MPI for Chemistry

The impact of a new Gas/Aerosol Chemistry module of MESSy/ECHAM5 on the hydrological cycle and the formation of clouds

Sensitivity model studies and model output evaluation using satellite data

Swen Metzger and Rüdiger Lang

EQSAM/M7 sensitivity studies and model evaluation

MPI for Chemistry

Current studies at the MPI-CHEM, Mainz:

Sensitivity studies using the Gas/Aerosol partitioning equilibrium model EQSAM (by Swen Metzger) coupled to GM7 and implemented in MESSy/ECHAM5

Goal

Evaluating the impact of modeled aerosol water content, CN activation, and CCN/ICN numbers on the evolution of clouds and the hydrological cycle in MESSy/ECHAM5 using satellite data (GOME water vapor and cloud statistics).

ECHAM5 Mz Version

Modular **E**arth **S**ubmodel **Sy**stem (MESSy)

>> resolution independent, e.g. 0D, 1D, 3D, BL, Tropos, Middle Atmosphere <<

MPI for Chemistry

http://www.messy-interface.org/

Gas-phase Chemistry (MECCA)

based on
ECHAM4-CHEM (Benedikt Steil) and
MATCH-NMHC8 (Rolf von Kuhlmann)
gas phase and heterogenous chemistry,
using the numerical integration package KPP
(Rolf Sander)

Lightning NOx

Price and Rind scaling
production [Tg/y]
(Patrick Jöckel)

MBL Chemistry

switchable extension to MECCA
with focus on halogens
(Roland von Glasow, Astrid Kerkweg)

Deposition

dry deposition of gases and aerosols based
on vegetation and land surface cover
(Laurens Ganzeveld)

Stratospheric Chemistry

polar stratospheric clouds (PSC),
micro-physics and sedimentation
(Joachim Buchholz, Stefanie Meilinger)

Cloud Scheme

Lohmann and Roeckner, 1996: Clim. Dyn. 557-572
Levkov et al., 1992: Beitr. Phys. Atm. 35-58. (ice phase)
Beheng, 1994: Atmos. Res. 193-206. (warm phase)
Lenderink et al., 1998; KNMI-REPORT 98-13 (condensation)
Tompkins 2000, J. Atmos. Sci. Submitted (cloud cover)
update is planned ...

Gas /Aerosol Chemistry

fast thermodynamical aerosol
composition module (EQSAM) coupled
with the size-resolving dynamical module (M7)
(as part of EU-Project PHOENICS).
Hygroscopic growth is considered for major inorganic
(NH4, SO4, NO3, SS, Dust)
and organic (BC, OC, SOA, POM) compounds,
and based
on gas/liquid/solid partitioning.
CCN Activation is based on the aerosol water content,
ICN will be based on aerosol water and ionic composition.
(Swen Metzger)

Photolysis

based on the
fast on-line scheme [Landgraf and Crutzen, 1998]
to calculate photolysis rates and solar heating rates
(further developed by Christoph Brühl)

^{14}CO / Radon

natural atmospheric tracer, evaluation
of tropospheric OH and stratosphere
troposphere exchange (STE)
(Patrick Jöckel)

Different Transport Schemes

semi-lagrangian, spitfire,
lagrangian scheme (Attila,
implemented by Michael Traub)

Scavenging

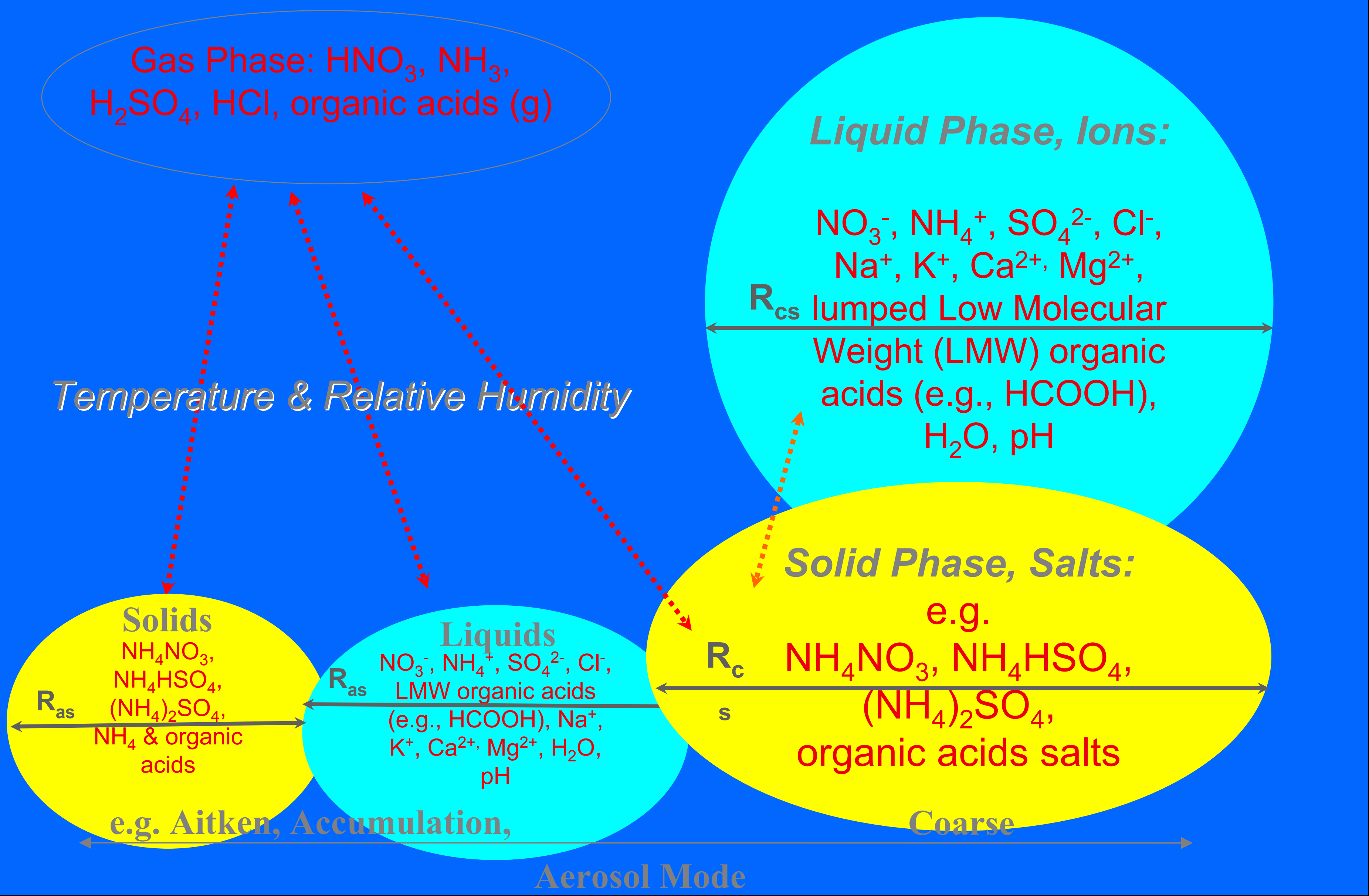
Below and in-cloud scavenging of gases and
aerosols
(Holger Tost, Laurens Ganzeveld)

Natural and Anthropogenic Emissions

biogenic surface emissions are calculated on-line (Laurens Ganzeveld)
anthropogenic emissions (EDGAR3.2) are calculated off-line and prescribed
(John van Aardenne)

