Global Source and Transport of Carbonaceous Aerosols

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Background

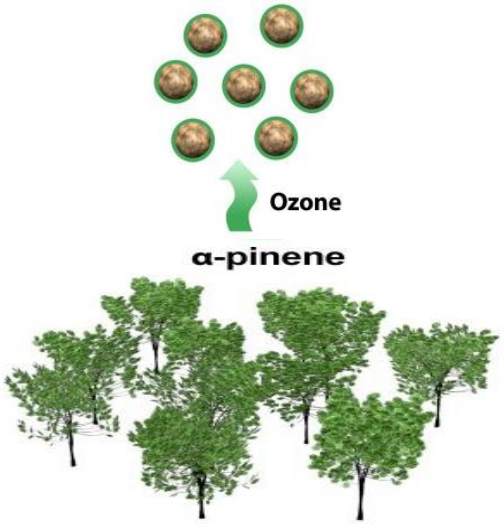
Primary Sources

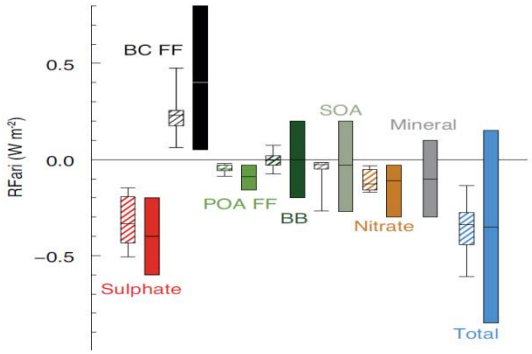
Black carbon (BC)

Primary OA (POA)

Secondary OA (SOA)

Organic aerosols (OA)





[*IPCC-AR5*]

Outline

- Long-range transport of black carbon aerosols to the Arctic and mid-Pacific regions
- Trans-boundary “transport” of black carbon emissions embodied in global trade system
- Global source of secondary organic aerosols (SOA) from gaseous and liquid phases

Long-range transport of black carbon aerosols to the Arctic

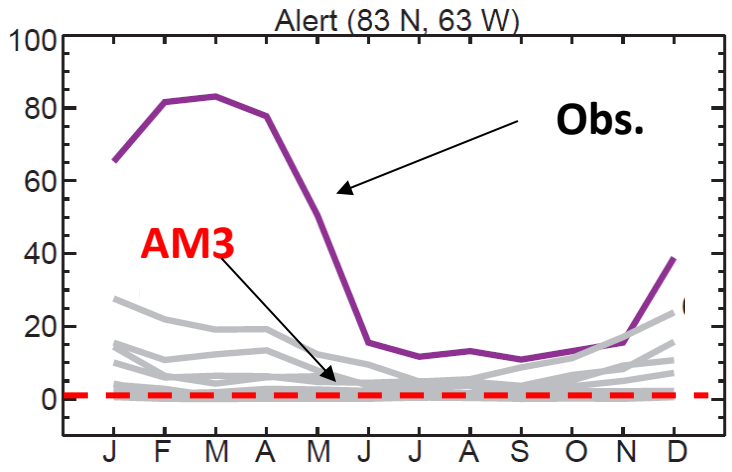
Model Improvements:

1. Wet deposition
   - BC aging (variable aging)

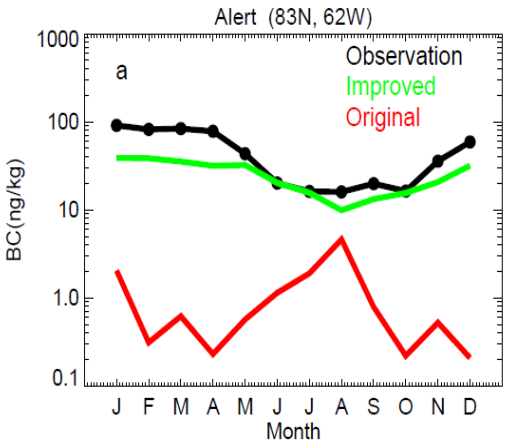
$$k_a = \beta \cdot [OH] + \delta$$

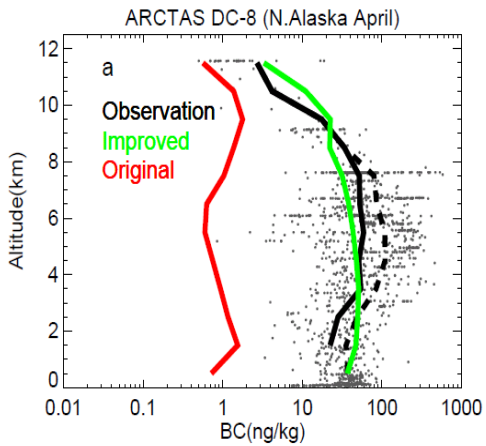
$\beta = 4.6 \times 10^{-12} cm^3 molecule^{-1} s^{-1}$; $\delta = 5.8 \times 10^{-7} s^{-1}$

   - Bergeron process
2. Dry deposition (surface dependent)



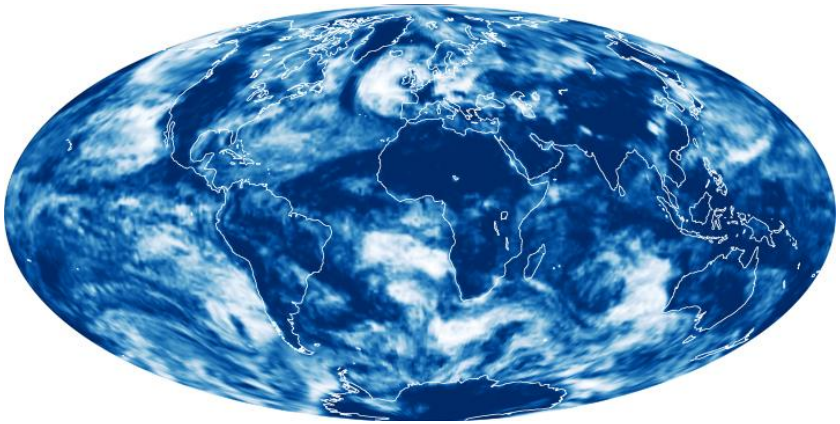
Shindell et al. [2008]





