Development of a multi-scale Eulerian aerosol chemistry and transport model and its application to transboundary air pollution issues in Asian outflow

Development of a new 3D-CTM to simulate aerosol properties, namely,

1. chemical compositions,
2. size distributions,
3. mixing states,
4. shapes,

that alter optical, CCN/IN properties & deposition processes, thus environmental issues.

The model simulated transport of contaminated air-mass in Asian outflow regions.

Mizuo KAJINO (Dr. Sci.)
RCAST, Univ. of Tokyo

EMTACS

<An Eulerian, Multi-scale, Tropospheric, Aerosol Chemistry and transport Simulator>

	Category	1	2	3	4	5	6	7	8	9	10	11	12	X
1	Aitken	M_0	M_2	M_3	Mass	-	-	-	-	SO_4^{2-}	NO_3^-	NH_4^+	H_2O	SOAs…
2	Soot	M_0	M_2	M_3	Mass	BC	OC	-	-	SO_4^{2-}	NO_3^-	NH_4^+	H_2O	SOAs…
3	Multi/unid. accum. mode	M_0	M_2	M_3	Mass	BC	OC	-	-	SO_4^{2-}	NO_3^-	NH_4^+	H_2O	SOAs…
4	Dust	M_0	M_2	M_3	Mass	-	-	Dust	-	SO_4^{2-}	NO_3^-	NH_4^+	H_2O	SOAs…
5	Sea salt	M_0	M_2	M_3	Mass	-	-	-	SS	SO_4^{2-}	NO_3^-	NH_4^+	H_2O	SOAs…
6	Multi/unid. coarse mode	M_0	M_2	M_3	Mass	BC	OC	Dust	SS	SO_4^{2-}	NO_3^-	NH_4^+	H_2O	SOAs…
7	Fog/cloud	M_0	M_2	M_3	Mass	BC	OC	Dust	SS	SO_4^{2-}	NO_3^-	NH_4^+	H_2O	SOAs…
Y	Other hydrometeors	M_0	M_2	M_3	Mass	BC	OC	Dust	SS	SO_4^{2-}	NO_3^-	NH_4^+	H_2O	SOAs…

52 prognostic variables (transport)

Diagnostic variables (equilibrium, met. model)

Future implementation

A modal/moment approach is used as it is computationally efficient

MAD-MS

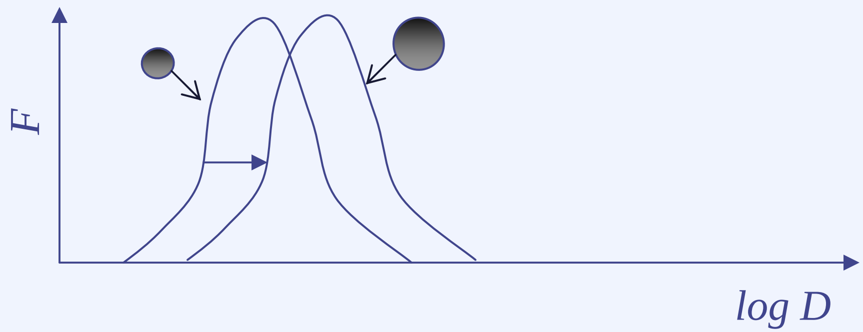
<Modal Aerosol Dynamics model for multiple Modes and arbitral Shapes>

MAD model (Whitby and McMurry, *AST*, 1997) is widely used for global/regional models

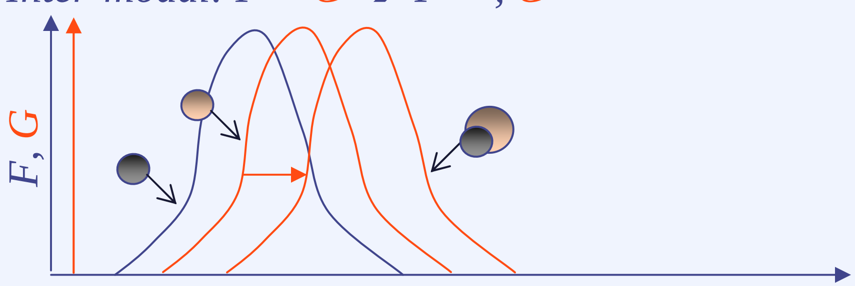
Spherical particles are assumed, only applicable for intra-modal coagulation, and inter-modal coagulation but difference in the size parameters (D_g, σ) should not be large.

<MAD approach>

Intra-modal: $F^t(N^t, D_g^t, \sigma^t) \rightarrow F^{t+\Delta t}$

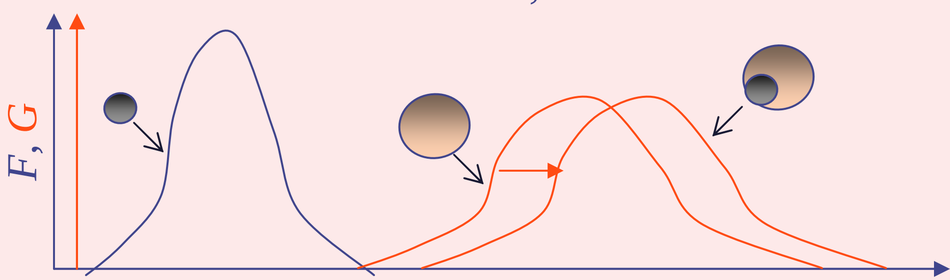


Inter-modal: $F^t \times G^t \rightarrow F^{t+\Delta t}, G^{t+\Delta t}$

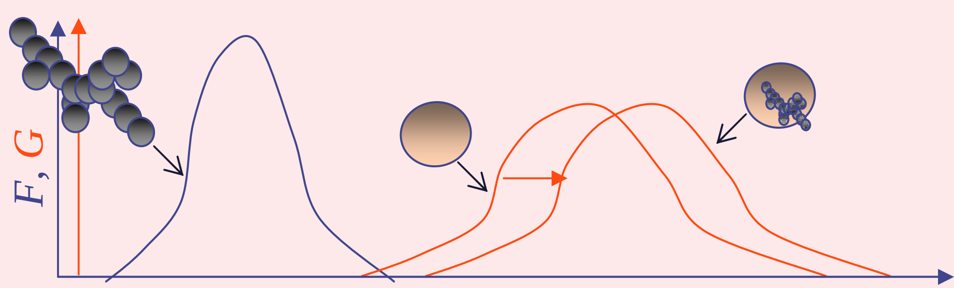


<MAD-MS approach>

Inter-modal: $F^t \times G^t \rightarrow F^{t+\Delta t}, G^{t+\Delta t}$



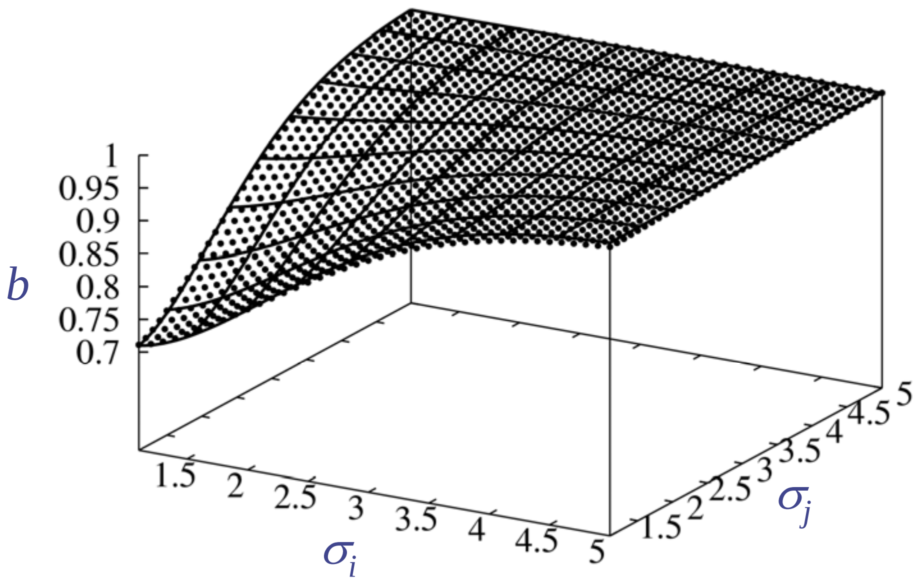
Fractal agglomerates: $N_0 = k_f (D_{chr} / d_p)^{D_f}$