Thermodynamical Aerosol Model, EQSAM: Gas/Liquid/Solid Partitioning

MAX-PLANCK-GESELLSCHAFT
MPI for Chemistry



Aerosol Dynamics and Equilibrium Thermodynamics

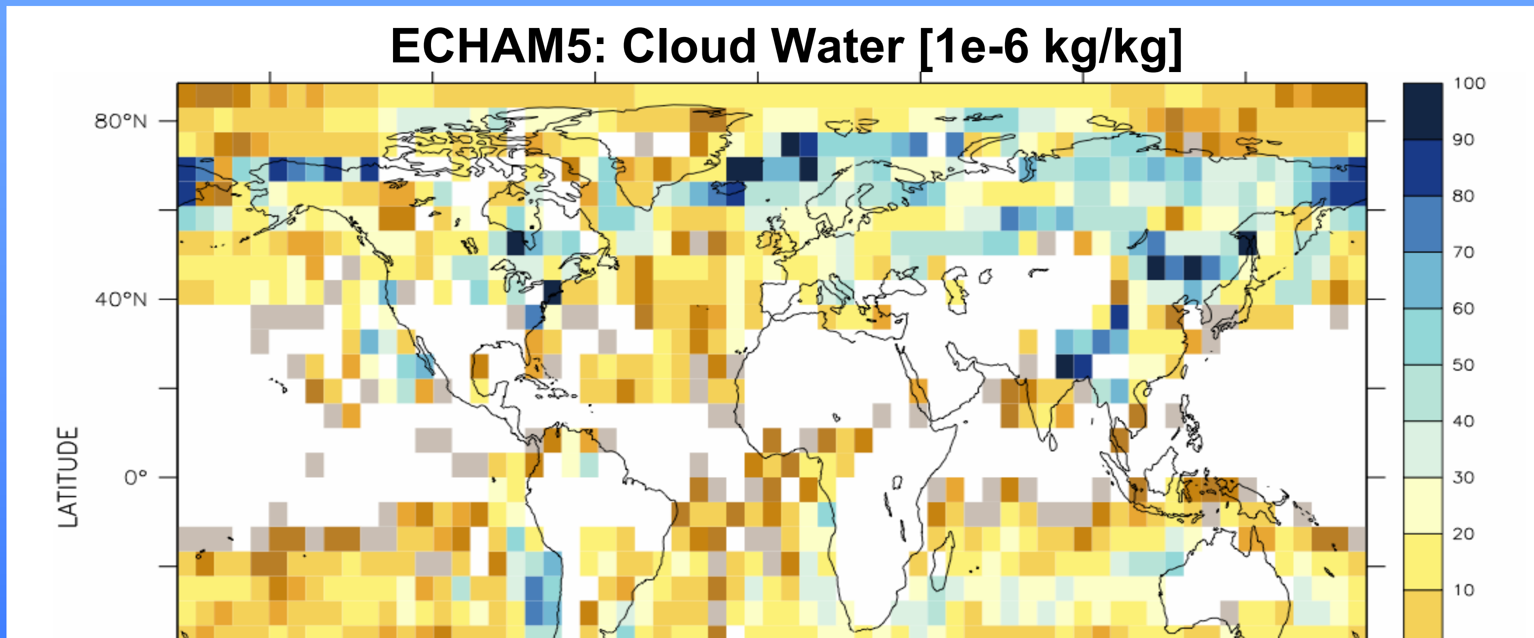
Mode			Domain		Domain dependent properties (output)
Nucleation	=>	Soluble	e.g. sulfate rich		
			sulfate neutral		
Aitken	=>		⋮	=>	aerosol composition, mass, number, radii, density, pH, optical properties, deposition parameters, supersaturation CN/CCN activation etc.
		Insoluble	BC-OC (primary)		
Accumulation	=>		DU3 (primary)		
Coarse	=>		⋮		

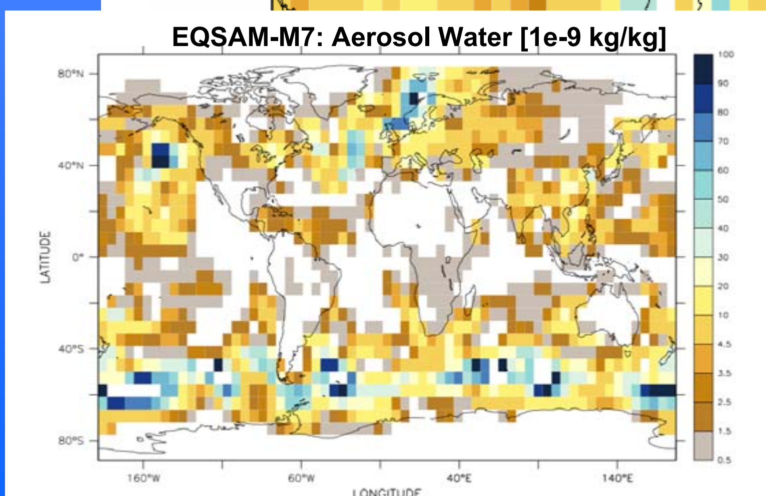
Note that the domain structure of EQSAM might be applied to all GM7 modes, e.g. to determine the coating of insoluble primary aerosols and the mass transfer to the soluble mode based on the condensation of aerosol precursor gases and the associated aerosol water mass.

