Estimation of the impact of uncertainties in meteo-rological parameters on modelled concentrations and wet deposition fluxes

S. Banzhaf[1], E. J. O'Connor[2,3], A. Kerschbaumer[1], M. Schaap and P. Builtjes[1,4]

1 Institute of Meteorology, Freie Universitaet Berlin, Germany
2 Department of Meteorology, University of Reading, United Kingdom
3 Finnish Meteorological Institute (FMI), Helsinki, Finland
4 Air Quality and Climate Team, TNO, Environment and Geosciences, The Netherlands

11th EMS/10th ECAM 2011, Berlin

Motivation

- Current projects over Europe (e.g. MAPESI) have shown that depositions still endanger our ecosystem

- Model intercomparisons show that modelled wet deposition fluxes still differ substantially from observations and show a large scatter among the models (Dentener et al., 2006; van Loon et al., 2004)
- Wet deposition is an important removal process in the pollution budget of the atmosphere
  → high relevance in order to obtain a correct chemical air pollution mass balance within CTMs
  → Model development concerning the description of wet scavenging processes as well as the sulphur and nitrogen budget in general is needed to improve modelling of wet deposition fluxes and thus the overall model performance

- An improved description of sulphate formation and scavenging processes has been implemented in the CTM REM-Calgrid (RCG) and sensitivities of the detailed process descriptions to meteorological input has been investigated

Chemistry Transport Model REM-Calgrid

- Advection scheme by Walcek (2000)
- Gaseous chemical reactions simulated using CBM-IV photochemical reaction scheme (Gery et al., 1989)
- **Aqueous-phase conversion of dissolved SO_2 to sulphate in cloud water via oxidation by H_2O_2 and oxidation by dissolved O_3**
- Equilibrium aerosol modules that treat the thermodynamics of inorganic aerosols (ISORROPIA: Nenes et al., 1999) and organic aerosols (SORGAM: Schell et al., 2001)
- Simple modules to treat the emissions of sea salt aerosols and wind blown dust particles are included
- Dry Depositions are simulated following a resistance approach - proposed by Erisman et al. (1994)
- **Wet scavenging scheme destinguishing between in-cloud and below-cloud scavenging for gases and particles**

- RCG was evaluated within many urban and regional applications and participated in several European model inter-comparison studies (Van Loon et al. 2004, Stern et al. 2008, Cuvelier et al. 2007, Thunis et al. 2007, Vautard et al. 2009)

Sulphate formation

- Aqueous phase sulphate formation by oxidation by H_2O_2 and O_3
  - Reaction rates are taken from Hoffmann and Calvert (1985) and McArdle and Hoffmann (1983)
  - The effective reaction rates are functions of variable cloud droplet pH and of **variable cloud liquid water content** (threshold sulphate formation: LWC > 0.04 g/m^3) and

Wet scavenging

- Wet scavenging scheme destinguishing between in-cloud and below-cloud scavenging for gases and particles

- Gas and particle in-cloud and below-cloud scavenging coefficients:

In-cloud threshold: $L_C > 0.04\ g/m^3$

Gases

$$\lambda_{aq} = 4.2 \cdot 10^{-7} \frac{E_c \cdot P \cdot H_{eff}(T, pH) \cdot c_g \cdot L_c}{d_d \cdot c \cdot \rho_w}$$

$$\lambda_g = 1.67 \cdot 10^{-6} \frac{K_c \cdot P}{d_d \cdot v_d}$$

$$\lambda_{ic_g} = \lambda_{aq} + \lambda_g$$

$$\lambda_{bc_g} = \lambda_g$$

Particles

$$\lambda_{ic_p} = \frac{4.2 \cdot 10^{-7} \cdot E_c \cdot P}{d_d}$$

$$\lambda_{bc_p} = \frac{4.2 \cdot 10^{-7} \cdot E_p \cdot P}{d_d}$$

Solubility limits on gas scavenging

$$\Delta c = [c_{eq}(H_{eff}) - c_0][1 - \exp(-\lambda \Delta t)]$$ (ENVIRON, 2010)

λ_{aq}=aqueous-phase scav. coeff.
λ_g=gas-phase scav. coeff.
λicg=in-cloud scav. coeff.
λbcg=below_cloud scav. coeff.
E_c=collection eff. (cloud)
E_p=collection eff. (particle)
P=precipitation rate
H_{eff}= eff. Henry Law const.
T=temperature
pH= pH value
c_g=gas conc.
c= total grid cell conc.
L_c=liquid water content
d_d=drop diameter
ρ_w=water density
v_d= mean drop fall velocity
c_{eq}=max. possible gas conc..
c_0= pre-existing gas in solution

Wet scavenging

- Wet scavenging scheme destinguishing between in-cloud and below-cloud scavenging for gases and particles
- Gas and particle in-cloud and below-cloud scavenging coefficients:

In-cloud threshold: $L_C > 0.04\ g/m^3$

Gases

$$\lambda_{aq} = 4.2 \cdot 10^{-7} \frac{E_c \cdot P \cdot H_{eff}(T, pH) \cdot c_g \cdot L_c}{d_d \cdot c \cdot \rho_w}$$

$$\lambda_g = 1.67 \cdot 10^{-6} \frac{K_c \cdot P}{d_d \cdot v_d}$$

$$\lambda_{ic_g} = \lambda_{aq} + \lambda_g$$

$$\lambda_{bc_g} = \lambda_g$$

Particles

$$\lambda_{ic_p} = \frac{4.2 \cdot 10^{-7} \cdot E_c \cdot P}{d_d}$$

$$\lambda_{bc_p} = \frac{4.2 \cdot 10^{-7} \cdot E_p \cdot P}{d_d}$$

Solubility limits on gas scavenging

$$\Delta c = [c_{eq}(H_{eff}) - c_0][1 - \exp(-\lambda \Delta t)]$$ (ENVIRON, 2010)

λ_{aq}=aqueous-phase scav. coeff.
λ_g=gas-phase scav. coeff.
λicg=in-cloud scav. coeff.
λbcg=below_cloud scav. coeff.
E_c=collection eff. (cloud)
E_p=collection eff. (particle)
P=precipitation rate
H_{eff}= eff. Henry Law const.
T=temperature
pH= pH value
c_g=gas conc.
c= total grid cell conc.
L_c=liquid water content
d_d=drop diameter
ρ_w=water density
v_d= mean drop fall velocity
c_{eq}=max. possible gas conc..
c_0= pre-existing gas in solution

