Possible feedbacks of aerosols on meteorology

2nd lecture: *Model realization examples*

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Lecture's Objective and Goal:

- **Description of the main feedback mechanisms of the chemical weather (atmospheric green-house gases and aerosols) impact on NWP and climate processes, in order to understand how important it is to include feedbacks from gases, aerosols, clouds, etc. in NWP and climate models.**
- **The goal is to give an orientation/understanding of which feedback processes are the most important: impact of feedbacks from gases, aerosols (direct, semi-direct, indirect effects), clouds, etc. on short and long time-range meteorological models.**
- **This subject is the main focus of the school. First part focuses on the physical processes behind these feedbacks. Second part focuses on model realization strategy and examples.**

Implementation of the feedback mechanisms into integrated models:

One-way integration (off-line):

- 1. Simplest way (no aerosol forcing): NWP meteo-fields as a driver for CTM (this classical way is used already by most of air pollution modellers);
- 2. CTM chemical composition fields as a driver for Regional/Global Climate Models (including the aerosol forcing on meteo-processes, it could also be realized for NWP or MetMs).

Two-way integration:

- 1. Driver + partly aerosol feedbacks, for CM or for NWP (data exchange in both directions with a limited time period coupling: off-line or on-line access coupling, with or without second iteration with corrected fields);
- 2. Full feedbacks included on each time step (on-line coupling/integration).

Coupling Air Quality and Meteorology/Climate Modeling Rationale and Motivation

- **Common deficiencies of a global climate-aerosol model**
  - Coarse spatial resolution cannot explicitly capture the fine-scale structure that characterizes climatic changes (e.g., clouds, precipitation, mesoscale circulation, sub-grid convective system, etc.) and air quality responses
  - Coarse time resolution cannot replicate variations at smaller scales (e.g., hourly, daily, diurnal)
  - Simplified treatments (e.g., simple met. schemes and chem./aero. treatments) cannot represent intricate relationships among meteorology/climate/AQ variables
  - Most models simulate climate and aerosols offline with inconsistencies in transport and no climate-chemistry-aerosol-cloud-radiation feedbacks
- **Common deficiencies of a urban/regional climate or AQ model**
  - Most AQMs do not treat aerosol direct and indirect effects
  - Most AQMs use offline meteorological fields without feedbacks
  - Some AQMs are driven by a global model with inconsistent model physics
  - Most regional climate models use prescribed aerosols or simple modules without detailed chemistry and microphysics

Coupling Air Quality and Meteorology/Climate Modeling
History and Current Status

- **Prior to 1990th: Separation of air quality, meteorology, climate**
- **1985-Present: Offline and online coupling**
  - » **Urban/Regional Models**
    - **The first attempt of online coupling meteorology/chemistry/aerosol models, developed in Novosibirsk scient. school in 1985**
    - **The first fully-coupled meteorology/chemistry/aerosol/radiation model,**
    - **GATOR-MMTD, was developed by Jacobson in 1994**
    - **The first community coupled meteorology/chemistry/aerosol/radiation/ clouds model: in USA - WRF/Chem (Grell et al., 2004), in Europe – Enviro-HIRLAM (Baklanov et al., 2006)**
    - **Most air quality models (AQMs) are still offline**
    - **Most AQMs do not treat aerosol direct and indirect effects**
    - **Most regional climate models use prescribed aerosols or simple modules**
    - **without detailed aerosol chemistry and microphysics**
  - » **Global Models**
    - **The first nested global-through-urban scale fully-coupled model, GATOR-GCMM, was developed by Jacobson in 2001**
    - **Most global AQMs (GAQMs) are still offline**
    - **Most GAQMs use an empirical sulfate-CCN relation for indirect effects**

Scientific hypotheses/questions to be tested/addressed

- **Hypothesis**
  - **Feedback mechanisms are important in accurate modeling of NWP/MM-ACT and quantifying direct and indirect effects of aerosols.**
- **Key questions**
  - **What are the effects of climate/meteorology on the abundance and properties (chemical, microphysical, and radiative) of aerosols on urban/regional scales?**
  - **What are the effects of aerosols on urban/regional climate/meteorology and their relative importance (e.g., anthropogenic vs. natural)?**
  - **How important the two-way/chain feedbacks among meteorology, climate, and air quality are in the estimated effects?**
  - **What is the relative importance of aerosol direct and indirect effects in the estimates?**
  - **What are the key uncertainties associated with model predictions of those effects?**
  - **How can simulated feedbacks be verified with available datasets?**

Processes/feedbacks to be considered

- **Direct effect - Decrease solar/thermal-IR radiation and visibility**
  - Processes needed: radiation (scattering, absorption, refraction, etc.)
  - Key variables: refractive indices, ext. coeff., SSA, asymmetry factor, AOD, visual range
  - Key species: cooling: water, sulfate, nitrate, most OC
    warming: BC, OC, Fe, Al, polycyclic/nitrated aromatic compounds
- **Semi-direct effect - Affect PBL meteorology and photochemistry**
  - Processes needed: PBL/LS, photolysis, met-dependent processes
  - Key variables: T, P, RH, Qv, WSP, WDR, Cld Frac, stability, PBL height, photolysis rates, emission rates of met-dependent primary species (dust, sea-salt, biogenic)
- **First indirect effect – Affect cld drop size, number, reflectivity, and optical depth via CCN**
  - Processes needed: aero. activation/resuspension, cld. microphysics, hydrometeor dynamics
  - Key variables: int./act. frac, CCN size/comp., cld drop size/number/LWC, COD, updraft vel.
- **Second indirect effect - Affect cloud LWC, lifetime, and precipitation**
  - Processes needed: in-/below-cloud scavenging, droplet sedimentation
  - Key variables: scavenging efficiency, precip. rate, sedimentation rate
- **All aerosol effects**
  - Processes needed: aero. thermodynamics/dynamics, aq. chem., precursor emi., water uptake
  - Key variables: aerosol mass, number, size, comp., hygroscopicity, mixing state

Implementation Priorities

- **Highest priority (urgent)**
  - Aerosol thermodynamics/dynamics, aq. chem., precursor emi., water uptake
  - Radiation, emission, PBL/LS schemes, photolysis, aerosol-CCN relation
  - Coding standard and users' guide for parameterizations
- **High priority (pressing)**
  - Aero. activation/resuspension, Brownian diffusion, drop nucleation scavenging
  - Other in-/below-cloud scavenging (collection, autoconversion, interception, impaction)
- **Important**
  - Hydrometeor dynamics, size representation, hysteresis effect, DMRH
- **Other**
  - Subgrid variability, multiple size distributions

ENVIRO-HIRLAM:
First indirect feedbacks of urban aerosols

For water clouds:

$r^3_{eff} = kr^3_v$

$r^3_{eff} = 3L/(4\pi\rho_l kN)$

(*Wyser et al. 1999*)

L : Cloud condensate content

N: Number concentration of cloud droplets

$\Delta N_{cont} = 10^{8.06}\ conc^{0.48}$

$\Delta N_{marine} = 10^{2.24}\ conc^{0.26}$

(*Boucher & Lohmann, 1995*)

Emission rate: 7.95 gs^{-1}; ETEX

Diameter: 1 μm

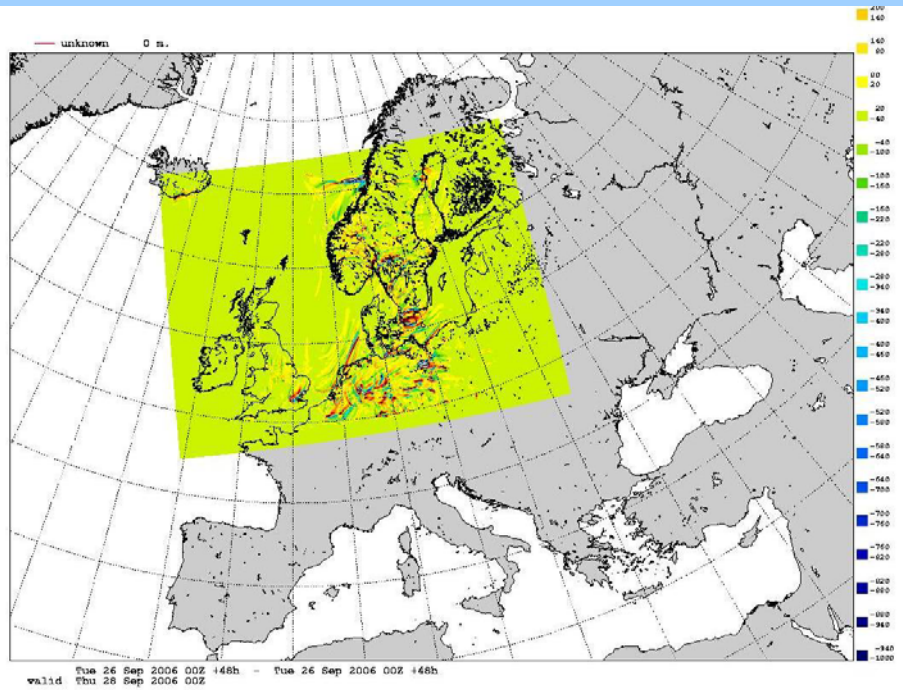
	k	N [m^{-3}]
Marine	0.81	10^8
Cont	0.69	$4x10^8$

Urban fractions [%; dark green – dark red]