MAD-MS

<Modal Aerosol Dynamics model for multiple Modes and arbitral Shapes>

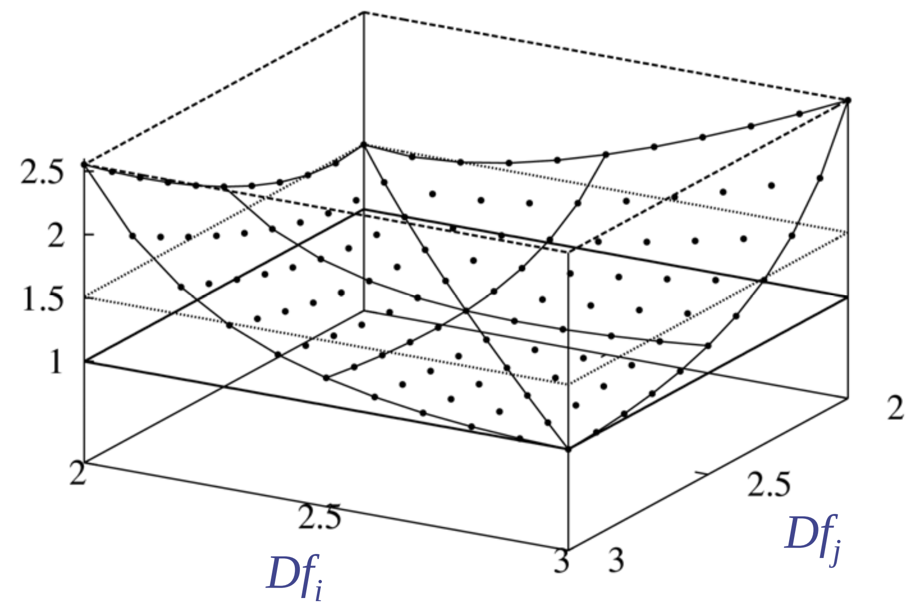
Approx. function *b*, MAD used in FM regime



$$b = 1 + 1.2\gamma \exp\left[-2\frac{\sigma_i + \alpha\sigma_j}{1+\alpha}\right] - 0.646\gamma \exp\left[-0.35\frac{\sigma_i^2 + \alpha\sigma_j^2}{1+\alpha}\right]$$

$$\gamma = \left[1 - \frac{\sqrt{1+\alpha^3}}{1+\sqrt{\alpha^3}}\right] \Big/ \left[1 - \frac{1}{\sqrt{2}}\right], \qquad \alpha = \frac{D_{gj}}{D_{gi}}$$

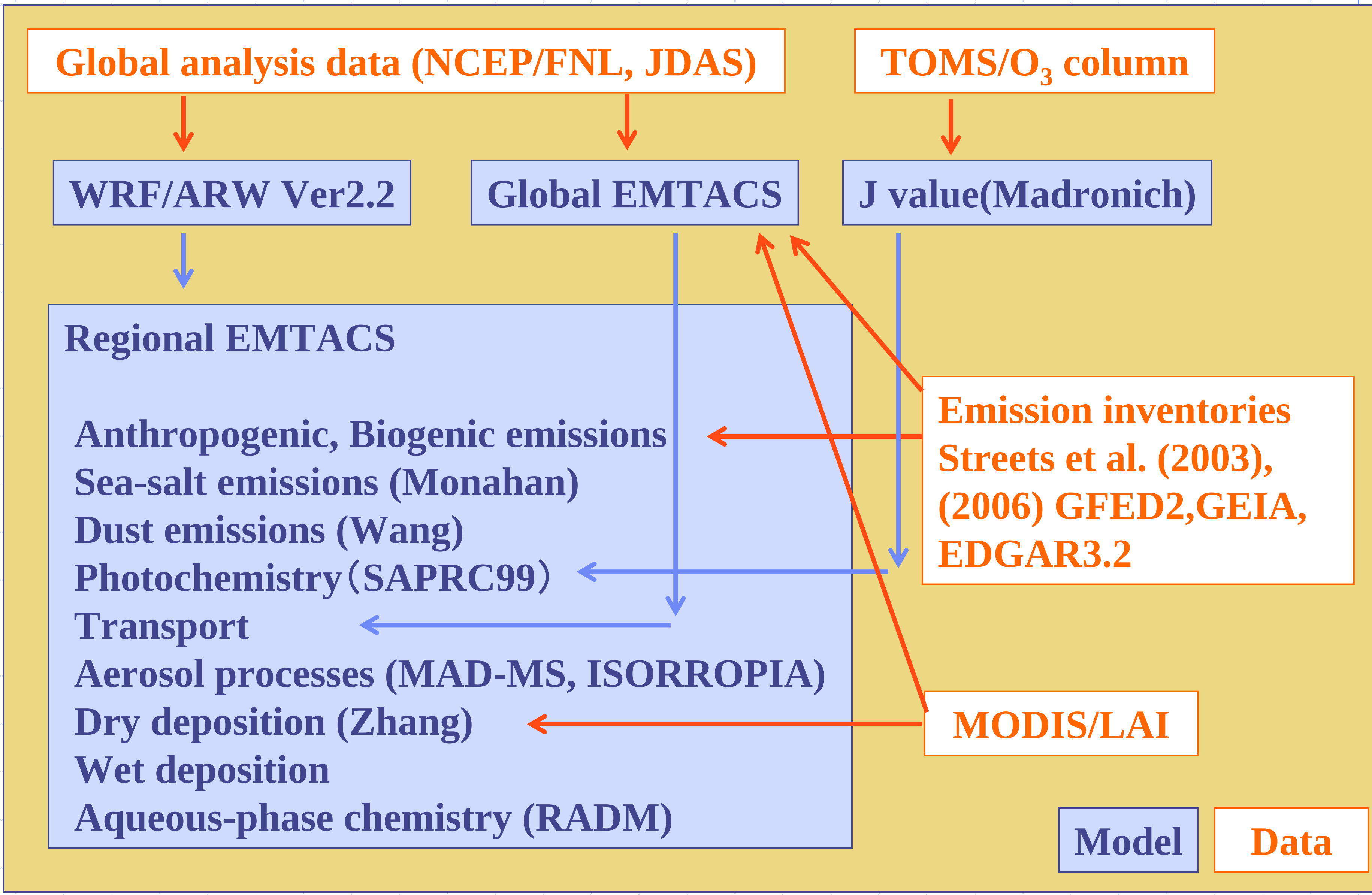
Ratio of collision frequencies btw. aggregates to btw. spheres in CO regime



Soot agglomerates (D_f=2) and other aerosols are spheres (D_f=3) $N_0 = k_f (D_{chr} / d_p)^{D_f}$