Wet scavenging

- Wet scavenging scheme destinguishing between in-cloud and below-cloud scavenging for gases and particles

- Gas and particle in-cloud and below-cloud scavenging coefficients:

In-cloud threshold: $L_C > 0.04\ g/m^3$

Gases

$$\lambda_{aq} = 4.2 \cdot 10^{-7} \frac{E_c \cdot P \cdot H_{eff}(T,pH) \cdot c_g \cdot L_c}{d_d \cdot c \cdot \rho_w}$$

$$\lambda_g = 1.67 \cdot 10^{-6} \frac{K_c \cdot P}{d_d \cdot v_d}$$

$$\lambda_{ic_g} = \lambda_{aq} + \lambda_g$$

$$\lambda_{bc_g} = \lambda_g$$

Particles

$$\lambda_{ic_p} = \frac{4.2 \cdot 10^{-7} \cdot E_c \cdot P}{d_d}$$

$$\lambda_{bc_p} = \frac{4.2 \cdot 10^{-7} \cdot E_p \cdot P}{d_d}$$

Solubility limits on gas scavenging

$$\Delta c = [c_{eq}(H_{eff}) - c_0][1 - \exp(-\lambda \Delta t)]$$ (ENVIRON, 2010)

λ_{aq}=aqueous-phase scav. coeff.
λ_g=gas-phase scav. coeff.
λicg=in-cloud scav. coeff.
λbcg=below_cloud scav. coeff.
E_c=collection eff. (cloud)
E_p=collection eff. (particle)
P=precipitation rate
H_{eff}= eff. Henry Law const.
T=temperature
pH= pH value
c_g=gas conc.
c= total grid cell conc.
L_c=liquid water content
d_d=drop diameter
ρ_w=water density
v_d= mean drop fall velocity
c_{eq}=max. possible gas conc..
c_0= pre-existing gas in solution

Wet scavenging

- Wet scavenging scheme destinguishing between in-cloud and below-cloud scavenging for gases and particles

- Gas and particle in-cloud and below-cloud scavenging coefficients:

In-cloud threshold: $L_C > 0.04\ g/m^3$

Gases

$$\lambda_{aq} = 4.2 \cdot 10^{-7} \frac{E_c \cdot P \cdot H_{eff}(T, pH) \cdot c_g \cdot L_c}{d_d \cdot c \cdot \rho_w}$$

$$\lambda_g = 1.67 \cdot 10^{-6} \frac{K_c \cdot P}{d_d \cdot v_d}$$

$$\lambda_{ic_g} = \lambda_{aq} + \lambda_g$$

$$\lambda_{bc_g} = \lambda_g$$

Particles

$$\lambda_{ic_p} = \frac{4.2 \cdot 10^{-7} \cdot E_c \cdot P}{d_d}$$

$$\lambda_{bc_p} = \frac{4.2 \cdot 10^{-7} \cdot E_p \cdot P}{d_d}$$

Solubility limits on gas scavenging

$$\Delta c = [c_{eq}(H_{eff}) - c_0][1 - \exp(-\lambda \Delta t)]$$ (ENVIRON, 2010)

λ_{aq}=aqueous-phase scav. coeff.
λ_g=gas-phase scav. coeff.
λicg=in-cloud scav. coeff.
λbcg=below_cloud scav. coeff.
E_c=collection eff. (cloud)
E_p=collection eff. (particle)
P=precipitation rate
H_{eff}= eff. Henry Law const.
T=temperature
pH= pH value
c_g=gas conc.
c= total grid cell conc.
L_c=liquid water content
d_d=drop diameter
ρ_w=water density
v_d= mean drop fall velocity
c_{eq}=max. possible gas conc..
c_0= pre-existing gas in solution

Results - Evaluation COSMO-EU Liquid Water Content

Model data

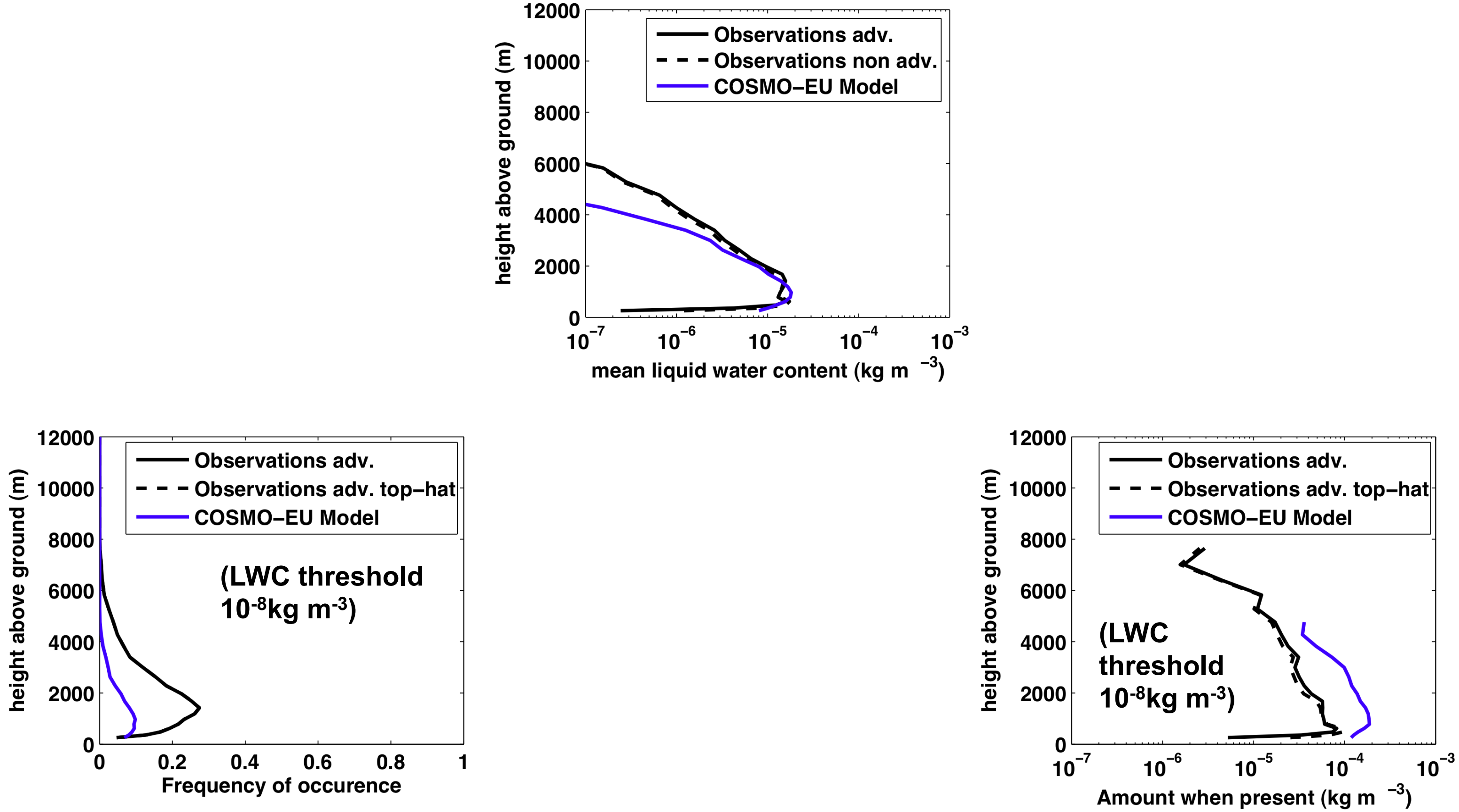
- COSMO-EU is the nonhydrostatic regional model of the German Weather Service (DWD) (Doms et al., 2008)
- Revised microphysics scheme is in operation since 31st January 2007 (Seifert and Crewell, 2008)