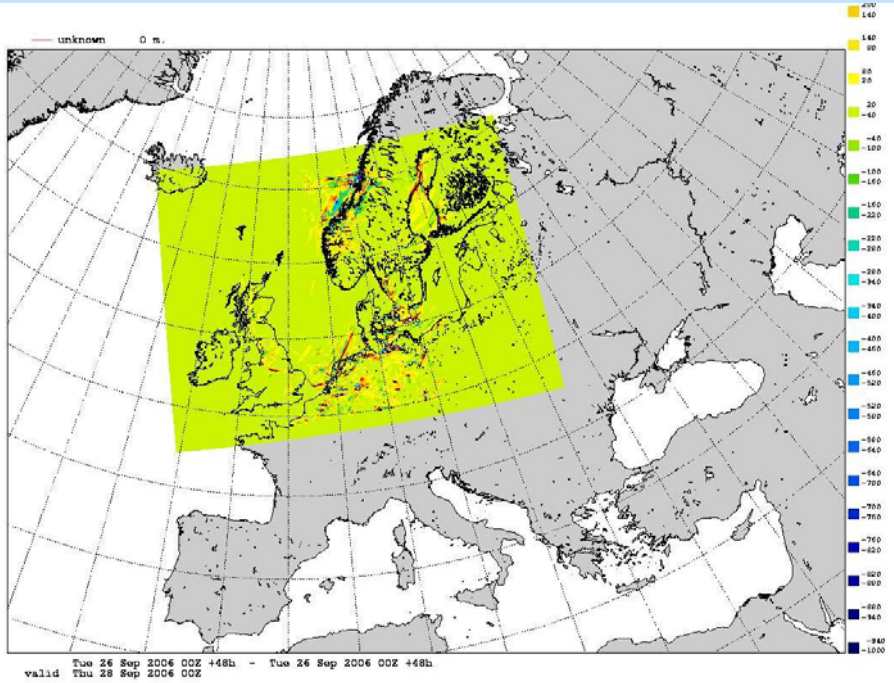
Korsholm & Baklanov, 2007

DMI-ENVIRO-HIRLAM: Feedbacks of urban aerosols

Korsholm & Baklanov, 2007



Difference (ref - perturbation)
in accumulated dry deposition [ng/m^2]



Difference (ref - perturbation)
in accumulated wet deposition [ng/m^2]

Feedbacks through the first indirect effect lead to modifications of the order 7 % in dry and wet deposition patterns over major polluted areas in Europe.

The effects of urban aerosols on the urban boundary layer height, h, could be of the same order of magnitude as the effects of the urban heat island (Δh is about 100-200 m for stable boundary layer).

ENVIRO-HIRLAM:
Second indirect feedbacks of urban aerosols

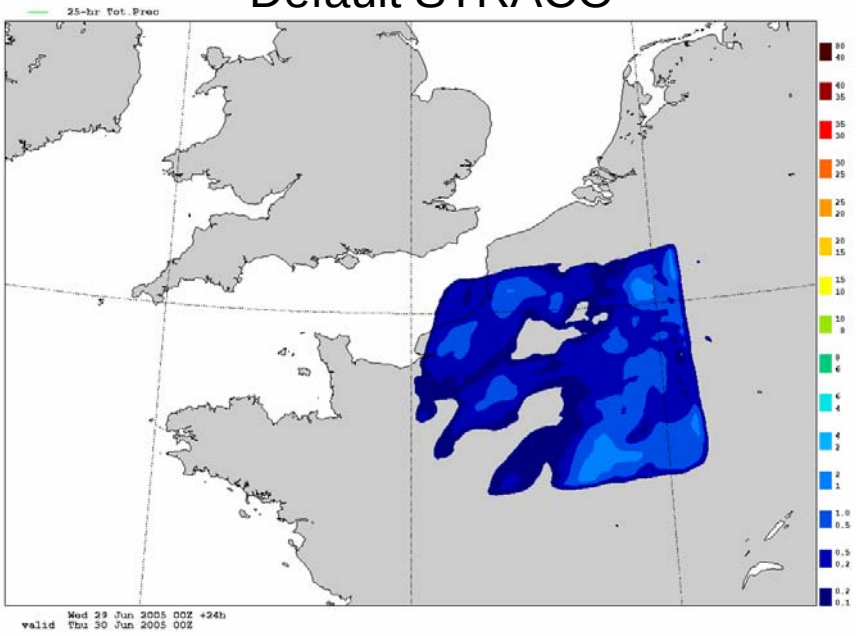
(recent simulation by U. Korsholm)

- Domain covering app. 500 x 400 km around Paris, France.
- Notes on the experiment: 0.05 x 0.05 degrees horizontal resolution, 40 vertical levels, 300 s time step, NWP-Chem chemistry (18 species),
- CAC-aerosol mechanism: homogeneous nucleation, condensation, coagulation
- Aerosols consists of H2O, HSO4-, SO4--, two log-normal modes: nuclei, accumulation
- Accumulation mode aerosols used as CCN's
- Case with low winds, convective clouds, little precipitation
- Reference run without feedbacks, Perturbed run with second indirect effect.
- Second indirect effect based on modified version of STRACO, where the autoconversion from Rasch-Kristjansson has been implemented.
- 24 hours spin-up, 24 hours forecast, all concentrations are at lowest model level,
- boundary zone shown but may be neglected, plots show reference and difference plots.

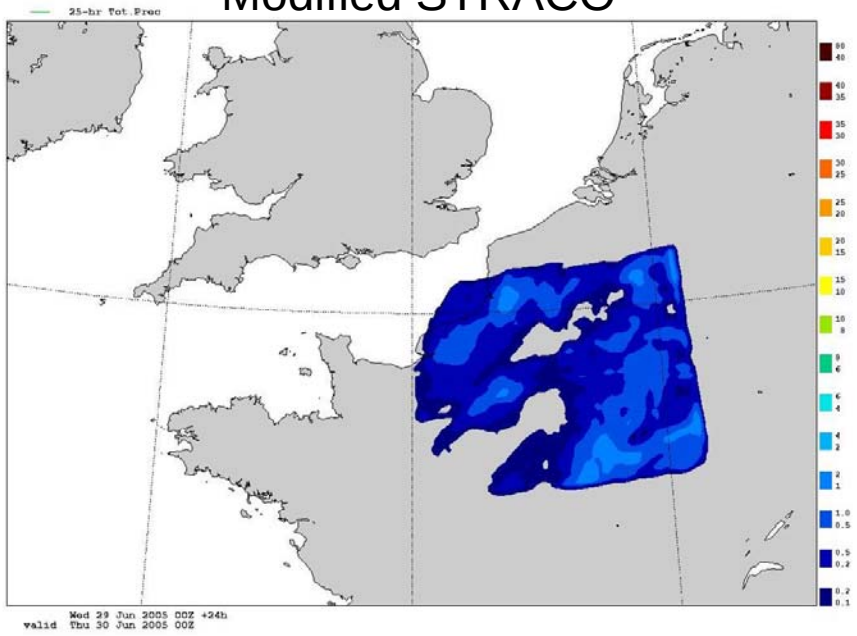
The reference and modified STRACO schemes

Accumulated precipitation +24h (mm)

Default STRACO



Modified STRACO

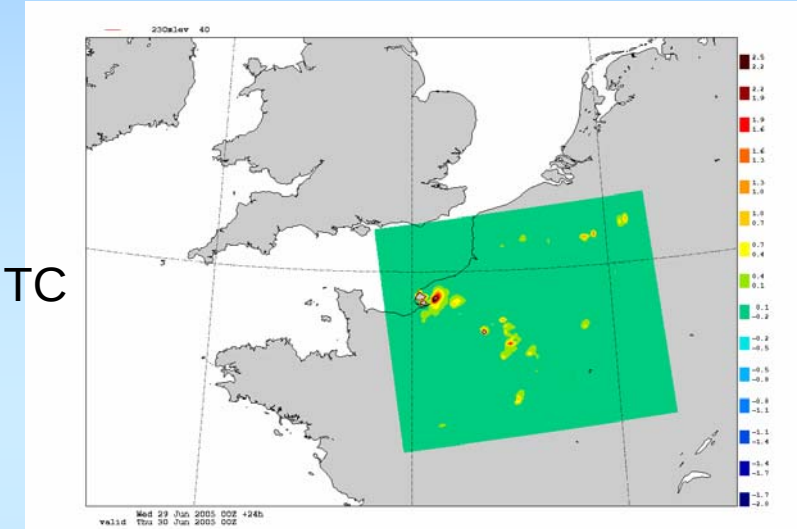


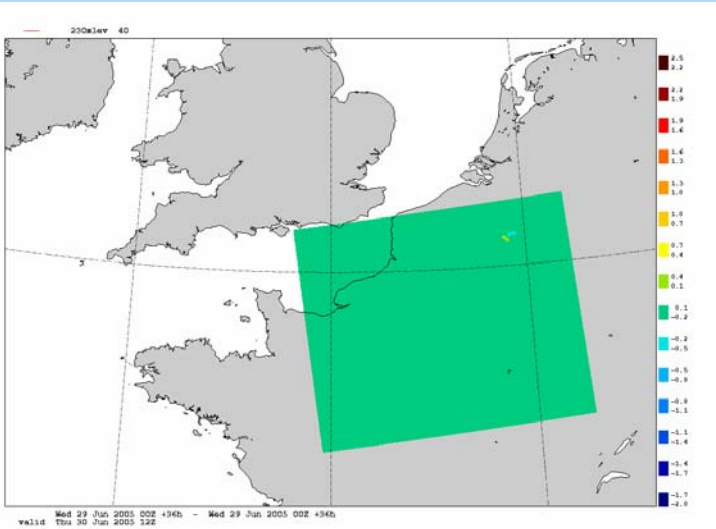
H2SO4 concentrations (micro g/m3)

