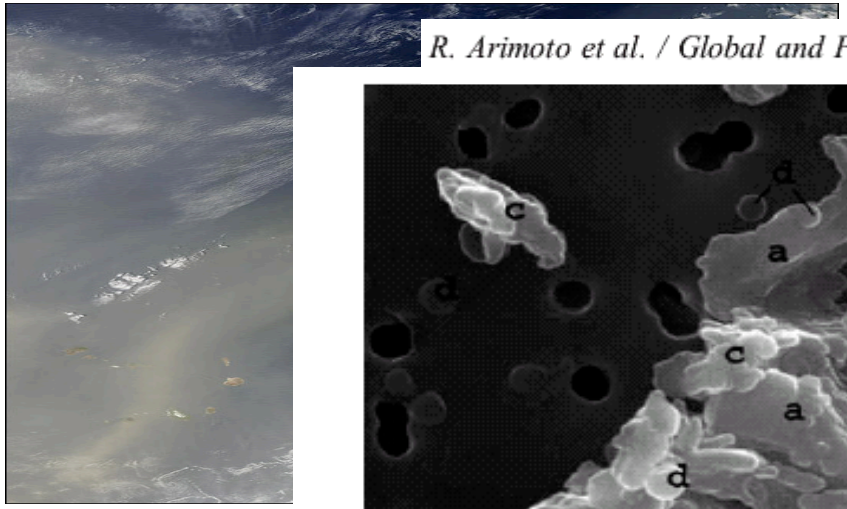
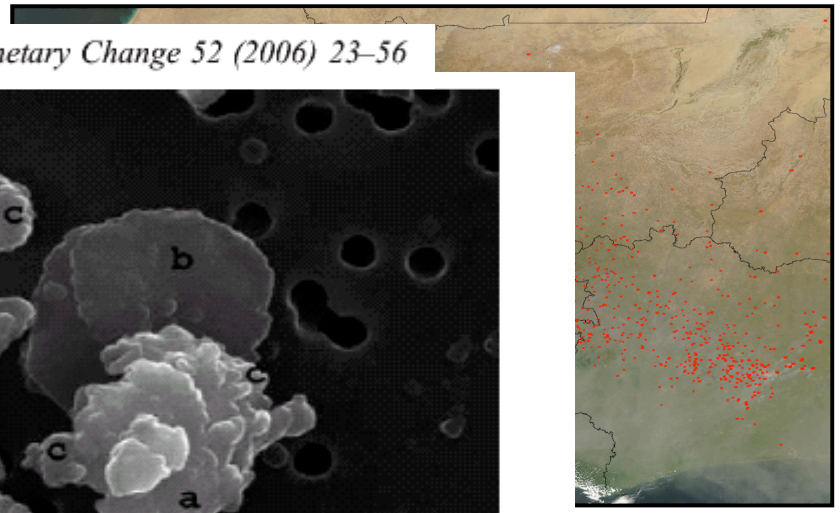
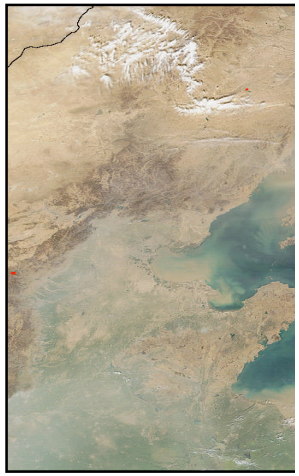
Coexistence of natural and anthropogenic particles at the regional scale







R. Arimoto et al. / Global and Planetary Change 52 (2006) 23–56

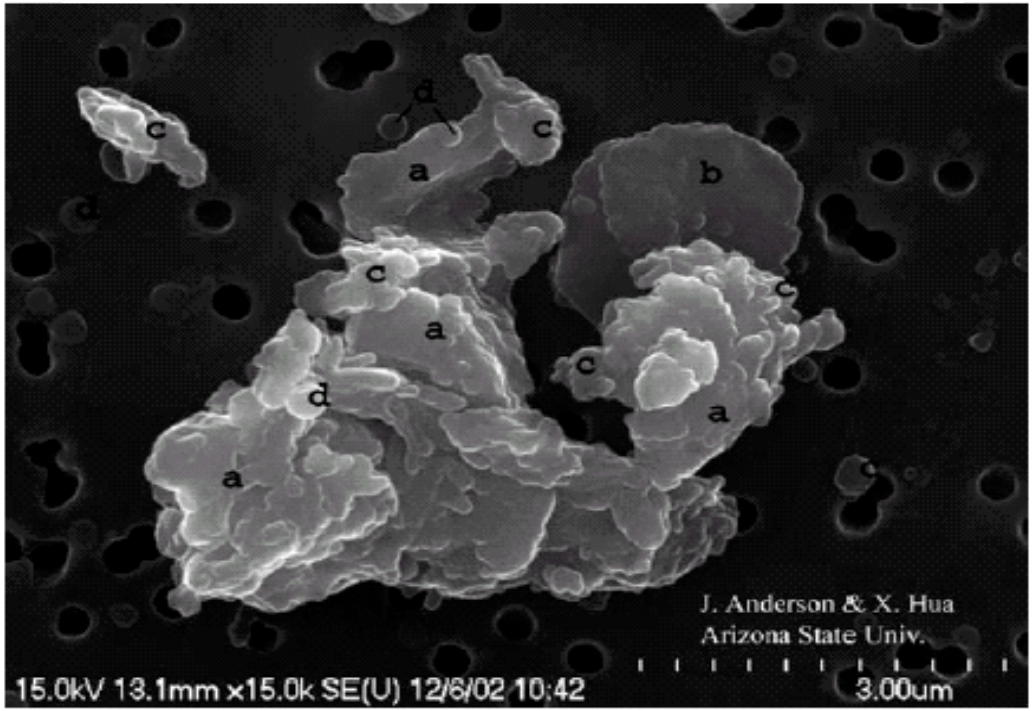


Fig. 9. Field emission SEM image of RF13 sample from 600 m leg over Yellow Sea. Large central particle is a silicate dust/black carbon aggregate consisting of: (a) Ca aluminosilicate, (b) Mg aluminosilicate, (c) black carbon spherule aggregates (soot), (d) separate carbon spheres.

N A S A / G S F C

- Experimental evidence of interactions btw particules and btw gas and particules
- Environmental impacts of the mixing

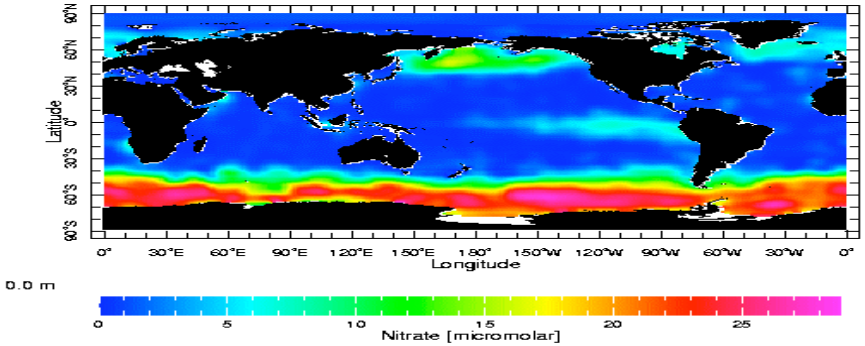
Ex 2 : Biogeochemical (and global climate) perspective.

« Iron cycle in the East Asia outflow, deposition to the North Pacific Ocean »

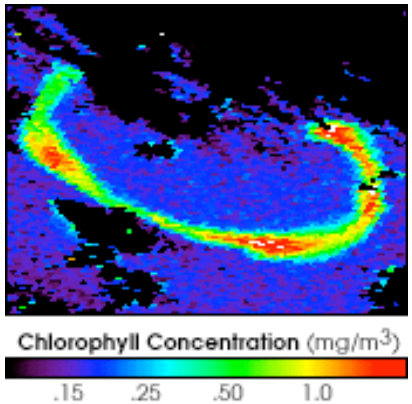
P. Chuang (UCSC), N. Meskhidze (NCSU)

Iron depositon in High Nutrients Low Chlorophyll regions

Annual average Nitrate concentration in surface water (levitus ocean atlas)



e.g / SOIREE expriment



Regional Iron Fertilisation experiment in different HNLC:

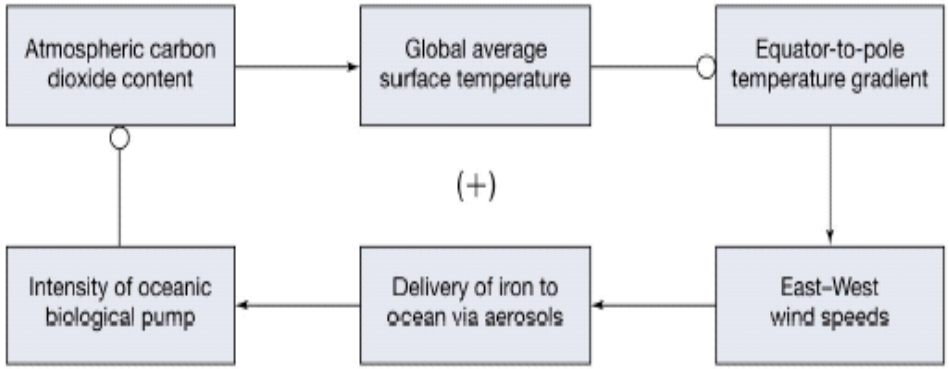
- **Bloom of biological activity and carbon sequestation**

A 30 to 90 µatm drawdown in surface pCO_2

- **Paleoclimate : 'The Iron hypothesis' (Martin)**

  Kohfeld et al., 2005; Archer et al., 2000; Mahowald et al.,1999 …

- **Climate change mitigation (!)**



The North Pacific HNLC

Source and bioavailability of iron

Atmospheric deposition ⇔ Dust is considered as the main sources of iron for open ocean

Rivers runoff

Upwelling

Dust iron is at the source mostly unsoluble i.e « not bioavailable »

Dissolved Iron Fraction DIF < 1 %

Measurments at remote sites show that DIF increases during atmospheric transport : DIF ~ 1-30 % with a large variability.

Modeling effort to better to characterize DIF (e.g Fan et al., 2006; Luo et al., 2007)

Possible anthropogenic influences on atmospheric processing , especially relevant in the North Pacific Ocean.

This study : Modeling approach to better charcterize soluble iron processes and deposition to the NPO.

Model developments

GEOS-CHEM coupling Meskhidze et al., 2005

Table 1. Species Simulated in Model

Symbol	Chemical Forms Allowed for Species[a]
SO_2[b]	$(SO_2)_g$
S(VI)[c]	$(SO_4^{2-})_{aq}$, $(HSO_4^{2-})_{aq}$, $(FeSO_4^{+})_{aq}$, $(AlSO_4^{+})_{aq}$, $(CaSO_4)_s$, $(Na_2SO_4)_s$, $(NaHSO_4)_s$, $((NH_4)_2SO_4)_s$, $(NH_4HSO_4)_s$, $((NH_4)_3H(SO_4)_2)_s$
NO_x[b]	$(NO)_g$, $(NO_2)_g$
N(V)[d]	$(HNO_3)_g$, $(NO_3^{-})_{aq}$, $(NH_4NO_3)_s$, $(NaNO_3)_s$
N(-III)[e]	$(NH_3)_g$, $(NH_4^{+})_{aq}$, $((NH_4)_2SO_4)_s$, $(NH_4HSO_4)_s$, $((NH_4)_3H(SO_4)_2)_s$, $(NH_4NO_3)_s$
Na[f,g]	$(Na^{+})_{aq}$, $(NaCl)_s$, $(NaNO_3)_s$, $(NaHSO_4)_s$, $(Na_2SO_4)_s$
Ca[g,h]	$(Ca^{2+})_{aq}$, $(CaCO_3)_s$, $(CaSO_4)_s$
Fe[g]	$(Fe^{3+})_{aq}$, $(Fe(OH)^{2+})_{aq}$, $(Fe(OH)_2^{+})_{aq}$, $(Fe(OH)_3^{0})_{aq}$, $(Fe(OH)_4^{-})_{aq}$, $(FeSO_4^{+})_{aq}$, $(Fe(OH)_3)_s$
Al[i]	$(Al^{3+})_{aq}$, $(Al(OH)^{2+})_{aq}$, $(Al(OH)_2^{+})_{aq}$, $(Al(OH)_3^{0})_{aq}$, $(Al(OH)_4^{-})_{aq}$, $(AlSO_4^{+})_{aq}$

$K_{0.6}Mg_{0.25}Al_{2.3}Si_{3.5}O_{10}(OH)_2$			
Smectite/Montmorillonite[c]	7	15	8
$Na_{0.6}Al_{1.4}Mg_{0.6}Si_4O_{10}(OH)_2 \cdot 4H_2O$			
Hematite[d]	5	8	5
Fe_2O_3			
Quartz	21	10	20
SiO_2			
Kaolinite	5	12	5
$Al_2Si_2O_5(OH)_4$			
Total	100	100	100