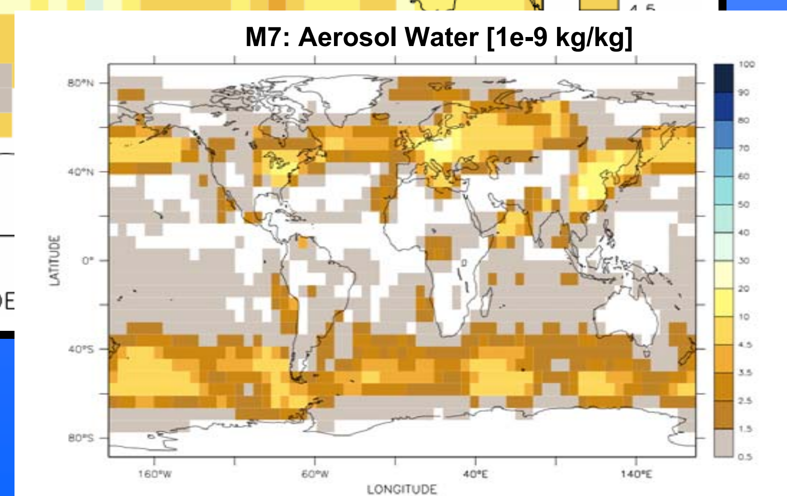
Coupled chemistry-GCM: Aerosol modeling

MPI for Chemistry

Qualitive comparison of cloud and aerosol water spatial distribution







MPI for Chemistry

Water Vapor

ECHAM-GOME

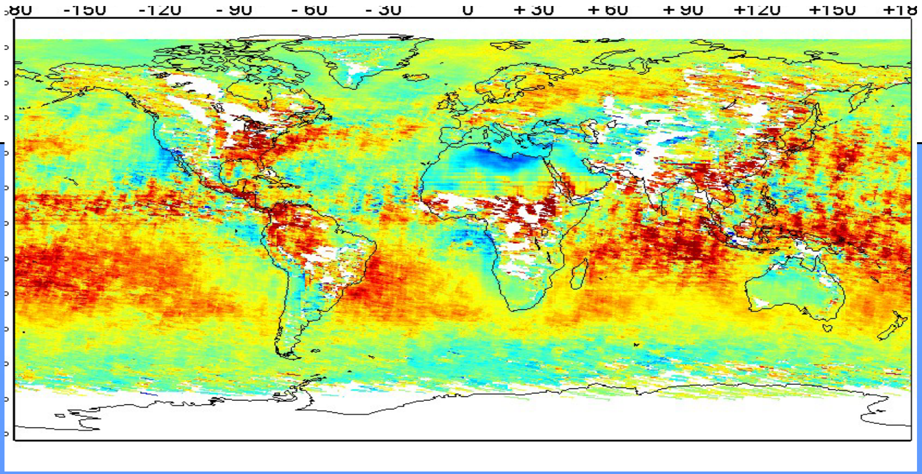
August 1998

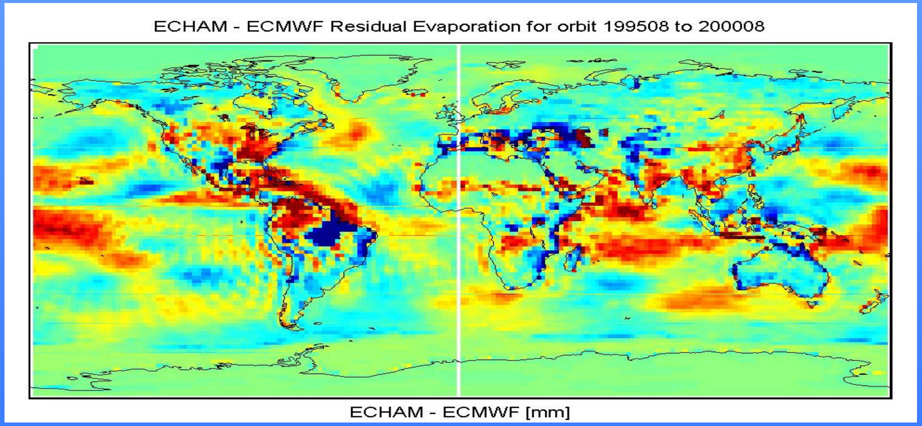
Evapo(transpi)ration

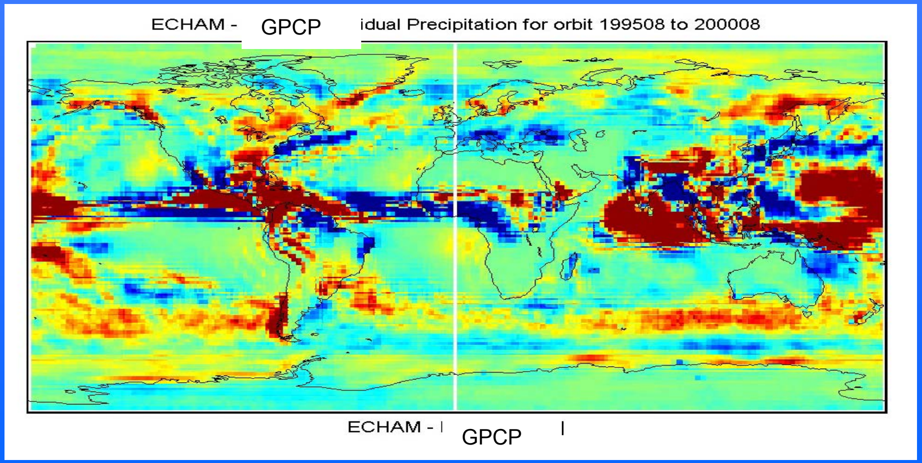
ECHAM-ECMWF

Precipitation

ECHAM-GPCP







Conclusion/Outlook

MPI for Chemistry

- There is a larger spatial variability in the global distribution of the aerosol water, if the actual aerosol composition is considered based on the gas/aerosol partitioning in addition to aerosol dynamics (EQSAM-M7).

- The spatial variability in the EQSAM-M7 aerosol water is qualitatively similar compared to the spatial variability in ECHAM5's cloud water, suggesting that the more detailed representation of the aerosol chemical composition in EQSAM-M7 will facilitate a direct link/coupling of the aerosol model to ECHAM5's cloud representation with respect to CCN activation.

- Linking CCN and ICN formation in MESSy-ECHAM5 by explicitly coupling the cloud- and aerosol water content based on the ionic composition that reflects the actual aerosol composition (incl. gas/liquid/solid aerosol partitioning).

- Evaluation of the ECHAM5 water/cloud fields, coupled to EQSAM-M7, with satellite observations (GOME) and radiosonde H_2O profiles.

For detailed questions/remarks on EQSAM: metzger@mpch-mainz.mpg.de

Summary ECHAM5.2.02 vs observations

global means, 1995-2000

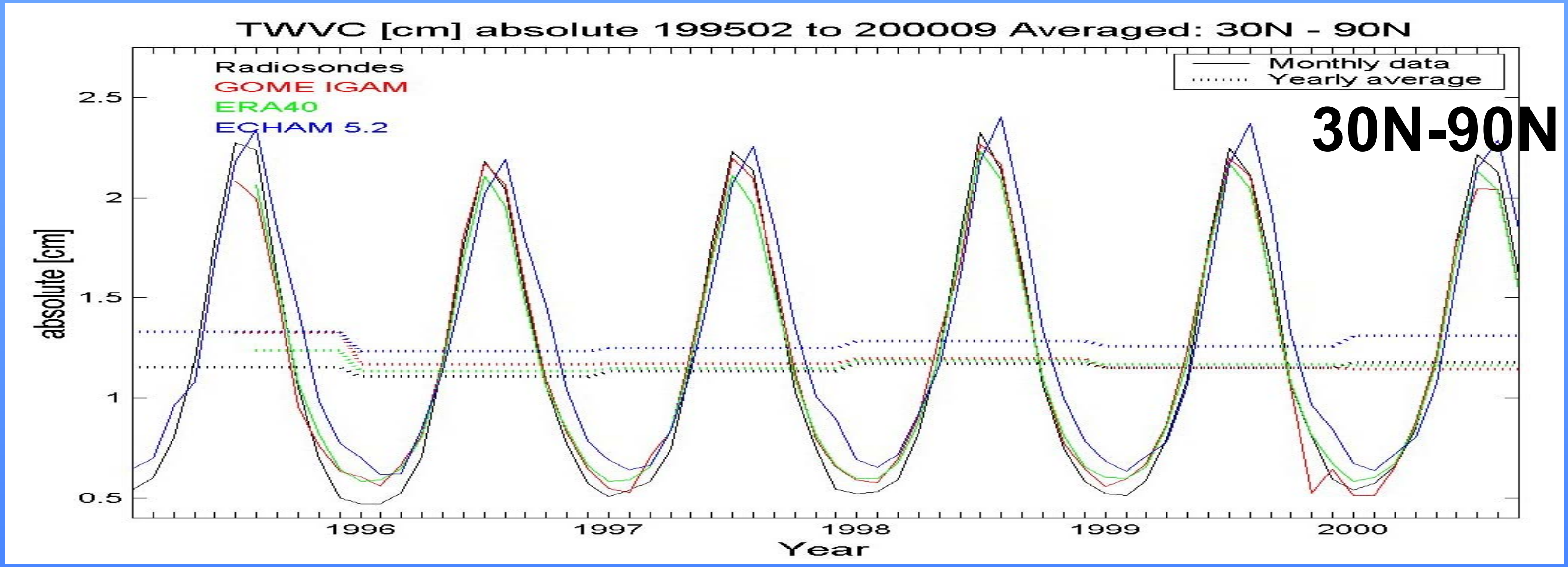
MAX-PLANCK-GESELLSCHAFT
MPI for Chemistry