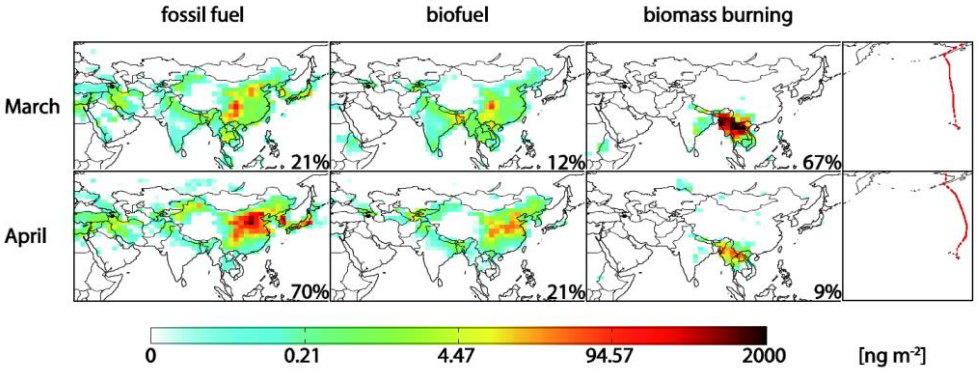
[*Liu et al. 2011,JGR*]

Long-range transport of black carbon to the Pacific Ocean



Low-altitude Cloud Fraction, December 27, 2008, from NASA Earth Observatory

GEOS-Chem adjoint sensitivity to HIPPO3 obs.

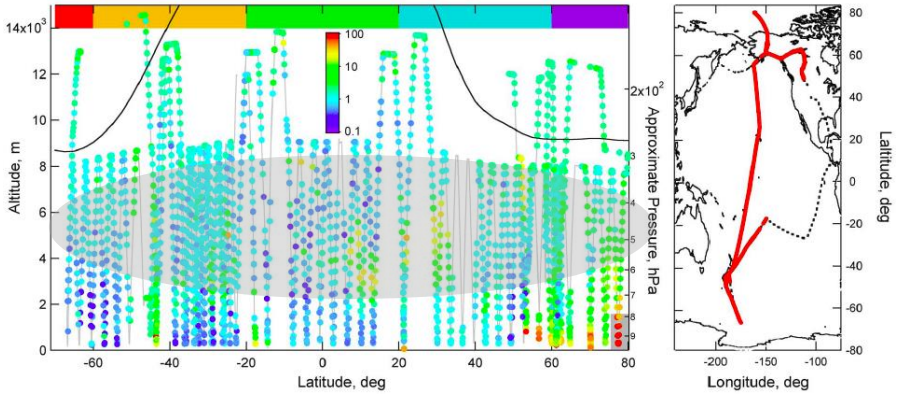


Idealized BC transport model:

$$C = Ee^{-\frac{T_N}{\tau}} + \sum_{n=1}^{N-1} Ee^{-\frac{T_n}{\tau}}\left(1 - e^{-\frac{T_{n+1}-T_n}{\tau}}\right)(1-R_{n+1})\cdots(1-R_N)$$

$$+ E\left(1 - e^{-\frac{T_1}{\tau}}\right)(1-R_1)\cdots(1-R_N)$$

HIAPER Pole-to-Pole Observations (HIPPO)



[*Schwarz et al.*, 2010]

	T_1 (d)	T_2 (d)	T_3 (d)	T_4 (d)	T_5 (d)
March	0.12	0.66	2.4	6.6	18.6
April	0.15	0.87	3.0	7.8	19.4
	R_1	R_2	R_3	R_4	R_5
March	9%	27%	51%	42%	26%
April	4%	18%	42%	37%	19%

[*Shen et al.*, 2014, *ACP*]

Source-receptor relationship of BC distribution

The 12 tagged continental regions

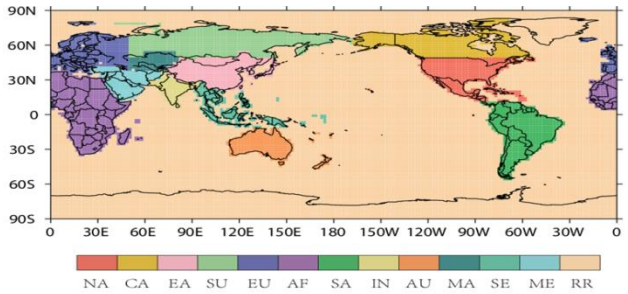
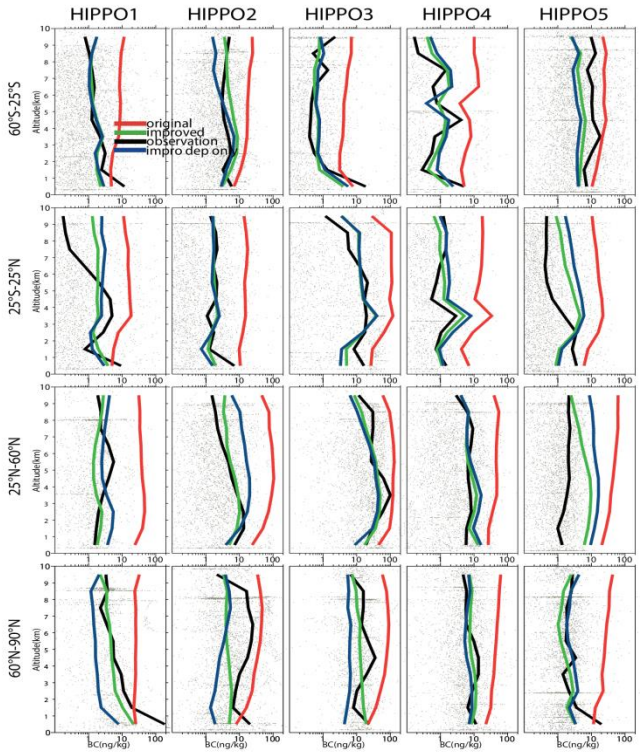


