Impact of the phase transition of sulfate particules of aerosols

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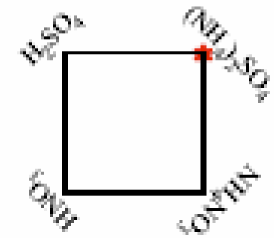
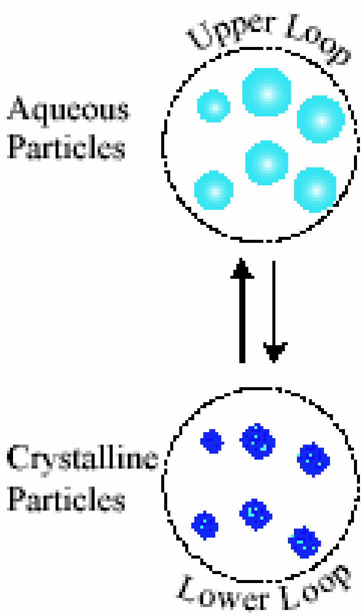
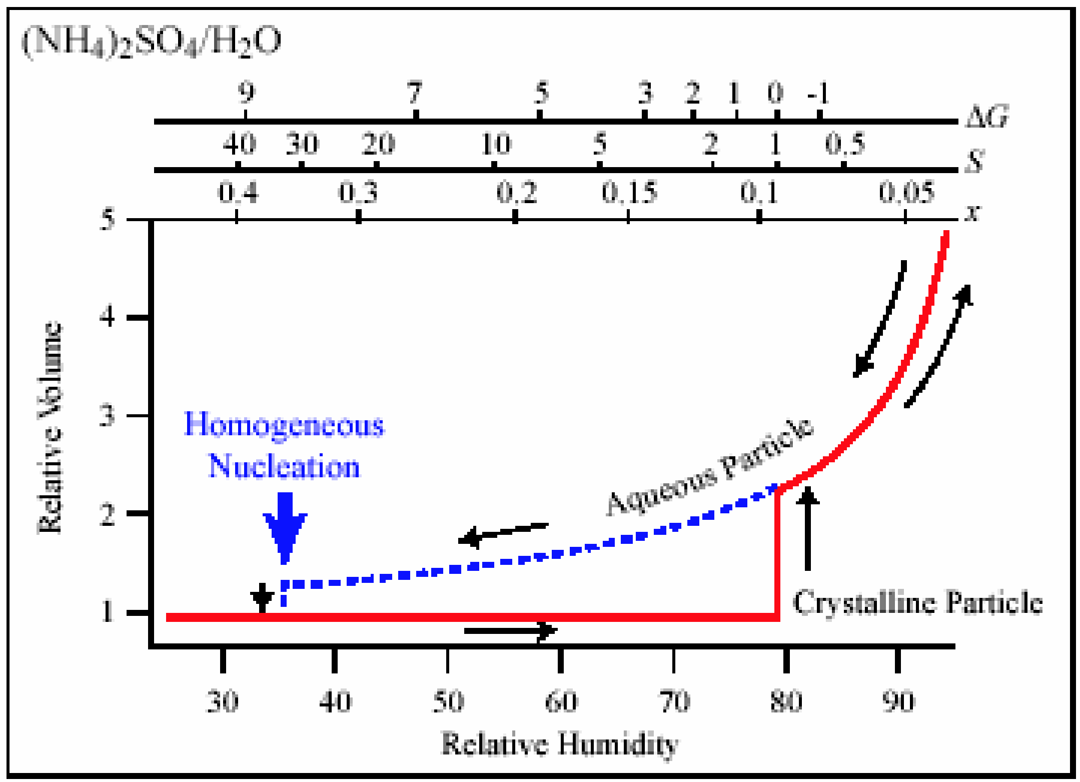
Hysteresis Effect

Particle phase depends on its relative humidity history







CRH=35% DRH=80%

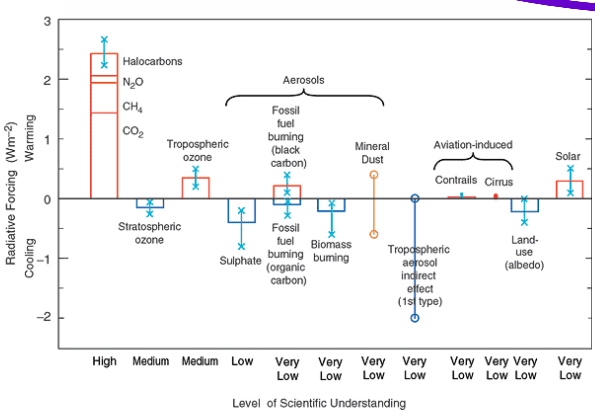
Atmospheric Implications

Particle phase affects radiative forcing

Full Sky TOA Forcing (W m^{-2}) Including Clouds

	F_L	F_U	$\%\Delta F_{U,L}$
DJF	-0.433	-0.501	16
MAM	-0.510	-0.632	24
JJA	-0.587	-0.764	30
SON	-0.408	-0.522	28
Annual	**-0.485**	**-0.605**	**25**