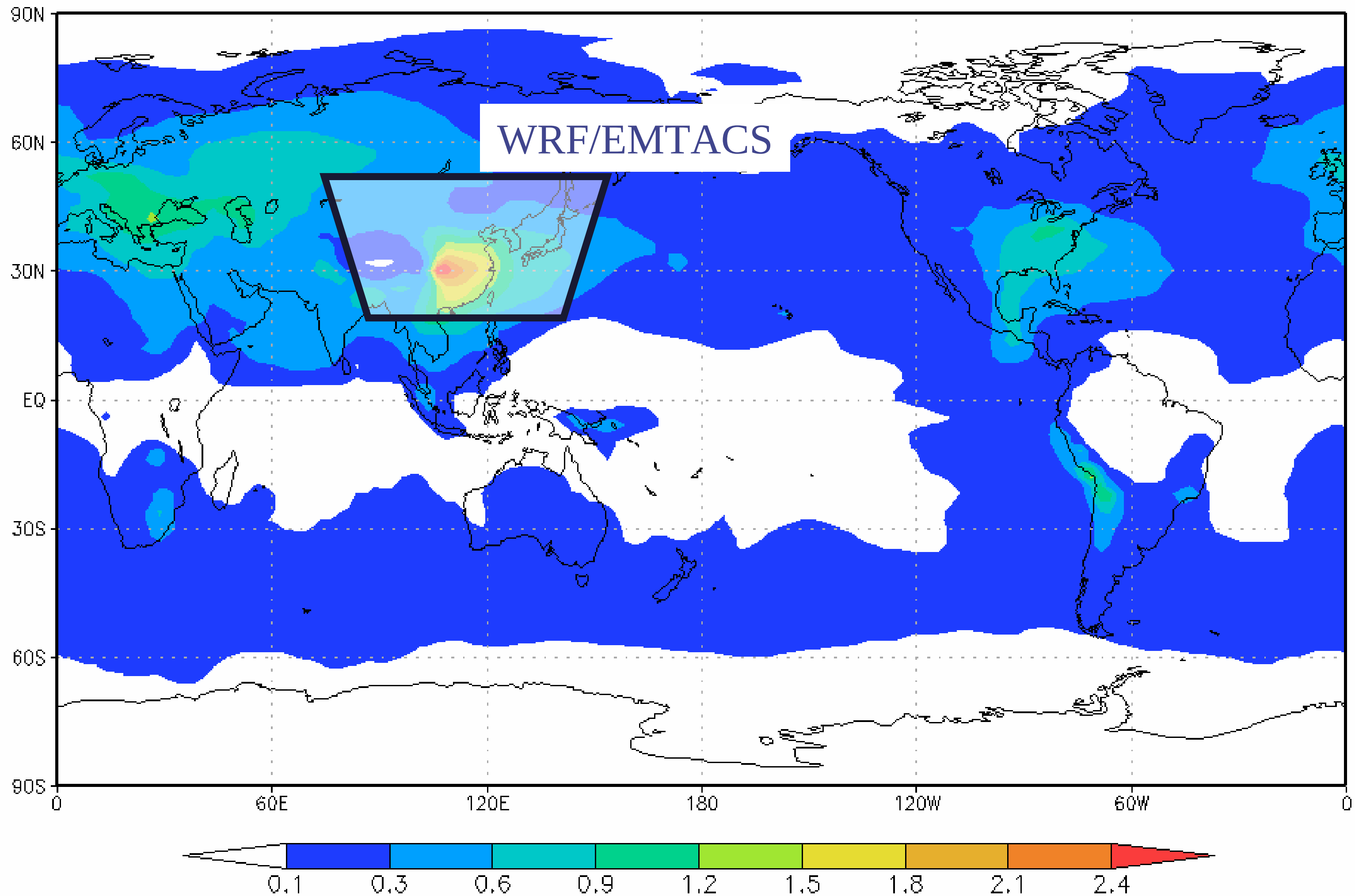
Only Brownian coagulation ➔ Turbulence & sedimentation should be implemented in future

Flow chart of calculation

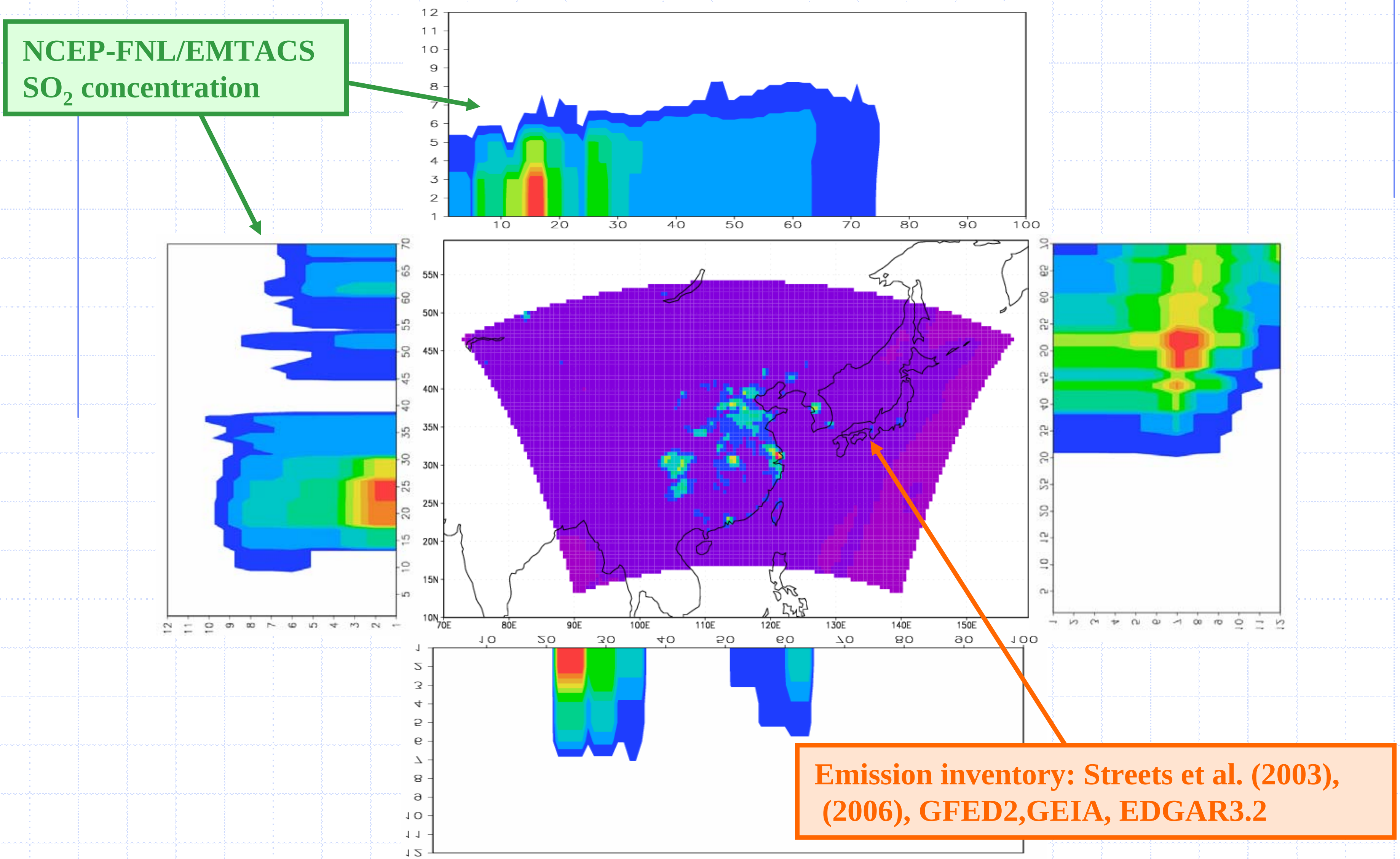


EMTACS driven by global analysis data, NCEP/fnl

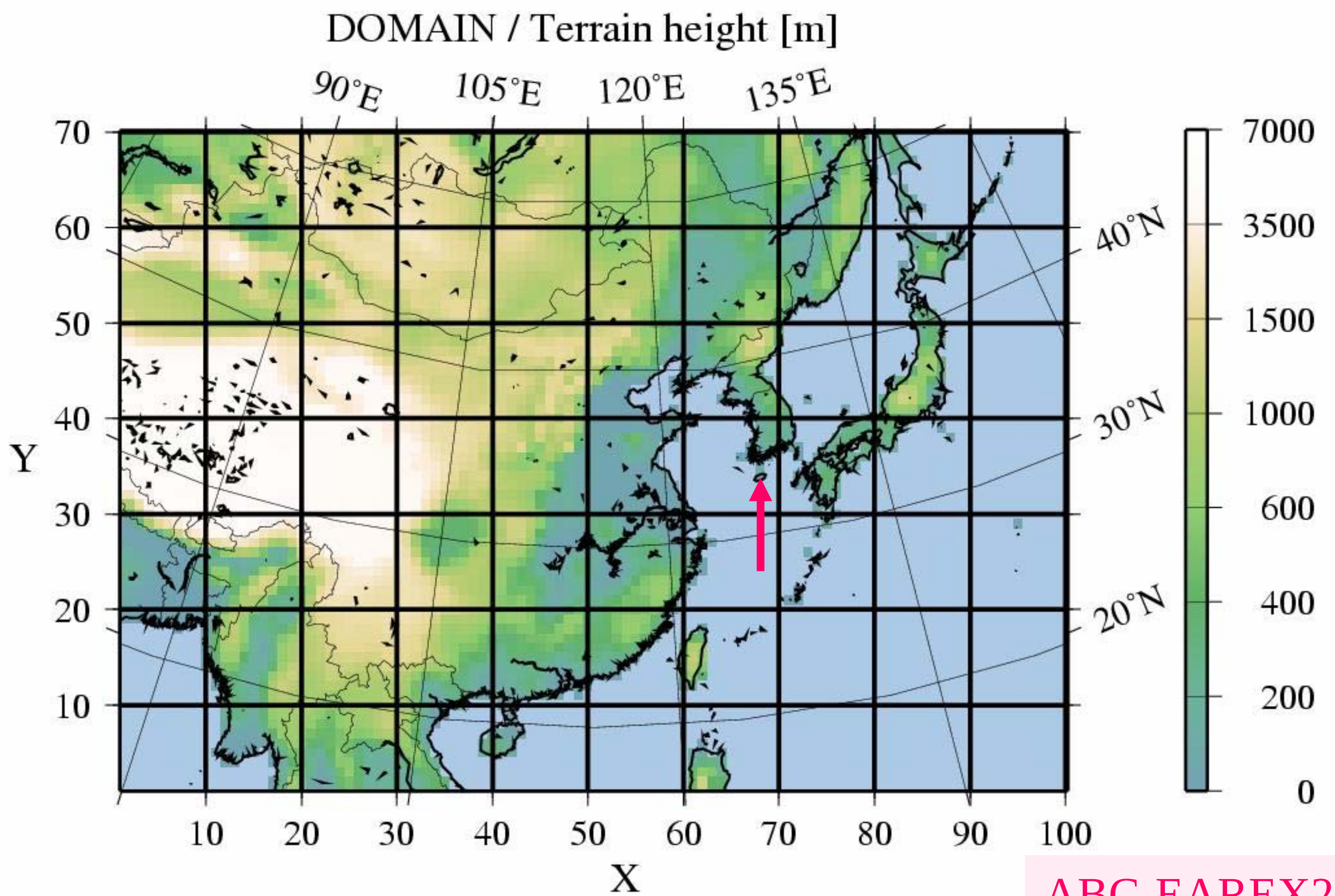
Monthly mean sulfate concentration in PBL in Mar 2005 [ppb]