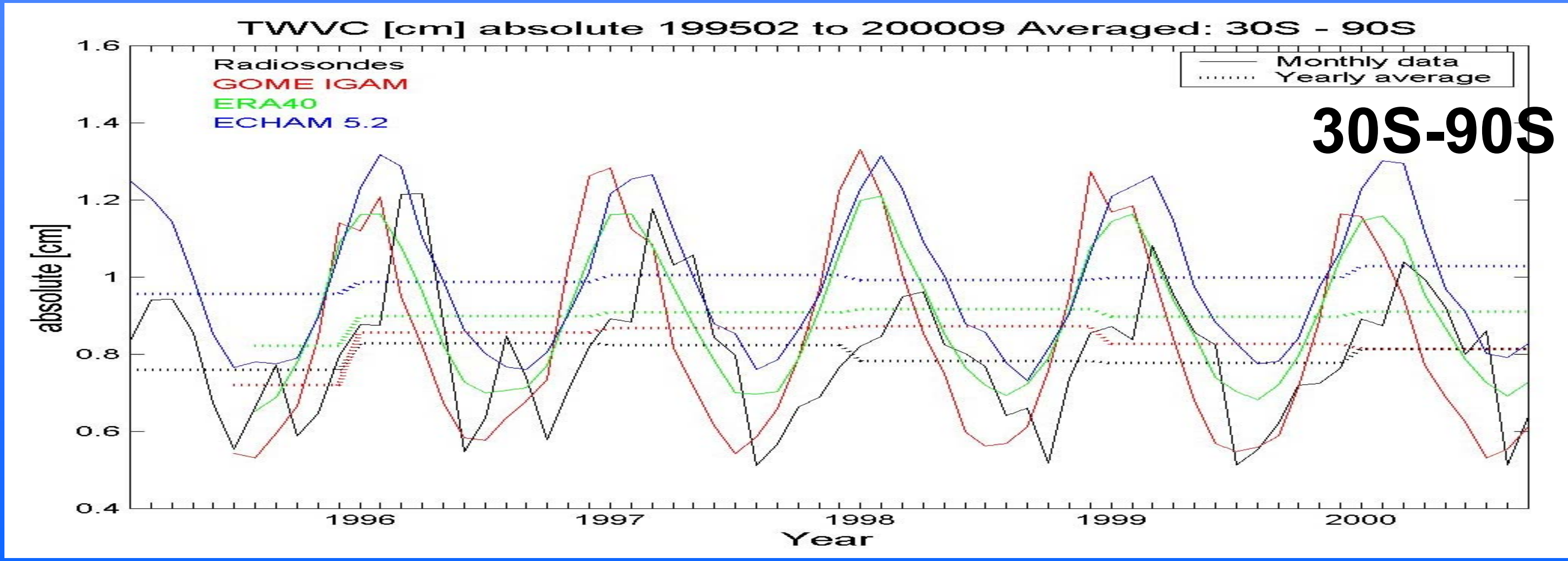
	GOME/GPCP/ECMWF	ECHAM	Abs. Diff	Rel. Diff [%]
Residence time [days]	7.49	8.8	1.31	14.89
E-P [mm]	-0.11	-0.34	-0.23	67.65

[mm]	GOME/GPCP/ECMWF	ECHAM	Abs. Diff	Rel. Diff [%]
Evaporation	2.64	2.74	0.1	3.65
Precipitation	2.75	3.08	0.33	10.71
Water Vapor	21.79	26.13	4.34	16.61

The GOME-IGAM/SSP WV data record

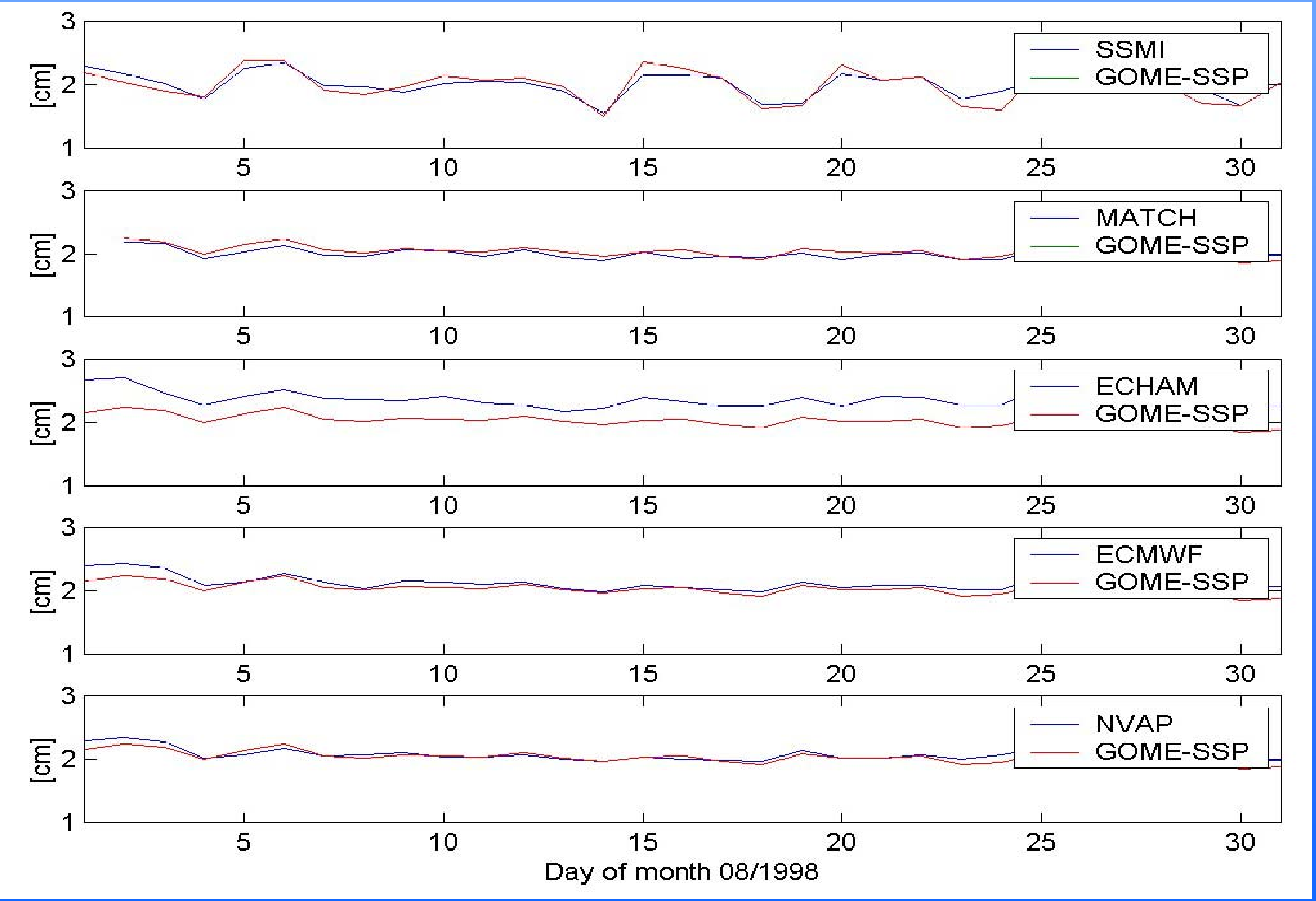
GOME-IGAM/SSP retrievals vs ECHAM5





GOME-SSP WV comparisons ECHAM5 and others with GOME-SSP (global daily means)

MPI for Chemistry



Conclusion/Outlook satellite observations

MAX-PLANCK-GESELLSCHAFT
MPI for Chemistry

- *ECHAM5.2 T63 L39, generally too moist with respect to observations (GOME, SSM/I)*
- the water vapor content is high with respect to GOME and SSMI
- the evaporation fluxes are high with respect to ECMWF
- the precipitation rate is high with respect to GPCP
- the WV residence time is increased by ECHAM by 15% w.r.t. the observations

⇘ the strong precipitation is not compensating for the high evaporation fluxes. More WV is stored by ECHAM than observations suggest.