Martin et al., 2004



→ Different effect according to the phase

→ Significant Cooling Effect

Objectives

• Determination of the phase : aqueous versus crystalline

Evaluate the effects of particle phase using GEOS- CHEM in conjunction with lab data

• Provide prognostic determination at monthly, seasonal and annual periods at the global scale

• Particles phase , including the hysteresis effect is treated prognostically

Predictions of aerosols particle phase require:

1) DRH
2) CRH } → Lab Data
3) RH history → GEOS-CHEM

Parametrization of the phase transition

Step 1: Ammonium Sulfate Scenario

- All particules are ammonium sulfate: 35%-80%

- 2 tracers : crystaline ammonium sulfate (AS) and aqueous ammonium sulfate (ASO4)

- All AS is sulfate

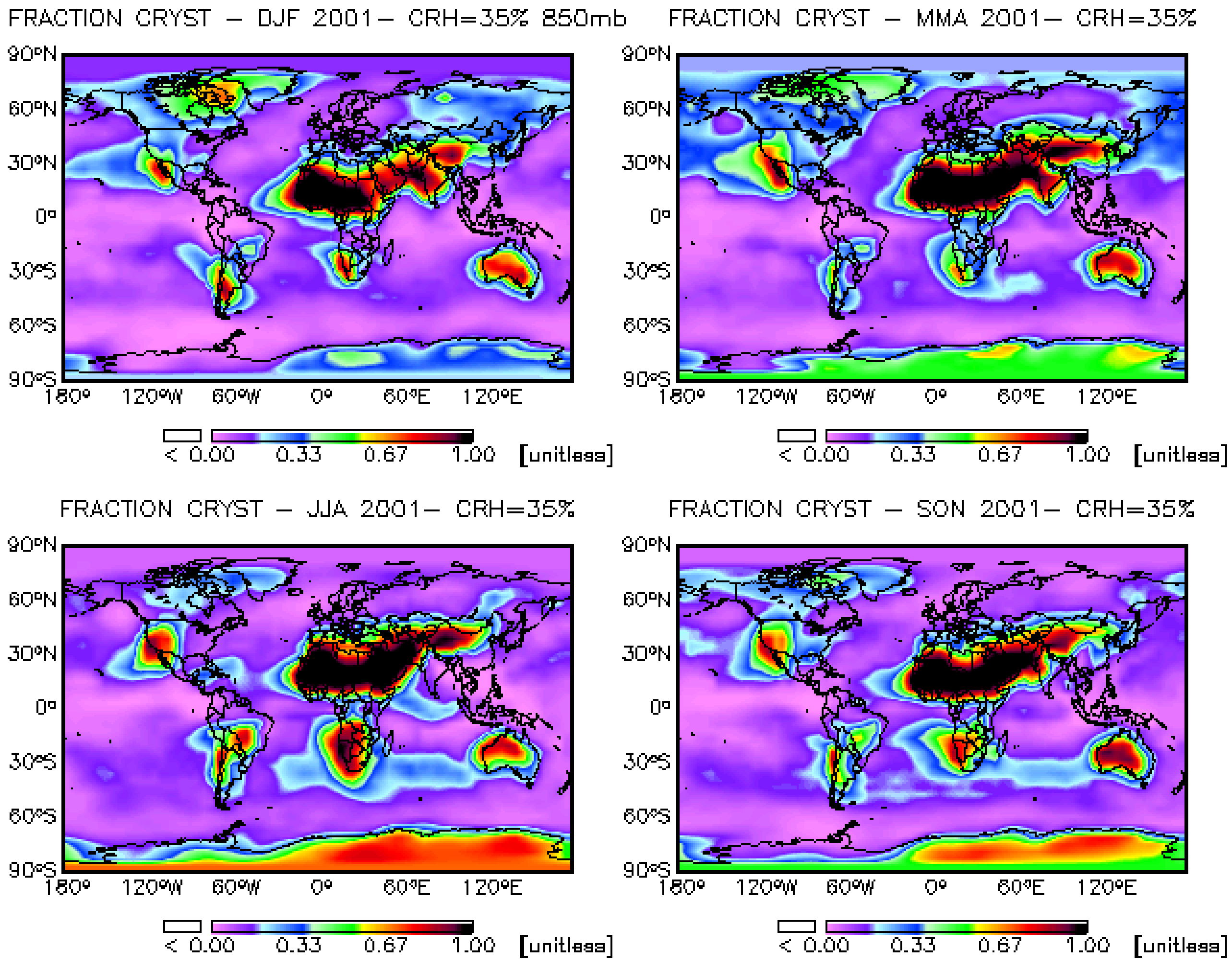
- Chemistry is a source only for the aqueous phase