NCEP-FNL/EMTACS used as initial & boundary conc. for regional calculations



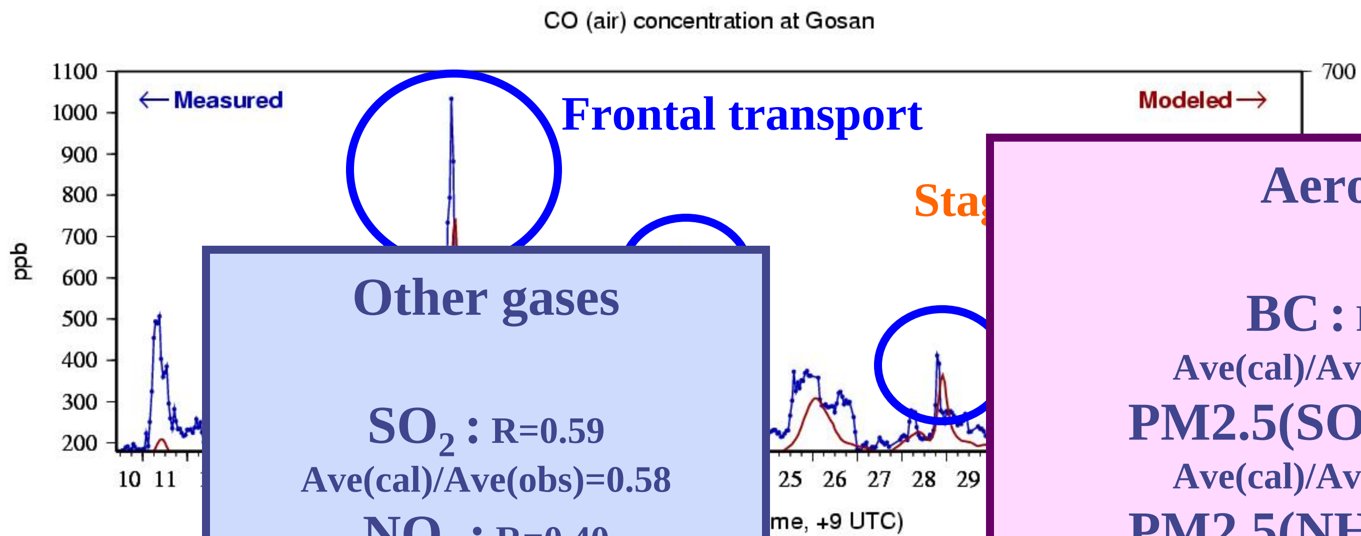
Offline coupled Met&Chem, WRF/EMTACS



ABC-EAREX2005
Jeju island, Korea

Horizontal resolution: 60km
Vertical resolution: 27 levels up to 12km
Temporal resolution: 1 hourly

ABC-EAREX 2005 Gosan
<Asian Brown Cloud / East Asian Regional EXperiment>

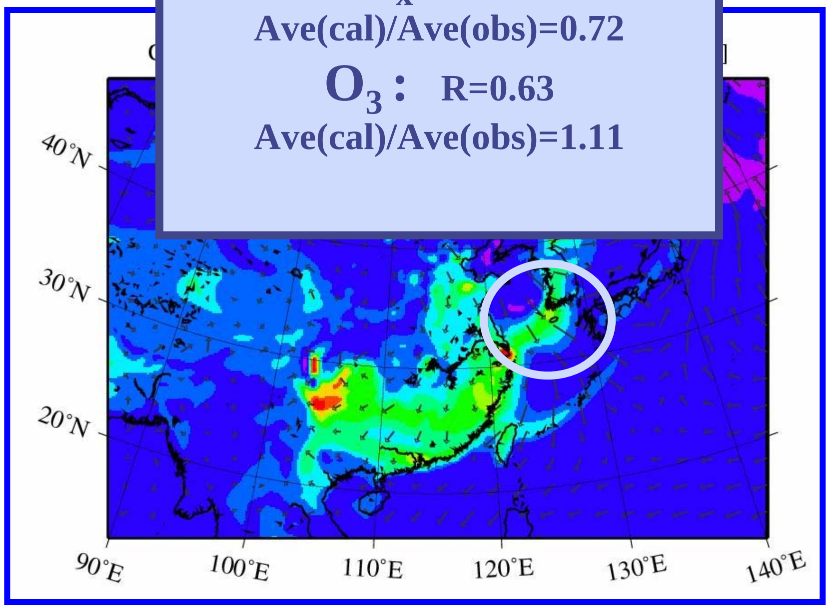


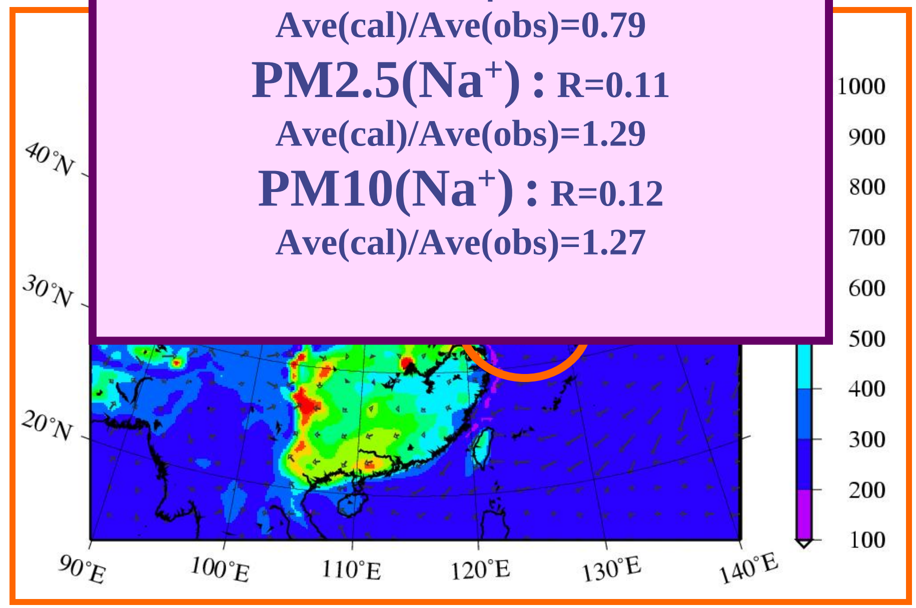
Other gases

SO_2 : R=0.59
Ave(cal)/Ave(obs)=0.58
NO_x : R=0.40
Ave(cal)/Ave(obs)=0.72
O_3 : R=0.63
Ave(cal)/Ave(obs)=1.11