- *ECHAM5.2 T63 L39, WV field sensitivity to SST forcing is estimated to be +-1cm locally*

References

1. Metzger, S. M., Gas/Aerosol Partitioning: *A simplified Method for Global Modeling*, Ph.D. Thesis, University Utrecht, The Netherlands, 2000. **http://www.library.uu.nl/digiarchief/dip/diss/1930853/inhoud.htm**

2. Metzger, S. M., F. J. Dentener, J. Lelieveld, and S. N. Pandis, *Gas/aerosol Partitioning I: A Computationally Efficient Model.* J Geophys. Res., 107, D16, 10.1029/2001JD001102, 2002. **http://www.agu.org/journals/jd/jd0216/2001JD001102/index.html**

3. Metzger, S. M., F. J. Dentener, A. Jeuken, and M. Krol, J. Lelieveld, *Gas/aerosol Partitioning II: Global Modeling Results.* J Geophys. Res., 107, D16, 10.1029/2001JD001103, 2002. **http://www.agu.org/journals/jd/jd0216/2001JD001103/index.html**

4. Metzger, S. M., *Gas/aerosol partitioning III: Model development (EQSAM) and comparison (MINOS Data)*, in preparation.
   The new version of EQSAM has been successfully applied within the EMEP modelling framework. Results are included in the EMEP reports, **http://www.emep.int/common_publications.html**, 2003.

5. Trebs, I., S. Metzger, F. X. Meixner, G. Helas, A. Hoffer, M. O. Andreae, M. A.L. Moura, R. S. da Silva (Jr.), J. Slanina, Y. Rudich, A. Falkovich, P. Artaxo, *The* NH^{4+}-NO^{3-}-Cl^{-}-SO^{42-}-H_2O *system and its gas phase precursors at a rural site in the Amazon Basin: How relevant are crustal species and soluble organic compounds?,* submitted to JGR.

MPI-CHEM Radiosonde climatology

(preliminary results!)

15 January 2003 LANZANTE ET AL.

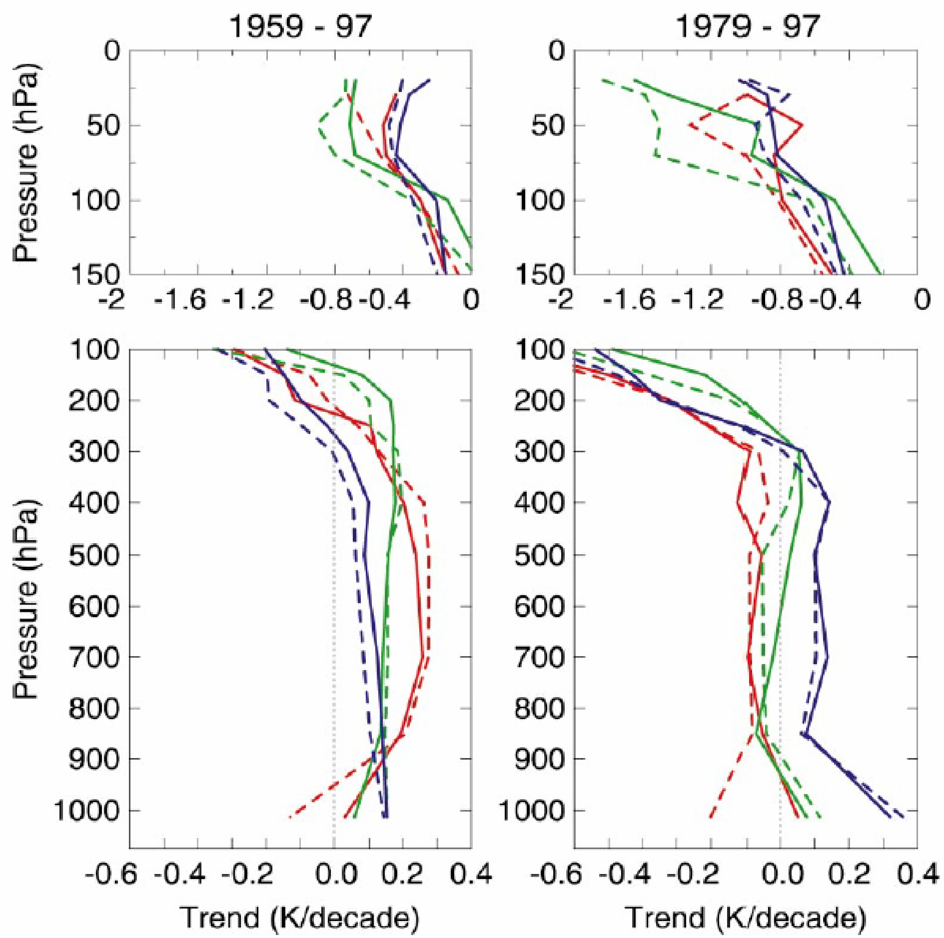
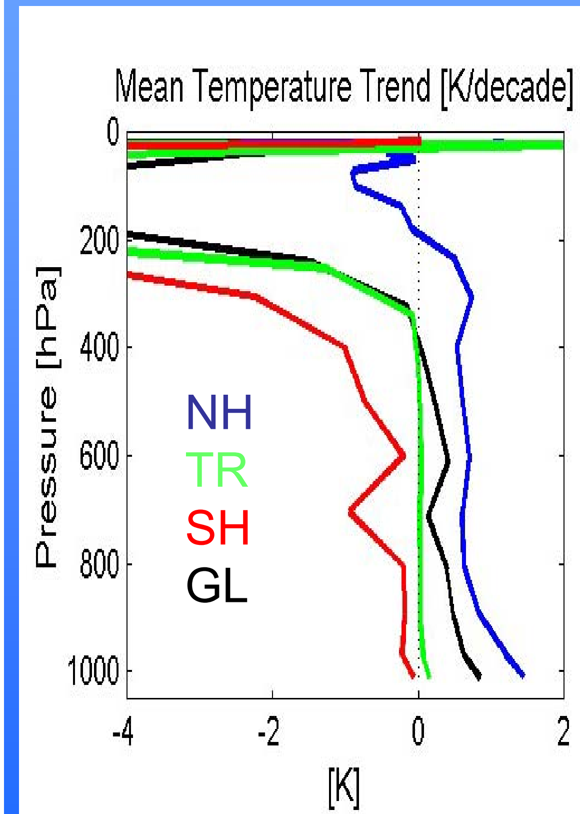


Fig. 4. Temperature trend (K decade^{-1}) as a function of pressure level (hPa) (left) for 1959–97 and (right) for 1979–97. Because of the different ranges, trend profiles have been plotted separately for (top) stratospheric and (bottom) tropospheric levels, with different increments on the abscissa. Trend is median over all stations in a particular latitude zone: NH (30°–90°N, blue), TRPC (30°N–30°S, green), and SH (30°–90°S, red). Each zone has curves based on unadjusted (UNADJ, dashed) and adjusted (LIBCON, solid) data.