Observational data

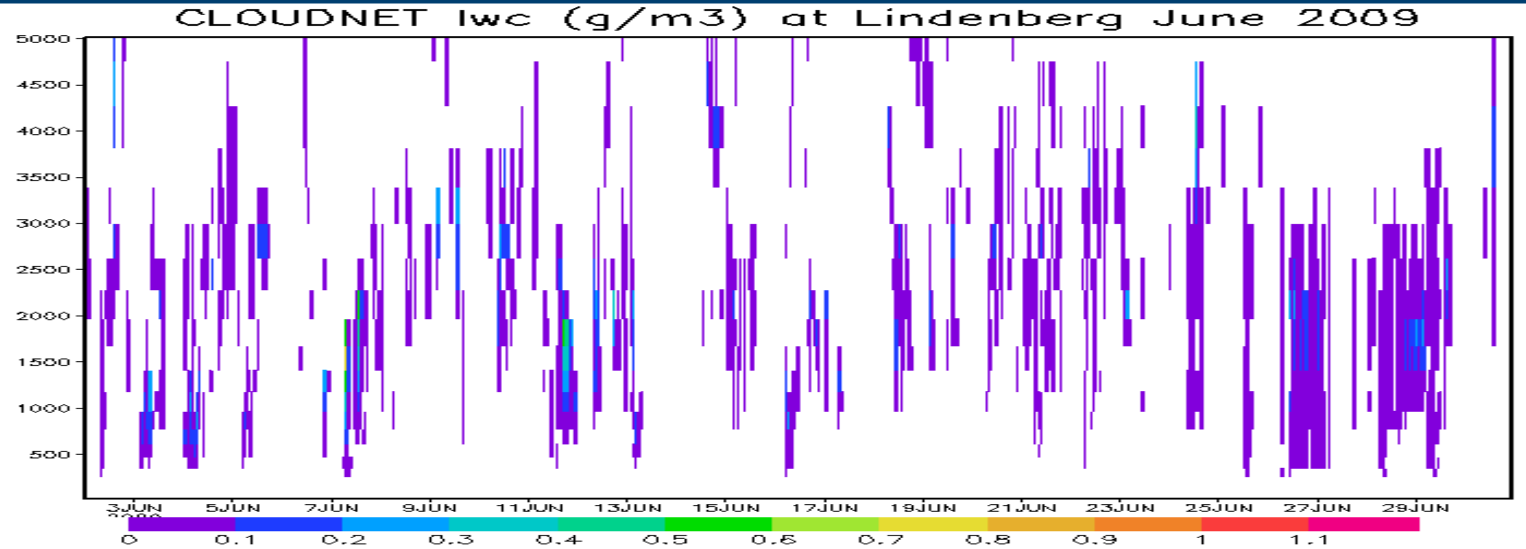
- CloudNet Project
  Aim: to retrieve continuously the crucial cloud parameters for climate and forecast models for evaluation purposes (Illingworth et al. 2007)
- Four main sites: Chilbolton (GB) Cabauw (NL), Palaiseau(F), Lindenberg (DE) (since 2005)
- LWC is derived using radar and lidar cloud boundaries and scaled using the liquid water path from the dual-wavelength microwave radiometer measurements

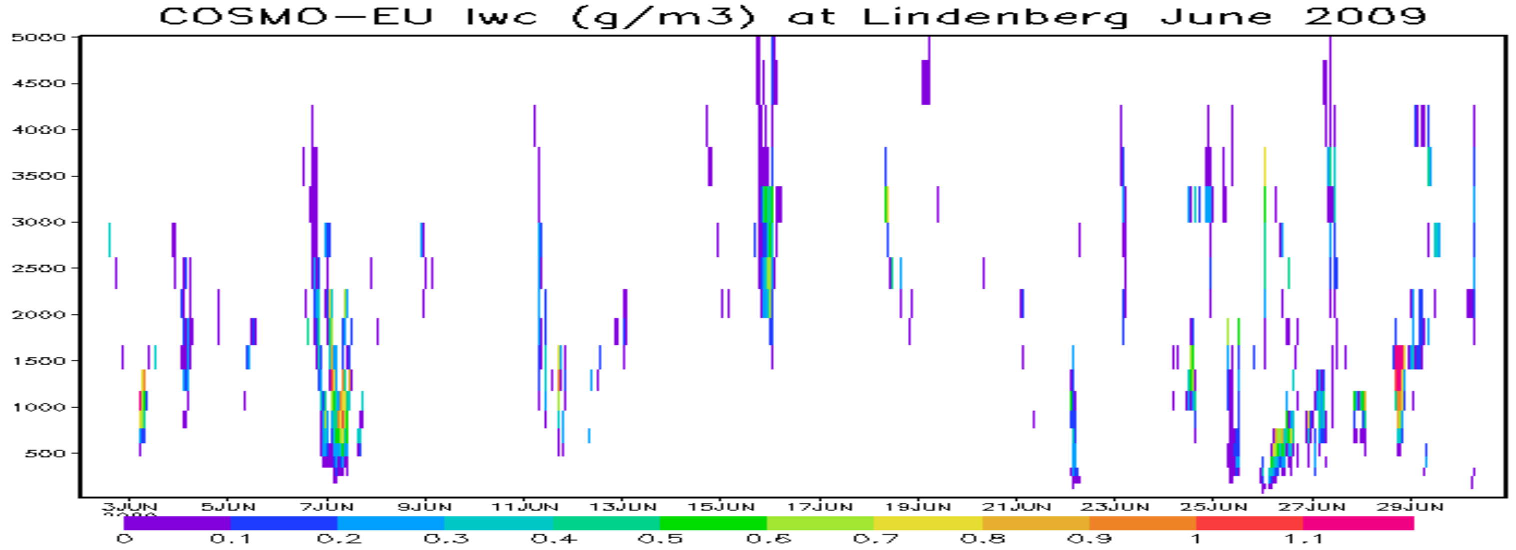
Results - Evaluation COSMO-EU Liquid Water Content