$3.5 \times 10^{-12}\exp[9.2 \times 10^{3}(1/298-1/T)]$

Dissolved iron modelling

- **11 new tracers in GC representing mineral species in the dust mode :**

  (Fe, Ca, Al, Na, Sil, K, Mg, SO_4^{2-}, NO_3^-, $NH_4)_{aq}$, $(CaCO_3)_s$

Symbol	Chemical Forms Allowed for Species[a]
SO_2[b]	$(SO_2)_g$
S(VI)[c]	$(SO_4^{2-})_{aq}$, $(HSO_4^{2-})_{aq}$, $(FeSO_4^+)_{aq}$, $(AlSO_4^+)_{aq}$, $(CaSO_4)_s$, $(Na_2SO_4)_s$, $(NaHSO_4)_s$, $((NH_4)_2SO_4)$ $(NH_4HSO_4)_s$, $((NH_4)_3H(SO_4)_2)_s$
NO_x[b]	$(NO)_g$, $(NO_2)_g$
N(V)[d]	$(HNO_3)_g$, $(NO_3^-)_{aq}$, $(NH_4NO_3)_s$, $(NaNO_3)_s$
N(-III)[e]	$(NH_3)_g$, $(NH_4^+)_{aq}$, $((NH_4)_2SO_4)_s$, $(NH_4HSO_4)_s$, $((NH_4)_3H(SO_4)_2)_s$, $(NH_4NO_3)_s$
Na[f,g]	$(Na^+)_{aq}$, $(NaCl)_s$, $(NaNO_3)_s$, $(NaHSO_4)_s$, $(Na_2SO_4)_s$
Ca[g,h]	$(Ca^{2+})_{aq}$, $(CaCO_3)_s$, $(CaSO_4)_s$
Fe[g]	$(Fe^{3+})_{aq}$, $(Fe(OH)^{2+})_{aq}$, $(Fe(OH)_2^+)_{aq}$, $(Fe(OH)_3^0)_{aq}$, $(Fe(OH)_4^-)_{aq}$, $(FeSO_4^+)_{aq}$, $(Fe(OH)_3)_s$
Al[i]	$(Al^{3+})_{aq}$, $(Al(OH)^{2+})_{aq}$, $(Al(OH)_2^+)_{aq}$, $(Al(OH)_3^0)_{aq}$, $(Al(OH)_4^-)_{aq}$, $(AlSO_4^+)_{aq}$

Dissolved iron FEDI (oxydation III)

- **One mode representative of dust (aggregation of bins PM10)**
- **Tracers are transported and removed (wet and dry dep) as dust in GC**
- **At the source : FETOT = 3.7 % * DUST ;**

  DIF = FEDI / FETOT = 0.45 % (from obs ACE-ASIA)

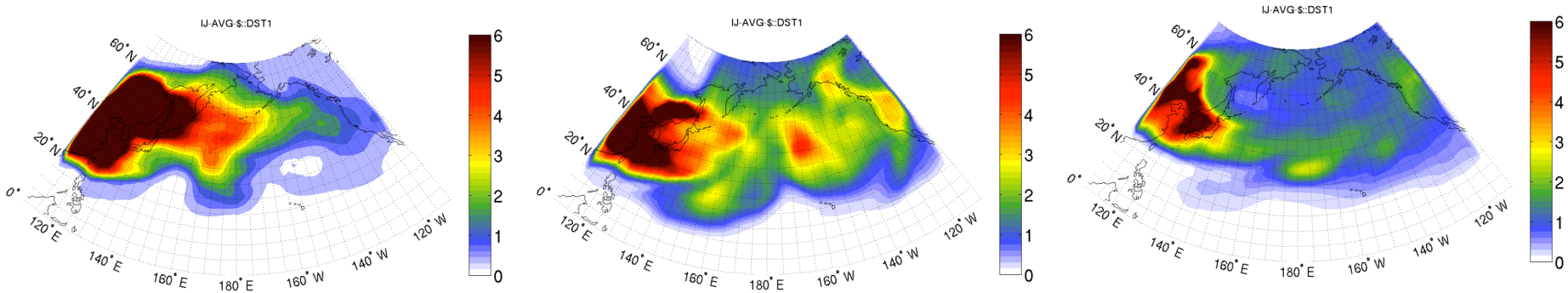
Test Case Simulation (2 x 2.5, Full chemistry) : MARCH-APRIL 2001

Apr 7-14 — **Week 2**

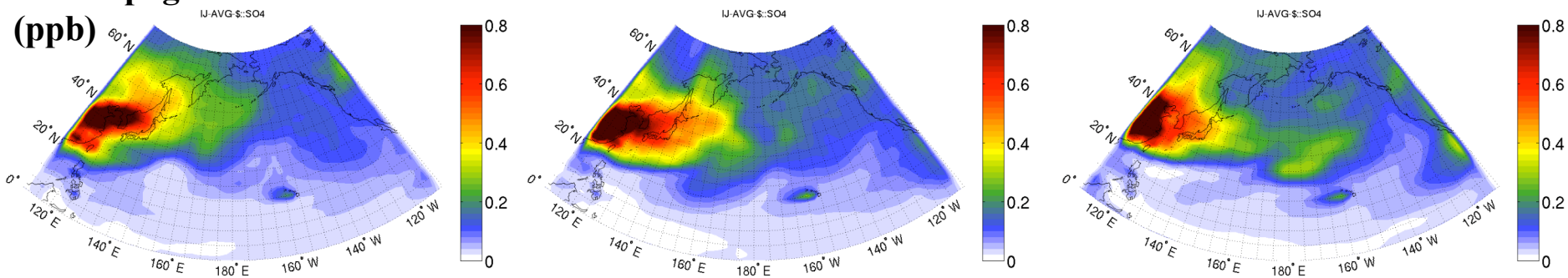
Apr 15-21 — **Week 3**

Apr 22-28 — **Week 4**

DUST (< 1µm) vert av. (µg.m-3)



Anthropogenic SO4 vert av. (ppb)



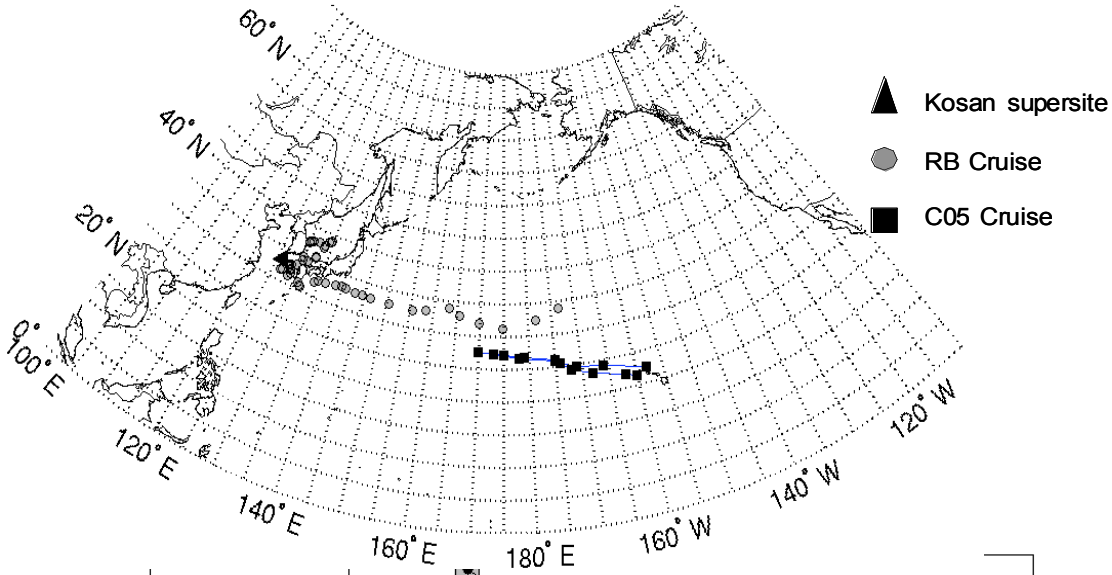
« HIDU »
High dust regime

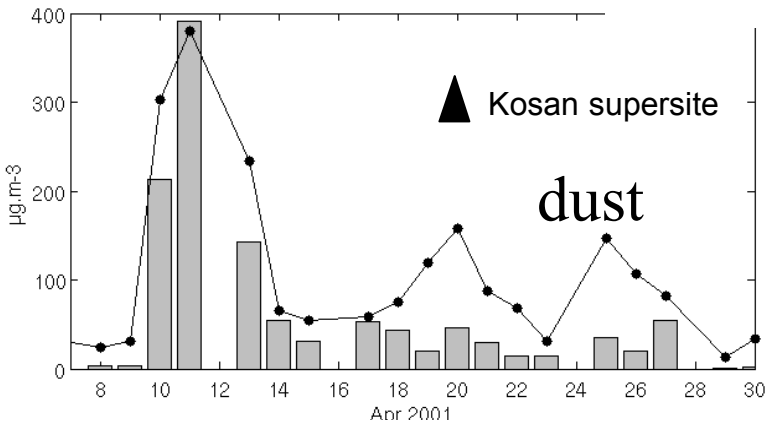
Validation of GC aerosol fields

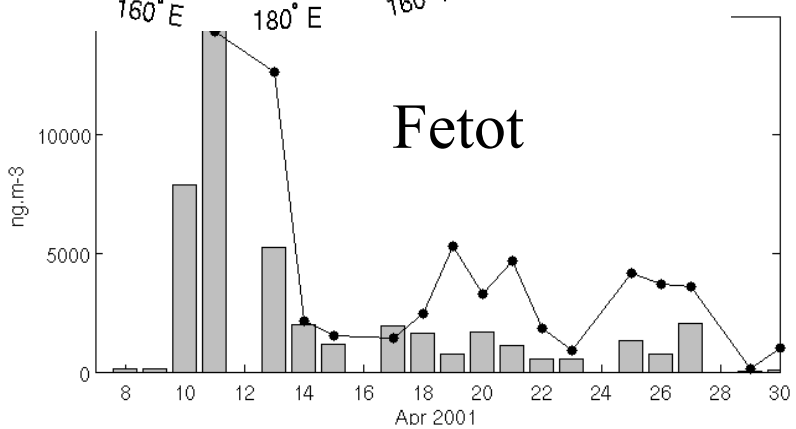
Heald et al., 2006

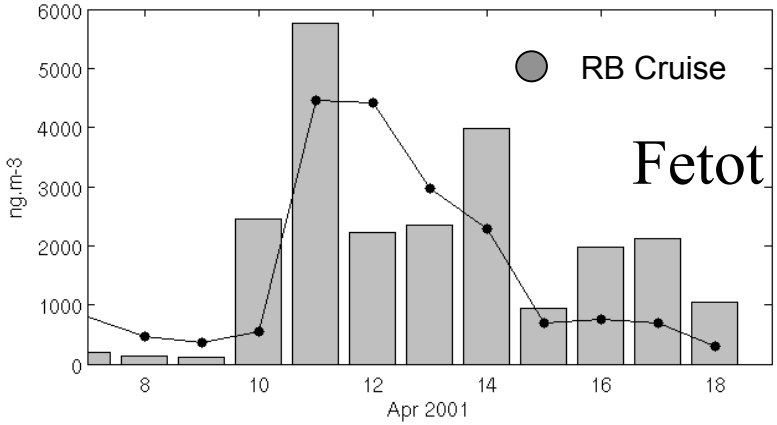
« LODU »
Low dust regime

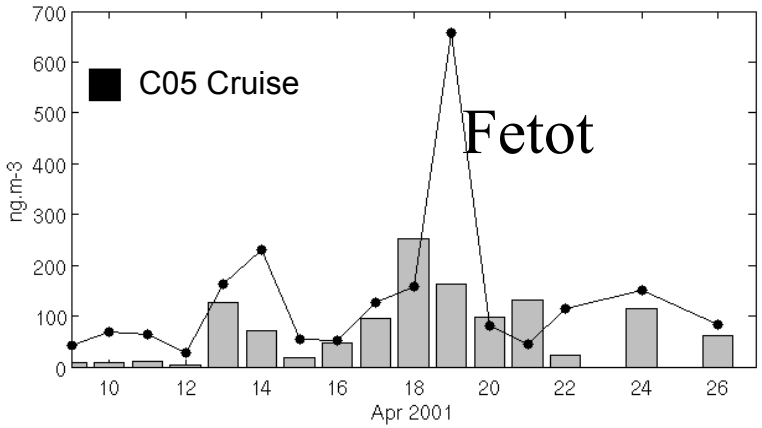
Model / data comparison during april 2001