Aerosols

BC : R=0.76
Ave(cal)/Ave(obs)=0.46
PM2.5(SO_4^{2-}) : R=0.77
Ave(cal)/Ave(obs)=0.71
PM2.5(NH_4^+) : R=0.79
Ave(cal)/Ave(obs)=0.79
PM2.5(Na^+) : R=0.11
Ave(cal)/Ave(obs)=1.29
PM10(Na^+) : R=0.12
Ave(cal)/Ave(obs)=1.27

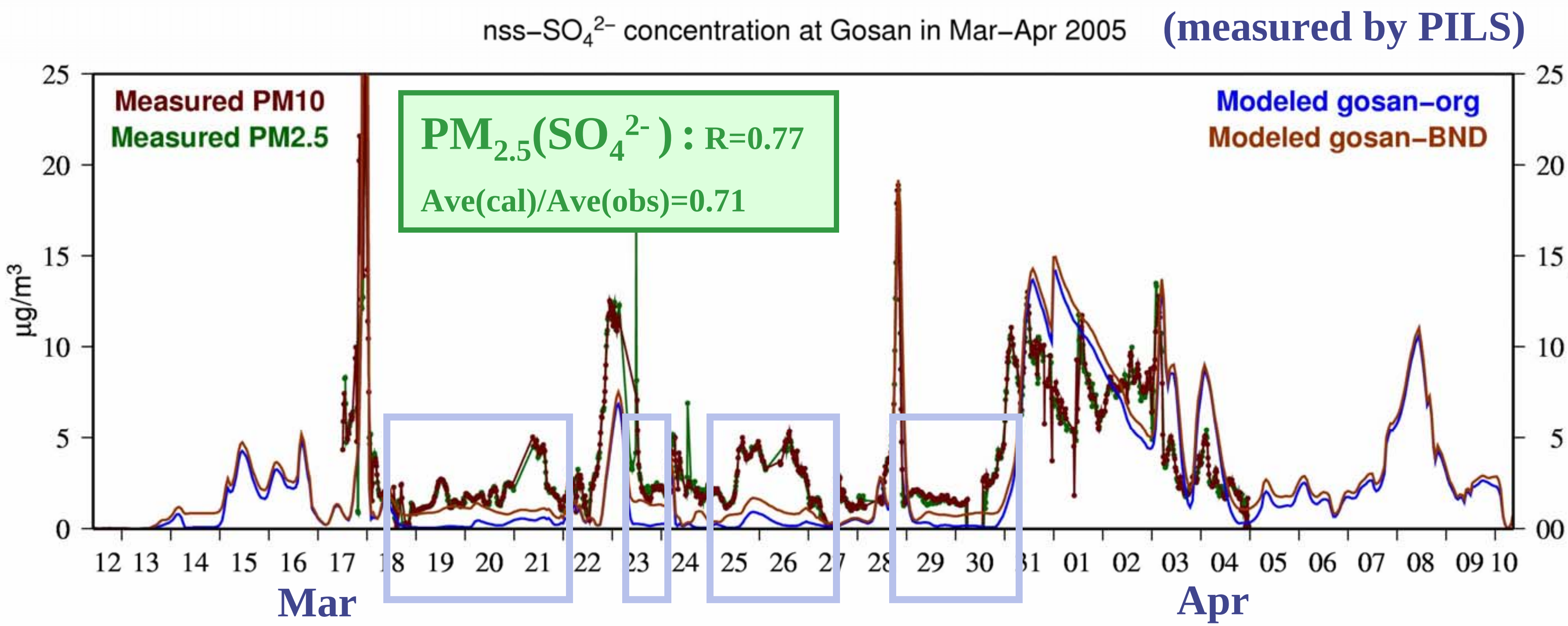




Topics

1. Contribution of boundary SOx on surface sulfate concentration at Gosan.
2. Sensitivity of surface O_3 concentration at Gosan to dry deposition parameterization.
3. Sensitivity of evolution of size distribution to particle morphology in Asian outflow.

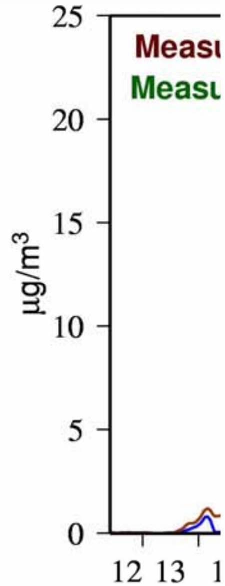
Topic 1. SO_x from outside of North East Asia



95% of sulfate during the event over 10 μg/m³ is explained by NEA emission.

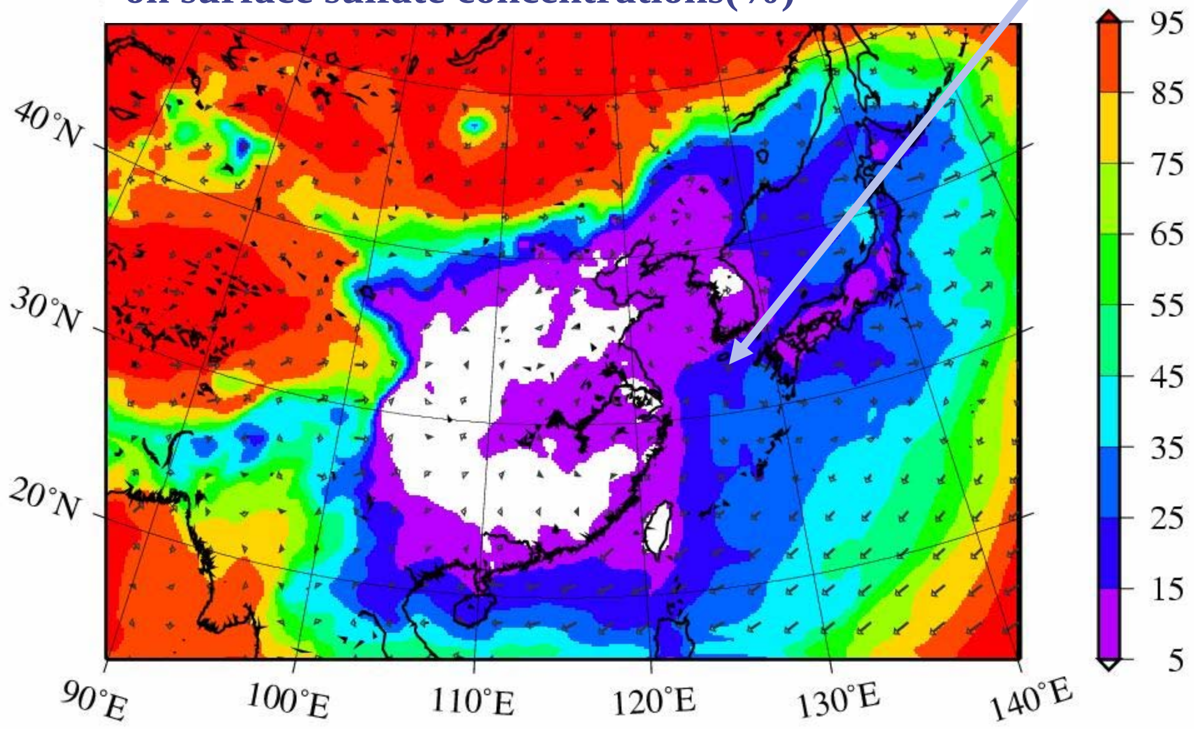
1~2 μg/m³ is explained by outside of NEA during low conc. period

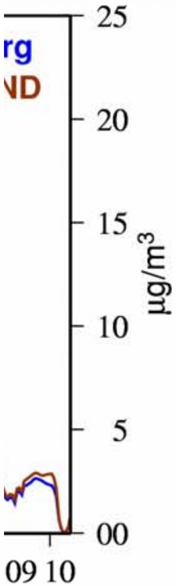
Topic 1. SO_x from outside of North East Asia



Monthly mean contribution of boundary SOx on surface sulfate concentrations(%)

16.2%





The contribution of sulfate originated from the boundary SOx on the surface sulfate conc. at Gosan is 16.2% (Mar. 10 to Apr. 10, 2005).