Evaluation of COSMO−EU Model liquid water content at Lindenberg for 2009 (Equivalent of 183.5 days of data)



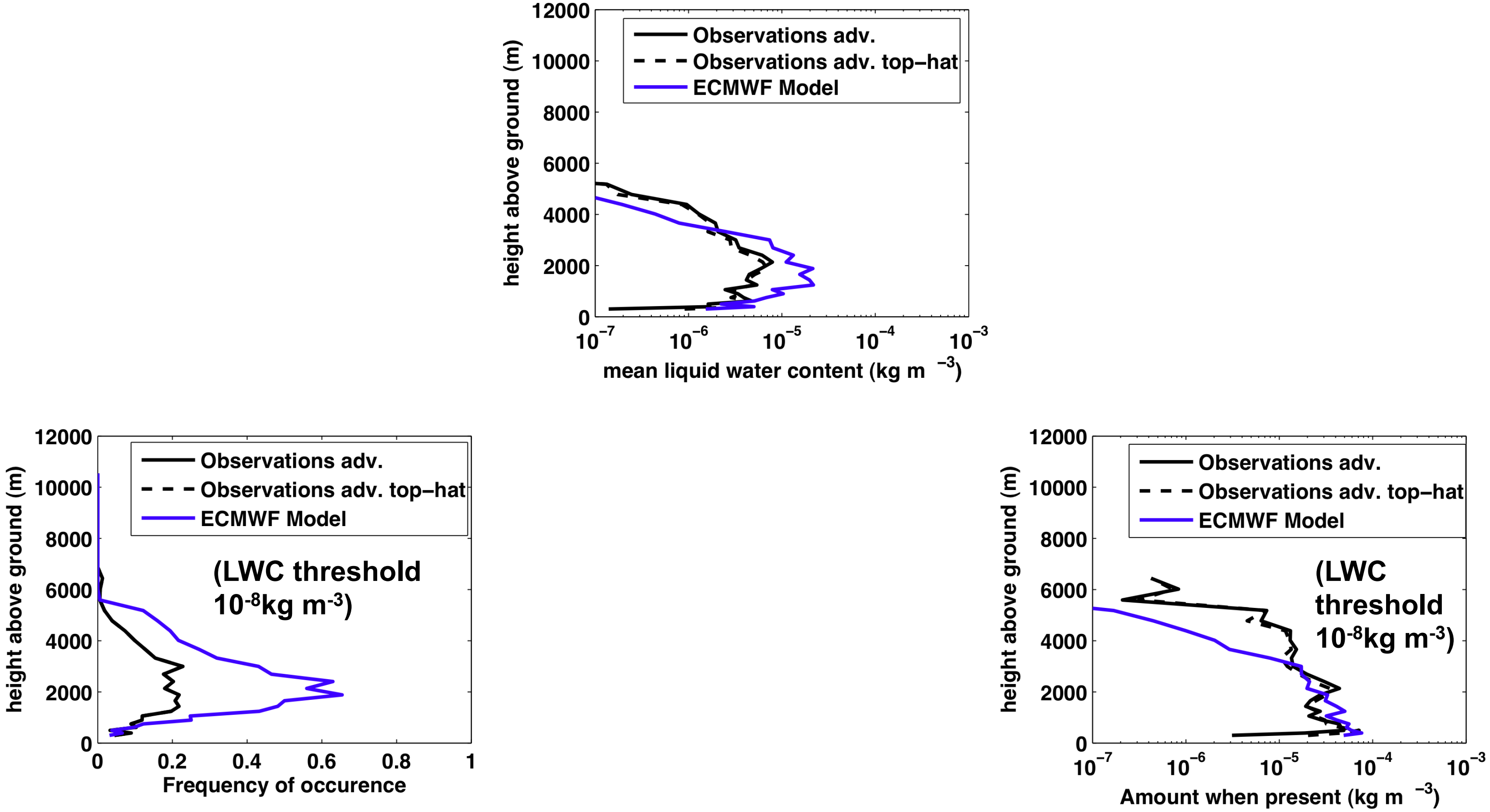
Results - Evaluation COSMO-EU Liquid Water Content





Results - Evaluation ECMWF Liquid Water Content

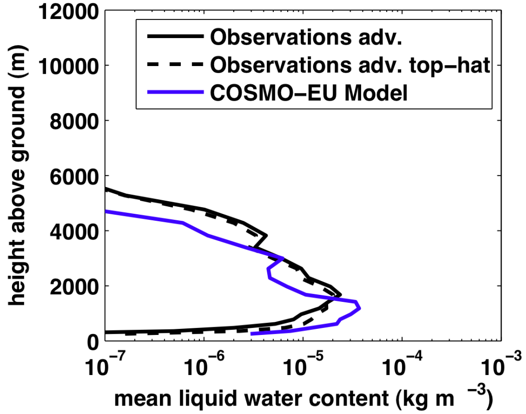
Evaluation of ECMWF Model liquid water content at Lindenberg for April 2009 (Equivalent of 10.4 days of data)