- Emissions only for crystaline phase

- Same dry and wet removal for both phases

Crystalline Sulfate Fraction

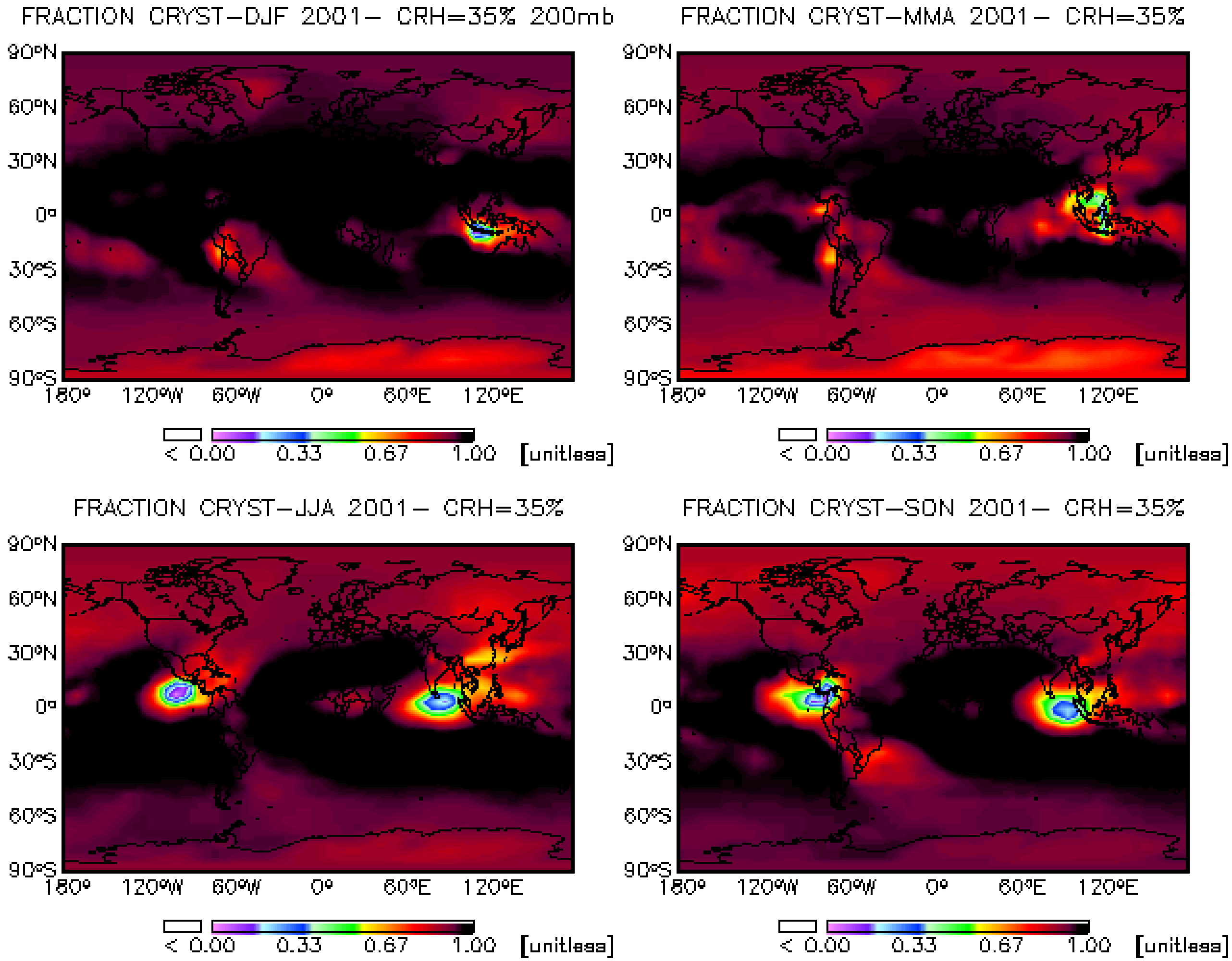
Seasonal Average in the lower troposphere ~ 850mb