Reference

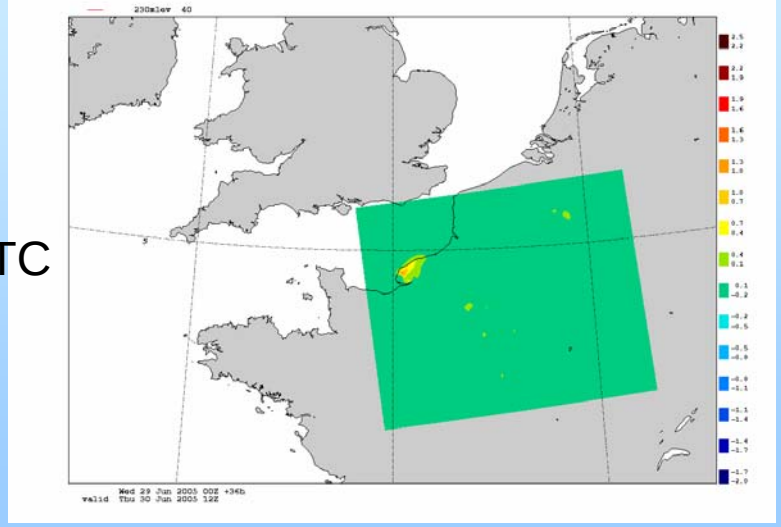
Including second indirect effect

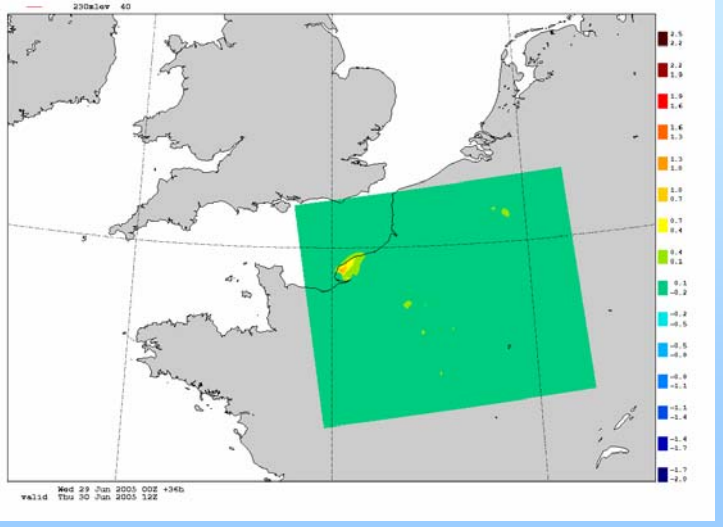
00 UTC



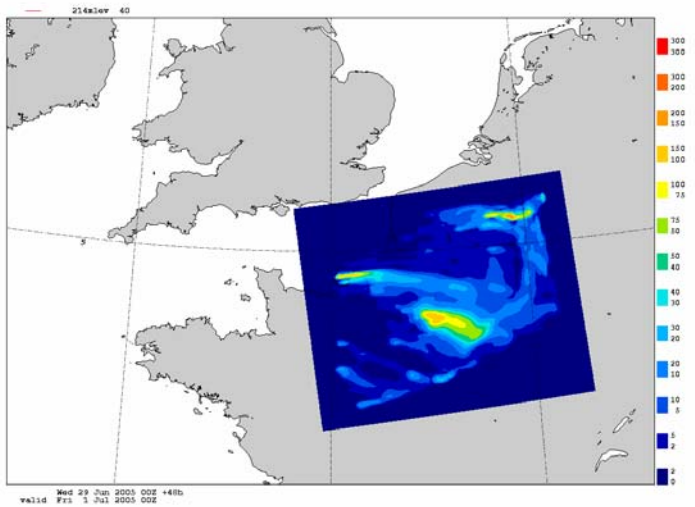
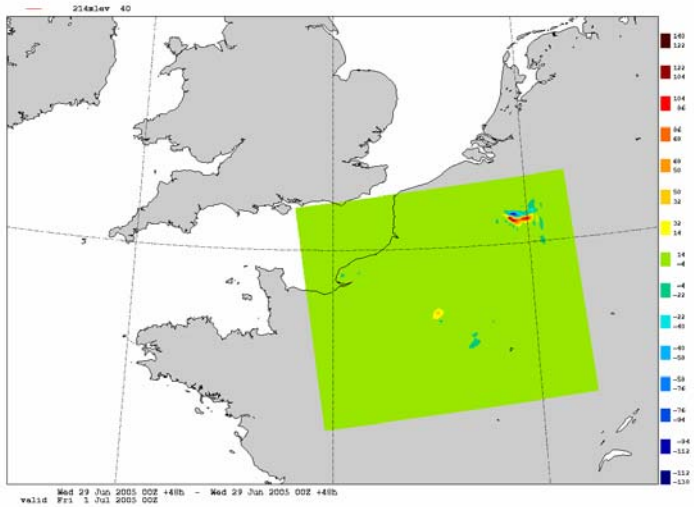
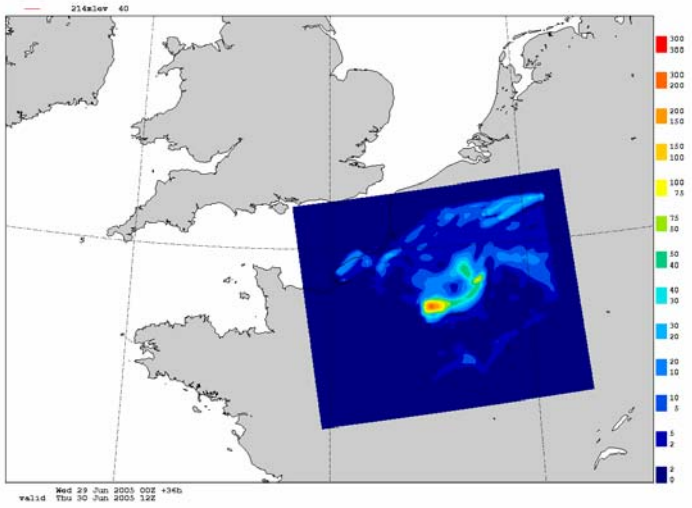
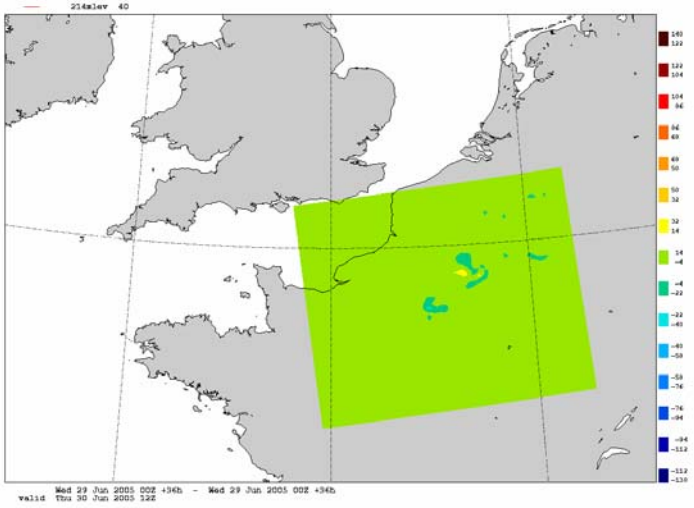


12 UTC





NO2 concentrations (micro g/m3)

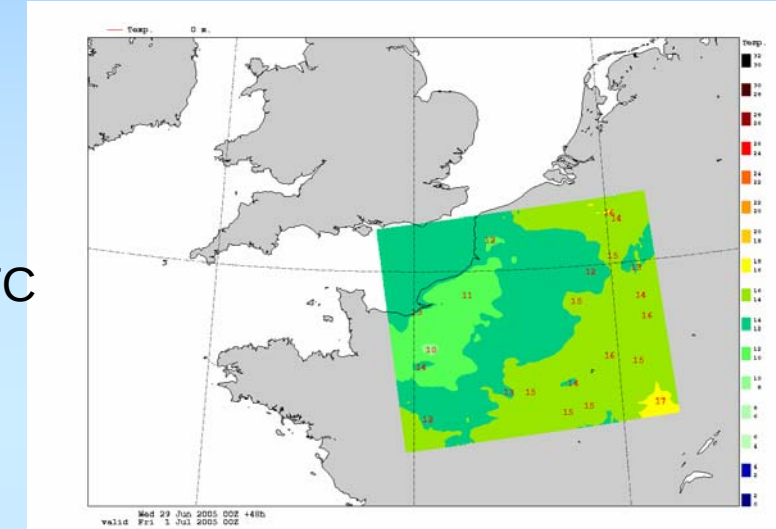
	Reference	Including second indirect effect
00 UTC		
12 UTC		

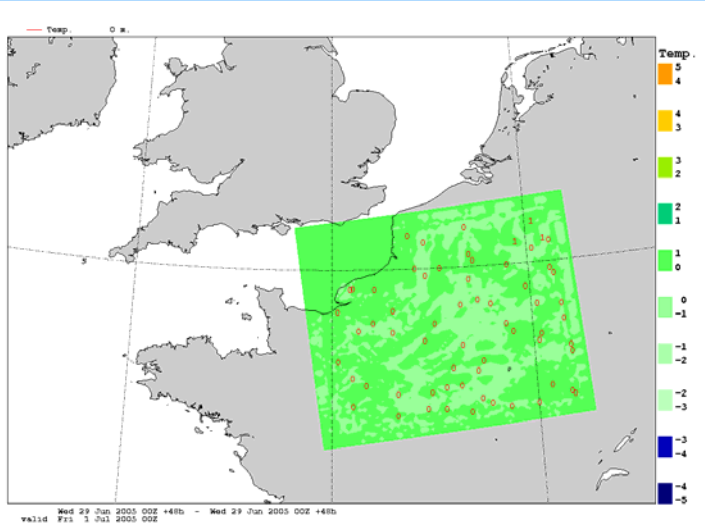
Surface temperature (C)