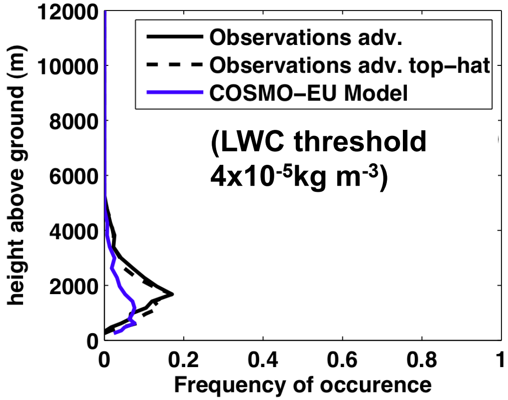


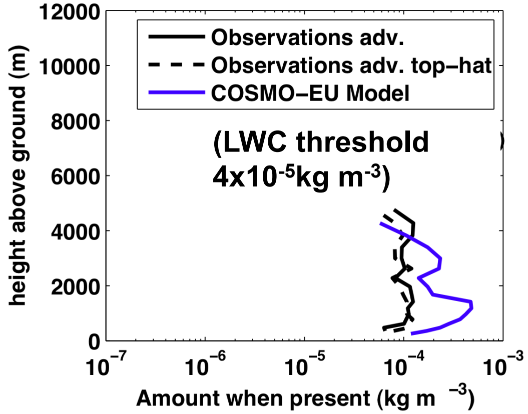
RCG Model setup

- Domain covering Germany (47.2N-55.1N; 5.4E-15.7E) with a horizontal resolution of approximately 7x7km^2 and 20 vertical layers up to 5000m
- A large scale RCG run covering Europe provided the Boundary Conditions
- Emissions from local and national inventories (Jörß et al., 2010) and EMEP data post-processed at TNO (Denier van der Gon et al., 2010)
- Hourly meteorological fields are provided by COSMO-EU (Doms et al., 2008)

Evaluation of COSMO–EU Model liquid water content at Lindenberg for June 2009 (Equivalent of 20.8 days of data)







RCG Model runs

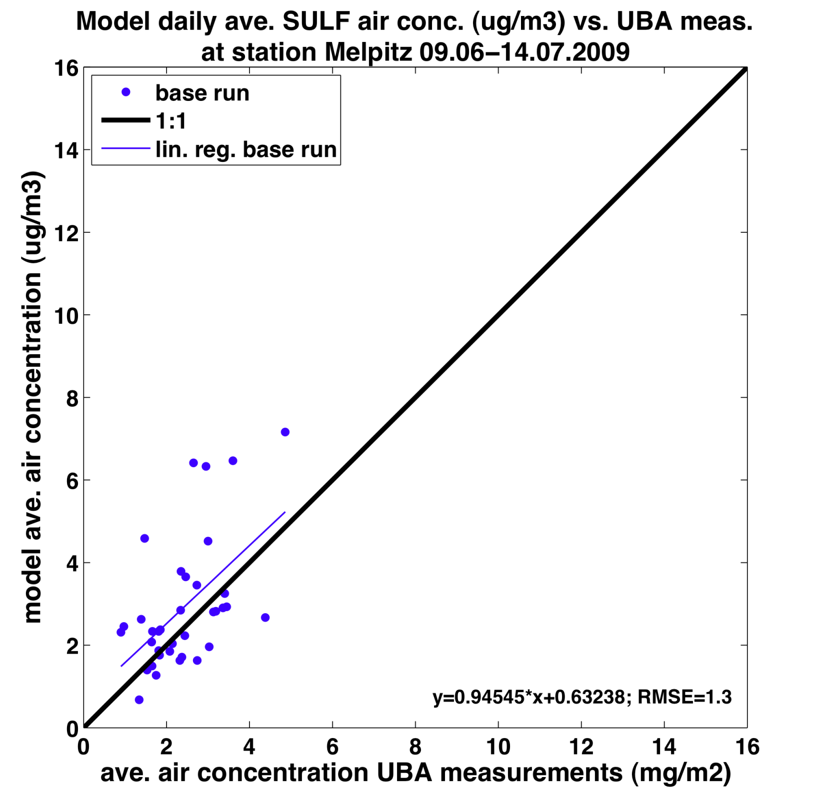
- Investigation period: 09.06.-14.07.2009

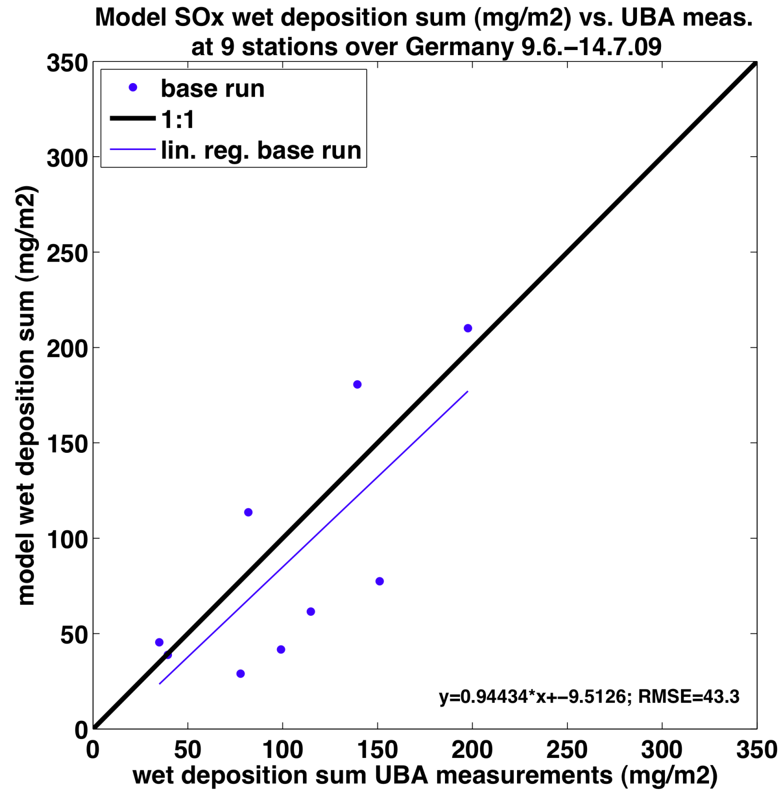
- Base run

- LWC/2.5 run
  with lower limit 0.04 (=threshold for sulphate formation and in-cloud scavenging)
  → decrease by 250%

- +60% cloud run
  increase cloud occurrence by creating non-precipitating clouds ($LWC=0.05g/m^3$) for RH>90% and T>270K
  → increase by 60%

Results - Model performance (base run)