Lazante et al., J. Climate, 2003

MPI-CHEM Radiosonde dataset 1980-2004

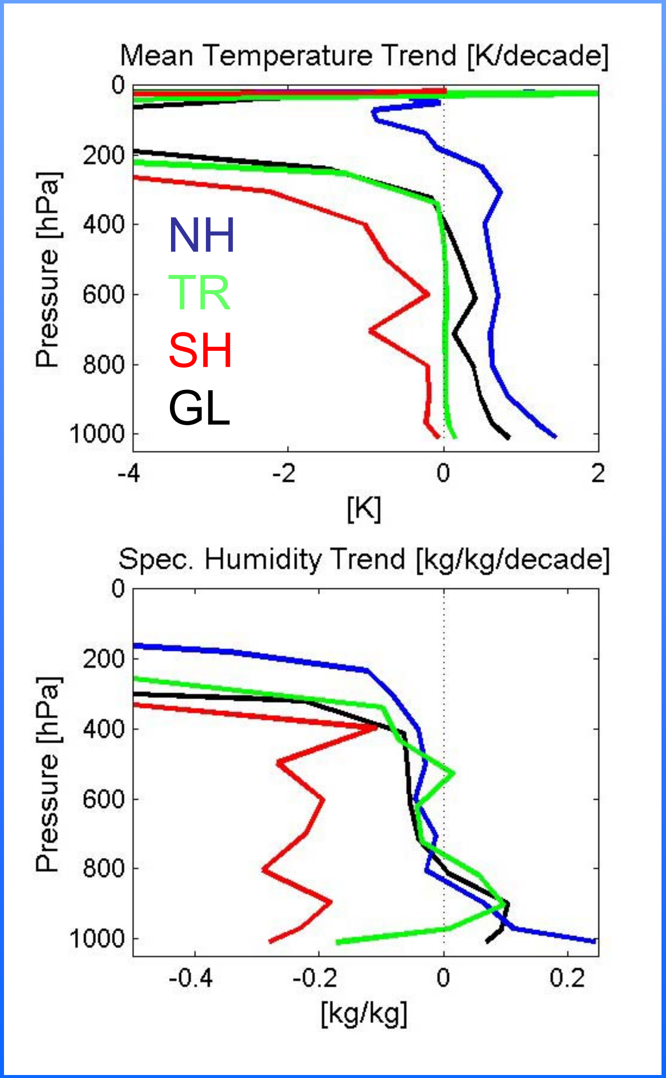


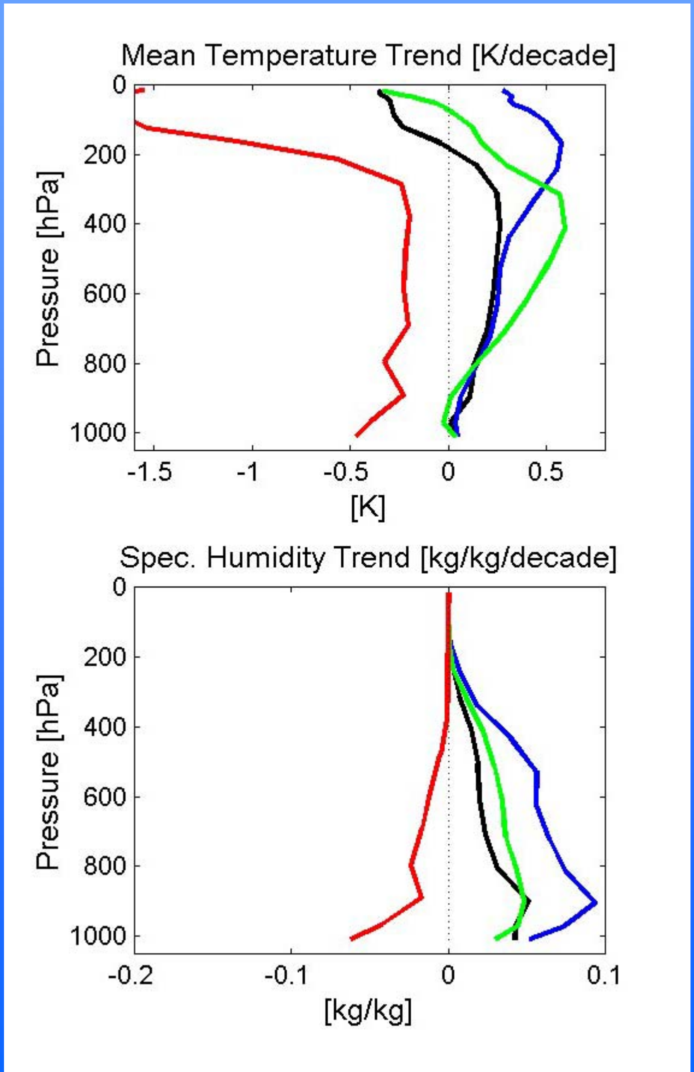
MPI-CHEM Radiosonde climatology

(preliminary results!)

MPI for Chemistry

Radiosondes 1980-2000 ECHAM 5





Coupled chemistry-GCM: Aerosol modeling

MPI for Chemistry

New M7/**EQSAM** Structure

Gasphase: H_2SO_4, **NH_3**, **HNO_3**, **SOA1**, **SOA2**, **HCl**

G: 6
N: 7
M: 34
total: 47

Nucleation

1.
1. N
2. $H2SO_4$

Aitken

2.
1. N
2. BC-OA1a
3. BC-OA2a
4. NO_3 5. NH_4 6. SO_4
7. SOA1 8. SOA2

5.
1. N
2. BC-OA1 (primary)
3. BC-OA2 (primary)

As an example:

Accumulation

3.
1. N
2. SO4
3. BC => **BC-OA1a, BC-OA2a**
4. OC => **SOA1 8. SOA2**
5. SS => **SS-Na 12.SS-Cl**
6. DU => **DU1, DU2**
7./8./9. **NO_3, NH_4, H_2O**

6.
1. N
2. DU3 (solid Si-core)