Reference

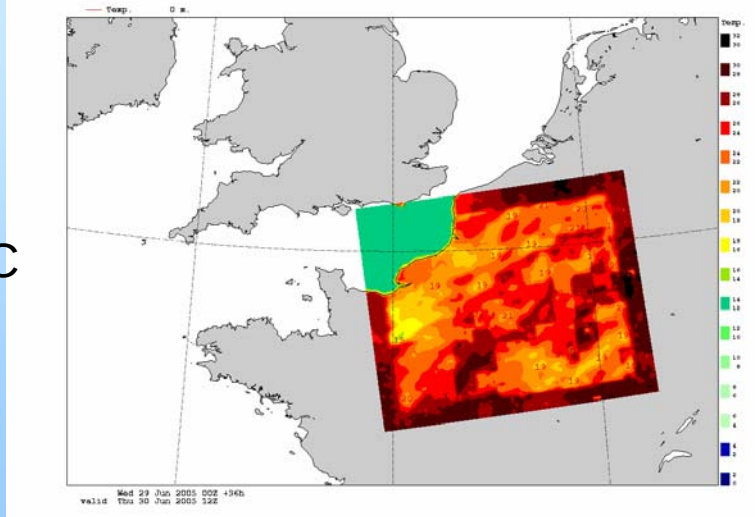
Including second indirect effect

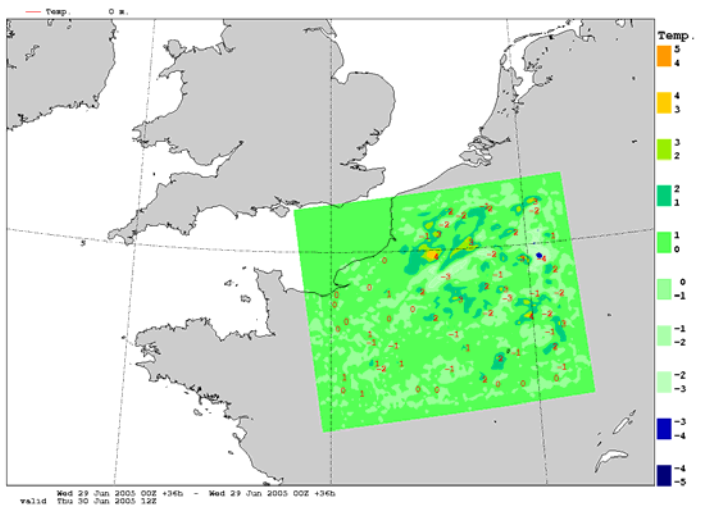
00 UTC





12 UTC





Temperature changes are up to 4º C

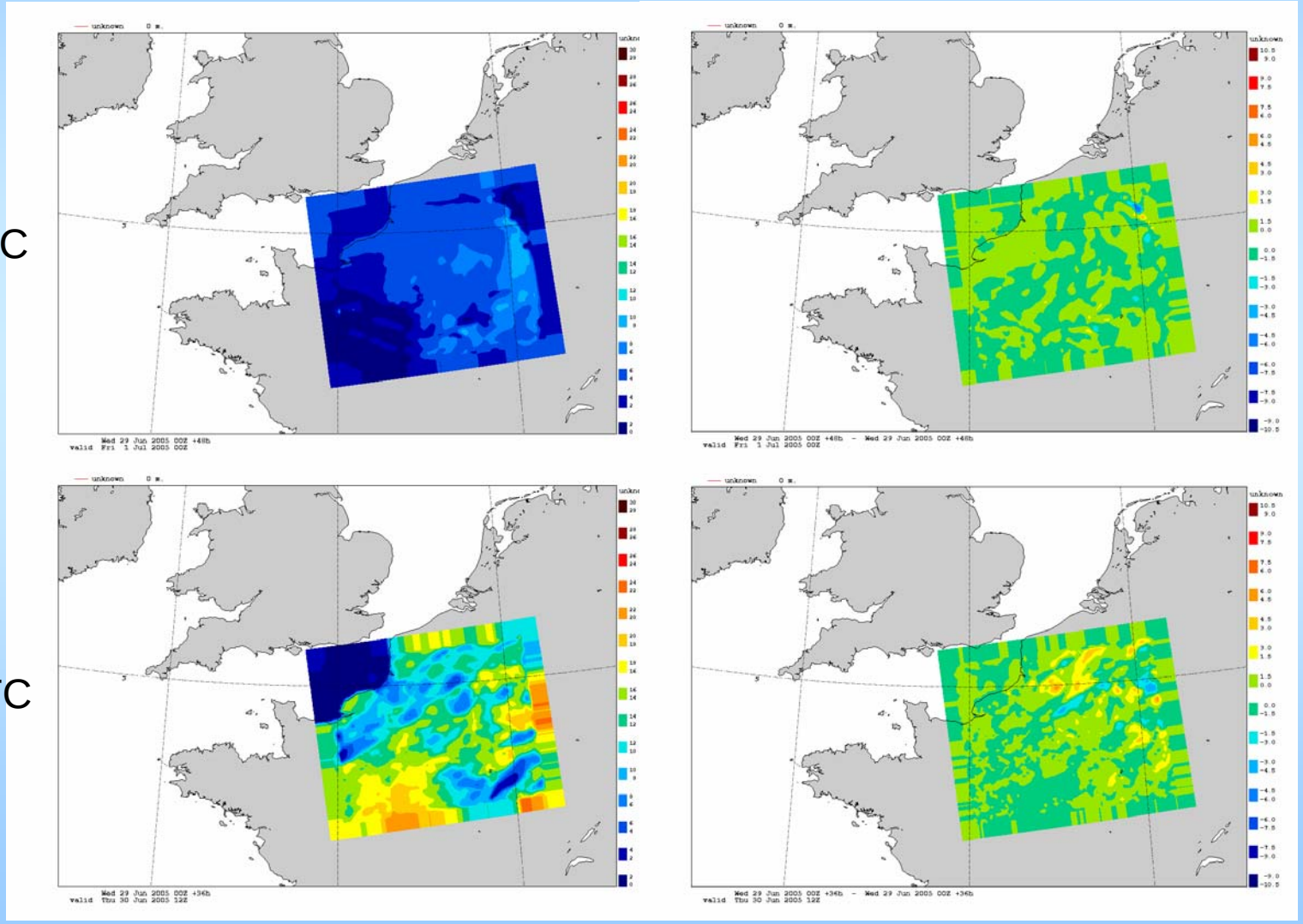
PBL height *(m) X 0.01*

Reference

Including second indirect effect

00 UTC

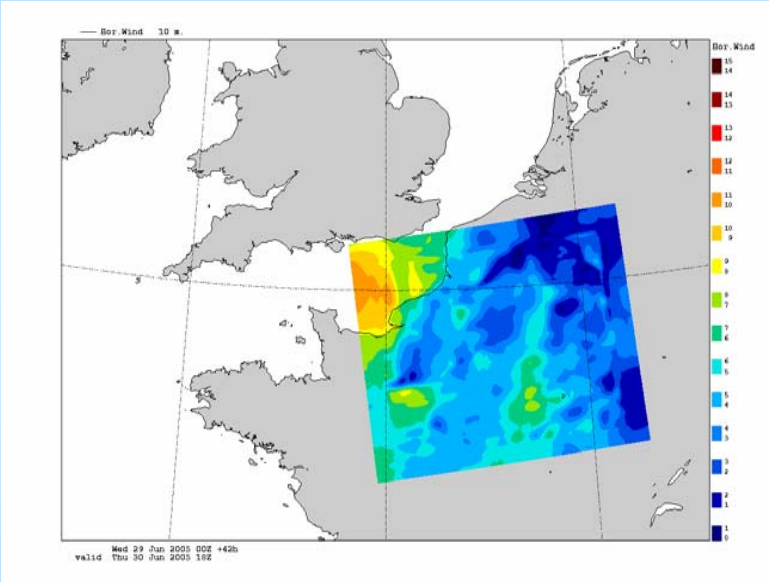
12 UTC



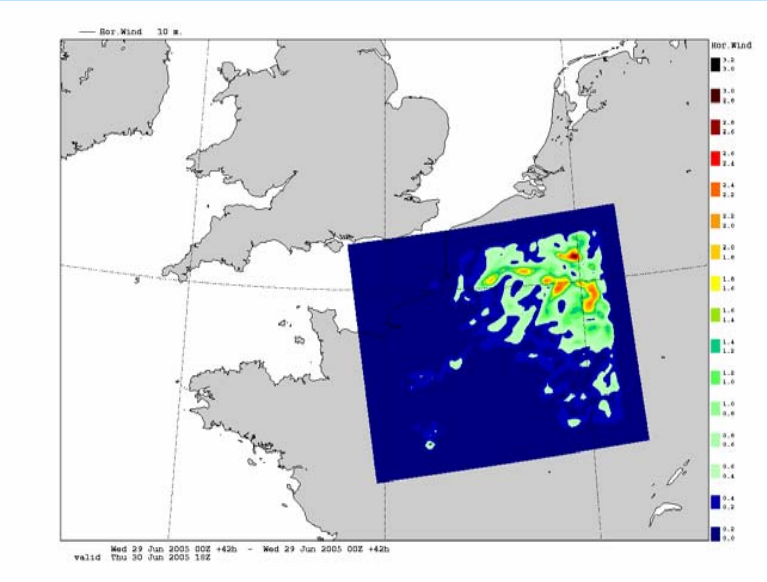
Changes in PBL height quite large (up to 600 m)