Fig.1 Thirteen defined source regions

The optimized aging timescale (hr) for different regional BC tracers

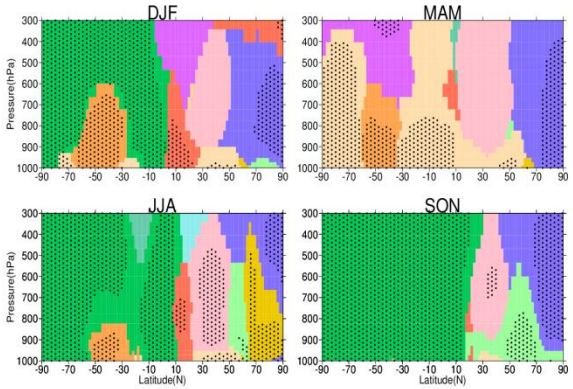
	Time	CA	SU	EU	MA	EA	ME	NA	SE	IN	AF	SA	AU	RR	Mean bias (Improved)	Mean bias (Original)
HIPPO1	Jan	200	120	60	120	4	48	60	4	4	4	60	4	4	3.4	26.4
HIPPO2	Nov	200	200	120	60	4	4	4	4	4	4	200	4	4	1.7	13.2
HIPPO3	Apr	200	200	200	200	24	60	4	24	38	48	4	4	200	1.4	6.6
HIPPO4	Jun	48	4	120	4	4	200	4	8	4	4	60	4	4	1.0	10.6
HIPPO5	Aug	60	4	12	4	4	4	4	4	4	60	4	27	4	2.2	18.7

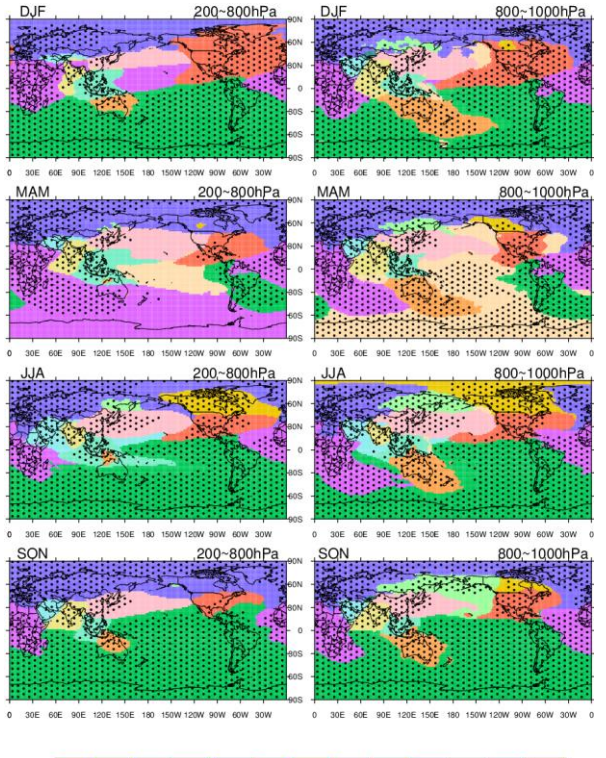
MOZART-4 results *vs* HIPPO obs.



The dominant regional contributors to column BC concentrations (right plots),

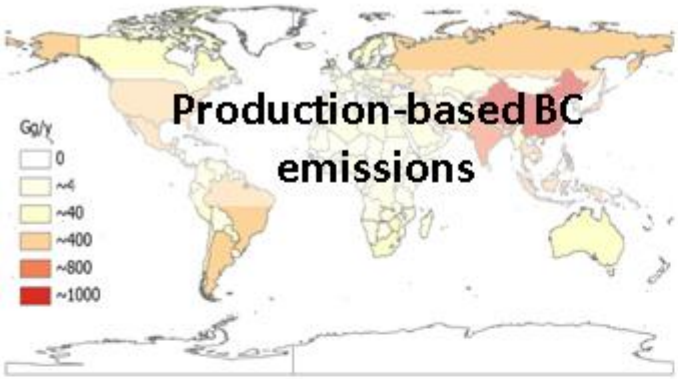
and zonal mean BC concentrations over the central Pacific (130 ℃W-150 ℃E, 2009-2011 average, below)