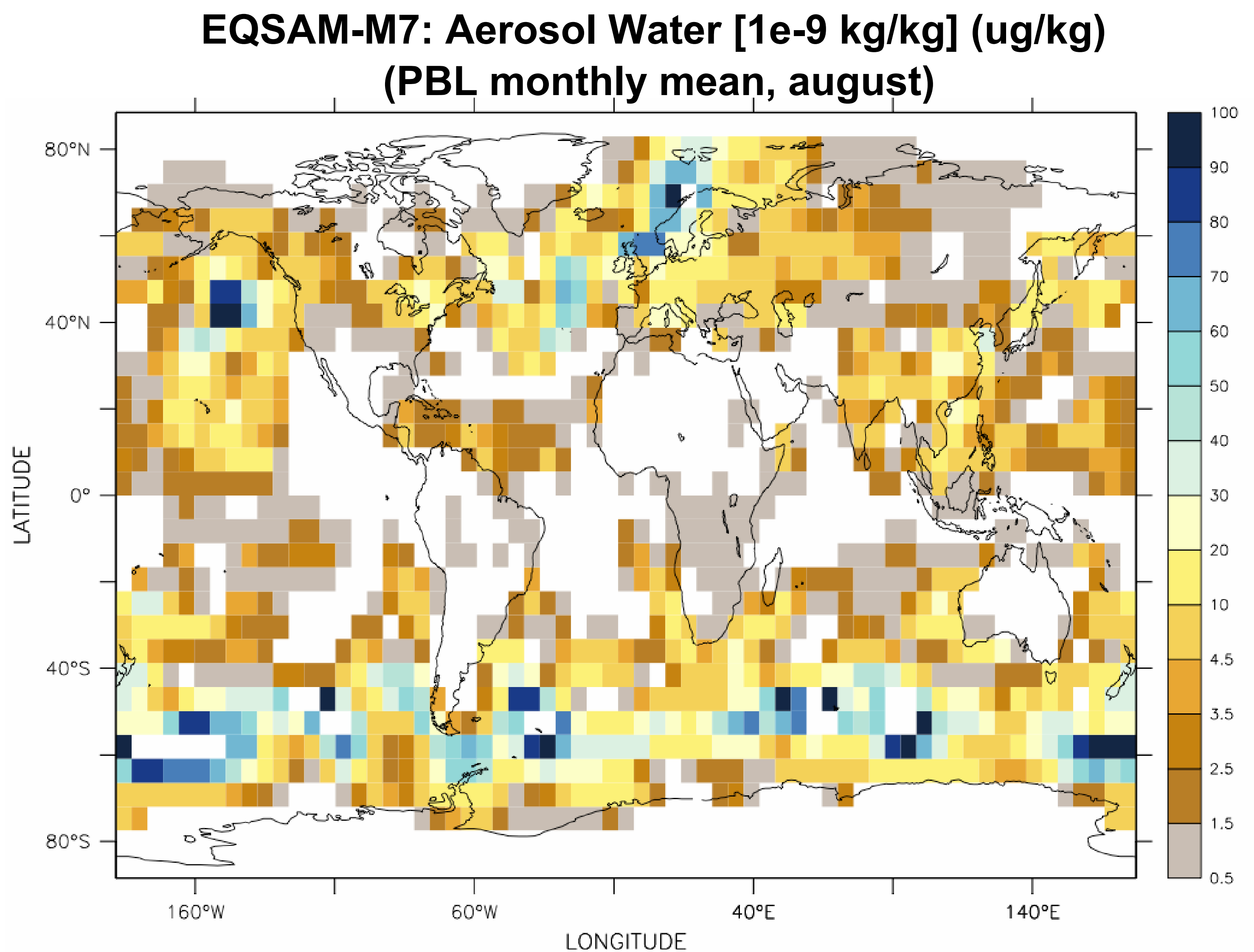
Coarse

4.
1. N
2. BC-OA1a
3. BC-OA2a
4. NO_3 5. NH4 6. SO_4
7. SOA1 8. SOA2
9. DU1 10.DU2 11. SS-Na 12.SS-Cl

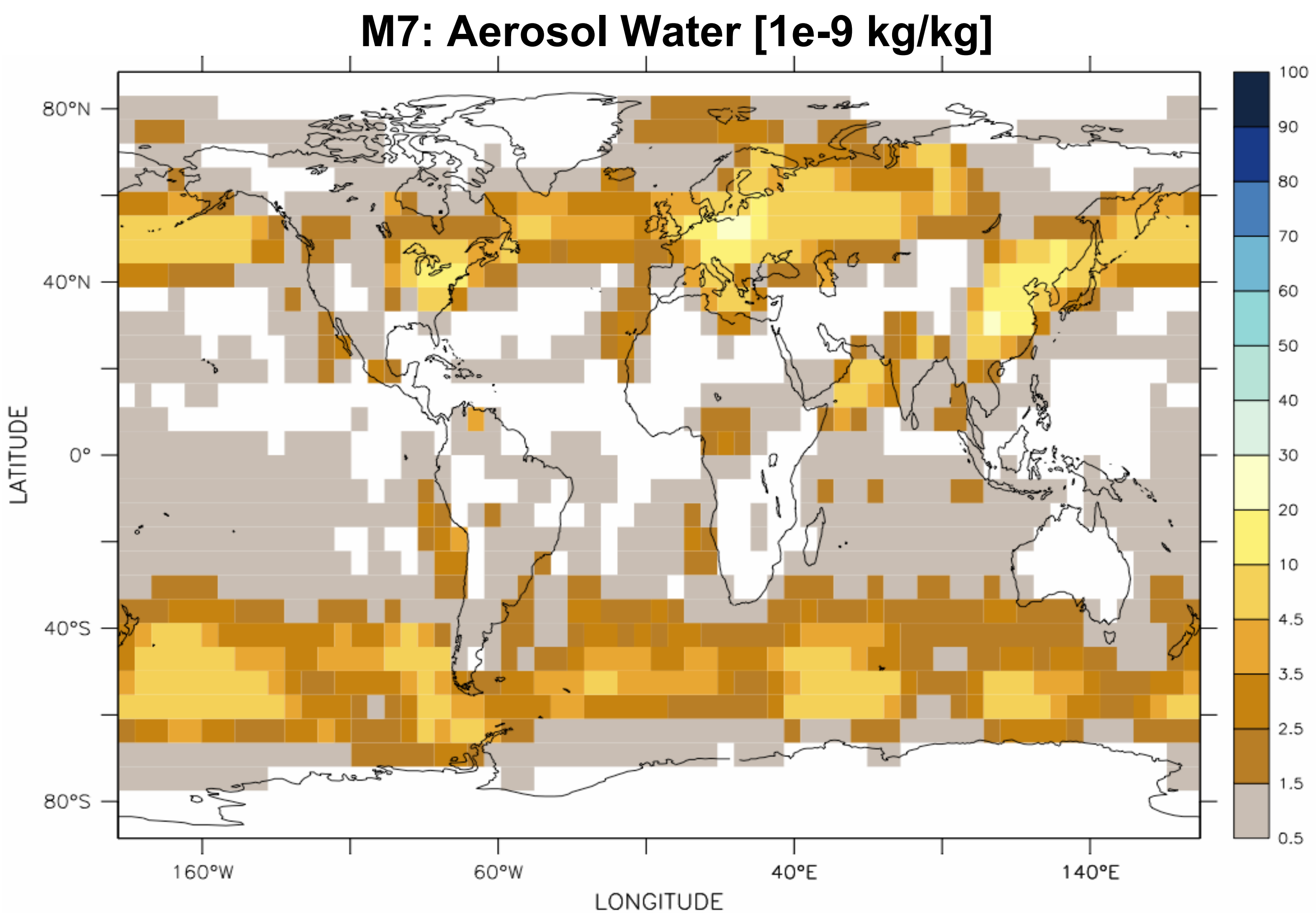
7.
1. N
2. DU3 (solid Si-core)

soluble (liquid/solid) <=> insoluble (solid)