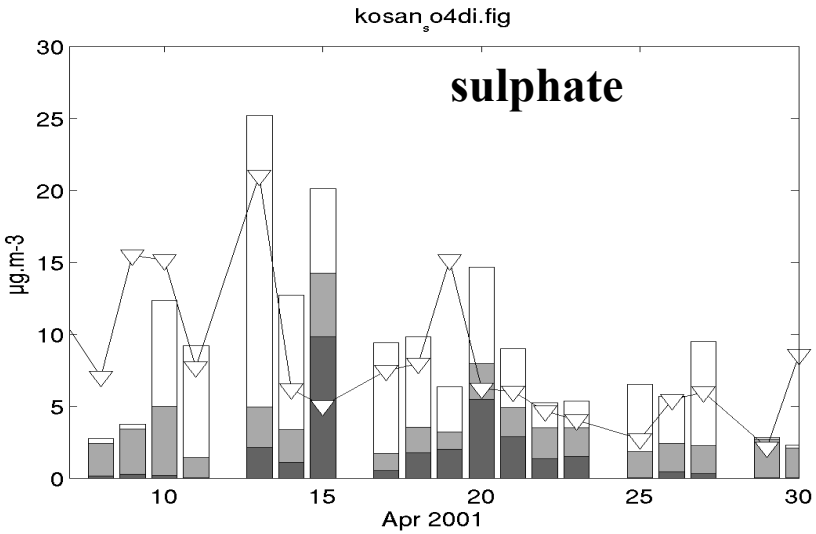


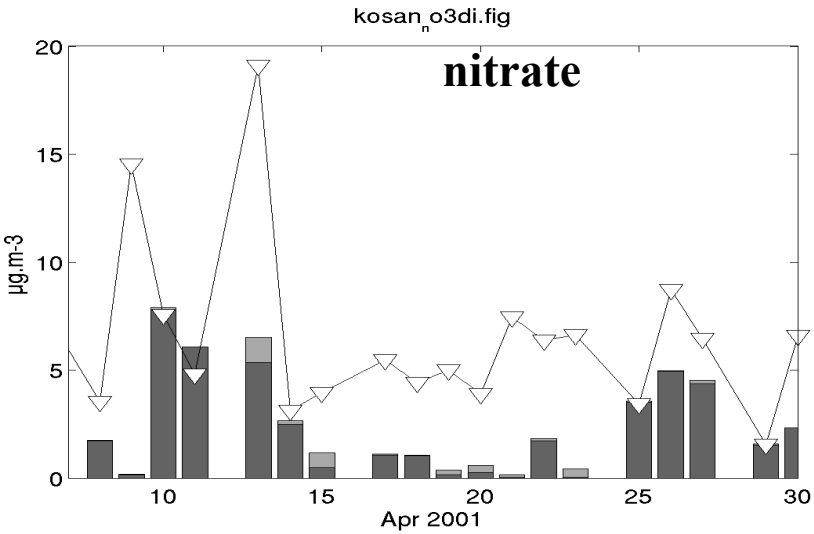


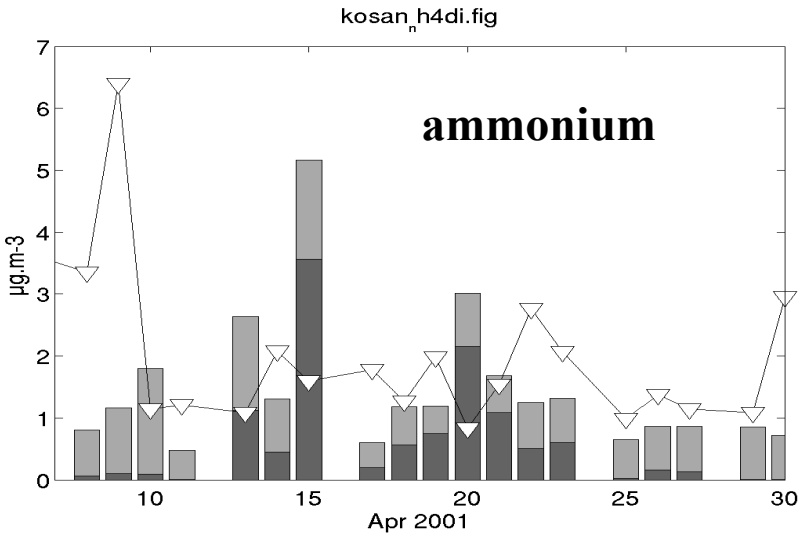


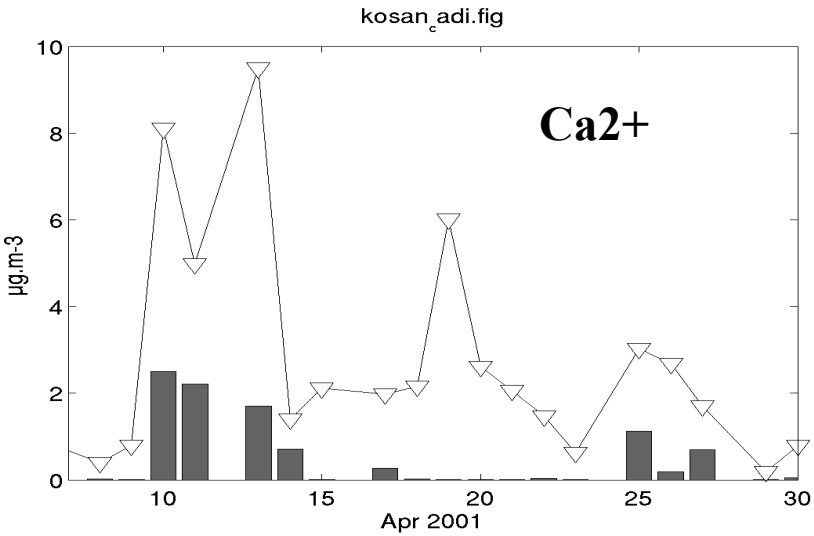
Scavenging of anthropogenic compounds by dust (Kosan)

dust mode
anthro mode
sulfate in solid CaSO4



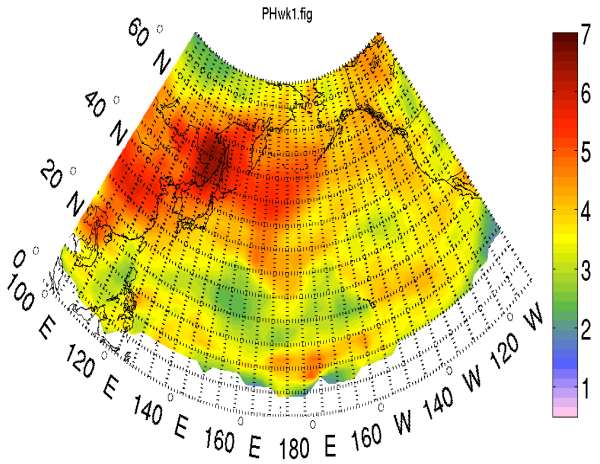




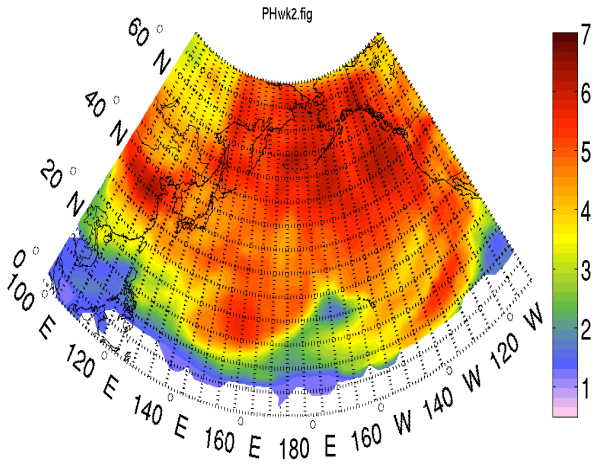


Dust mode pH evolution

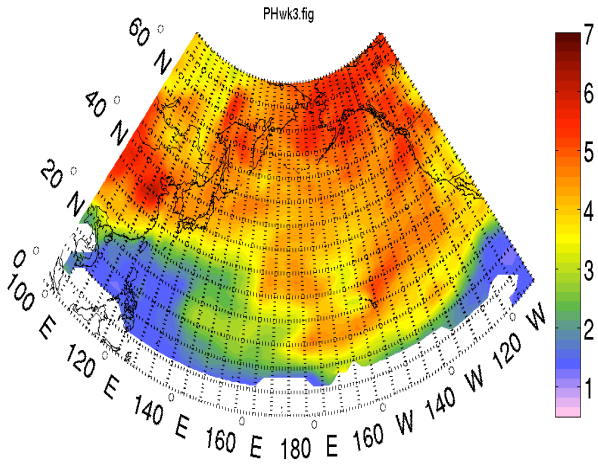
HIDU **LODU**



Week 2



Week 3



Week 4

$CaCO_3$ dissolution

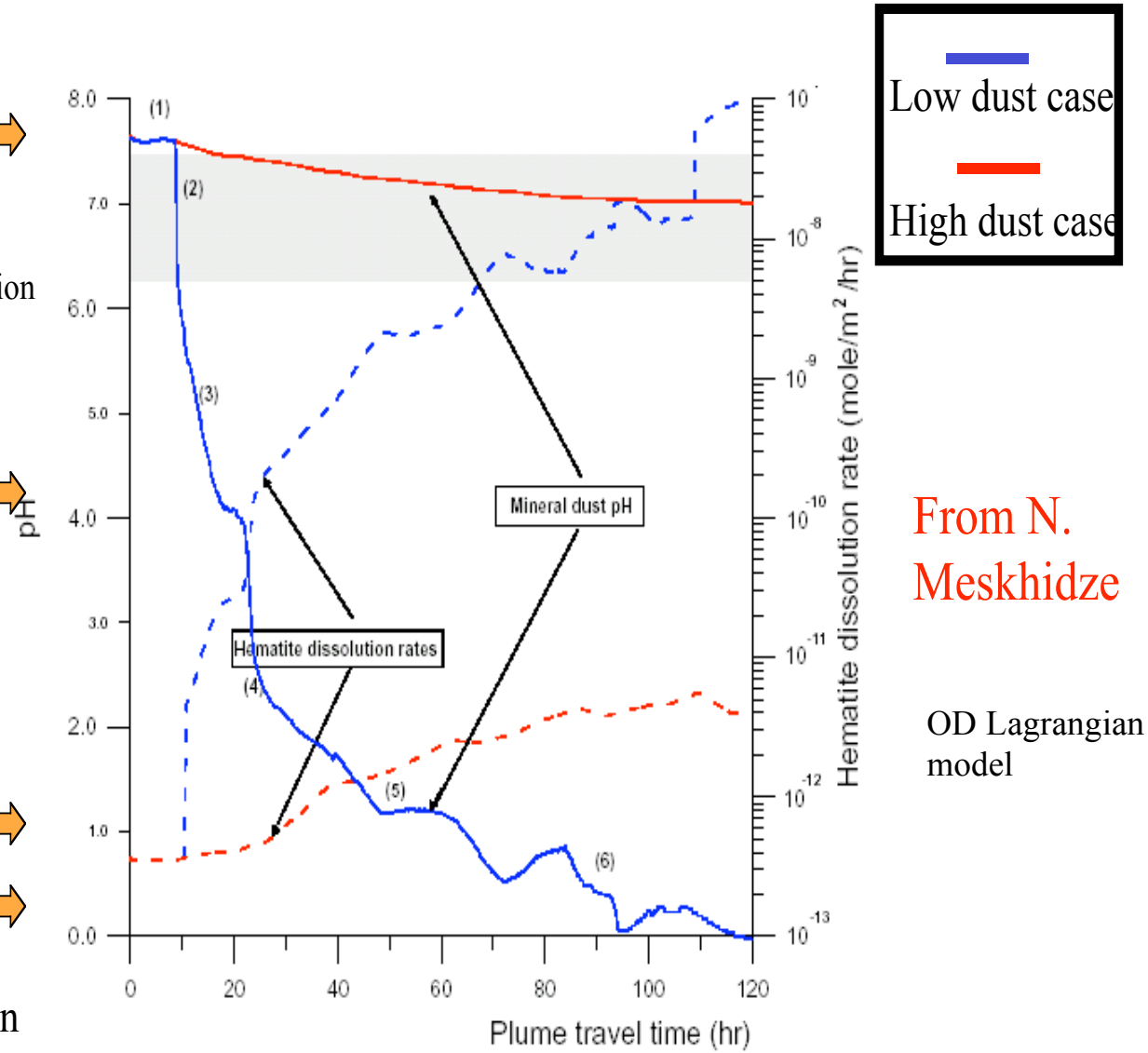
$CaSO_4$ formation
Carbonate volatilisation

NH_3 solubilisation.