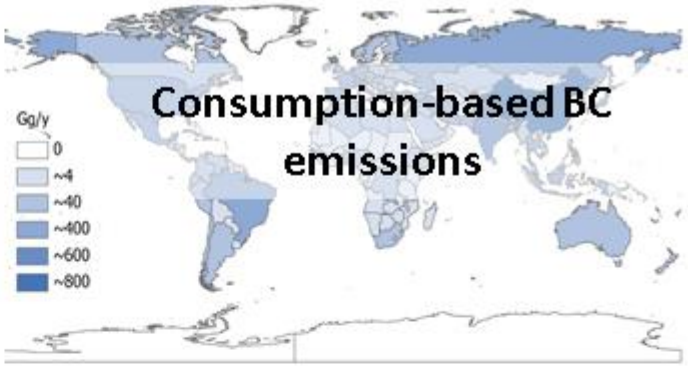


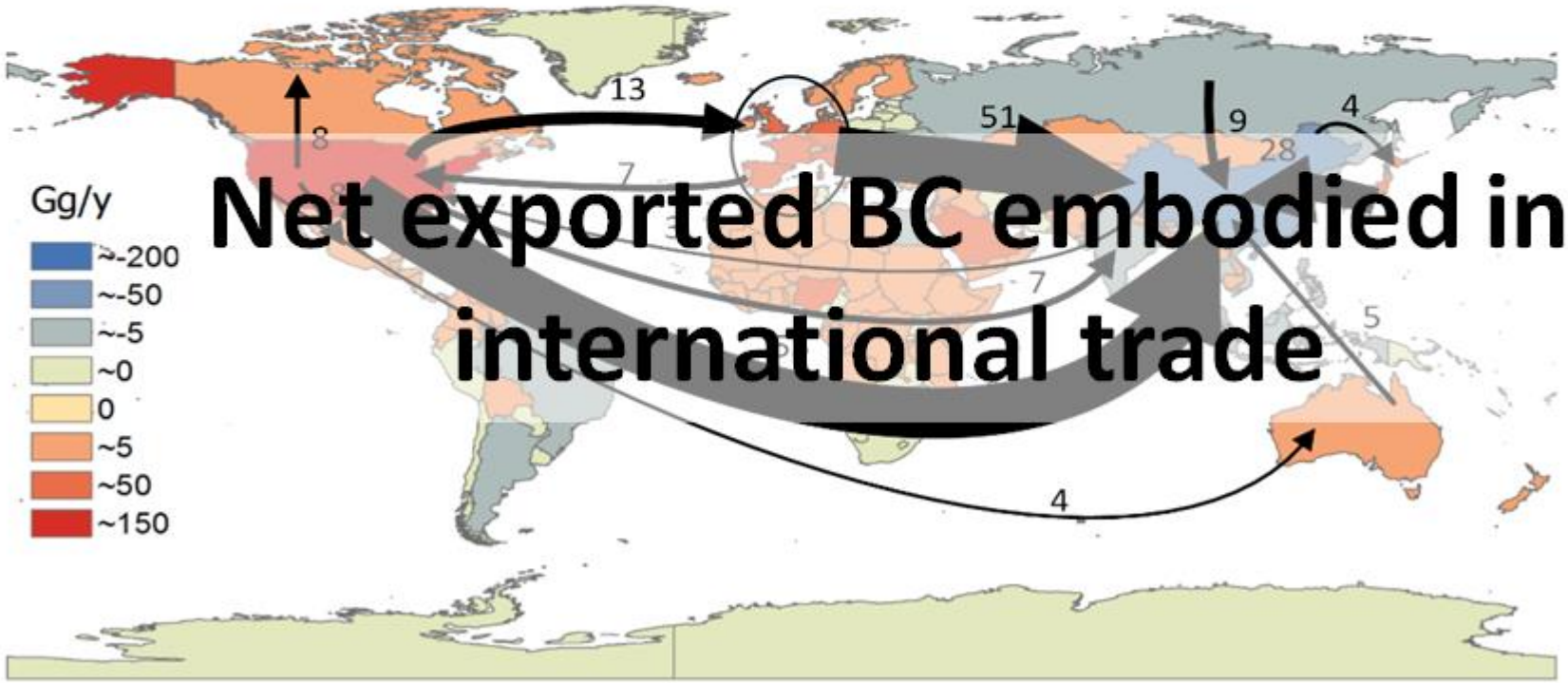


[*Zhang et al., in preparation*]

Production *vs* Consumption

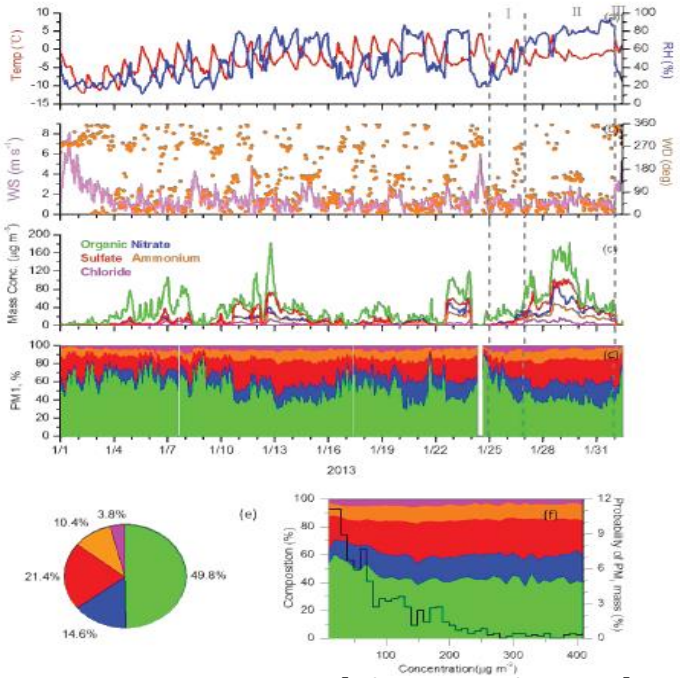




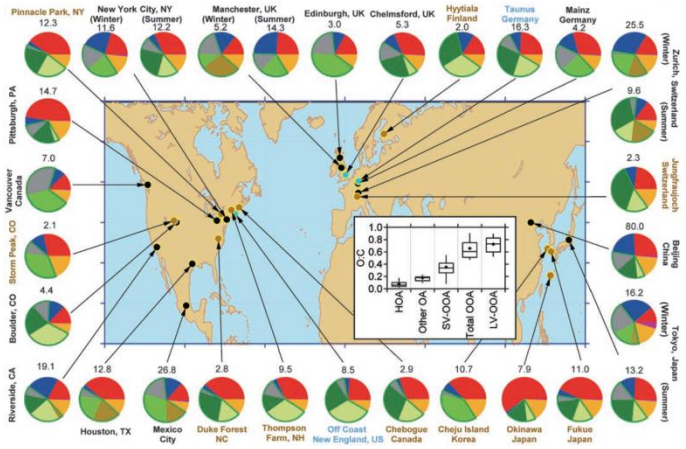


[*Meng et al., in preparation*]