10 meter wind (m/s) at 18 UTC

Reference



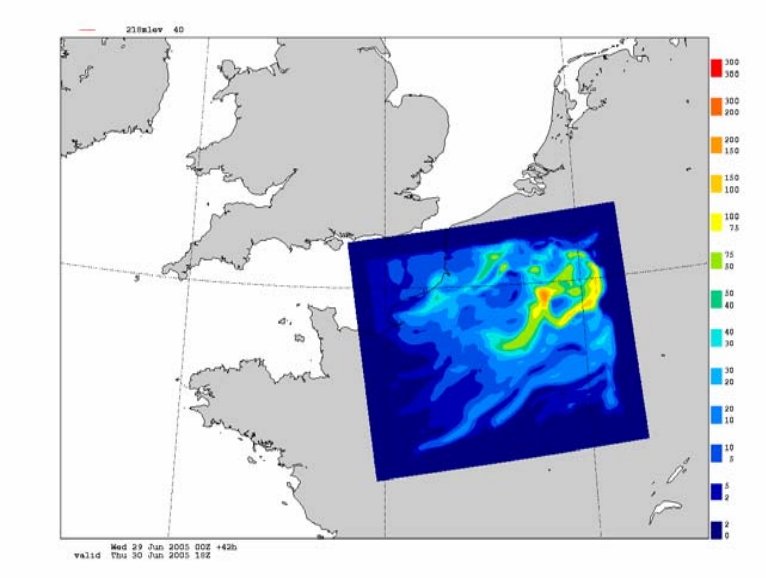
Including second indirect effect



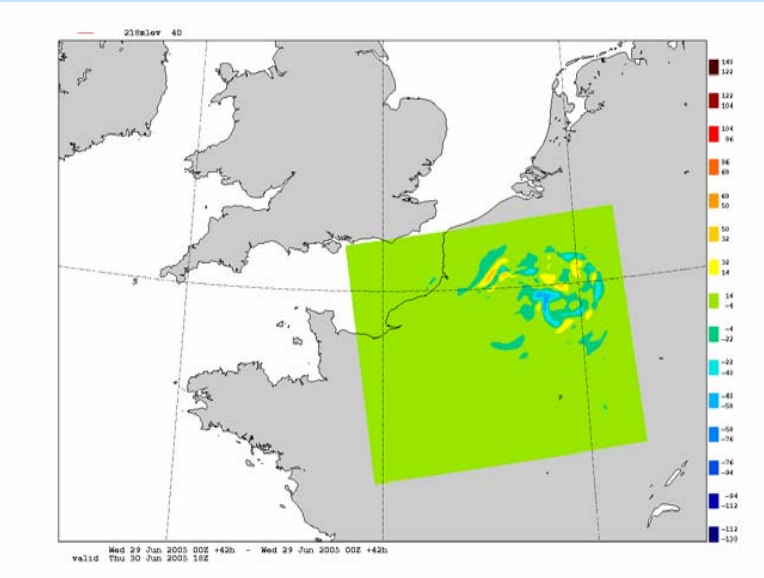
wind changes up to 3m/s

O3 concentartion (micro g/m3) at 18 UTC

Reference

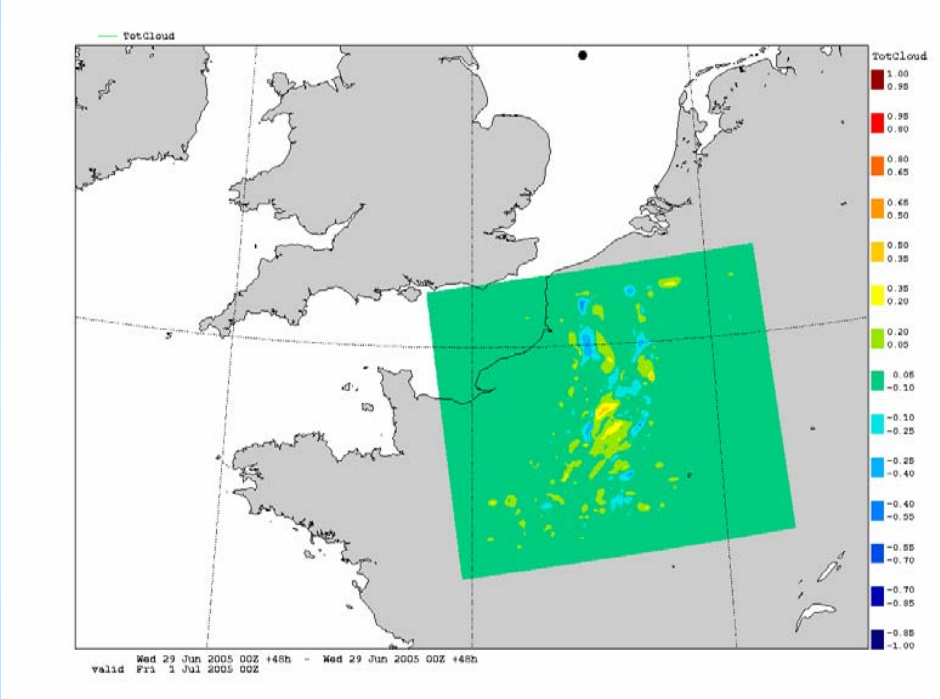


Including second indirect effect



Changes in O3 conc. happened primarily in upper right hand quadrant
O3 not driven by emissions, therefore more dependent on met. factors.

Change in total cloud cover at 00 UTC (%)



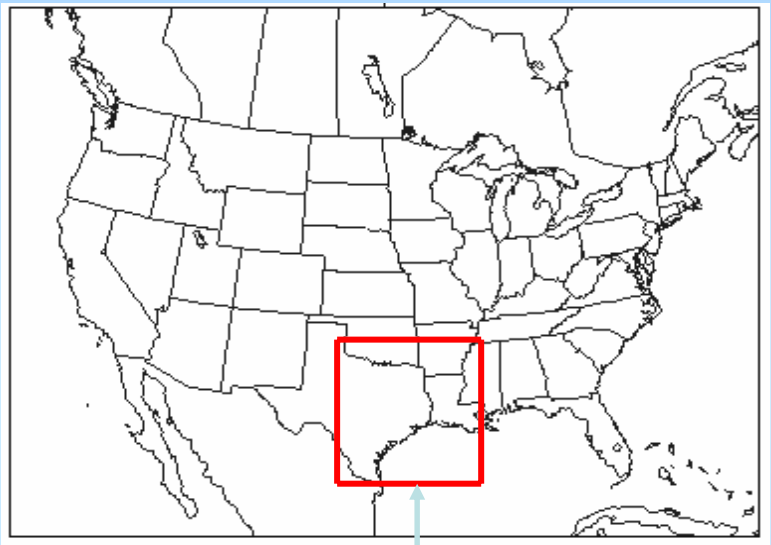
Changes in cloud cover is accounted for by changes in low cloud and no change in high or medium Clouds: seems consistent, since there is no convection of the tracers.

WRF/Chem-MADRID model Study *(Y. Zhang, 2007)*

Model Configurations

July 1-7 2001 CONUS

- **Horizontal resolution: 36 km (148 × 112)**
- **Vertical resolution:**
  - **MM5 (L34), CMAQ (L14)**
  - **WRF/Chem (L34)**
- **Emissions:**
  - **SMOKE: US EPA NEI'99 (v3)**
- **Initial and boundary conditions:**
  - **The same ICs/BCs for WRF/ MM5 and for CMAQ and WRF/Chem**
- **Gas-phase chemical mechanism:**
  - **CMAQ: CB05**
  - **WRF/Chem: CB05 or CBMZ**
- **Data for model evaluation:**
  - **CASTNet and SEARCH**

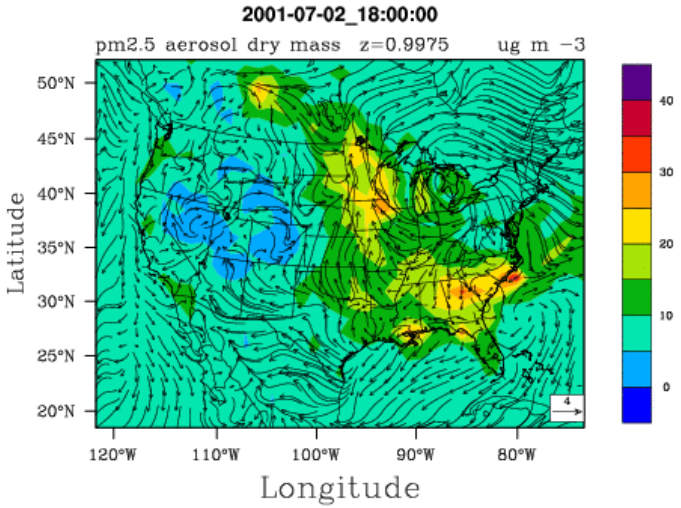


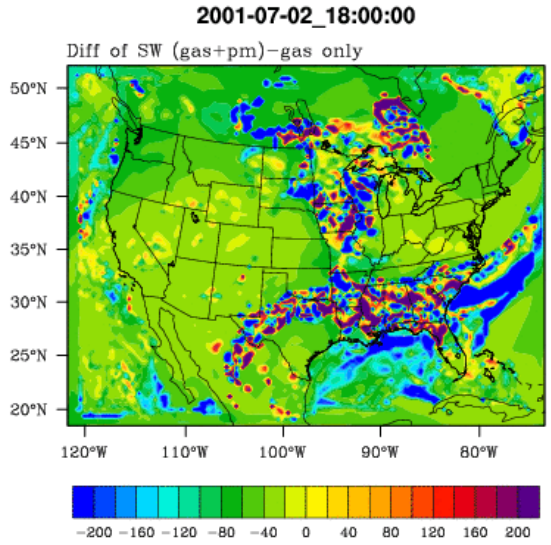
Aug. 28-Sept. 2, 2000 TeXAQS

- **Horizontal resolution: 12 km (88 × 88)**
- **Vertical grid spacing: L57, 15-m at L1**
- **Emissions**
  - **Gases from TCEQ**
  - **PM based on EPA's NEI'99 V. 3 + online s.s.**
- **Initial/boundary conditions**
  - **3-hr N. Amer. reg. reanal. for met.**
  - **Horizontally homogeneous ICs**
- **Gas-phase chemical mechanism: CBMZ**
- **Data for model evaluation**
  - **CASTNet, IMPROVE, AIRS, STN, TeXAQS**

WRF/Chem-MADRID-CBMZ
Effects of Aerosols on Meteorology and Radiation

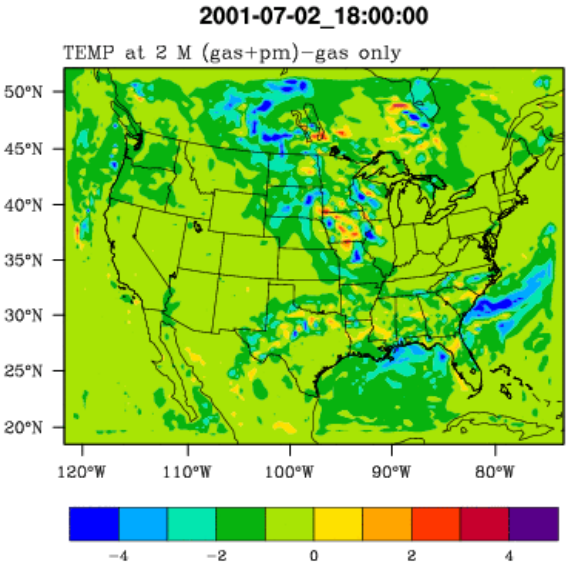
PM2.5

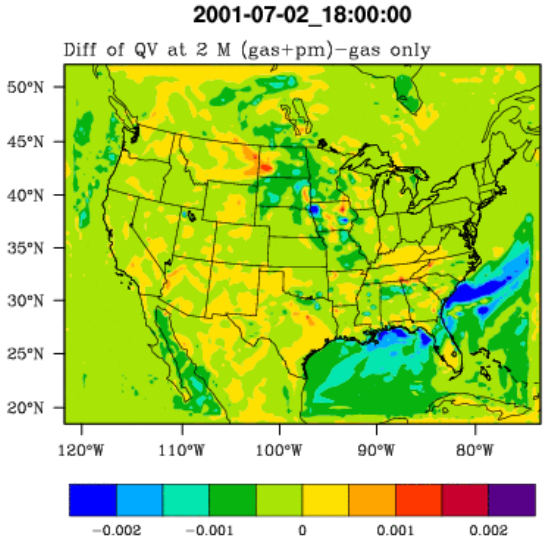




SW Radiation (-20 to 20%)