NO_3 volatilisation

SO_4

Self neutralisation



From N. Meskhidze

OD Lagrangian Box model

Increasing interaction with acidic compounds

>

Soluble iron formation

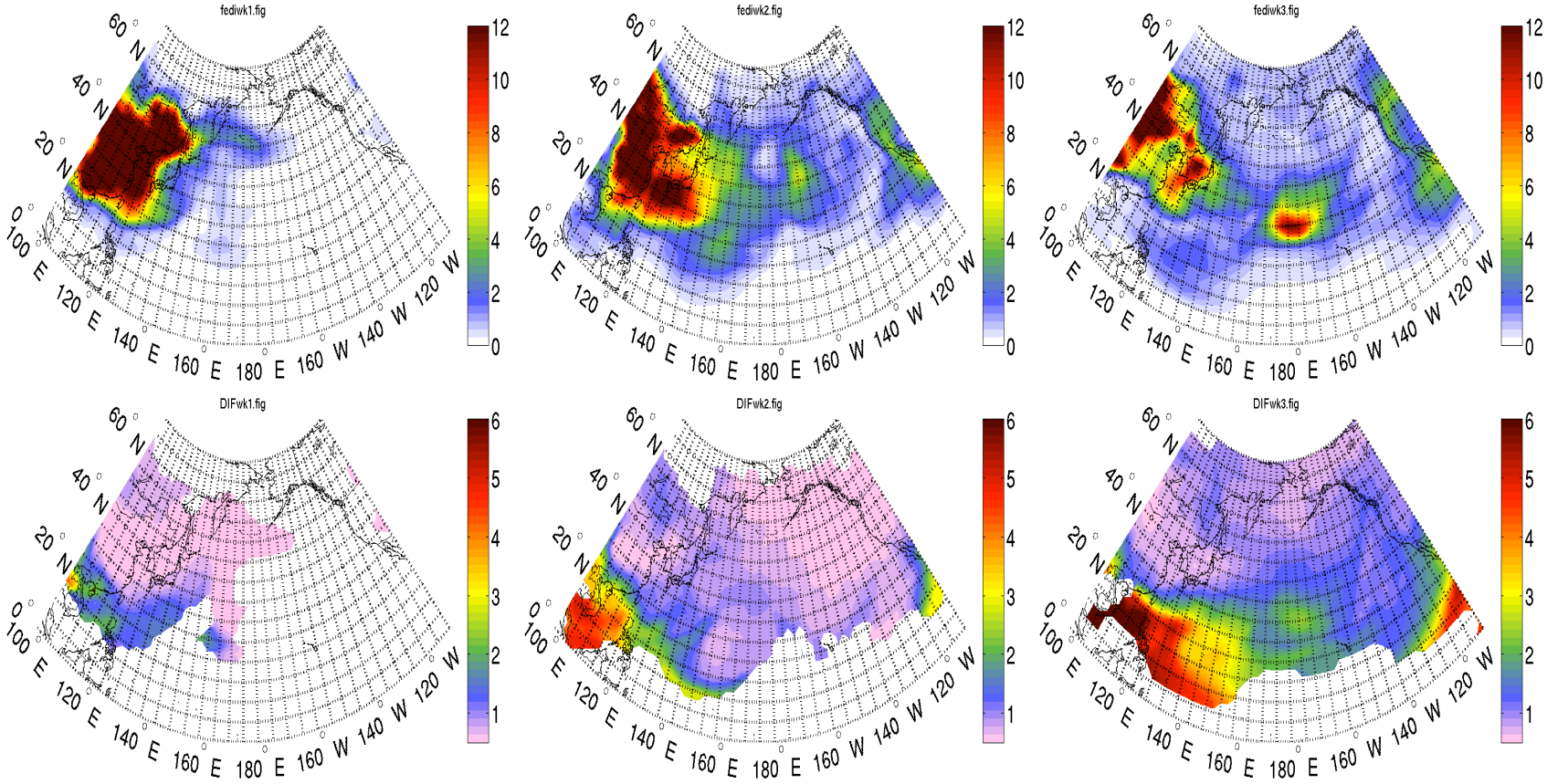
HIDU **LODU**

vert av.

FEDI

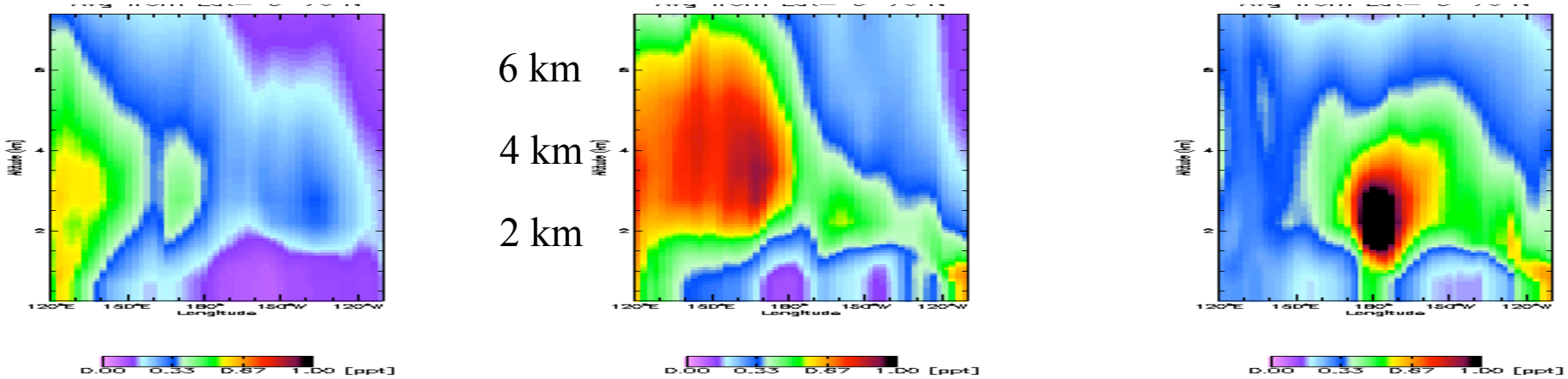
(ng.m-3)

DIF %

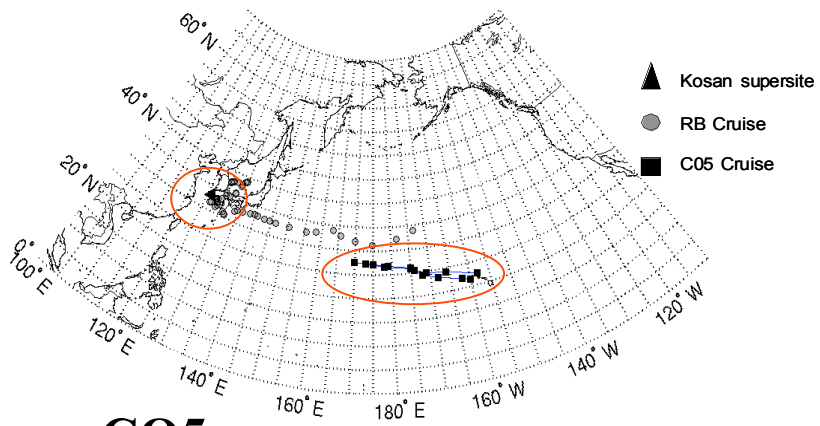


- **Large dust event are not necessarily the most FEDI productive (consistency with the 0D scheme, Meskhidze et al., 2005)**