Topic 2. Sensitivity of surface O_3 concentration to dry deposition parameterization in Asian outflow region

1. Wesely's parameterization

		Seasonal category				
		1	2	3	4	5
Landuse type	1	r_{1-1}	r_{1-2}	r_{1-3}	r_{1-4}	r_{1-5}
	2	r_{2-1}	r_{2-2}	r_{2-3}	r_{2-4}	r_{2-5}
	3	r_{3-1}	r_{3-2}	r_{3-3}	r_{3-4}	r_{3-5}
	4	…	…	…	…	…
	5	…	…	…	…	…
	6	…	…	…	…	…
	7	…	…	…	…	…
	8	…	…	…	…	…
	9	…	…	…	…	…
	10	…	…	…	…	…
	11	r_{11-1}	r_{11-2}	r_{11-3}	r_{11-4}	r_{11-5}

Seasonal category
1: Midsummer
2: Autumn
3: Late autumn
4: Winter
5: Spring

← Difficult to apply to Asia due to its various seasonality (wide range for latitude)

2. Zhang's parameterization

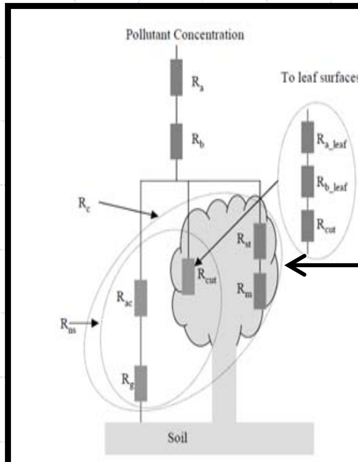
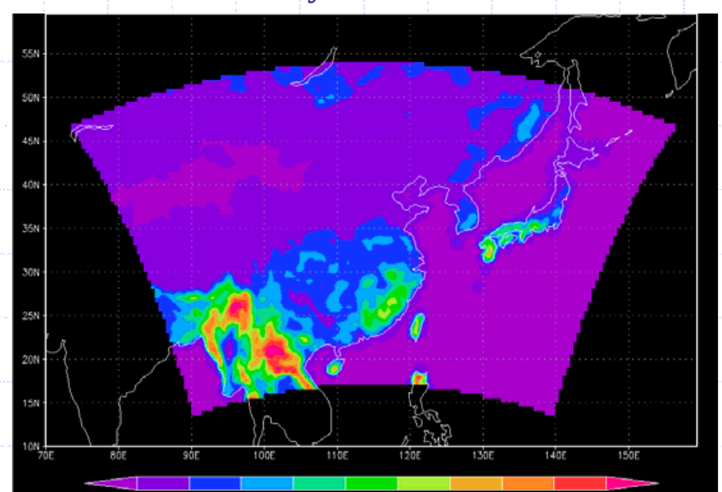


Fig. 1. Scheme of resistance analogy.

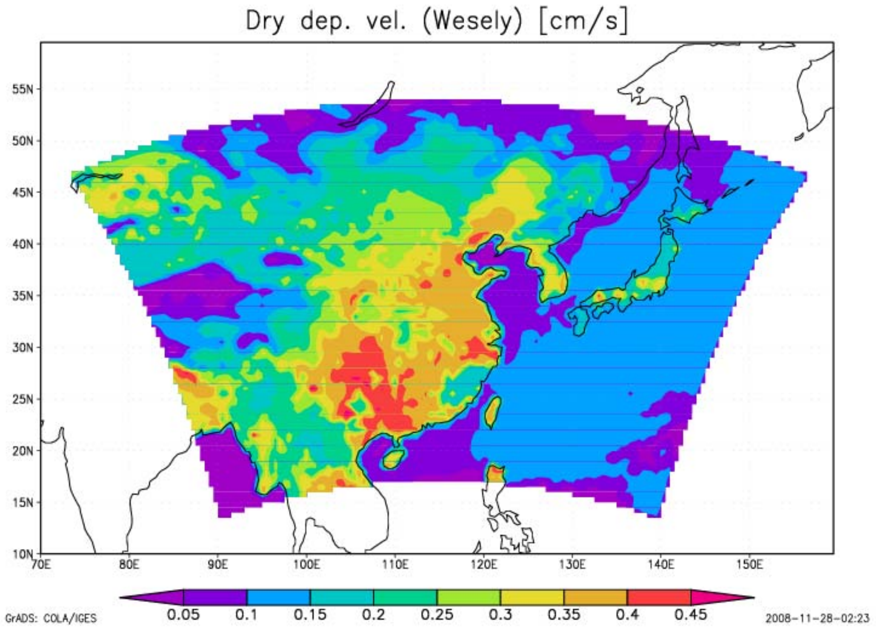
Some of surface resistances are function of LAI (Leaf Area Index)

Monthly MODIS/LAI

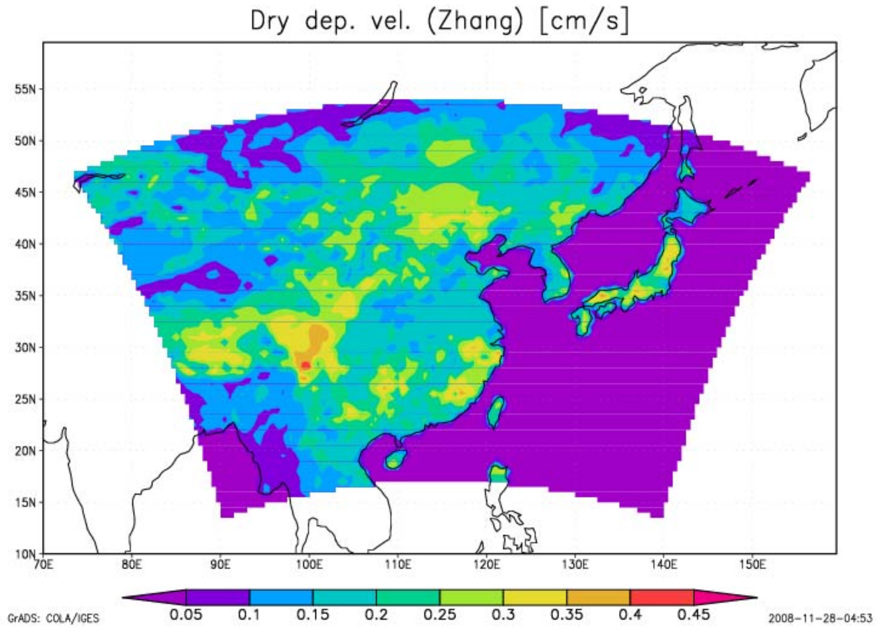


Topic 2. Sensitivity of surface O_3 concentration to dry deposition parameterization in Asian outflow region

Monthly mean dry deposition velocity for O_3 [cm/s]



1. Wesely's parameterization

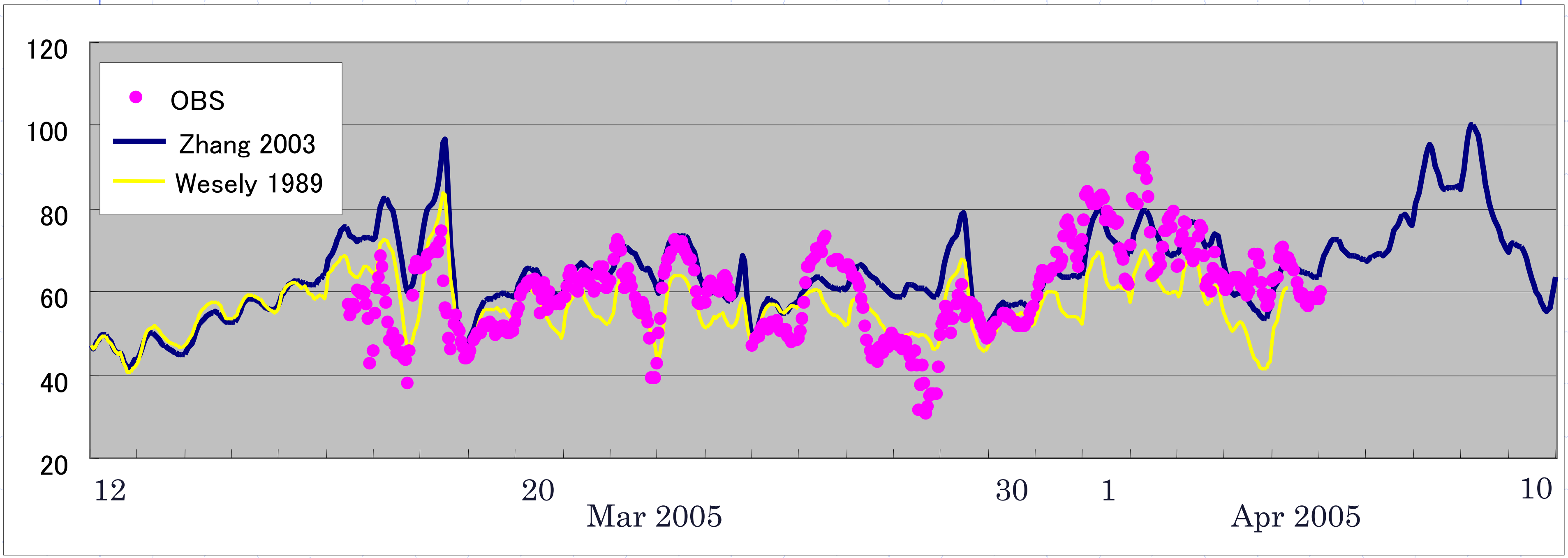


2. Zhang's parameterization

V_d for O_3 of Wesely is at most twice as large as Zhang's on land, larger on the ocean as well ➔