Ammonium Sulfate Scenario

Crystalline Sulfate Fraction

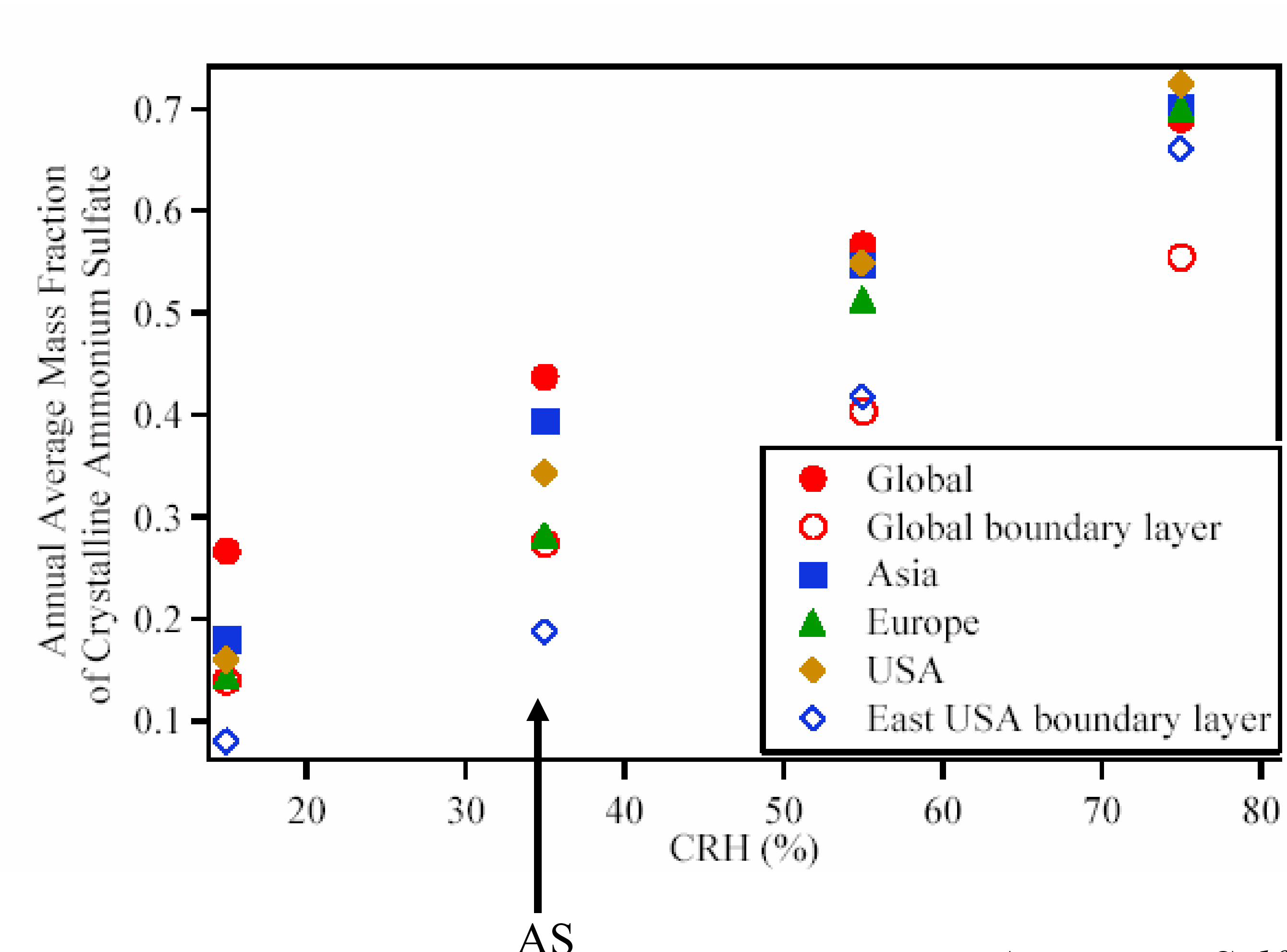
Seasonal Average in the upper troposphere ~ 200mb



Ammonium Sulfate Scenario

Impact of CRH value on Crystalline Fraction of Ammonium Sulfate

Annual Tropospheric Column Average shown for different regions



Ammonium Sulfate Scenario

Conclusion on Ammonium Sulfate Scenario

Worl only composed of AS

GLOBALLY

• 42 % of crystaline tropospheric fraction

• 25 % of crystalline fraction in the BL

REGIONALLY

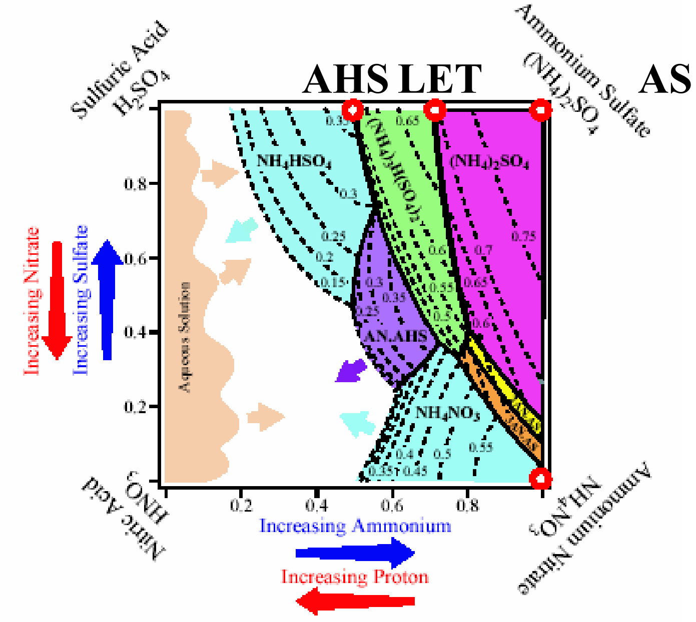
• 40 % of Asian crystalline tropospheric fraction