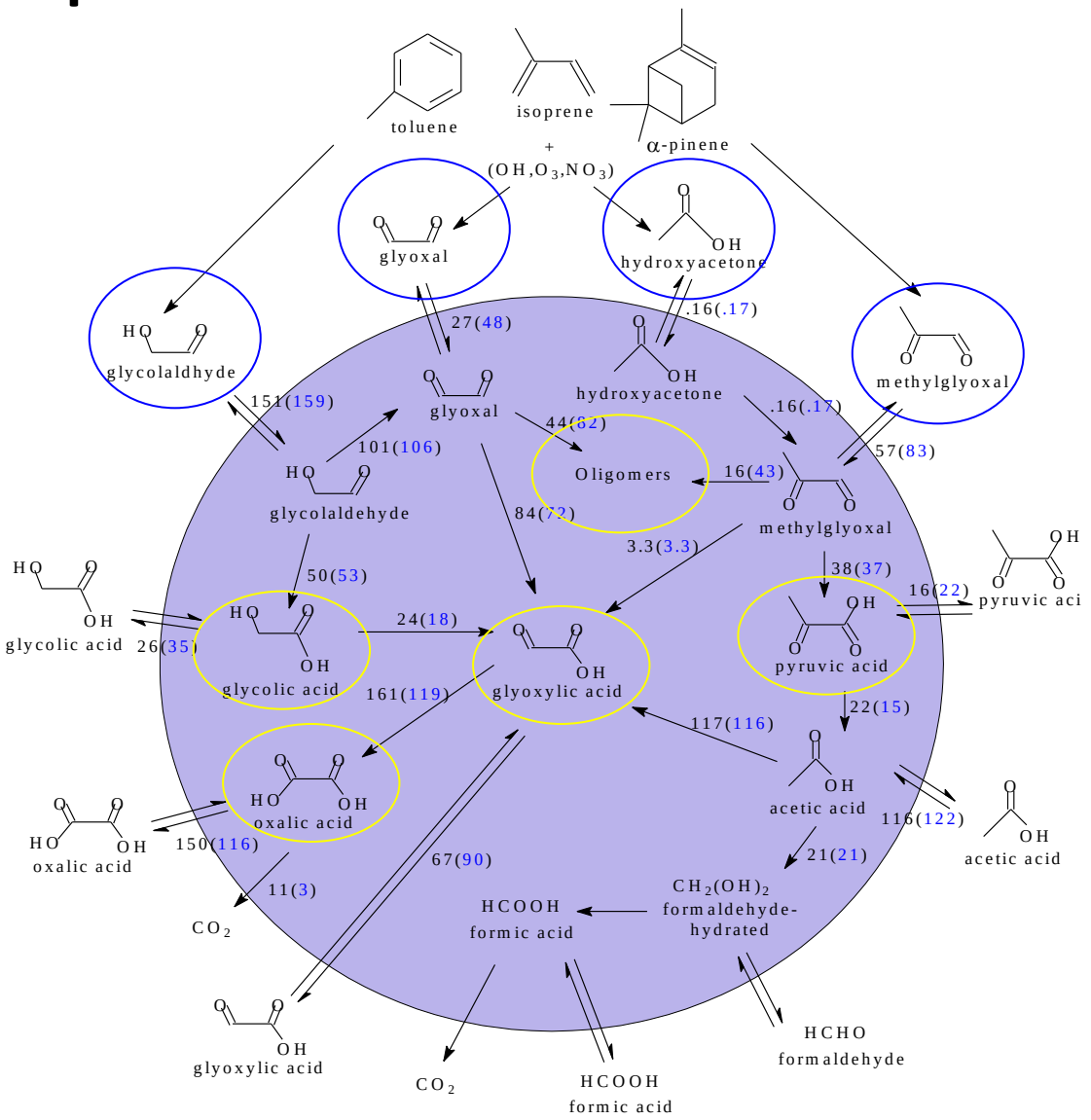
Liquid-phase production of SOA



[*Zhang et al., 2013*]



[*Jimenez et al., 2009*]

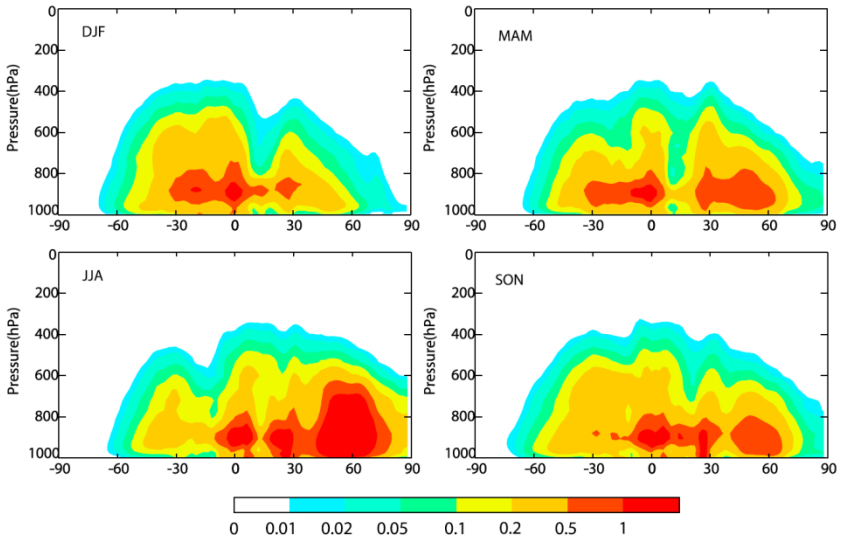


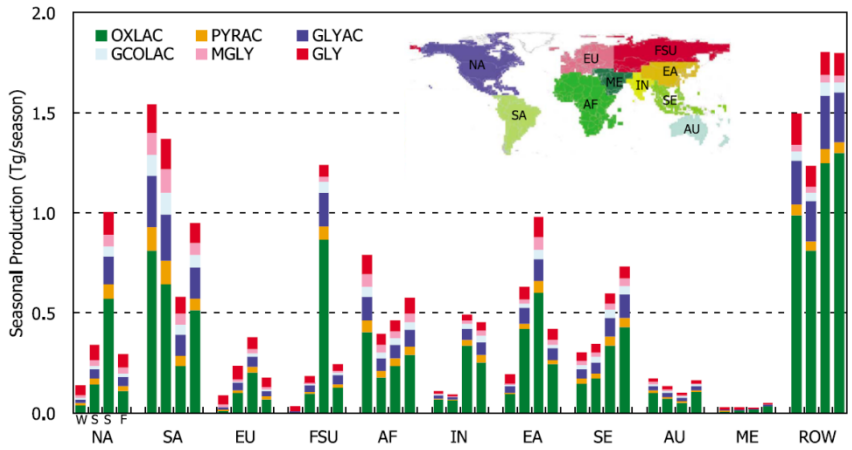
Global total in-cloud production of SOA is **21-30** Tg/yr, comparable to total gas-phase SOA production based on two-product models.