FeDI Vertical distribution



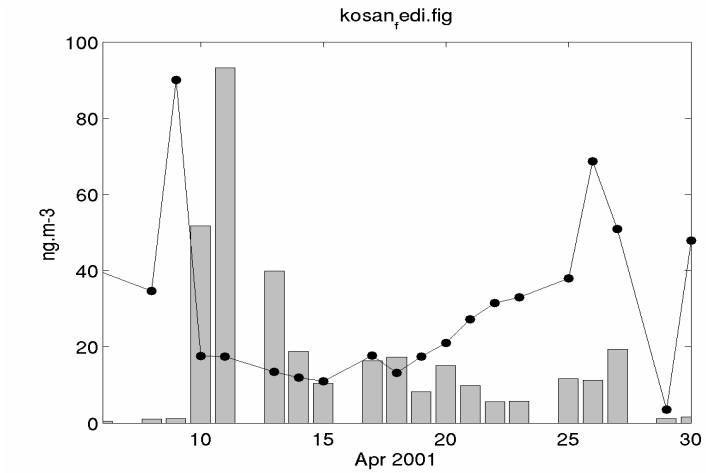
Simulated vs measured dissolved iron

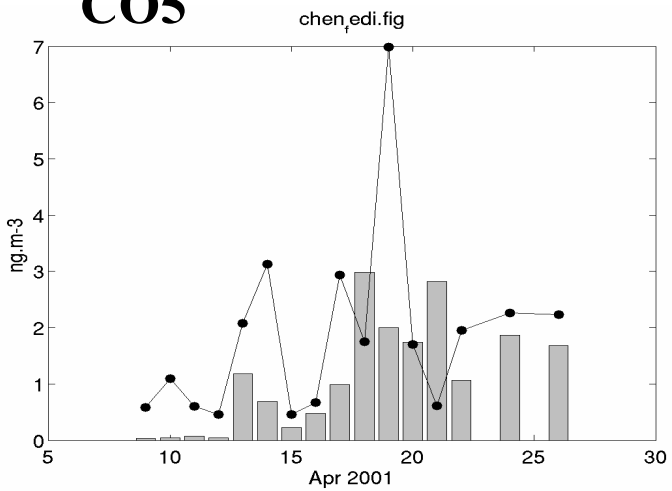


Kosan

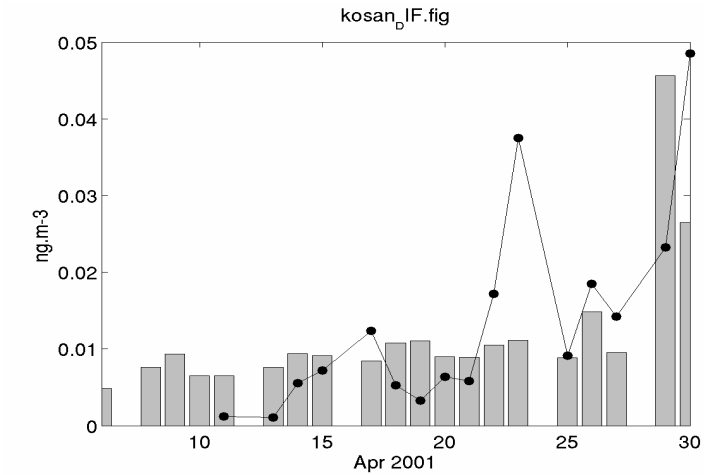
CO5

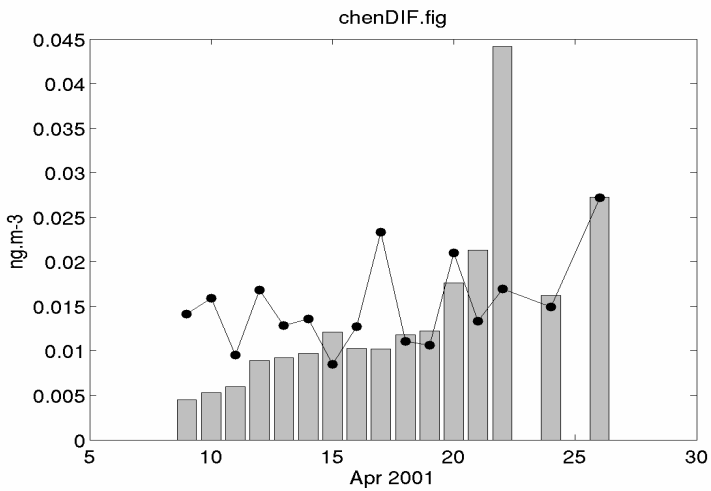
FEDI





DIF

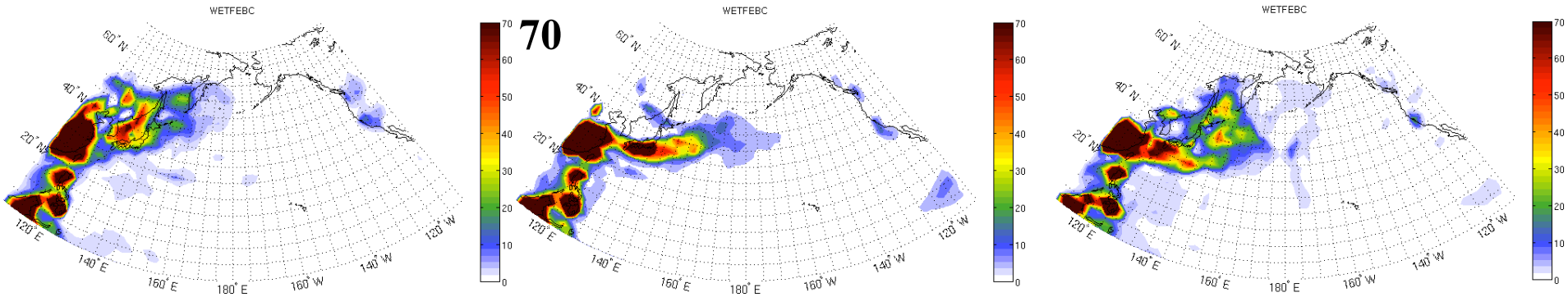




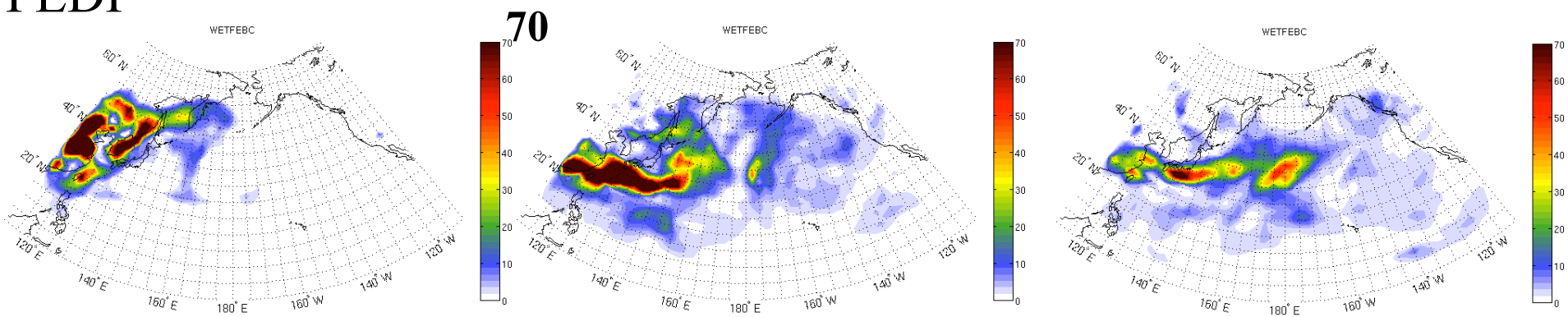
Wet deposition of soluble iron

FEbc

ng.m^{-2}.d^{-1}



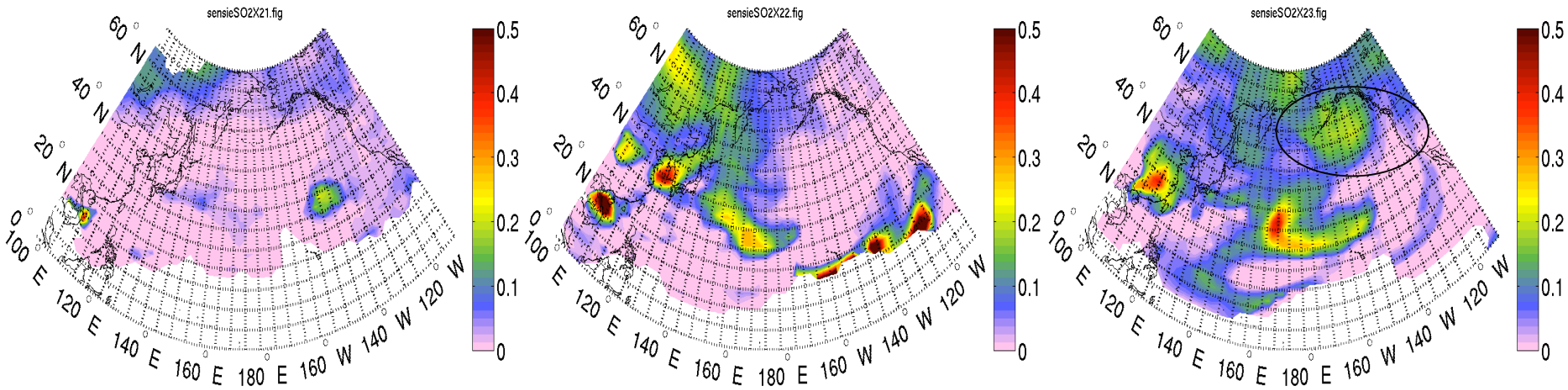
FEDI



« HIDU » « LODU »

Sensitivity studies : SO2 x 2 , April 2001

HIDU **LODU**



Relative change (fraction) in mean soluble iron concentration (and dissolved iron fraction)

2001 seasonal results

FEDI (ng.m-3)

DIF(%)

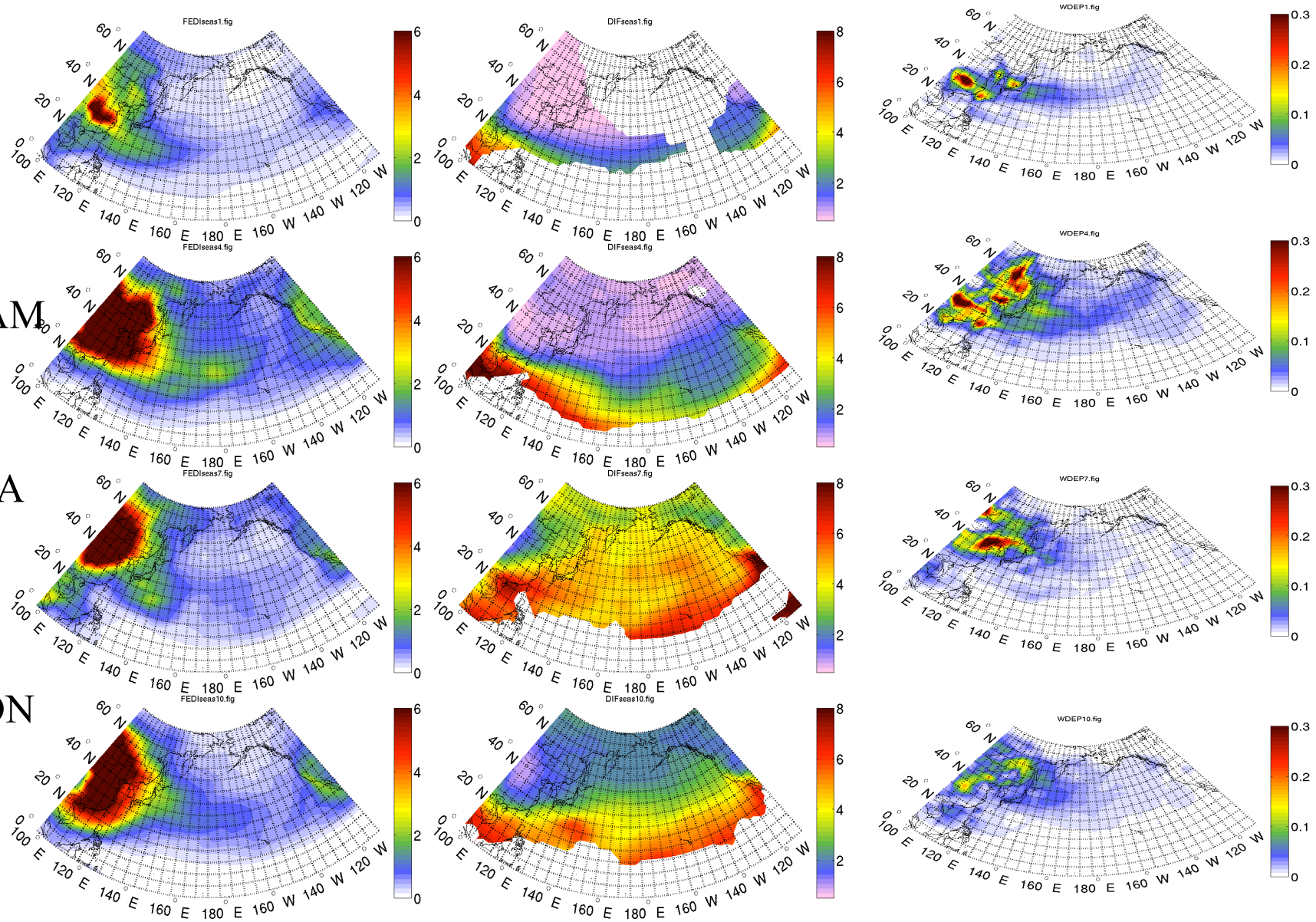
WETDEP (ng.m-2.s-1)

JF

MAM

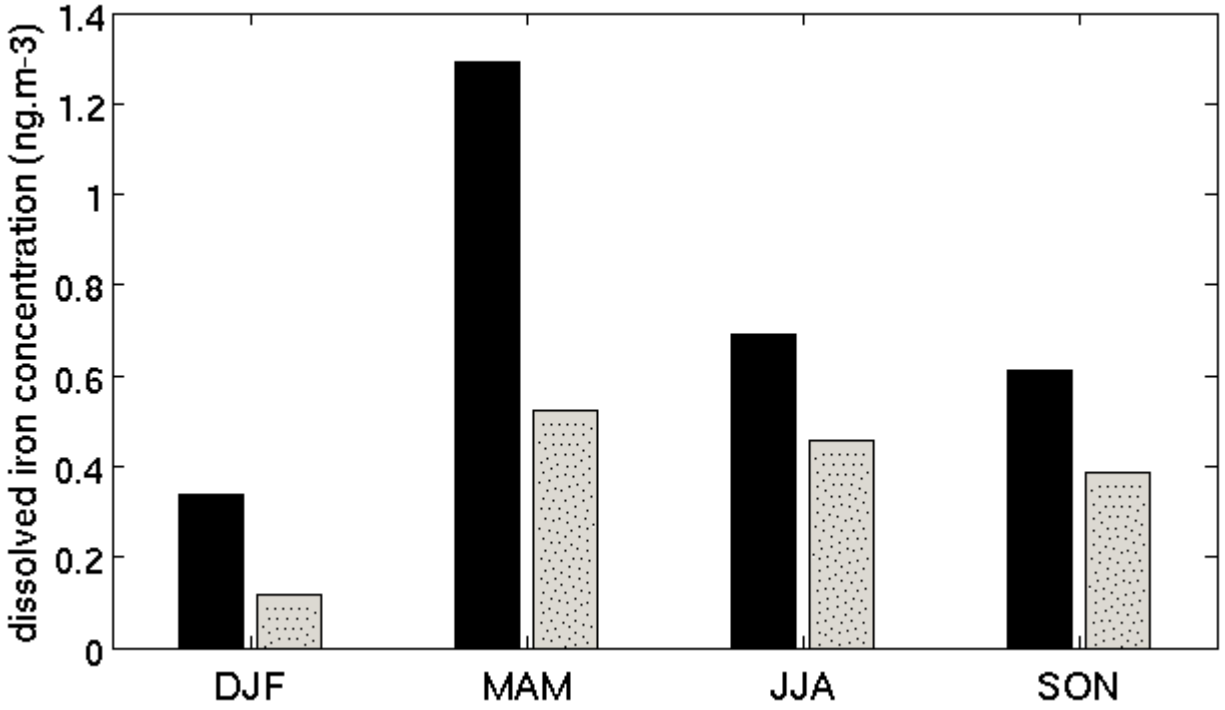
JJA

SON



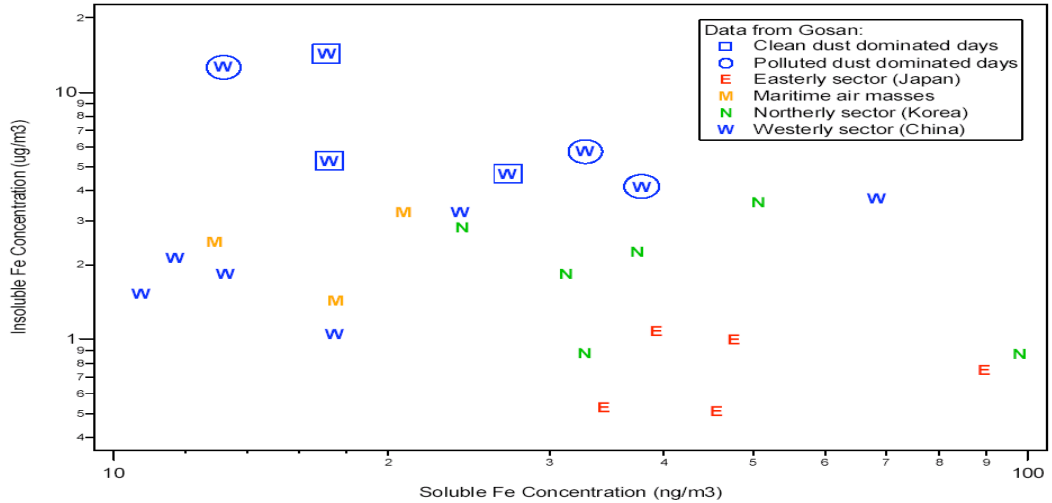
Chemical production of soluble iron via dust pollution interactions

(averaged concentrations 140E – 130 W , 20N-60N)

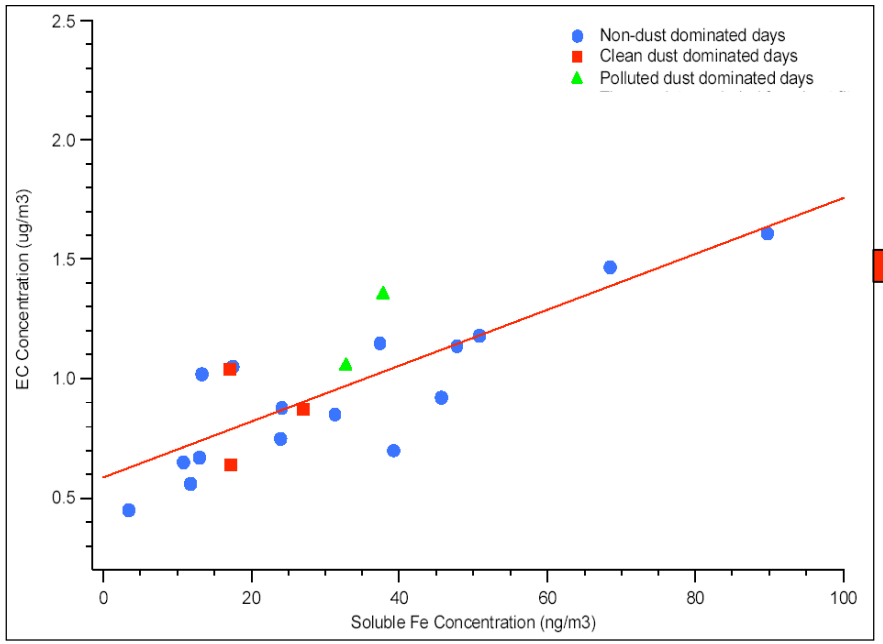


Anthropogenic direct contribution ?

Chuang et al., 2005



No significant correlation with total iron carried by dust



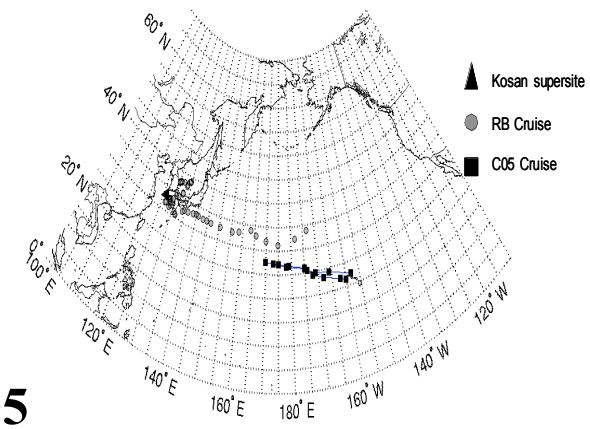
Good correlation (R=0.67) with BC particles (anthropogenic activities)

Combustion processes as significant source of dissolved iron ?