• 35 % of US crystalline tropospheric fraction

• 18 % of East US BL crystalline tropospheric fraction

Parametrization of the phase transition

Step 2: Effect of the acidity on the phase partitioning



•4 tracers : ammonium sulfate (AS), ammonium bisulfate (AHS), letovicite (LET) and aqueous sulfate (ASO4)

• DRH from lab data

•CRH polynomial rules (Martin et al., 2003)

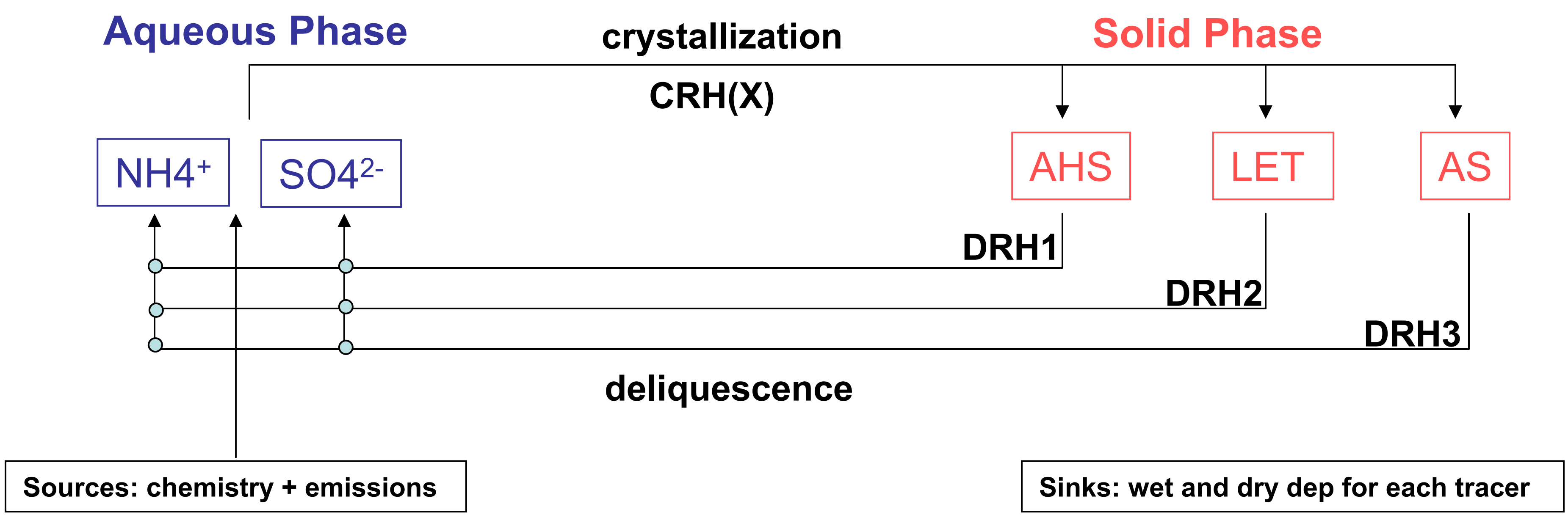
•Chemistry is a source only for the aqueous phase