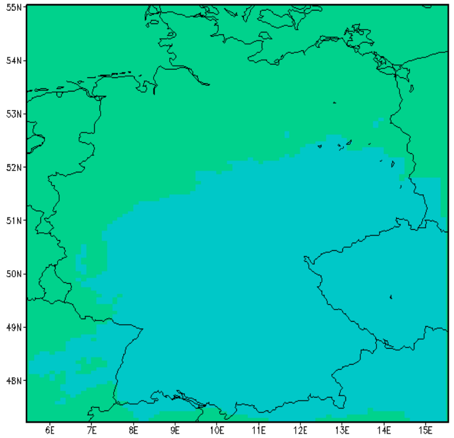


Results – Impact on concentrations

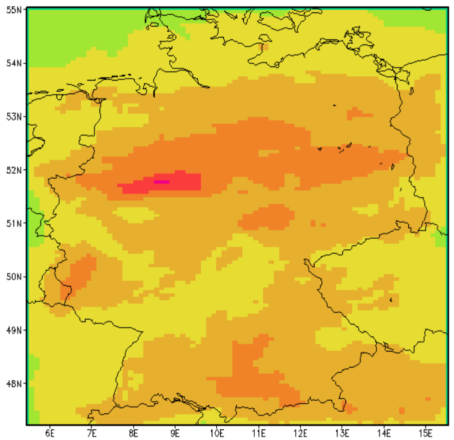
average concentration deviation from base run (%)

SO^4

LWC/2.5 run



+60% cloud run



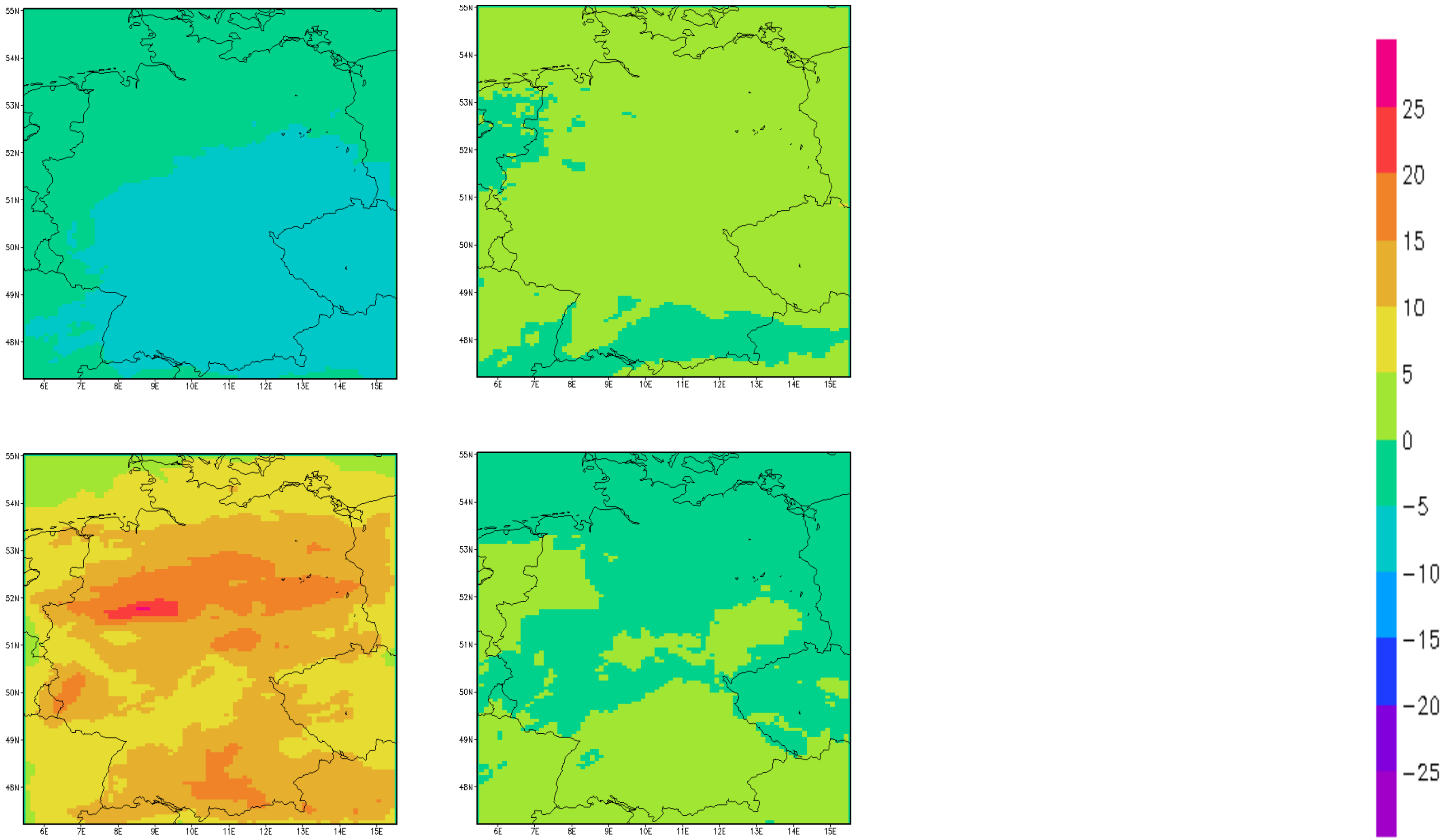
Results – Impact on concentrations

average concentration deviation from base run (%)