Anthropogenic DIF = 10 %

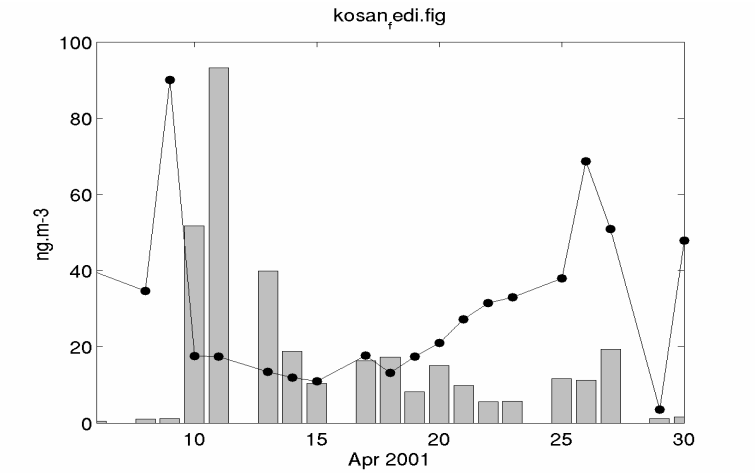
Simulated vs measured dissolved iron

Kosan



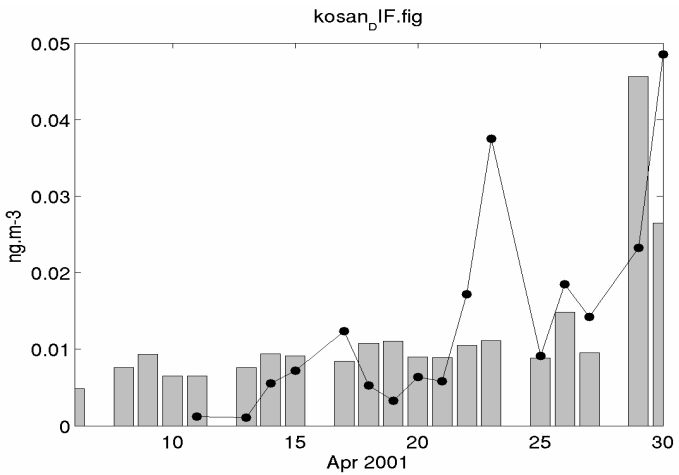
CO5

FEDI



Direct Contribution of pollution (not seen by the model)

DIF



Increase of DIF : Indirect Contribution of pollution via interaction with dust

Conclusions

- **Impact of anthropogenic pollution on soluble iron carried by dusts in the East Asian outflow.** Longer term simulations, further validations (global) and sensitivity studies are required.

- **Importance of chemical buffering effects : low intensity events (more frequent) are more efficient to produce soluble iron compared to big storms.**

  =>Validation and further development of dust/anthro heterogeneous chemistry and aerosol µ-physic in GEOS-CHEM is an important issue for iron modelling.

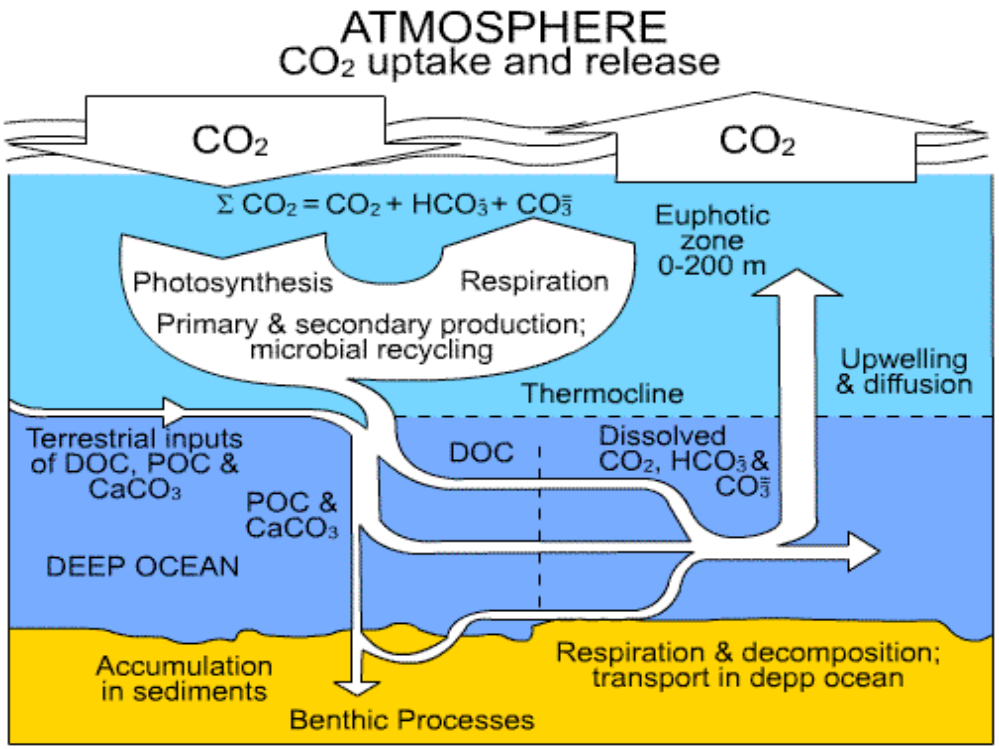
- **Other mechanisms for dust iron processing and DIF increase (chlorine, iron III photoreduction / dissolution promoted by organic acids in clouds).**

- **Potential importance of continuous anthropogenic emission of soluble iron (Luo et al., 2007). Experimental characterisation of combustion iron and processing is an issue.**

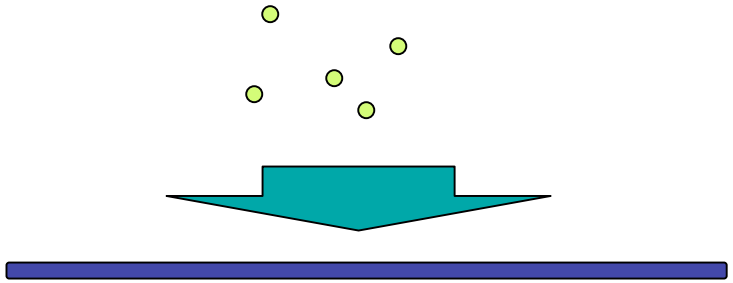
How will soluble iron deposition and ecosystem response evolve in the future ?

… Toward possible climatic impacts ?

What's going on in the ocean



FeDi deposition

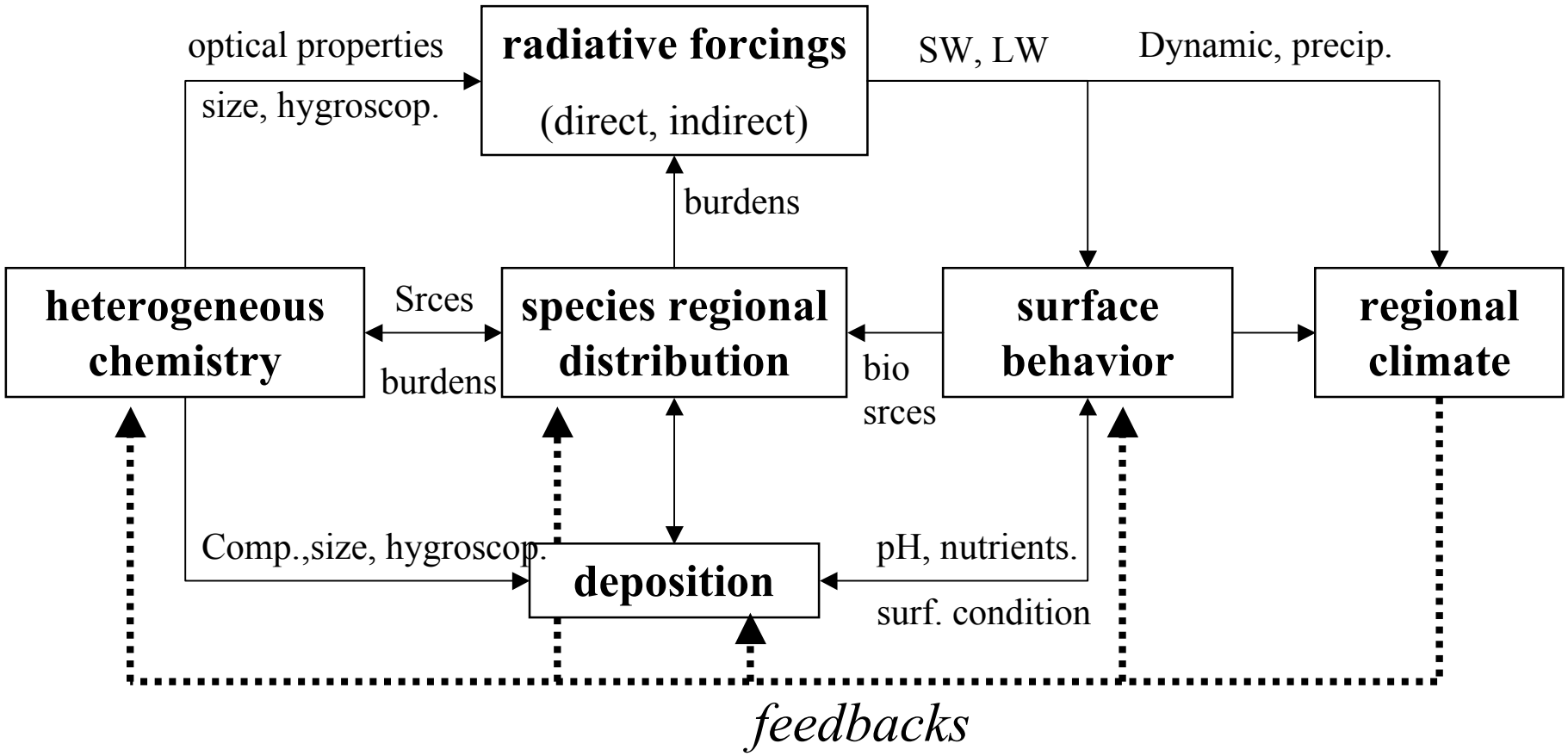


Iron Chemistry in the marine boundary layer

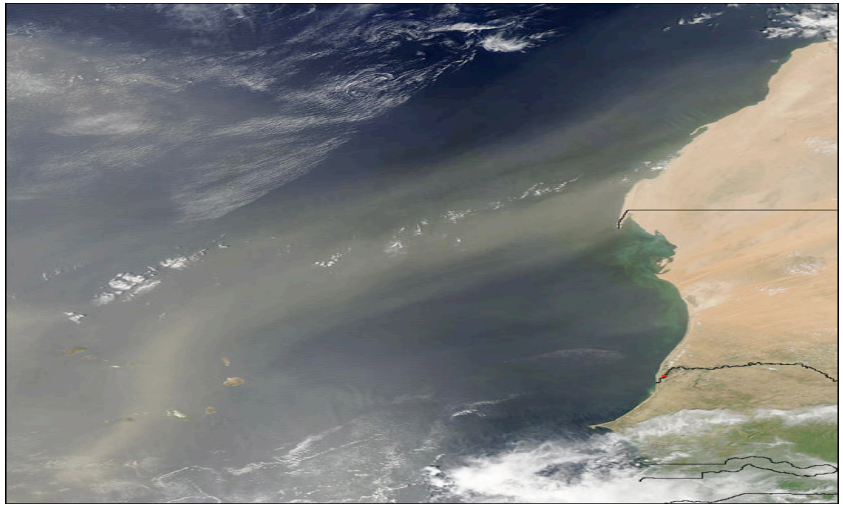
Availability of other nutrients

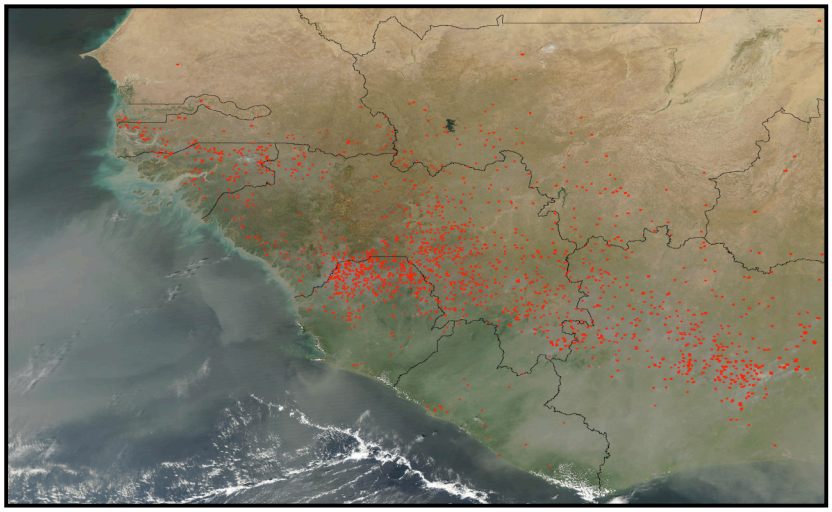
Ecosystem dynamics

Heterogenous chemistry / regional climate



Over west Africa ….