Topic 2. Sensitivity of surface O_3 concentration to dry deposition parameterization in Asian outflow region

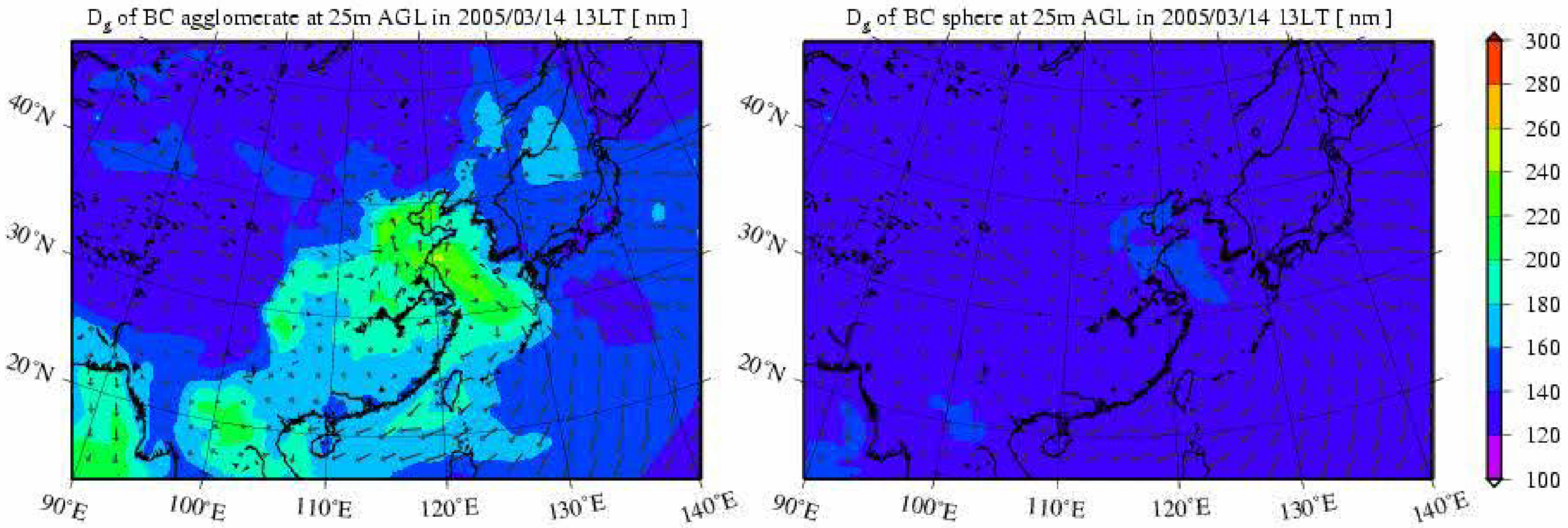
Surface O_3 concentration at Gosan (ppb)



Pink	Observation	57 ppb
Blue	Zhang et al (2003)×MODIS/LAI	64 ppb (+7), R=0.63
Yellow	Wesely with seasonal categories	57 ppb (+0), R=0.53

Topic 3. Significant change in mean diameter of uncoated soot particles, while the mass remains unchanged

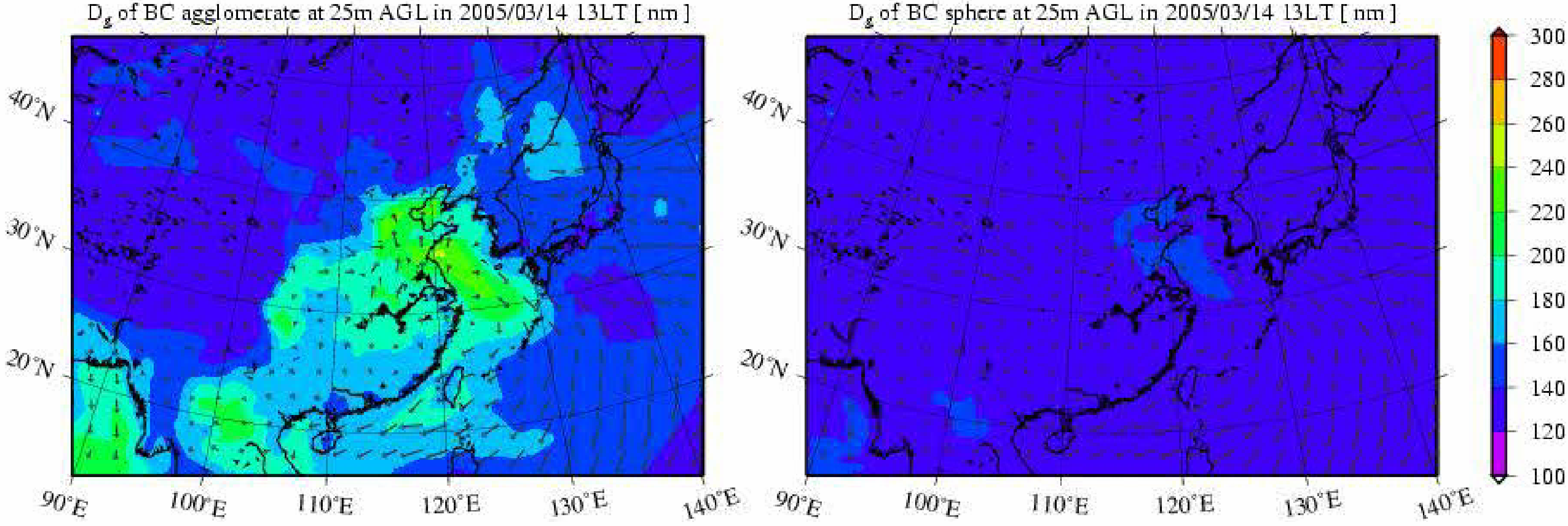
Volume-equivalent geometric mean diameters of soot [nm]



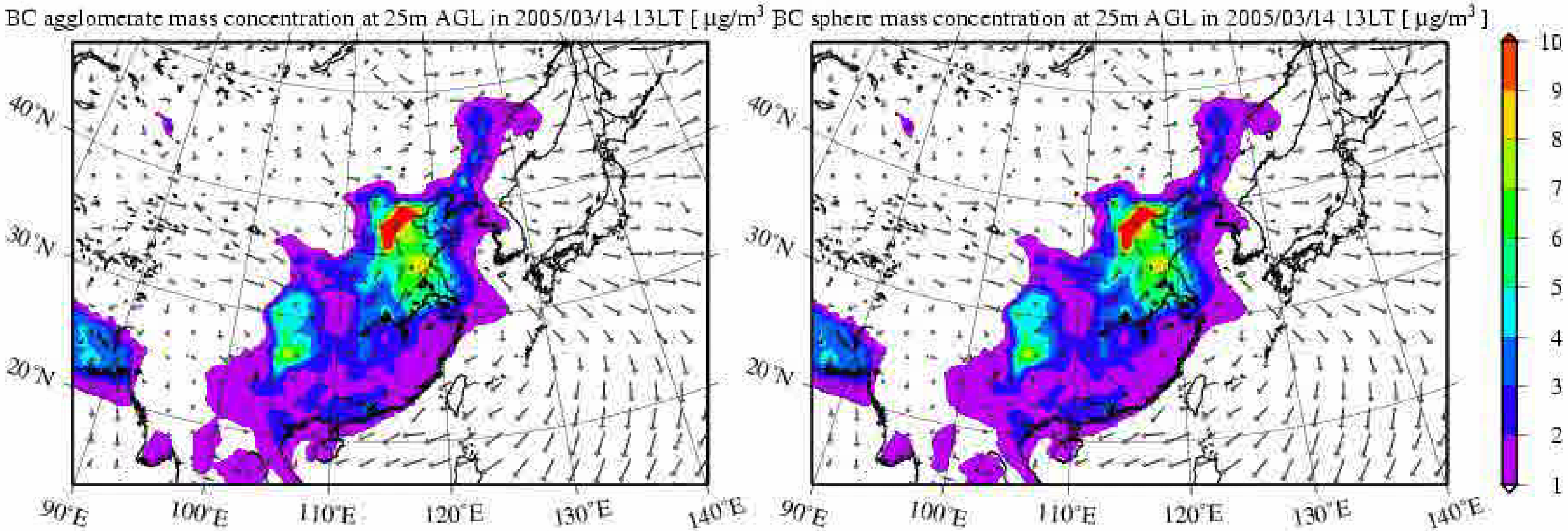
Soot agglomerates ($D_f = 2$)   Soot spheres ($D_f = 3$)