[*Liu et al., 2012, JGR*]

Parameterization of SOA_{cld} prod. in AM3

Distribution of SOA_{cld} production



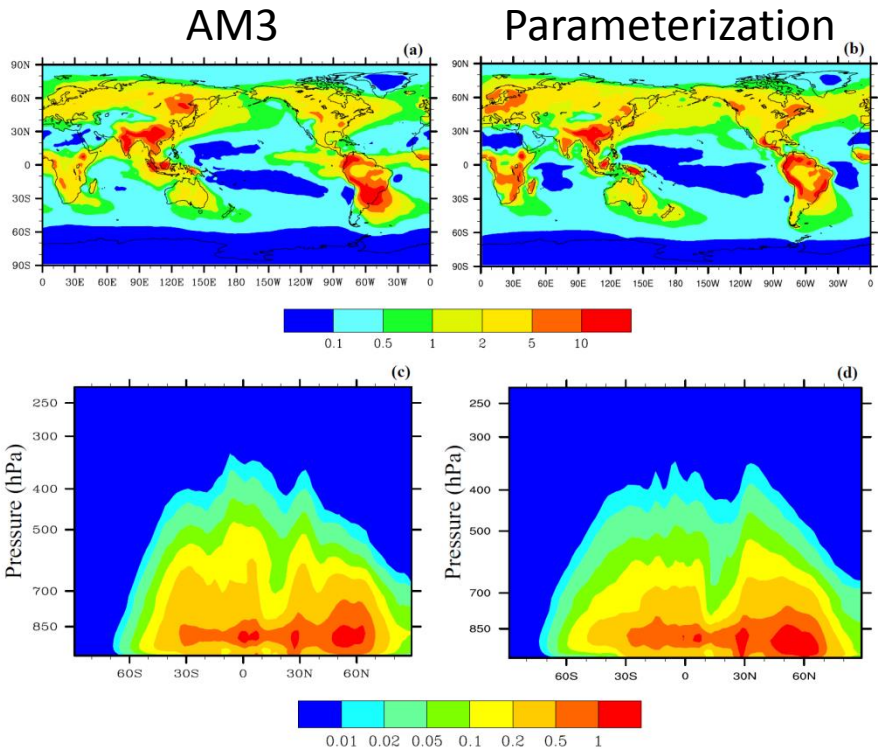


[*Liu et al., 2012, JGR*]

Parameterization of SOA_{cld} production based on the detailed multiphase chemistry in AM3:

$$P_{SOAcld} = \alpha \times LWC \times TC_{loss}^{\gamma} + \beta$$

Where γ=0.4, independent on season.



[*He et al., 2013, ACP*]

Parameterizations of SOA production in gaseous and liquid phases

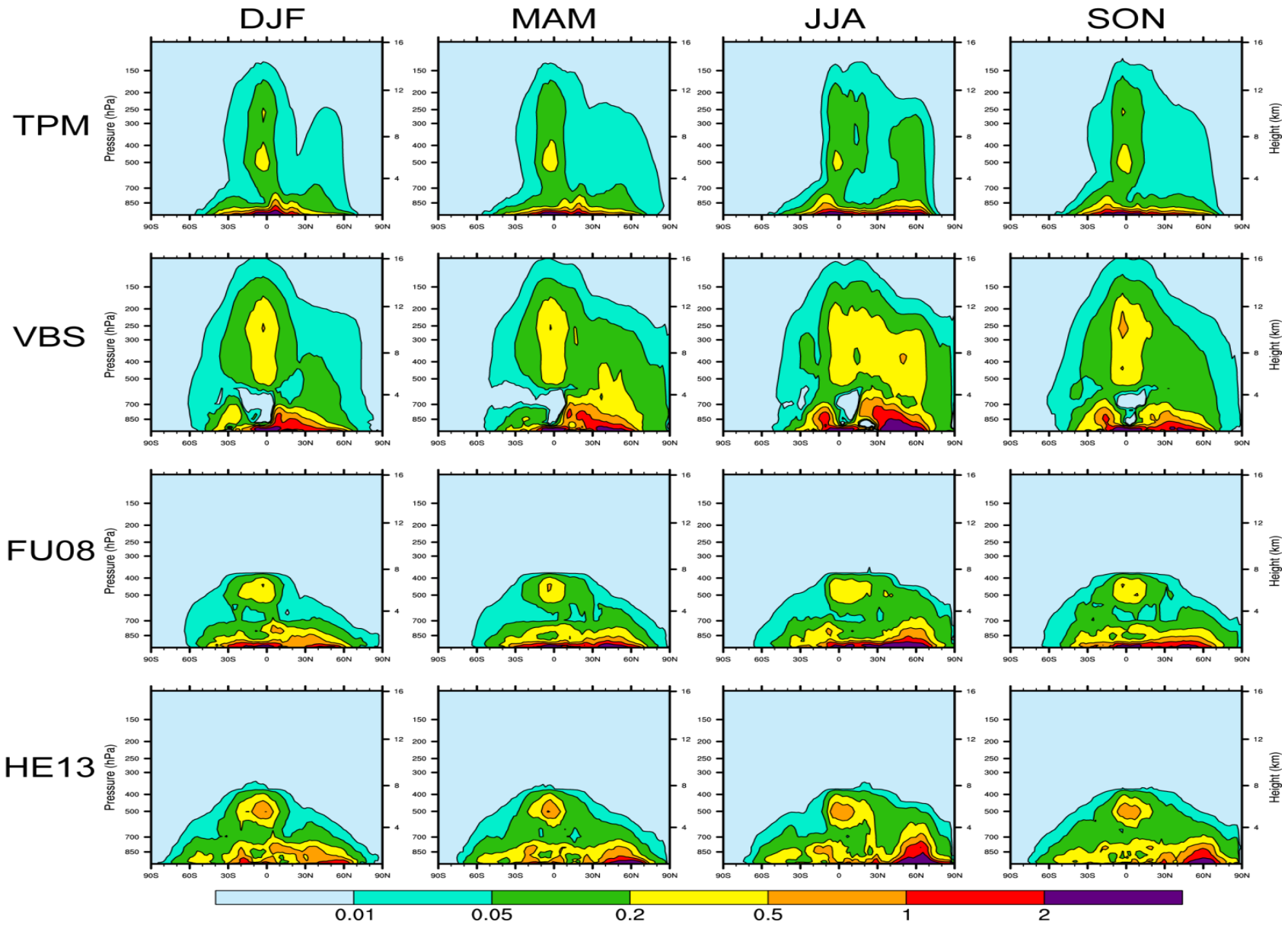
Gas-phase SOA parameterizations: Two-Product Model (**TPM**)
Volatility Basis Set (**VBS**)

Liquid-phase SOA parameterizations: Uptake model (**FU08**)
Global model parameterization (**HE13**)