Effect of aerosol mixture (DUST, OC , BC …) on West African regional climate ?

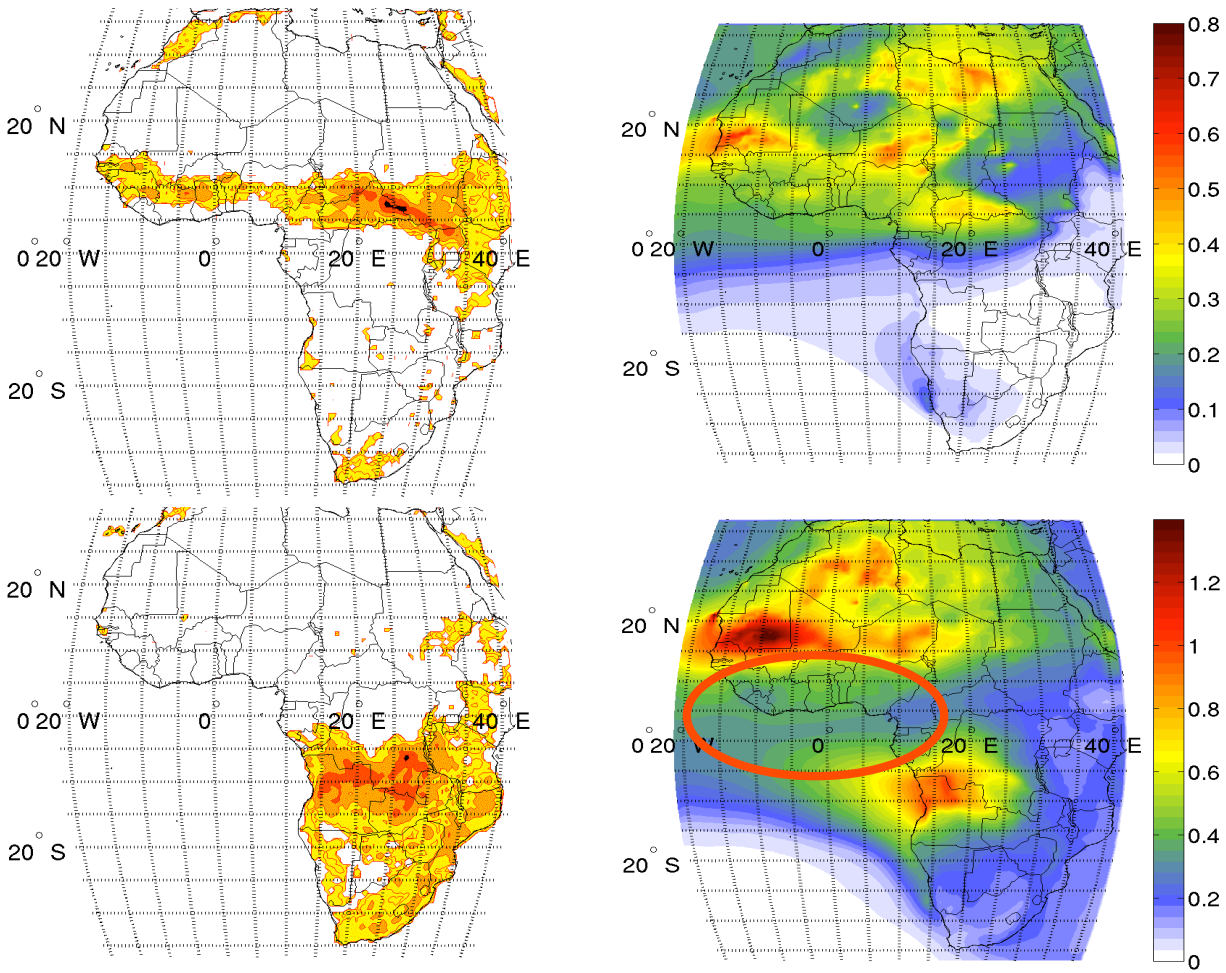
Seasonal BC (DJF – JJA 2006) emissions

contour =[0 0.01 0.1 1 5 10 15] mg.m-2.day

RegCM AOD

DJF

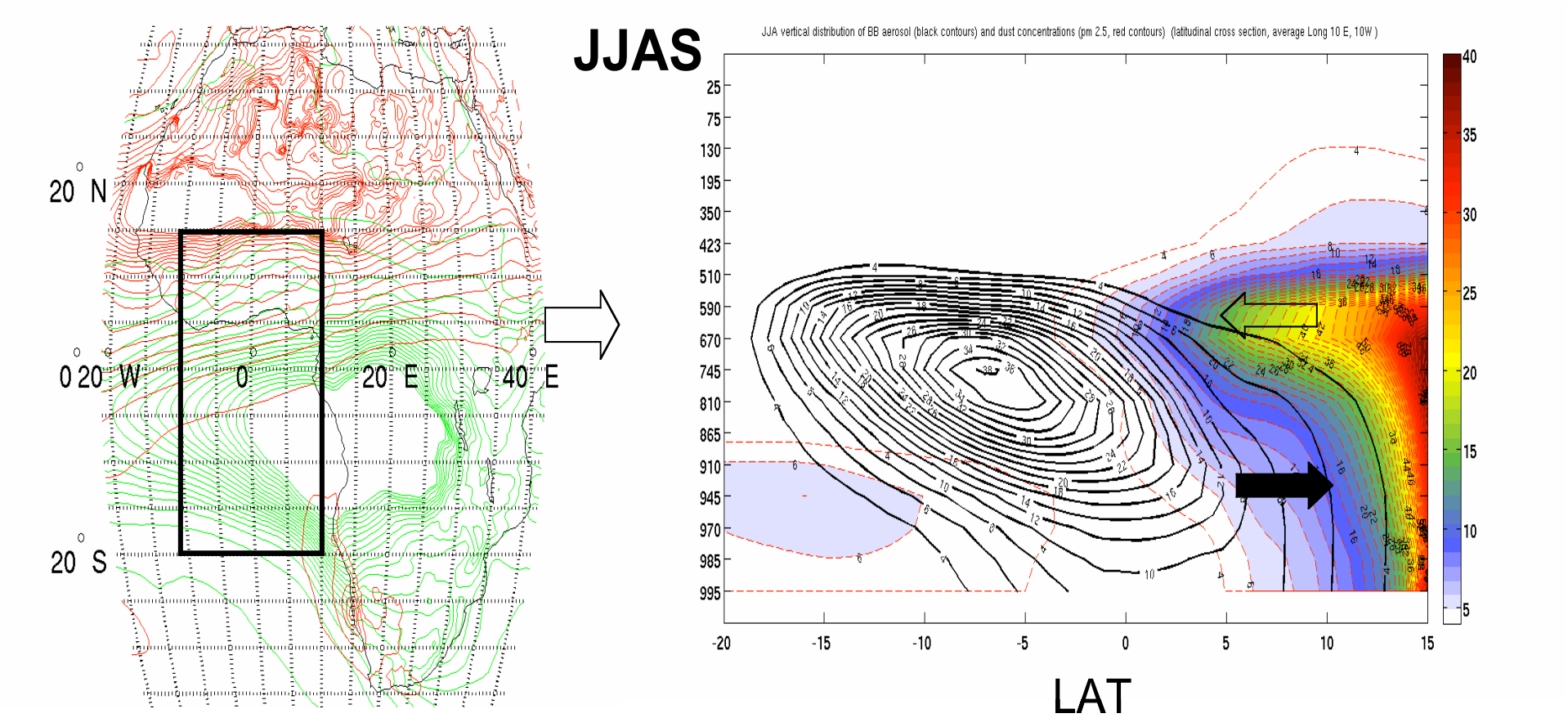
JJAS



Climatic signal of BB aerosol ?

Megacities in West Africa, which impacts ?

Aerosol mixing

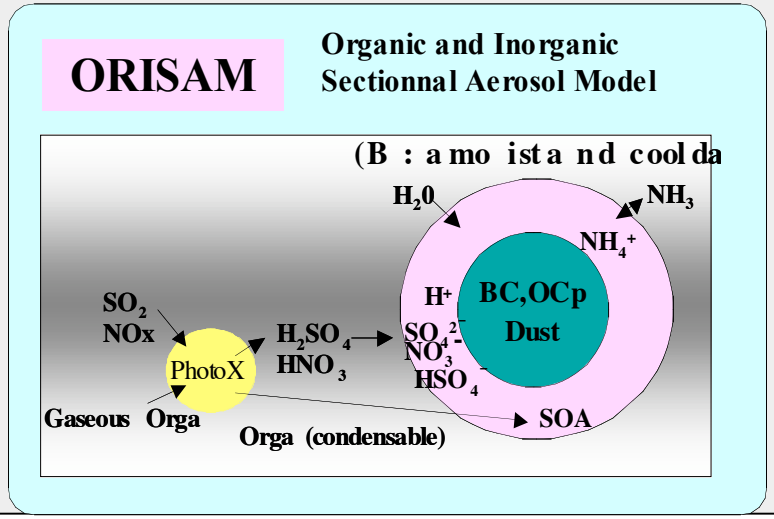


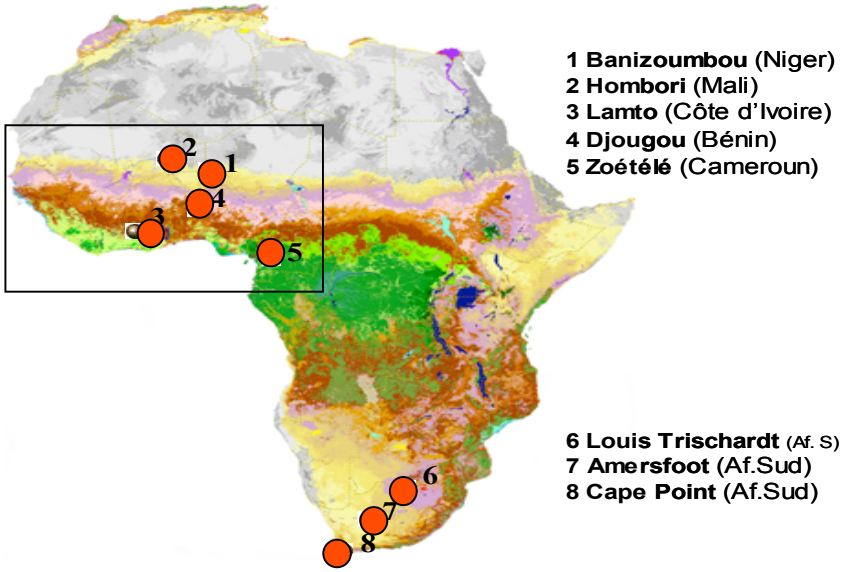
RegCM , dust + BB aerosols, JJAS 2005-2006

Mixing state and optical properties ? Needs more a more detailed chemstry scheme compatible with regional climate modelling

On going actvities at Laboratoire d'Aérologie

- Coating formation
- Coating composition (mineral dissolution, …)
- Optical and CCN properties