<u>**Initial size is 130nm**</u>

Volume-equivalent geometric mean diameters of soot [nm]



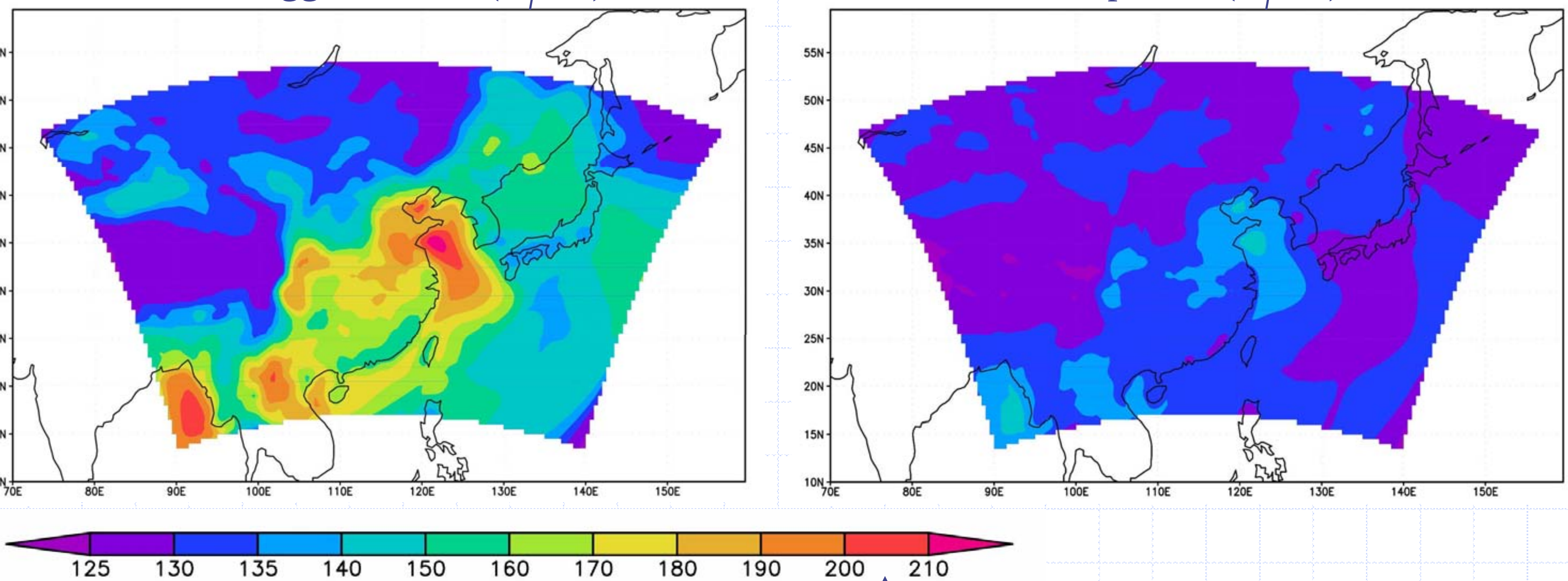
Surface mass concentration of soot [μg/m³]



Soot agglomerates ($D_f = 2$) Soot spheres ($D_f = 3$)

Monthly mean volume-equivalent mean diameter of uncoated soot (conc.>0.5ug/m³)

Soot agglomerates ($D_f = 2$) Soot spheres ($D_f = 3$)



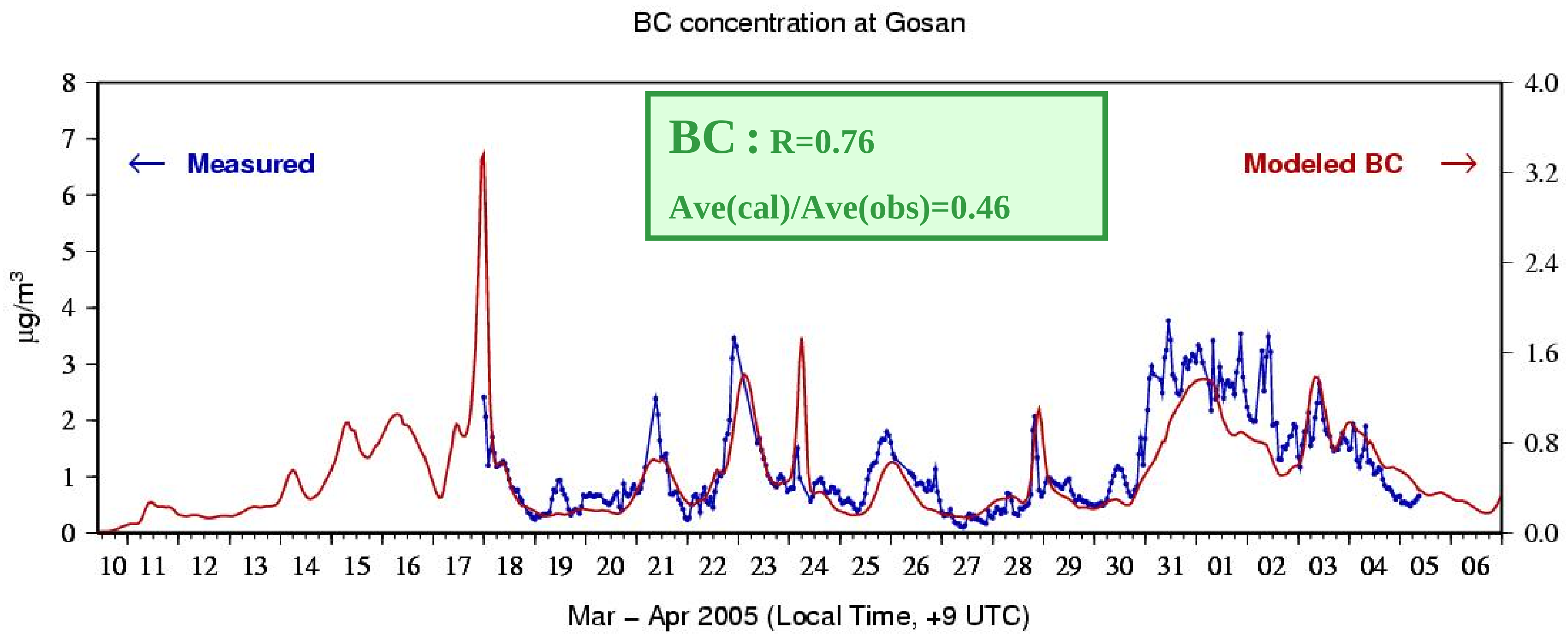
[nm]

Urban site: ~130nm
(Tokyo)[Kondo et al. 2006]

Num. Conc.: 10^8 ~10^9[particles/m³]
Mass Conc.: 10^0~10^1 [ug/m³]

Remote site: 200—220nm
(Fukue) [Shiraiwa et al. 2008]

Temporal variation of surface soot concentration at Gosan



Conclusions & future plans

0. CTM is now being developed to solve aerosol properties that affect environment, such as chemical composition, size distribution, mixing state and shape.

1. Boundary conditions affects even for surface <u>sulfate</u> by 15% in Asian outflow region in spring.

2. Surface <u>ozone</u> concentration can be altered by dry deposition parameterizations by 10~15 ppb.

3. Mean diameter can be changed significantly, considering mass-fractal shape of particles.

In future…

4. Implement secondary organic aerosols

5. Link to mixed-phase cloud microphysics & radiation processes