SOA budget production(Tg/y)	**TPM**	**VBS**	**FU08**	**HE13**
Alkanes and aromatics*	2.40	13.42	1.69	4.90
Isoprene	11.00	8.19	13.82	15.78
Terprene	9.15	19.58	9.61	7.53
Total	**22.55**	**41.19**	**25.12**	**25.60**

[*Chen et al., in preparation*]

Zonal mean distribution of SOA production



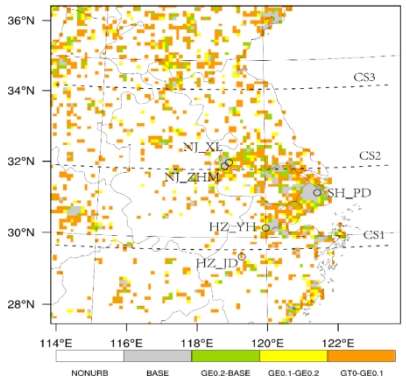
[*Chen et al., in preparation*]

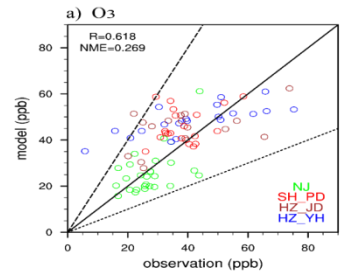
Conclusions

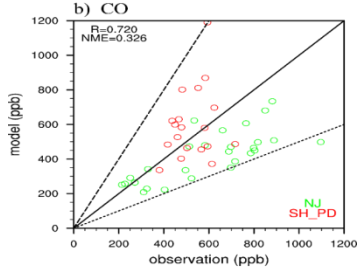
- Aerosol-cloud interaction is the key process controlling global aerosol distribution.
- Global trade system relocates substantial BC emissions between continents.
- Large discrepancies exist in estimating the global SOA sources.
- More HIPPO-like, species-oriented BC and OA measurements (e.g., near cloud, or over remote regions) are needed.

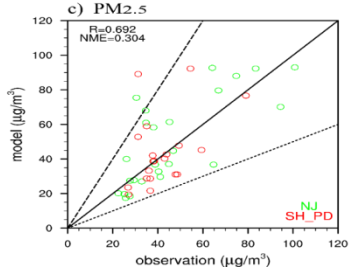
Thank you !