DEBITS IDAF activities

Validation and constraints

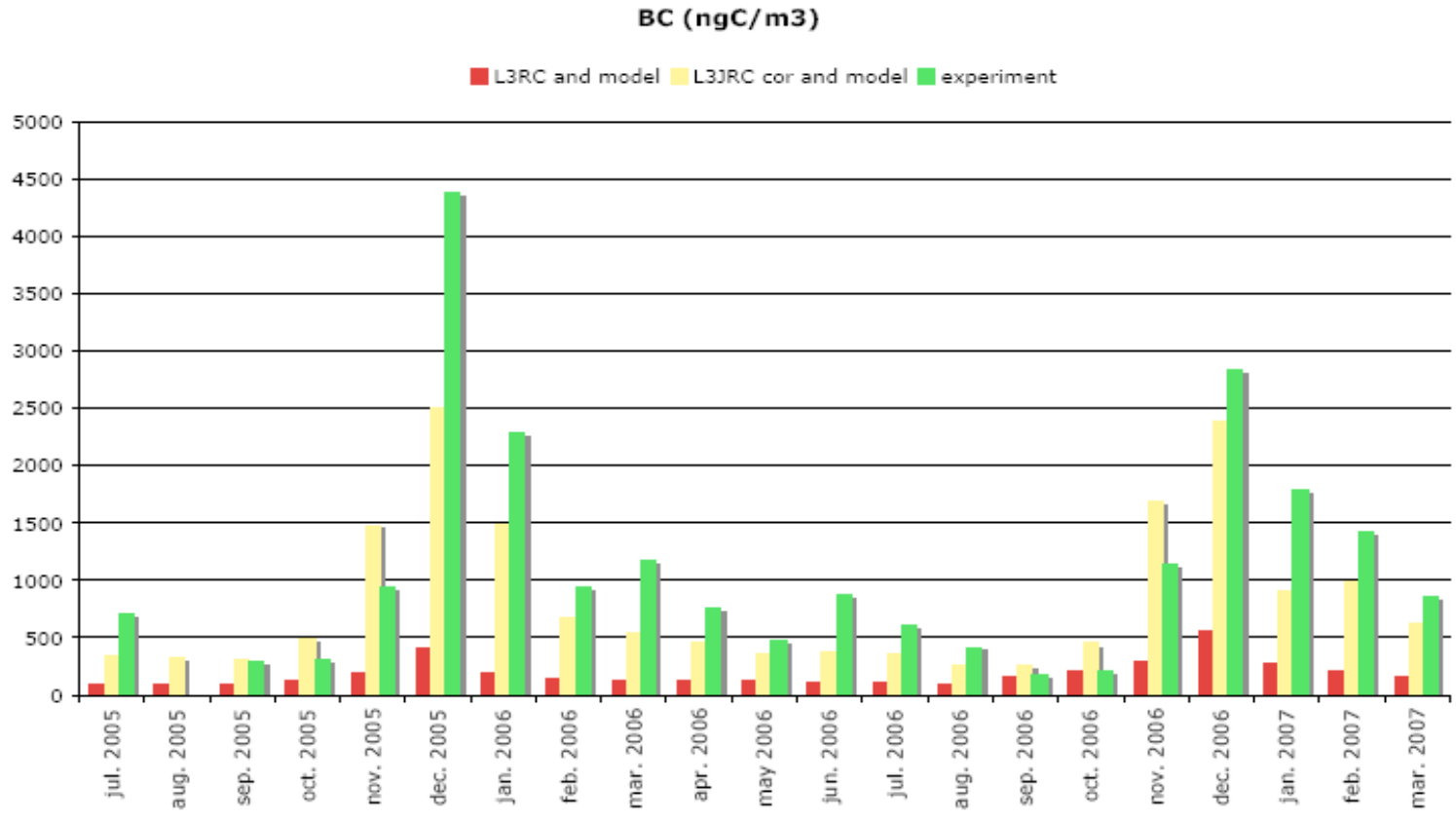
Regionalisation and impact studies

Promotion of modelling activities in Africa

RegCM

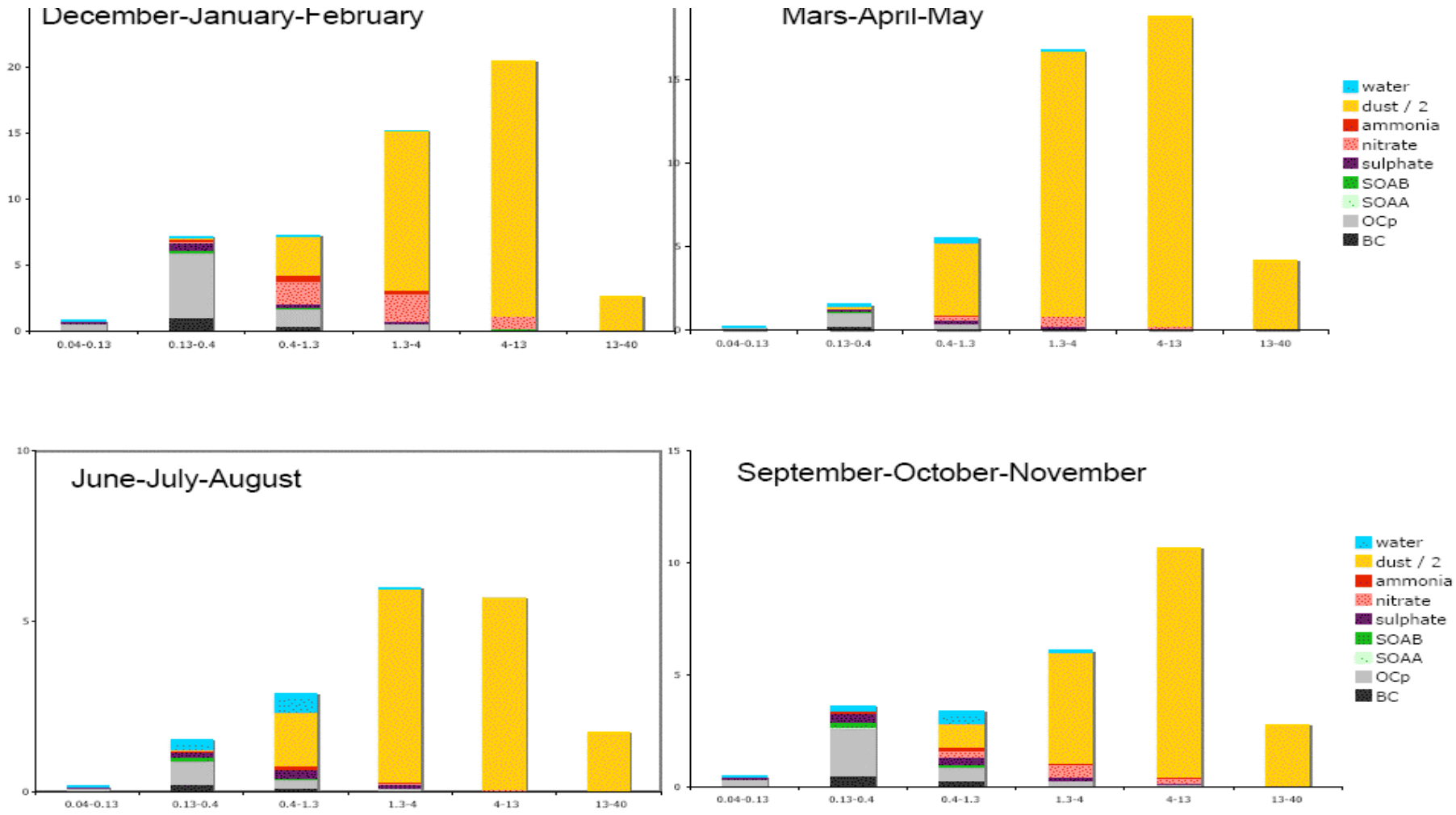
TM4 - ORISAM

Djougou (Benin)



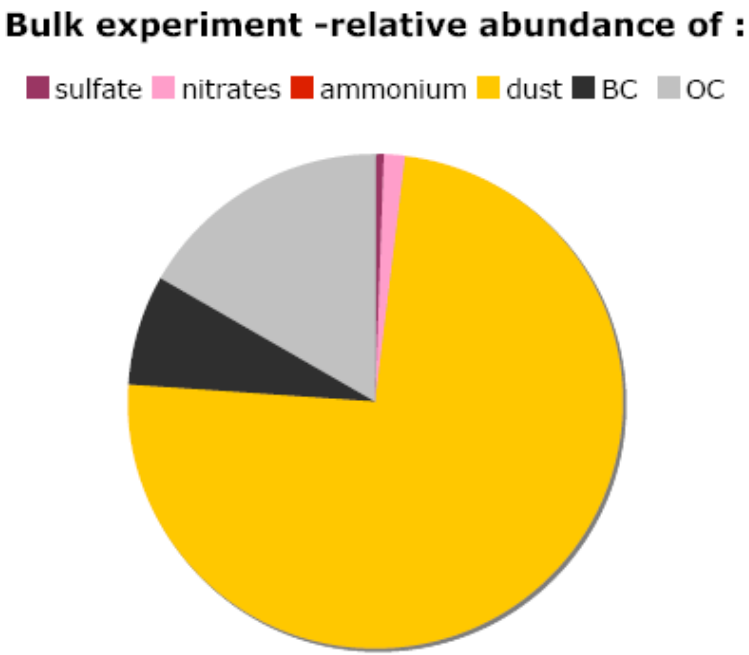
TM4 – ORISAM

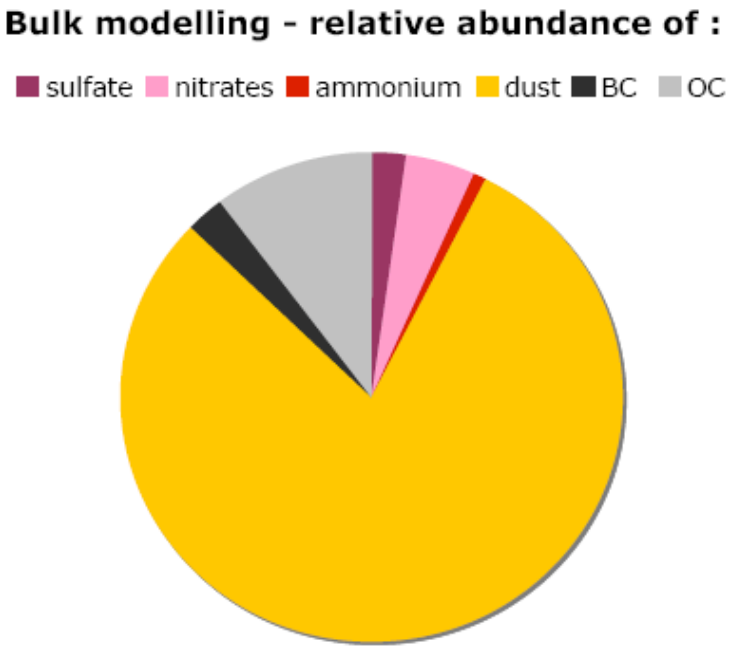
Djougou (Benin)



Djougou (Benin)

TM4 –ORISAM vs OBS



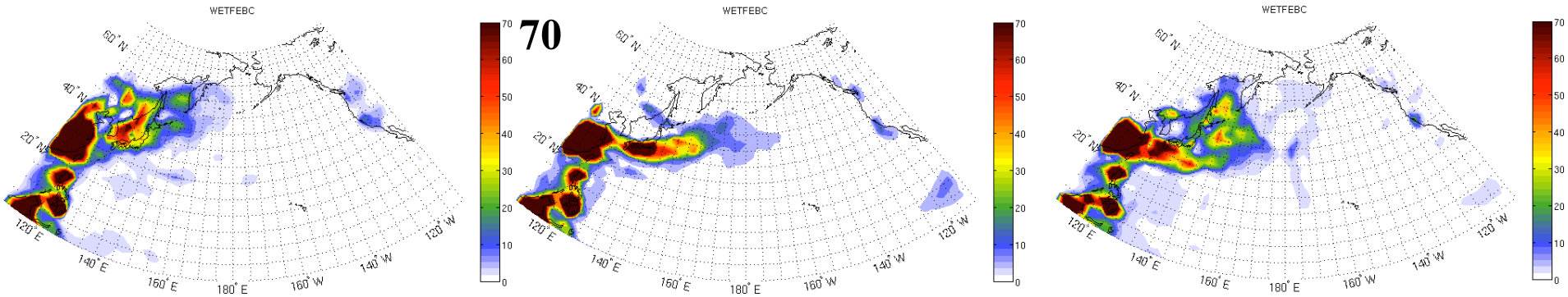


Thank you !

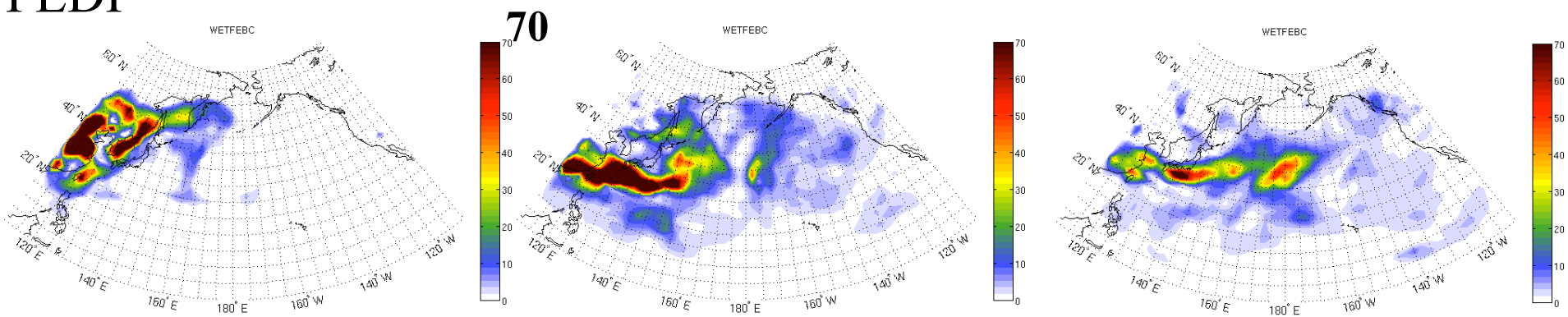
Wet deposition of soluble iron

FEbc

ng.m^{-2}.d^{-1}



FEDI



« HIDU » « LODU »

Iron dissolution modelling

GEOS-CHEM

Meskhidze et al., 2005

1: Assume an initial mineral composition for the dust

Table 3. Concentration of Major Minerals in the Soil and Clay Fractions of Surface Soils in the Gobi Desert and in Mineral Dust Originating From These Soils

Mineral	In Soil,[a] % wt		In Mineral Dust and Used as Initial Condition for Model Simulation,[b] % wt
	In Silt	In Clay	
Anhydrite $CaSO_4$	6	0	6
Calcite $CaCO_3$	12	0	11
Albite $NaAlSi_3O_8$	18	8	17
Microcline $KAlSi_3O_8$	8	5	8
Illite[c] $K_{0.6}Mg_{0.25}Al_{2.3}Si_{3.5}O_{10}(OH)_2$	18	42	20
Smectite/Montmorillonite[c] $Na_{0.6}Al_{1.4}Mg_{0.6}Si_4O_{10}(OH)_2 \cdot 4H_2O$	7	15	8
Hematite[d] Fe_2O_3	5	8	5
Quartz SiO_2	21	10	20
Kaolinite $Al_2Si_2O_5(OH)_4$	5	12	5
Total	100	100	100

ISORROPIA