Coupled chemistry-GCM: Aerosol modeling



Coupled chemistry-GCM: Aerosol modeling



Equilibrium Constants

MPI for Chemistry

Equilibrium relation	Constant expression	Equilibrium constant[a] K (298.15)	a	b	Units
NH_3 (g) = NH_3 (aq)	$\frac{[NH_3(aq)]\ \gamma_{NH_3}}{p_{NH_3}}$	57.639	13.79	- 5.39	mol / kg atm
HNO_3 (g) = H^+ (aq) + NO_3^- (aq)	$\frac{[H^+][NO_3^-]\ \gamma_{H^+}\ \gamma_{NO_3^-}}{p_{HNO_3}}$	2.511×10^{6}	29.17	16.83	mol^2 / kg^2 atm
HCl (g) = H^+ (aq) + Cl^- (aq)	$\frac{[H^+][Cl^-]\ \gamma_{H^+}\ \gamma_{Cl^-}}{p_{HCl}}$	1.971×10^{6}	30.20	19.91	mol^2 / kg^2 atm
H_2O (aq) = H^+ (aq) + OH^- (aq)	$\frac{[H^+][OH^-]\ \gamma_{H^+}\ \gamma_{OH^-}}{a_w}$	1.010×10^{-14}	-22.52	26.92	mol^2 / kg^2
NH_3 (aq) + H_2O (aq) = NH_4^+ (aq) + OH^- (aq)	$\frac{[NH_4^+][OH^-]\ \gamma_{NH_4^+}\ \gamma_{OH^-}}{[NH_3(aq)]\ \gamma_{NH_3} a_w}$	1.805×10^{-5}	-1.50	26.92	mol / kg
HSO_4^- (aq) = H^+ (aq) + SO_4^{2-} (aq)	$\frac{[H^+][SO_4^{2-}]\ \gamma_{H^+}\gamma_{SO_4^{2-}}}{[HSO_4^-]\ \gamma_{HSO_4^-}}$	1.015×10^{-2}	8.85	25.14	mol / kg
NH_4Cl (s) = NH_3 (g) + HCl (g)	$p_{NH_3}\ p_{HCl}$	1.086×10^{-16}	-71.00	2.40	atm^2
NH_4NO_3 (s) = NH_3 (g) + HNO_3 (g)	$p_{NH_3} p_{HNO_3}$	5.746×10^{-17}	-74.38	6.12	atm^2
$(NH_4)_2SO_4$ (s) = 2 NH_4^+ (aq) + SO_4^{2-} (aq)	$[NH_4^+]^2[SO_4^{2-}]\ \gamma^2_{NH_4^+}\ \gamma_{SO_4^{2-}}$	1.817	-2.65	38.57	mol^3 / kg^3
NH_4HSO_4(s) = NH_4^+(aq)+HSO_4^-(aq)	$[NH_4^+][HSO_4^-]\ \gamma_{NH_4^+}\ \gamma_{HSO_4^-}$	1.383×10^{4}	-2.87	15.83	mol^2 / kg^2
$(NH_4)_3H(SO_4)_2$(s) = 3 NH_4^+(aq) + HSO_4^-(aq) + SO_4^{2-}(aq)	$[NH_4^+]^3[HSO_4^-][SO_4^{2-}]\ \gamma^3_{NH_4^+}\ \gamma_{HSO_4^-}\gamma_{SO_4^{2-}}$	29.72	-5.19	54.40	mol^5 / kg^5
NaCl (s) = Na^+ (aq) + Cl^- (aq)	$[Na^+][Cl^-]\ \gamma_{Na^+}\ \gamma_{Cl^-}$	37.661	-1.56	16.90	mol^2 / kg^2
$NaNO_3$ (s) = Na^+ (aq) + NO_3^- (aq)	$[Na^+][NO_3^-]\ \gamma_{Na^+}\ \gamma_{NO_3^-}$	11.971	-8.22	16.01	mol^2 / kg^2
Na_2SO_4 (s) = Na^+ (aq) + SO_4^{2-} (aq)	$[Na^+]^2[SO_4^{2-}]\ \gamma^2_{Na^+}\ \gamma_{SO_4^{2-}}$	4.799×10^{-1}	0.98	39.75	mol^3 / kg^3
$NaHSO_4$(s) = Na^+(aq) + HSO_4^-(aq)	$[Na^+][HSO_4^-]\ \gamma_{Na^+}\ \gamma_{HSO_4^-}$	2.413×10^{4}	0.79	14.746	mol^2 / kg^2
KCl (s) = K^+ (aq) + Cl^- (aq)	$[K^+][Cl^-]\ \gamma_{K^+}\ \gamma_{Cl^-}$	8.680	-6.902	19.95	mol^2 / kg^2
KNO_3 (s) = K^+ (aq) + NO_3^- (aq)	$[K^+][NO_3^-]\ \gamma_{K^+}\ \gamma_{NO_3^-}$	0.872	-14.08	19.39	mol^2 / kg^2
K_2SO_4 (s) = 2K^+ (aq) + SO_4^{2-} (aq)	$[K^+]^2[SO_4^{2-}]\ \gamma^2_{K^+}\ \gamma_{SO_4^{2-}}$	1.569×10^{-2}	-9.585	45.81	mol^3 / kg^3
$KHSO_4$(s) = K^+(aq) + HSO_4^-(aq)	$[K^+][HSO_4^-]\ \gamma_{K^+}\ \gamma_{HSO_4^-}$	24.016	-8.423	17.96	mol^2 / kg^2
$CaCl_2$ (s) = Ca^{2+} (aq) + 2Cl^- (aq)	$[Ca^{2+}][Cl^-]^2\ \gamma_{Ca^{2+}}\ \gamma^2_{Cl^-}$	7.974×10^{11}	--	--	mol^3 / kg^3
$Ca(NO_3)_2$(s) = Ca^{2+}(aq) + 2NO_3^-(aq)	$[Ca^{2+}][NO_3^-]^2\ \gamma_{Ca^{2+}}\ \gamma^2_{NO_3^-}$	6.067×10^{5}	--	--	mol^3 / kg^3
$CaSO_4 \cdot 2H_2O$ (s) = Ca^{2+}(aq) + SO_4^{2-} (aq) + 2 H_2O	$[Ca^{2+}][SO_4^{2-}]\ \gamma_{Ca^{2+}}\ \gamma_{SO_4^{2-}}\ a_w^2$	4.319×10^{-5}	--	--	mol^2 / kg^2
$MgCl_2$ (s) = Mg^{2+}(aq) + 2Cl^- (aq)	$[Mg^{2+}][Cl^-]^2\ \gamma_{Mg^{2+}}\ \gamma^2_{Cl^-}$	9.577×10^{21}	--	--	mol^3 / kg^3
$Mg(NO_3)_2$(s) = Mg^{2+}(aq)+2NO_3^-(aq)	$[Mg^{2+}][NO_3^-]^2\ \gamma_{Mg^{2+}}\ \gamma^2_{NO_3^-}$	2.507×10^{15}	--	--	mol^3 / kg^3
$MgSO_4$ (s) = Mg^{2+}(aq) + SO_4^{2-} (aq)	$[Mg^{2+}][SO_4^{2-}]\ \gamma_{Mg^{2+}}\ \gamma_{SO_4^{2-}}$	1.079×10^{5}	--	--	mol^2 / kg^2

[a] Constants *a* and *b* are in $K = K(T_0) \cdot \exp\left[a\left(\frac{T_0}{T} - 1\right) - b\left(1 + \ln\left(\frac{T_0}{T}\right) - \frac{T_0}{T}\right)\right]$, where $T_0 = 298$ K.

LMW organic acids

- lumped low molecular weight (LMW) organic acids include:

  malonic acid, methylmalonic acid, benzoic acid, glyoxilic acid, maleic acid, succinic acid, methylsuccinic acid, glyceric acid, fumaric acid, tartronic acid, glutaric acid, malic acid, adipic acid, vanillin, 3-hydroxybenzoic acid, pimelic acid, 2-ketoglutaric acid, 4-hydroxybenzoic acid, tartaric acid, tricarballylic acid, vanillic acid, azelaic acid, 3,4-dihydroxybenzoic acid, syringic acid

- more informationen under: http://www.mpch-mainz.mpg.de/~metzger

MPI for Chemistry

The GOME-IGAM/SSP WV data record (I)