•Emissions only for aqueous phase

•Same dry and wet removal for each phases

Parametrization of the phase transition

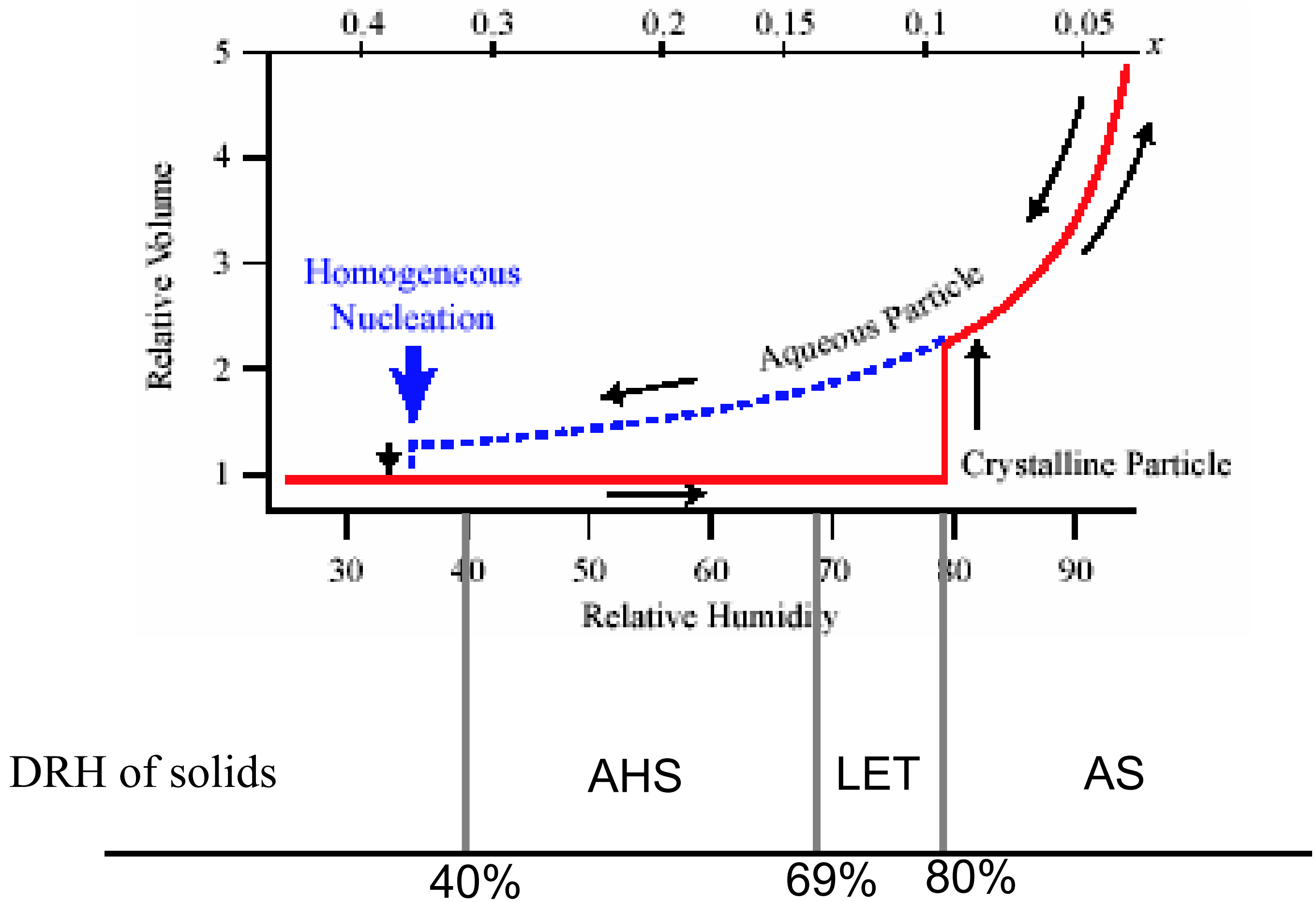
Step 2: Effect of the acidity on the phase partitioning



Deliquescence:

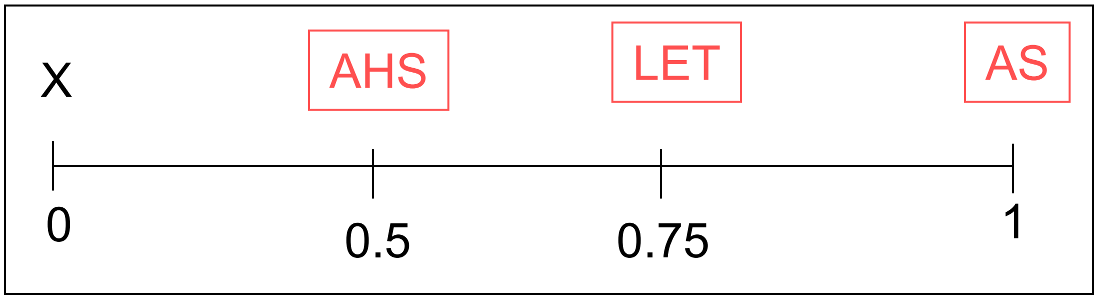
External mixing in solid phases

Deliquescence is total according to the DRH lab values



Crystallization

Ratio of neutralization $$X = \frac{NH_4^+}{2*SO_4^{2-}}$$



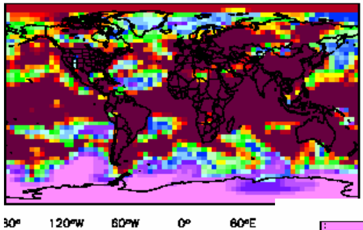
Internal mixing in aqueous phase
Total crystallization following the polynomial rules

Turn off the titration by nitrate CRH(X,Y) with Y=0 -> CRH(X)

CRH(X) = 0	for	$X < 0.5$	no crystallization
CRH(X)=CRH	for	$0.5 < X < 0.75$	AHS and LET can be formed
CRH(X)=CRH	for	$0.75 < X < 1$	LET and AS can be formed

Assumptions

Martin et al., 2004	This work
• Internal mixing • Phase transition off line • Nitrate	• External mixing • Phase transition on line • Transport • No Nitrate
↓	↓
•impossible for AS to co-exist with acidified aqueous solution the same grid box • AS does not form from acidified aqueous solution • More acidity, less AS	• possible co-existence of AS with acidified aqueous solution the same grid box • solids transported + formation of solids from acidified aqueous solution •Less acidity, more AS