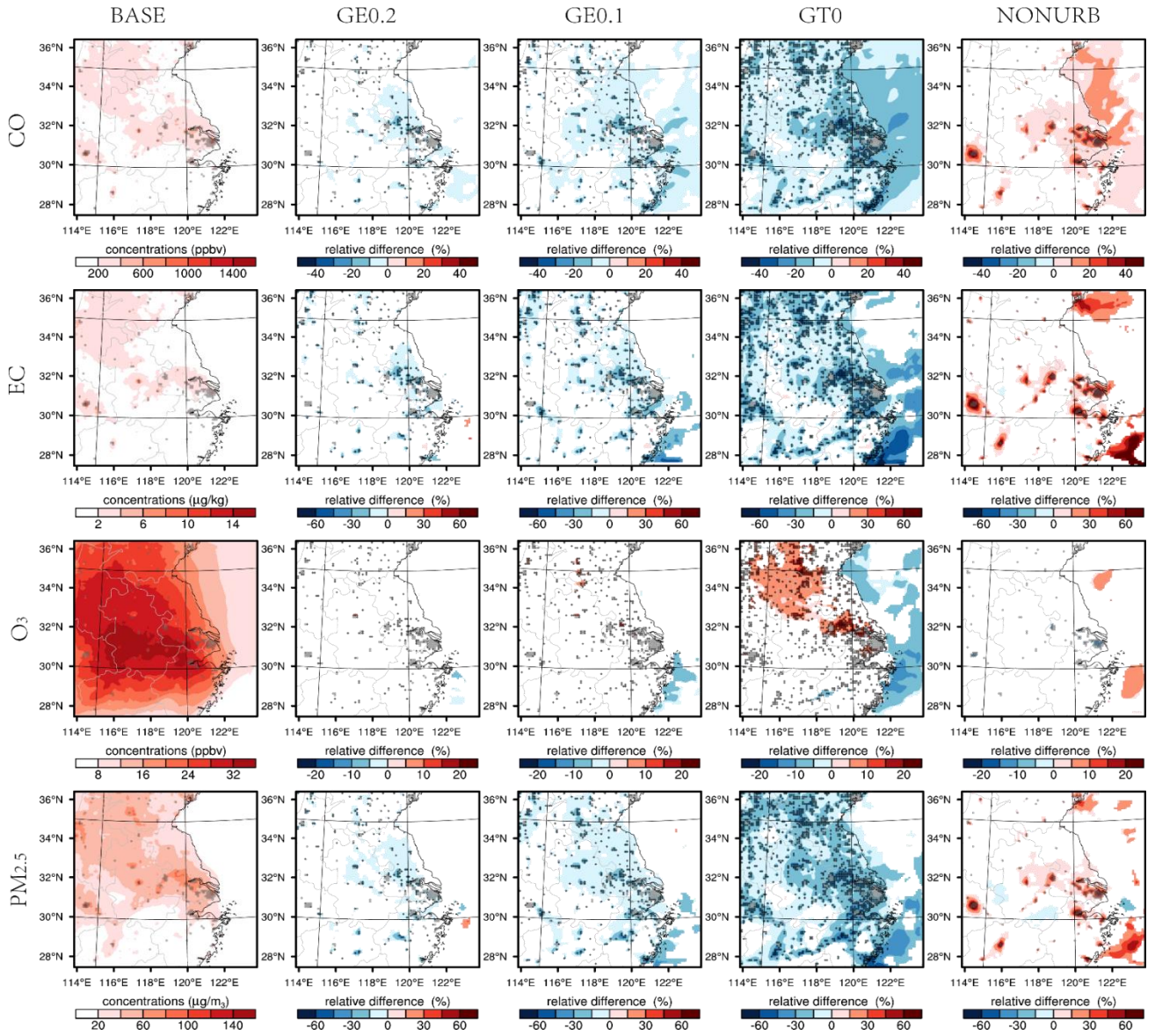
Effects of urbanization on air quality





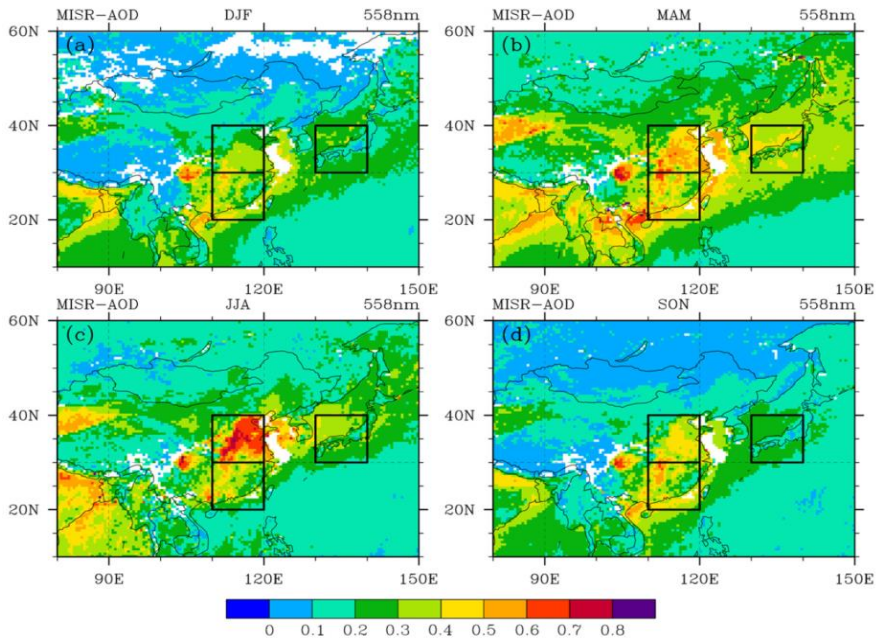


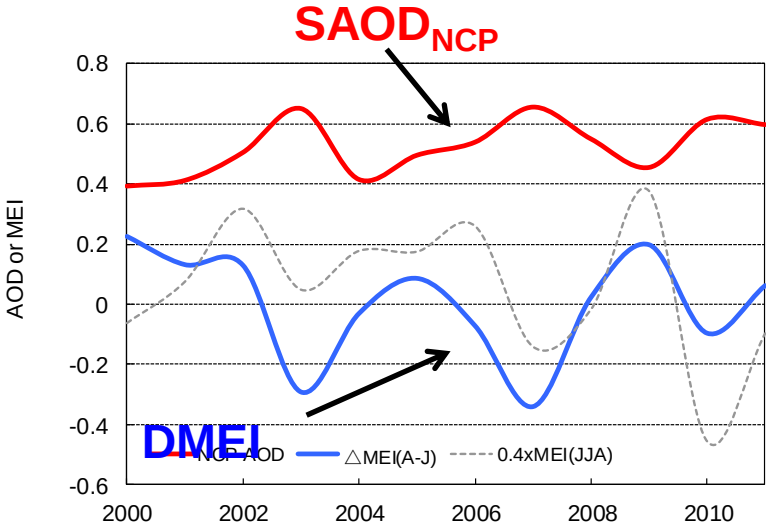


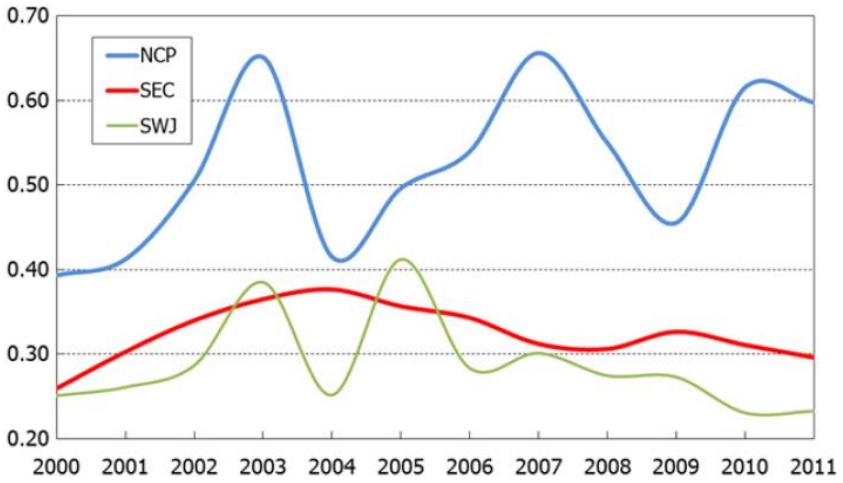


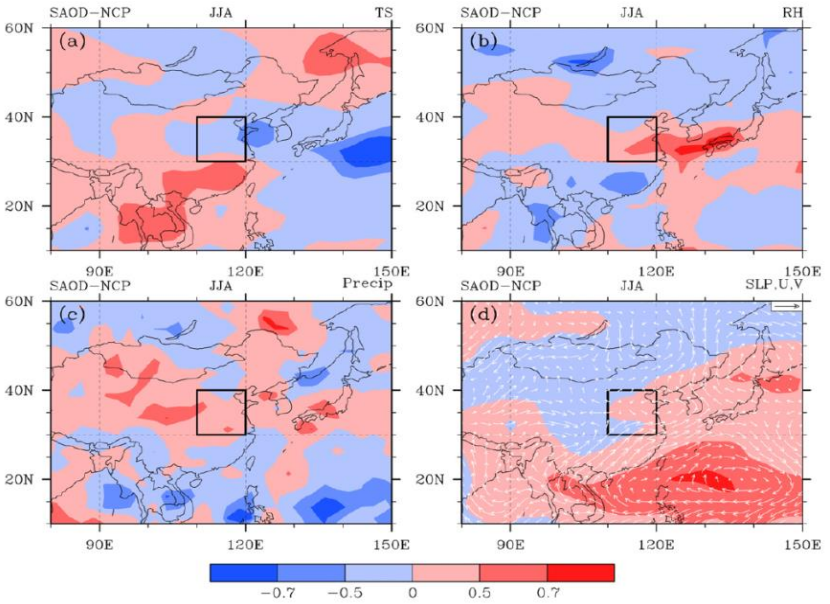
[*Tau et al., in preparation*]

Effects of ENSO on regional air quality









[*Liu et al., 2013, ERL*]