2-m Temp (-20% to 10%)



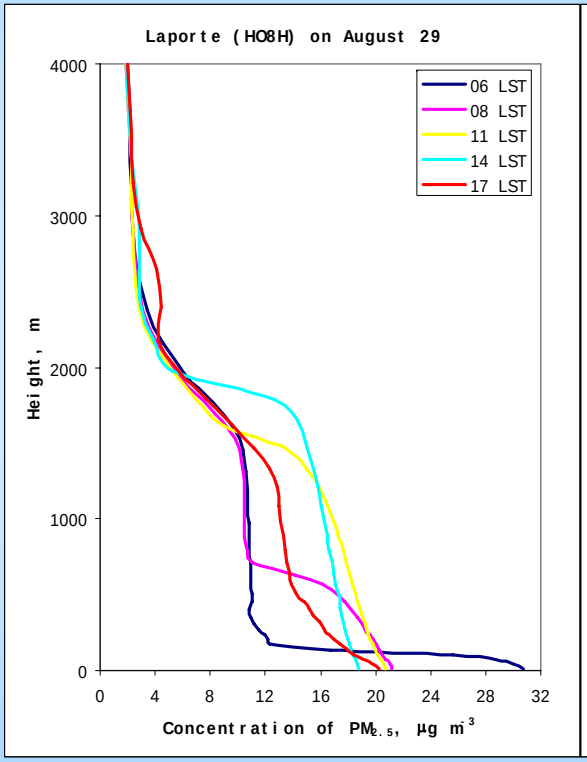


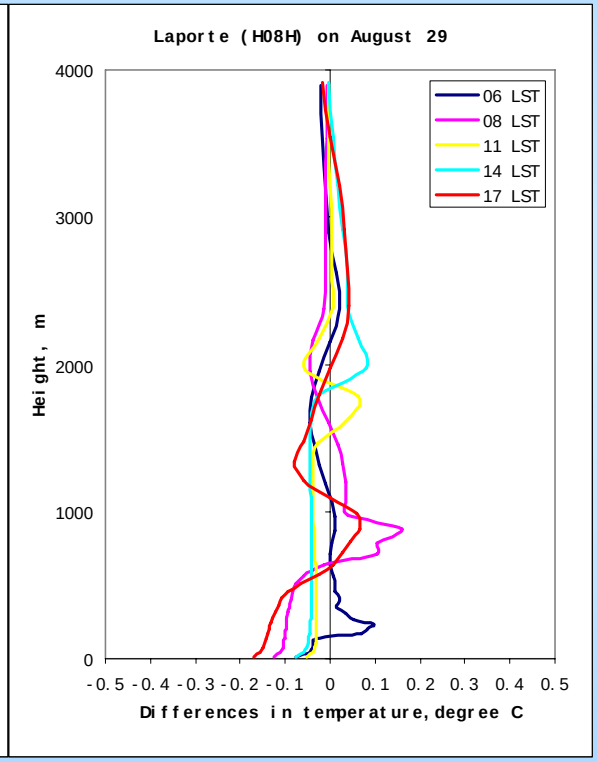
2-m Water Vapor (-10% to 10%)

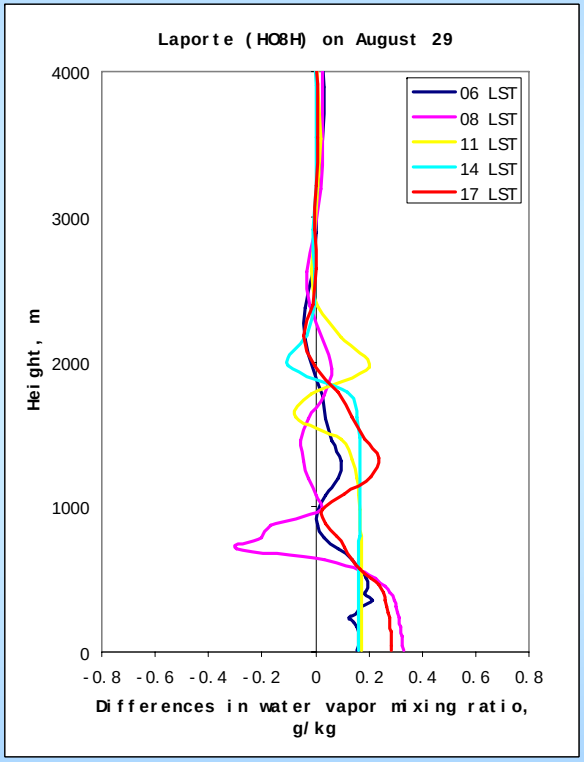
WRF/Chem-MADRID-CBMZ

Feedbacks of Aerosols to T and Q_v at LaPorte, TX

$PM_{2.5}$ (Gas+PM - Gas only)







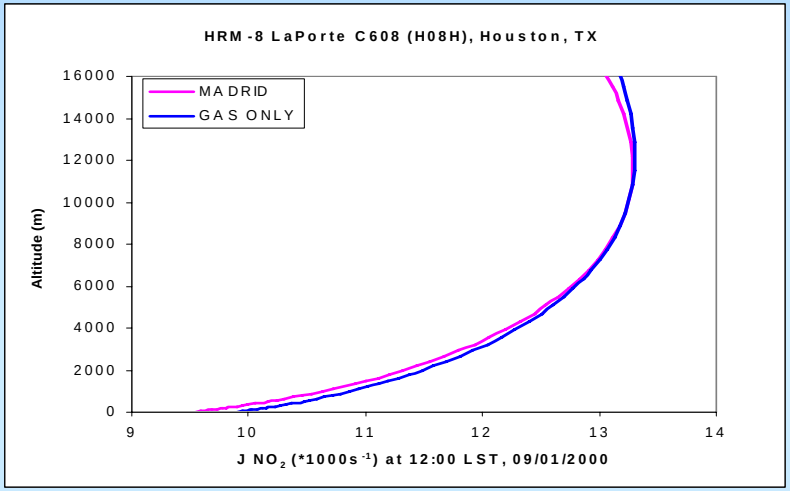
Diff > 0, $T(Q_v)$ increases (decreases) due to aerosol feedbacks