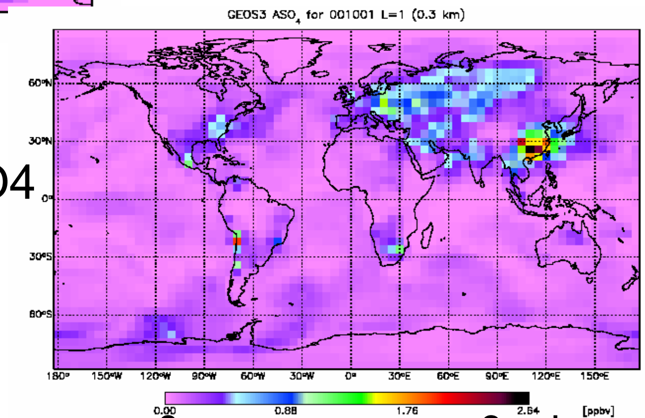


Mixing Ratios at the Surface

- October 2000

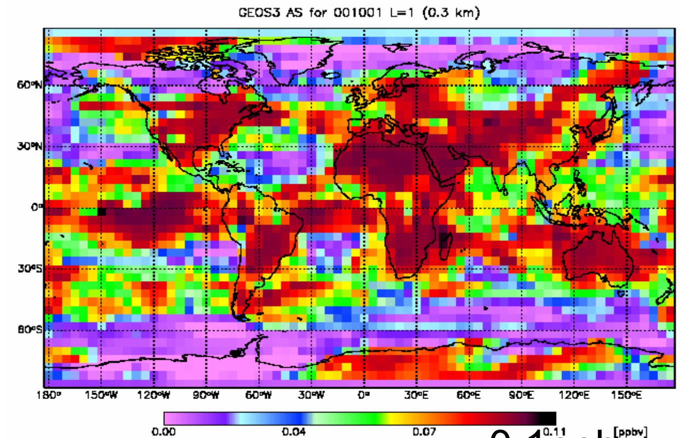
Formation of solids regulated by composition, RH, T