SO^4 | NO^3

LWC/2.5 run

+60% cloud run



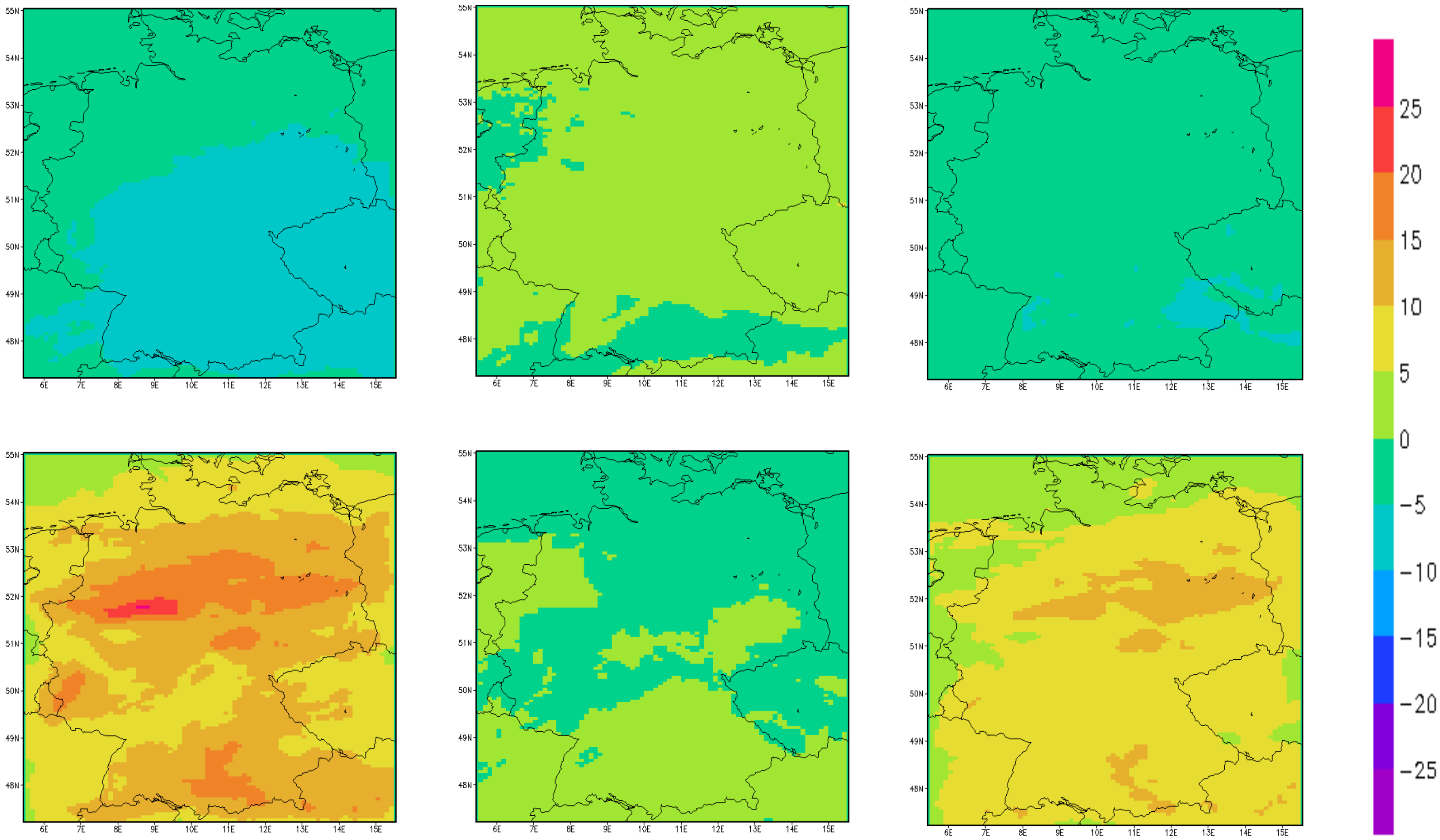
Results – Impact on concentrations

average concentration deviation from base run (%)

SO^4 | NO^3 | NH^4

LWC/2.5 run

+60% cloud run

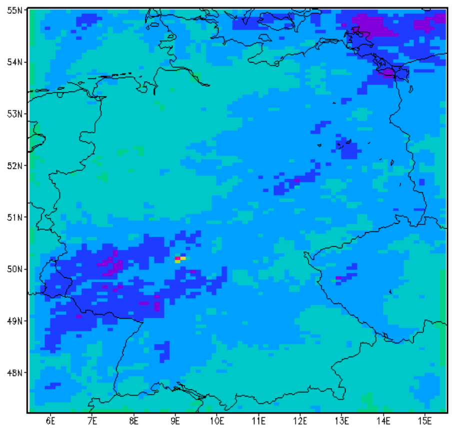


Results – Impact on wet deposition fluxes

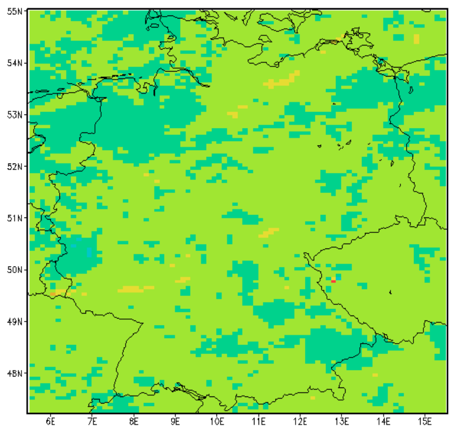
wet deposition sum deviation from base run (%)

SOx

LWC/2.5 run



+60% cloud run



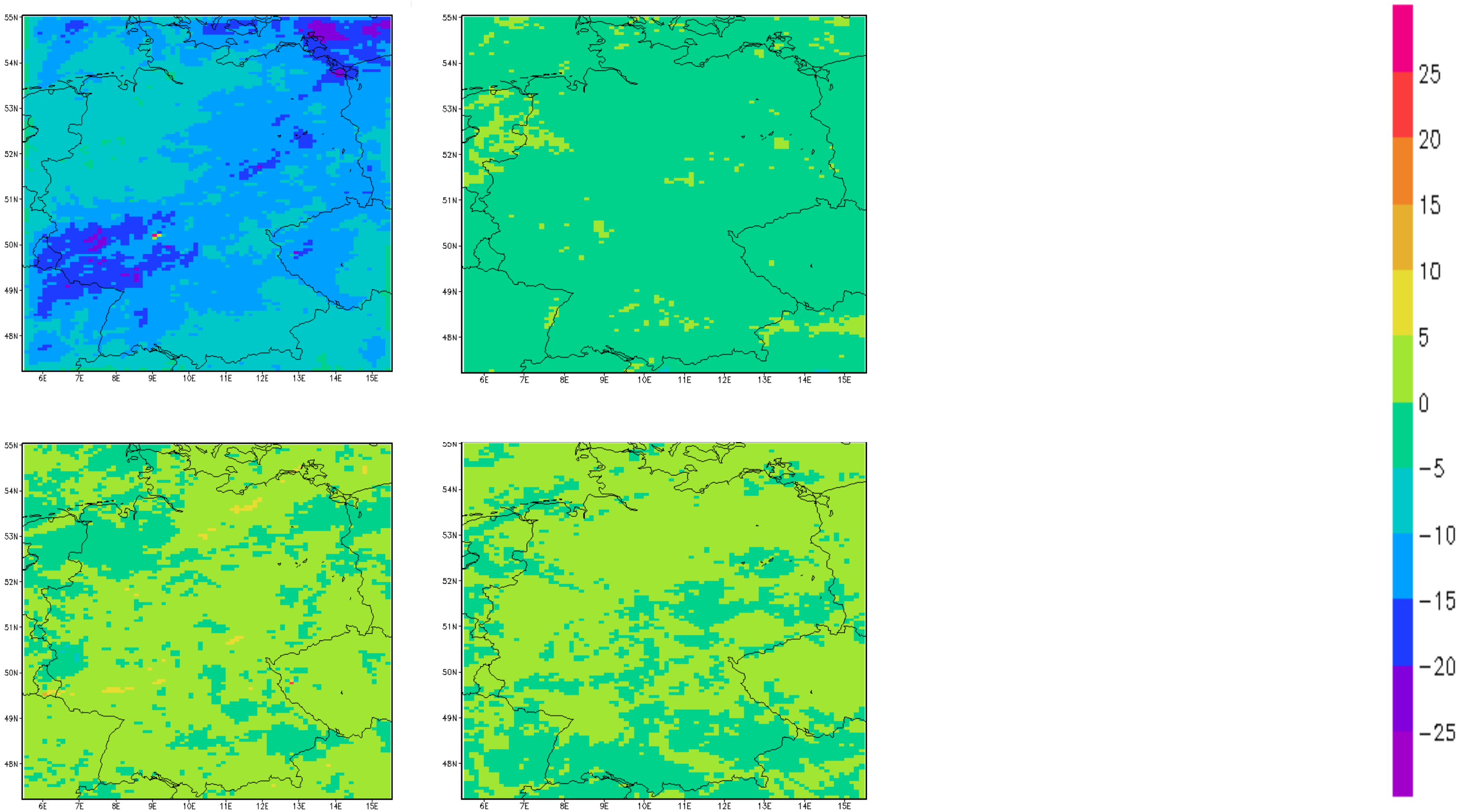
Results – Impact on wet deposition fluxes