Diff < 0, $T(Q_v)$ decreases (increases) due to aerosol feedbacks

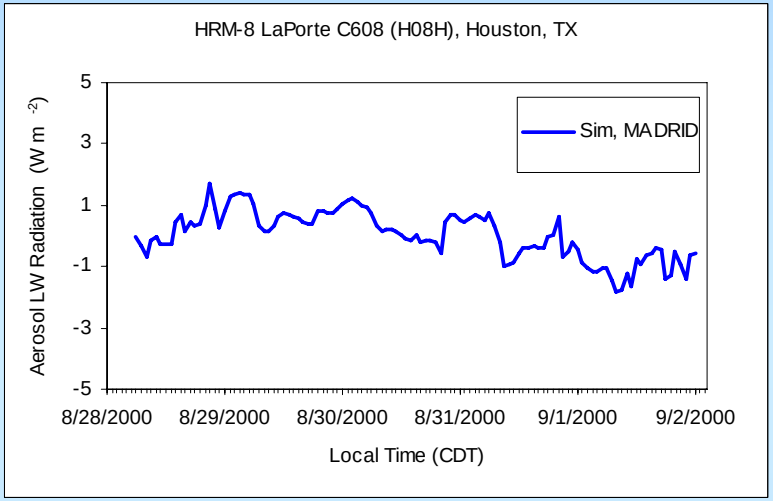
WRF/Chem-MADRID-CBMZ

Feedbacks of Aerosols to NO_2 Photolysis and Radiation

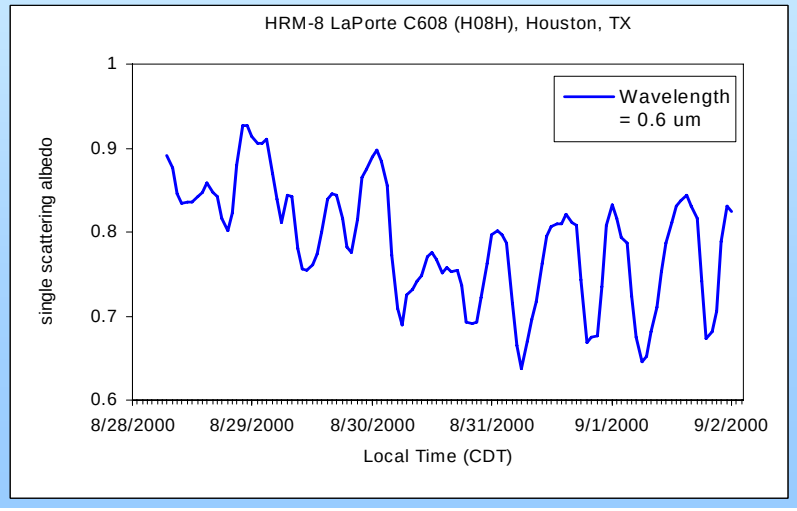
NO_2 Photolysis



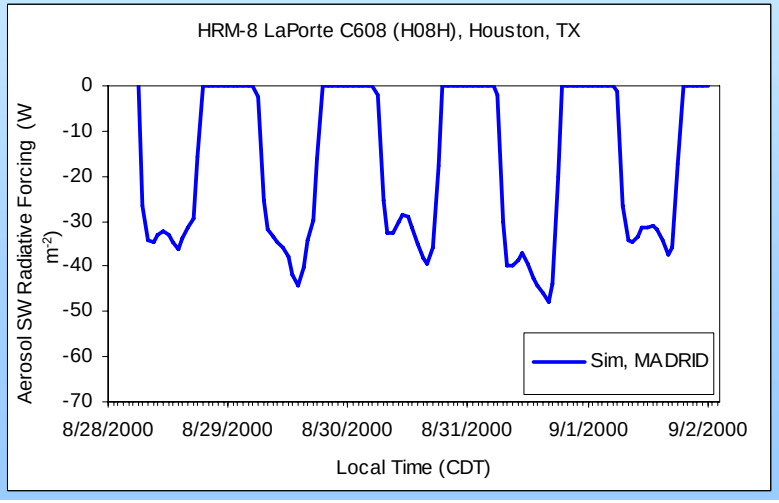
LW Radiative Forcing



Single Scattering Albedo

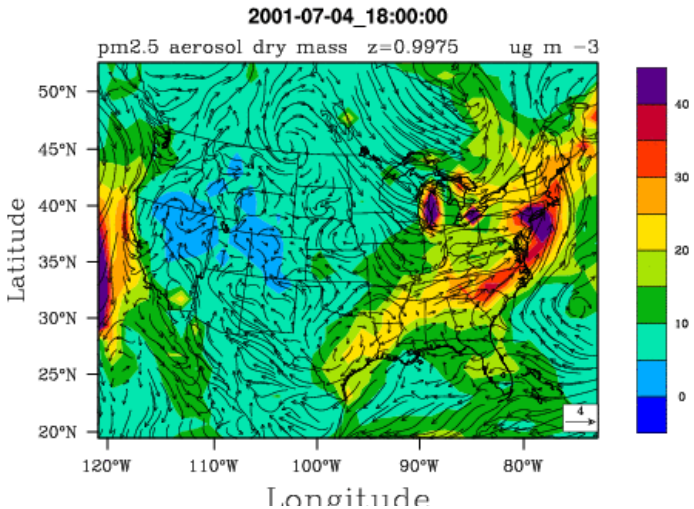


SW Radiative Forcing

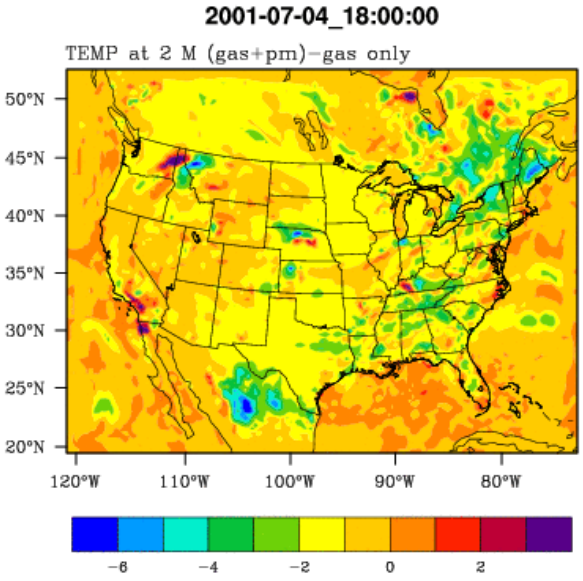


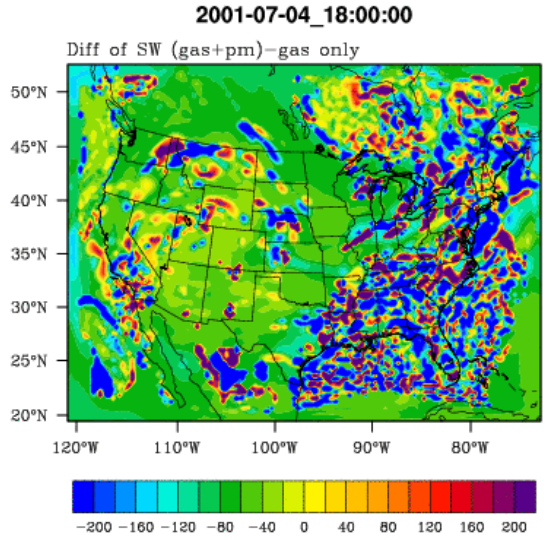
WRF/Chem-MADRID-CBMZ (old):
Effects of Aerosols on Meteorology and Radiation

PM2.5

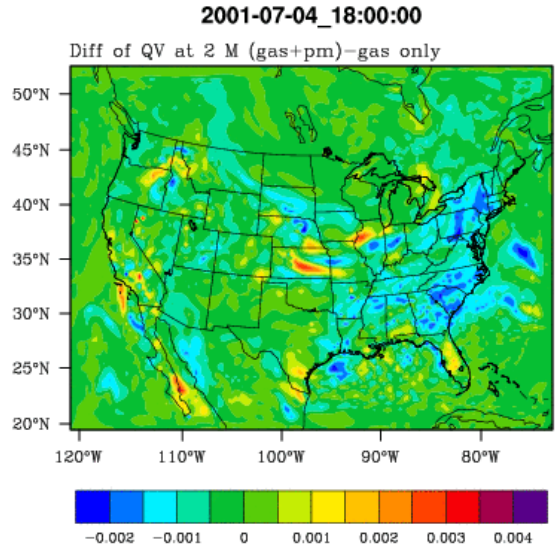


2-m Temp (up to -2.5%)





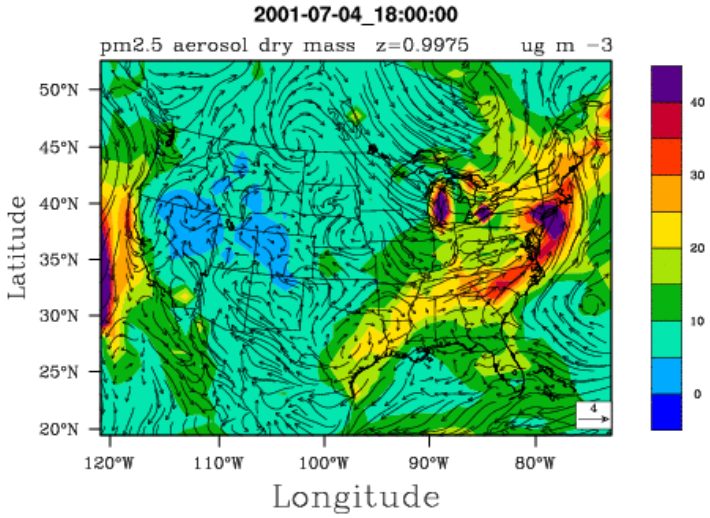
SW Radiation (-20 to 20%)



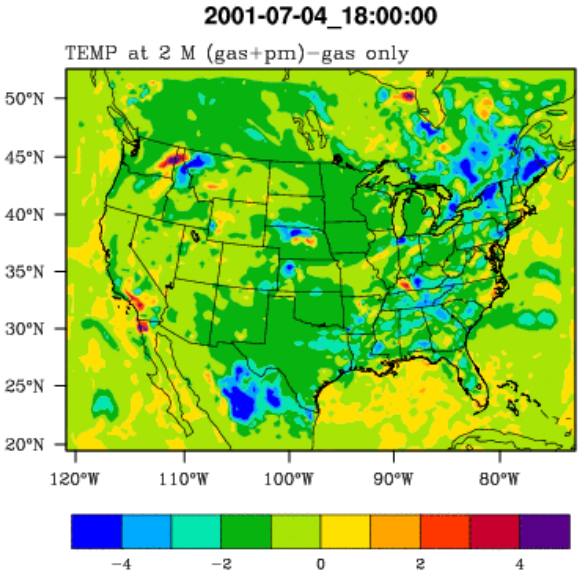
Water Vapor (up to -15%)

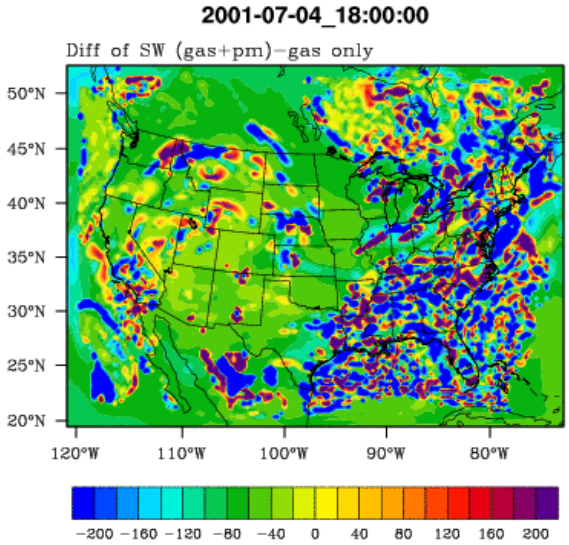
WRF/Chem-MADRID-CBMZ (old): Effects of Aerosols on Meteorology and Radiation

PM2.5

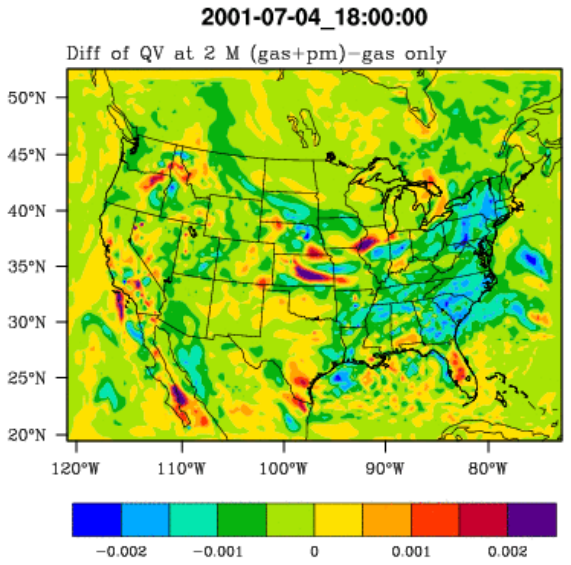


2-m Temp (up to -2%)





SW Radiation (-20 to 20%)



Water Vapor (up to -15%)

Aerosol Production in the Marine Boundary Due to Emissions from DMS

(Gross & Baklanov, 2004)

dmi.dk
DMI

- DiMethyl Sulphide (DMS) is a product of biological processes involving marine phytoplankton.
- DMS is estimated to account for approximately 25% of the total global sulphur released into the atmosphere.
- DMS can be transfered into aqueous-phase aerosols or oxidized to several other gas-phase species which can contribute to aerosol formation and growth, e.g. SO_2, H_2SO_4, dimethyl sulphoxide, dimethyl sulphone methane and sulphinic acid.