ASO4



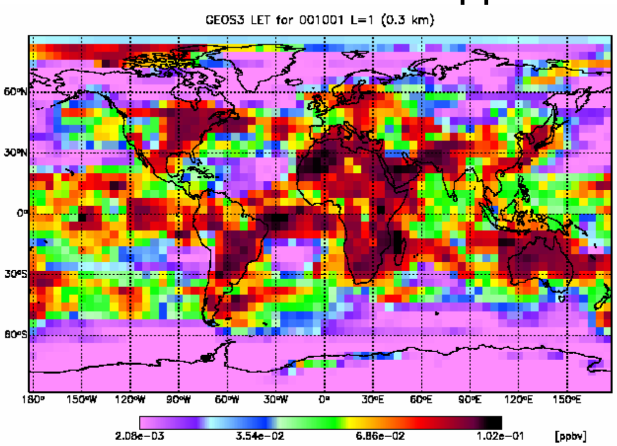
0 3ppbv

AS

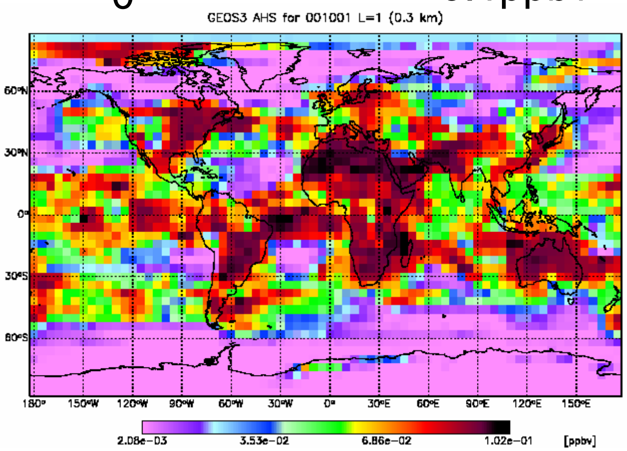


0 0.1ppbv

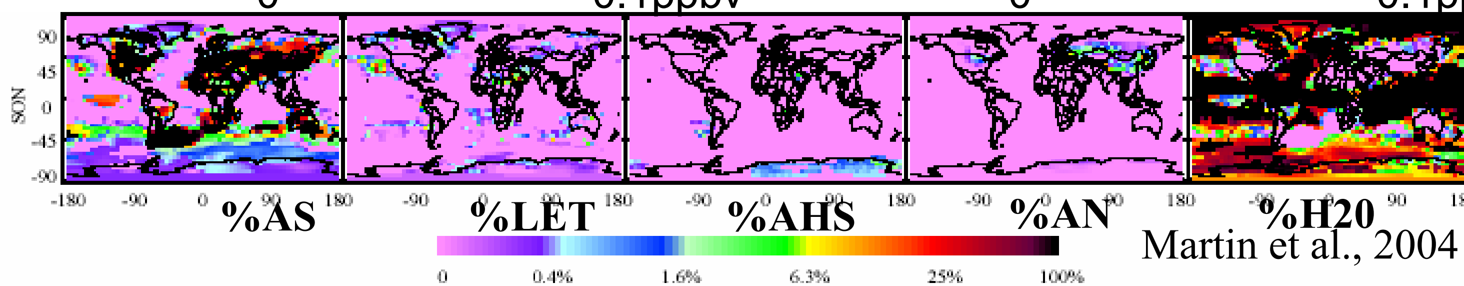
LET



AHS



0 0.1ppbv 0 0.1ppbv

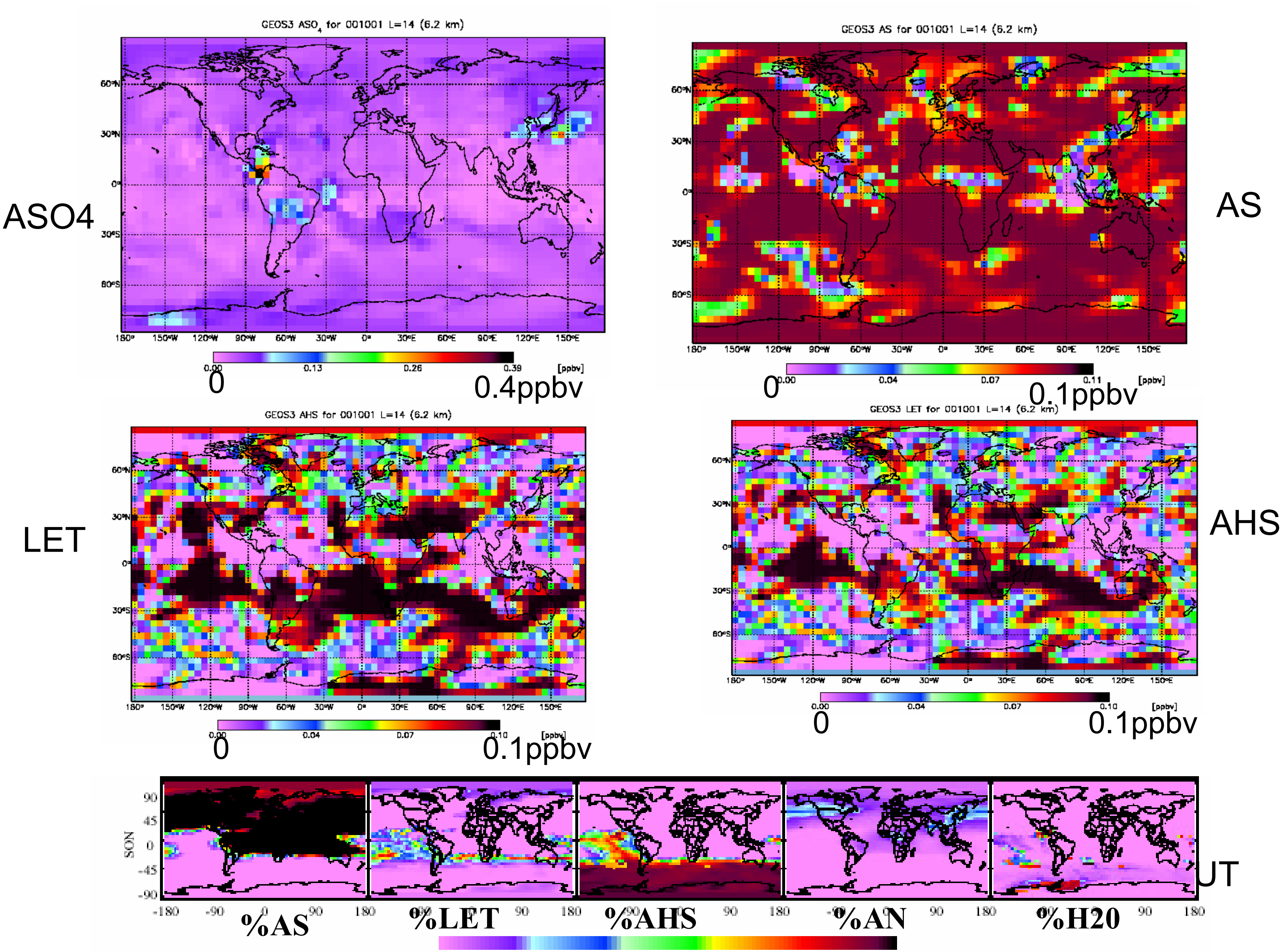


BL

Martin et al., 2004

Mixing Ratios at 500 hPa

- October 2000-



Martin et al., 2004

Future work

Phase transition + transport + effect of the acidity =

improvment in the realism of the phase transition in the model

•Use of a simplified thermodynamical model

(updated RP_MARES/ ISORROPIA/ own code?)

• Magnitude of the each phase on the radiative effect

•To implement nitrate phase transition