Elliot Atlas - POSIDON**
  - Organic Halogens, VSLs,  $\text{CH}_3\text{I}$ ,  $\text{Cl}$ , ....
  - Large variabiities in VSLs compounds – how will ACTM perform?
  - Methane and NMHCs showed a few days of high values and a peak in concentration at around 15 km

# ACAM-2017 notes

- **Tuesday afternoon -2**
- **Shih-Chun Candince Lung:**
  - Emission from incense sticks burning in the temples on air quality, PM2.5 exposure.
  - Based on the research results some temples in Taiwan have banned incense burning from vendors, but still allows by the visitors.
- **Guy Brasseur:**
  - Model underestimated O3 in day, overestimate NO2 in the night. This is because of the representation of PBL in the night
  - In another day, when PM2.4 reached 400 ug, but no model predicted that – underestimated by half or so
  - LES (large eddy simulation) combined with the regional models
- **Jordan Schnell**
  - Model simulation using CMIP5 underestimate PM2.5, but the results using CMIP6 emissions increased modeled concentration and compared better with observations. But many a times still underestimate the observations
  - The event of Diwali is completely missed by model, because emissions are not known/inputed to model.
  - Air Stagnation Index doesn't seem to be a great predictor for air quality in the northern-central IGP region, may be a bit of link is seen over the eastern IGP
- **Mary Barth**
  - Clear improvement in WRF-Chem over CAM-chem global model, but WRF simulations at two resolutions of 60km and 12km were pretty similar.
  - Previous speaker said you need more emission (CMIP6) to simulate PMs over the IGP, but WRF-Chem is fine with CMIP-5 emission inventory (unless Rajesh did some trick!)
- **Prashant Dave**

# ACAM-2017 notes

- **Wednesday morning -1**
- **Ru-Shan Gao:**
  - South Asian monsoon anticyclone keeps the airmass within or transport towards the East and South-east Asia region
  - Aerosol layer from ATAL could penetrate up to 2 km into the stratosphere by the monsoon anticyclone ; CESM with CARMA aerosol module does well in simulating the observations, at 2km above the tropopause, although there is an overestimation of aerosol at the UT region
  - ATAL contributes 15% of the total column aerosol surface area in the stratosphere, but much less than the overall tropical contributions of tropospheric aerosols to the stratosphere.
- **Duncan Fairlie: the BATAL campaigns**
  - Balloons are launched from NARL (with A. Jayaraman)
- **Michael Schwartz : MLS CO analysis using EOFs**
  - This analysis showed different modes of variabilities in CO – east-west, north-south dipoles etc.
  - Now that MLS CO is now longer than 10 years (also MLS on UARS) – have you looked into effect of Asian Monsoon on CO trends & variability in stratosphere?
- **Jiali Luo : MLS & IASI CO data analysis**
  - Daily maps are created using 5x5 degree Gaussian interpolation for analysis of variability
- **Michelle Santee : MLS based gases climatology**
  - High O3 in the UT region in MLS data is coming from the stratosphere by the Rossby wave breaking in the midlatitudes, and being transported to the ASM region
  - No correlation between IWC (convection tracer), CO and O3 variability – my impression is that the timescale and sources are not comparable (agreed in the Q&A session)
- **Jianchun Bian : SWOP – research opportunity**
- **Sachin Gude: WIFEX – IGIA measurements and modelling**
  - How is the source of water in to the system
- **Arnico Pandey : ICIMOD's atmospheric science activities**
  - New sites : Lumbini, Chitwan and Ratnank
  - Long-term climate observatory : Gedu in Bhutan (building is complete) and Imphyakamana in Nepal for GHGs, Aerosols
  - Yala glaciers site : BC of 1100 ug/m3 in April (BB??), but the values are quite high in other months too
  - Emission study for motor vehicles, before and after servicing : 1-2% of the vehicles emit 90% of BC emission; so a 10\$ servicing can reduce emissions by 90%  
800,000 2-wheel and 150,000 4 wheel drive in Kathmandu. Pulling out smoking-gun vehicles from the road will make huge impact (90% reduction)

# ACAM-2017 notes

- **Thursday morning -1 (No Indians arrived the venue until 8:40, even after delaying the start, as audience)**
- Simone Brunamonti, Balloon-borne measurements
  - Aerosols, water vapour, ice, very thin cirrus clouds are observed
  - The ECMWF PV plots show clearly that the surf zone is located just over Nainital in November, which was located more north in August.
  - Measurements were also made from Lasa (at the same latitude) and afterwards from Nainital. The delay in
  - ECMWF overestimate H<sub>2</sub>O by 0.5 ppm compared to the observations. Similarly temperature also high-biased
  - Two ways to analyse the balloon profiles for climatology; 1) just bin by pressure/alti and take mean or 2) adjust the cold-point temperature and then take the mean with reference to CPT
- Federico Fierli: How pollutants and water vapour enter the anticyclone?
  - Role of conduit (Bergeman, Fierli, 2013)
  - ISSCP gives OLR and compared with the WRF model run for the July
- Alina Fiehn (Fiehn et al., ACP, 2017)
  - Stemmler et al., 2015 (CHBr<sub>3</sub> emission using HMMOC model), include seasonal cycle; Indian ocean is a strong source of CHBr<sub>3</sub>
  - They find the transport in to the stratosphere did not change much for the different emission maps, from HAMMOG and Ziska et al. even though the emission were 3 times in HAMMOG!
- Paul Konopka: Chimney vs blower (see Ploeger et al., ACP, 2017)
  - Strong vertical transport across the tropopause (chimney) and strong isentropic transport (blower) mainly occurs above the tropopause
  - How sensitive are the results for heating rates, because there are uncertainties in heating rates between the reanalysis data, ECMWF, MERRA, JRA55 ...
- Klaus Gottschaldt : Analysis of HALO data using CLaMS model
- Bärbel Vogel :
  - HCFC-22 tag tracers simulations are compared with MIPAS measurements. She used HCFC-22 emission maps looks similar to CH<sub>4</sub>/FF CO<sub>2</sub> – distribution using population proxy. How will the transport efficiency change with a more realistic emission distribution??? I understand HCFC-22 has a long lifetime, but the horizontal transport may redistribute the emission signals differently before transporting up by convective transport.
- Rolf Muller : looks so similar to Vogel's presentation
- Abhishek Misra : Isoprene emission in the IGP, using measurements from Mohali
  - Isoprene emissions are sensitive to temperature and solar radiation; WRF-Chem underestimates observed Isoprene concentrations using emissions from EDGAR, Guenther & FINN
  - How is the OCHO (Jim) OH (PKP) concentrations ? They have measurements of O<sub>3</sub> and OH – will have a look how is the OH in model
- Pallavi Saxena:
  - Too low NO<sub>x</sub>, (~20ppb?), and too low O<sub>3</sub> (80 ppb peak) – which doesn't seem to be quite right for a megacity like Delhi?
- Tuan Nguyen Dinh
  - Benzene emissions increased by 3 times in Vietnam due to increase of motor vehicle from 3 mil in 2010 to 9 mil in 2015 or some such period