wet deposition sum deviation from base run (%)

SOx NOy

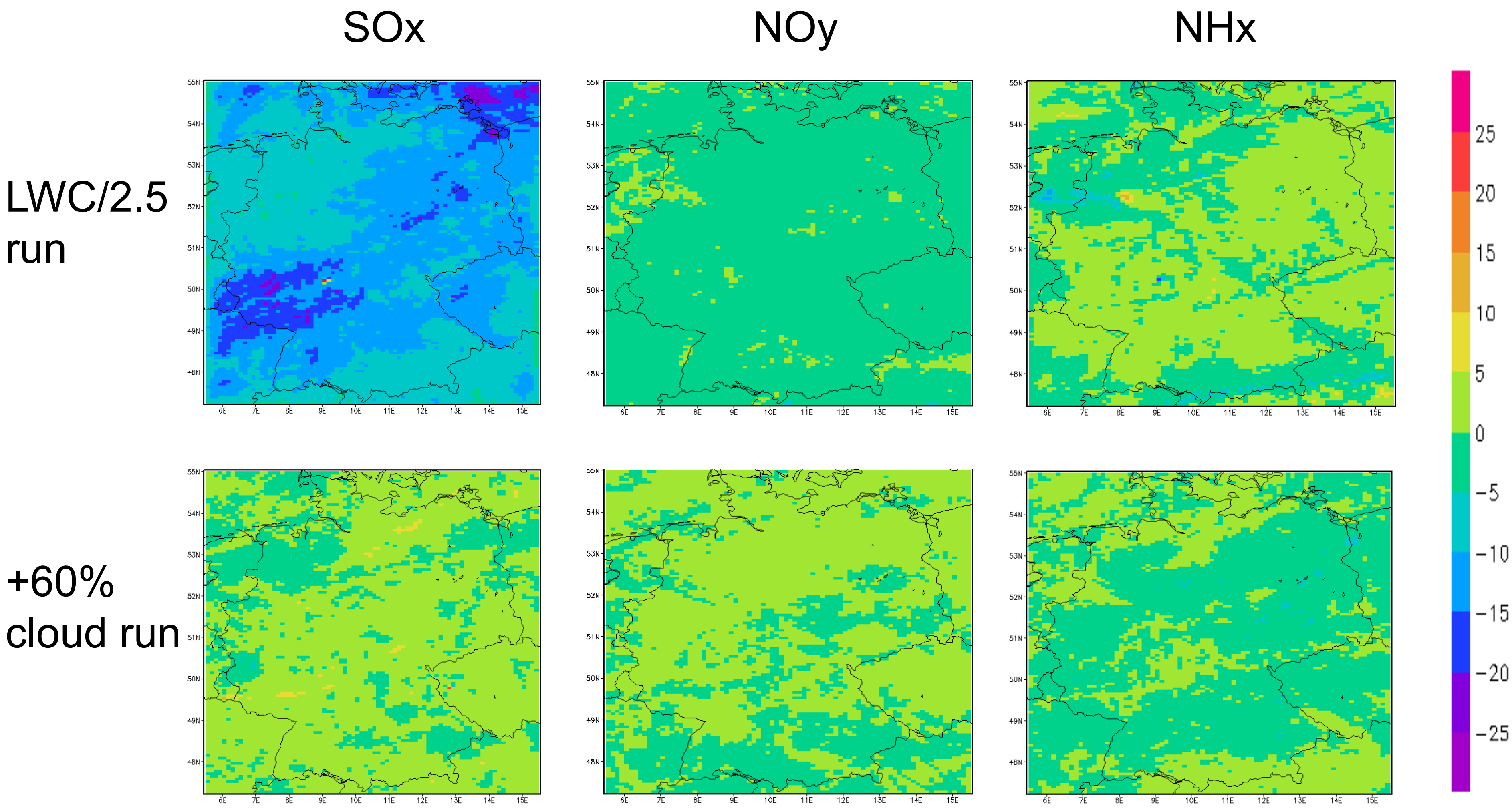
LWC/2.5 run

+60% cloud run



Results – Impact on wet deposition fluxes

wet deposition sum deviation from base run (%)



Conclusions

- Mean LWC COSMO-EU compares well with measured mean but
  - frequency of occurrence of LWC underestimated and
  - amount when LWC is present overestimated
    → compensating errors when averaging LWC

- CTM sensitivity runs
  - An increase of frequency of occurrence of LWC leads to a significant increase of sulphate concentrations
  - A decrease of LWC amount when present leads to a decrease of SOx wet deposition fluxes
    → Compensating error with a dominant impact on sulphate concentrations when LWC occurrence is increased

- The more detailed the physical and chemical descriptions of aqueous phase chemistry and scavenging processes are the more accurate meteorological input is required

Outlook

- Impact of uncertainties in

  → precipitation

  → cloud fraction

  on modelled concentrations and wet deposition fluxes

Thank you very much for your attention!

Acknowledgements

- CLOUDNET Project, (observed LWC profiles)
- UBA (observed wet deposition fluxes)
- Ralf Wolke, Gerald Spindler, Konrad Müller from IFT Leipzig (observed sulphate concentrations)