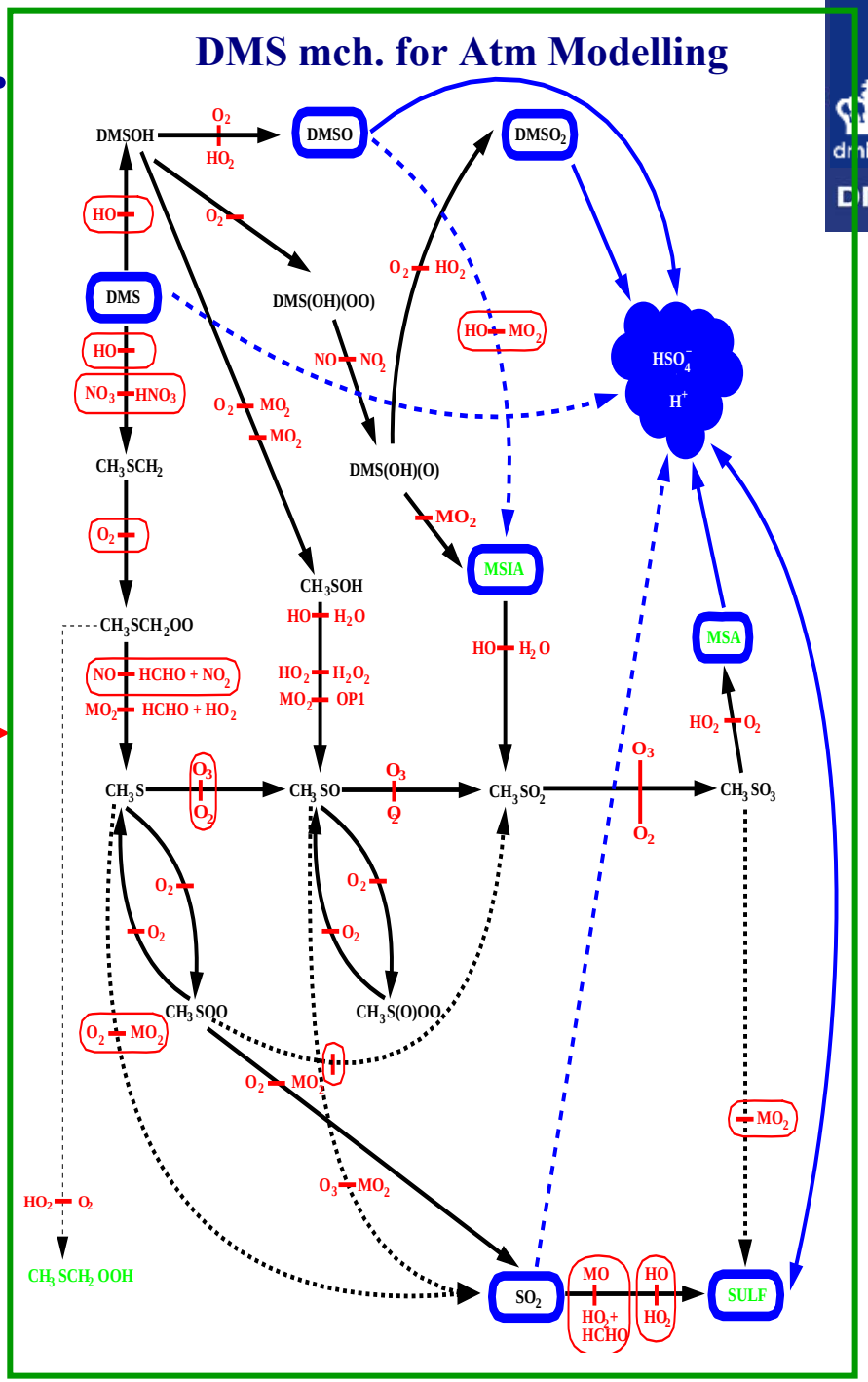
Therefore, it has been postulated that emission of DMS from the oceans can contribute to production of new condensation nuclei and eventually Cloud Condensation Nuclei (CCN). Thus, DMS may have a significant influence on the Earth's radiation budget.

The ELCID gas-phase mch.

A gas-phase DMS mch. was developed during the EU-project period. This DMS mch. included 30 sulphur species and 72 reactions (49 guessed & 23 experimental rates).

Based on clean MBL scenarios the DMS ELCID mch. was reduced to 21 sulphur species and 34 reactions (22 guessed & 12 experimental rates).

The ELCID mch. was further reduced by lumping to 15 sulphur species and 20 reactions. This mechanism was used for 3D modelling in the ELCID project.



The Atmospheric Box-model
(it's a part of Enviro-HIRLAM now)

In the box the following processes are solved for species *i* (which can be either a liquid or gas phases species):

$dC_i/dt =$

+ chemical production – chemical loss

+ emission

– dry deposition – wet deposition

+ entraiment from the free troposphere to the boundary layer

+ aerosol model

+ CCN model + cloud model

Gross and Baklanov, IJEP, 2004, 22, 52

Aerosol Formation and Transformation Processes

The aerosol dynamic model is base on the modal description of the particle distributions suggested by Whitby et al. (1997), i.e. lognormal distributions are used for particle size in each mode.

Analytical solutions are found using the suggestions by Whitby et al. (1997) and Binkowski et al. (2000). These solutions are used in the model.

The present model has three modes: nuclei, accumulation and coarse (coarse mode is not included in this study).

Aerosol Formation and Transformation Processes, Cont.

dmi.dk
DMI

The following aerosol physical processes are solved

Accumulation mode (j):

- **condensation growth, G(j),**
- **intramodal coagulation, C(j→j),**
- **intermodal transfer of moment from nuclei to accumulation mode, C(i→j),**
- **primary emission, E(k,j)**

$$d\,M(k,j)/dt = G(j) - C(j \rightarrow j) + C(i \rightarrow j) + E(j)$$

Nuclei mode (i):

- **nucleation, N(i),**
- **condensation growth, G(i),**
- **intramodal coagulation C(i→i)**
- **intermodal loss of nuclei particles to accumulation mode, C(i→j),**

$$d\,M(i)/dt = N(i) + G(i) + C(i \rightarrow i) + C(i \rightarrow j)$$

Analytical solutions are found using the suggestions by *Whitby et al. (1997)* and *Binkowski et al. (2000)*, and these solutions are used in the model.

Clean MBL Scenarios Simulated in the Study

Meteorological Conditions:	
Ground Albedo	0.10
Pressure (mbar)	1013.25
Relative Humidity	90 %
Cloud Frequency	1 d^{-1}
Precipitation Frequency	0.1 d^{-1}
Temperature (K)	288.25

Initial Aerosol Distribuitions:	
Nuclei Mode:	
Number conc.	133 cm^{-3}
log(σ)	0.657
Geo. Mean Dia .	0.8×10^{-6} cm
Accumulation mode:	
Number conc.	66.6 cm^{-3}
log(σ)	0.21
Geo. Mean Dia.	$.266\times10^{-4}$ cm

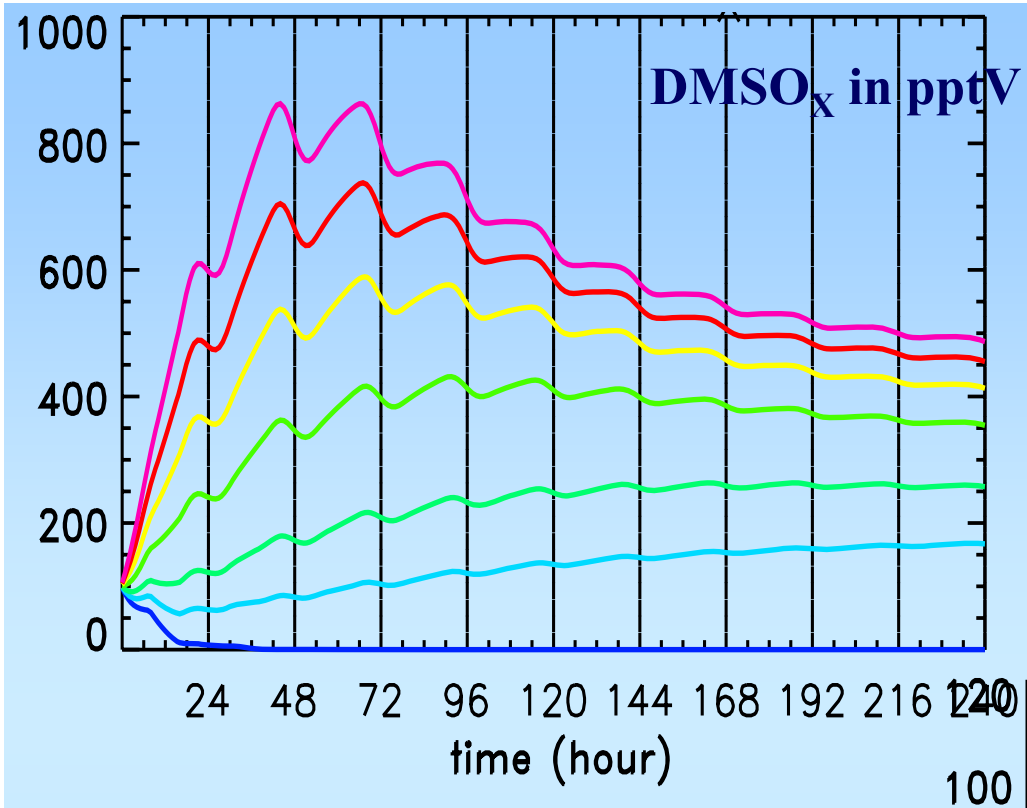
Initial Gas-Phase Conc.:	
H_2	2 ppmV
CH_4	1.7 ppmV
CO	0.14 ppmV
H_2O	3 %
N_2	78%
O_2	20 %
NO_2	400 pptV
H_2O_2	1 pptV
HO_2	0 pptV
CH_3O_2	0 pptV
HNO_3	150 pptV
O_3	40 ppbV
HCHO	10 pptV
VOC	5.5 ppbC
SO_2	2 pptV
DMS	100 pptV
MSA	1 pptV

Emission of SO_2 in pptV/min: 0.014

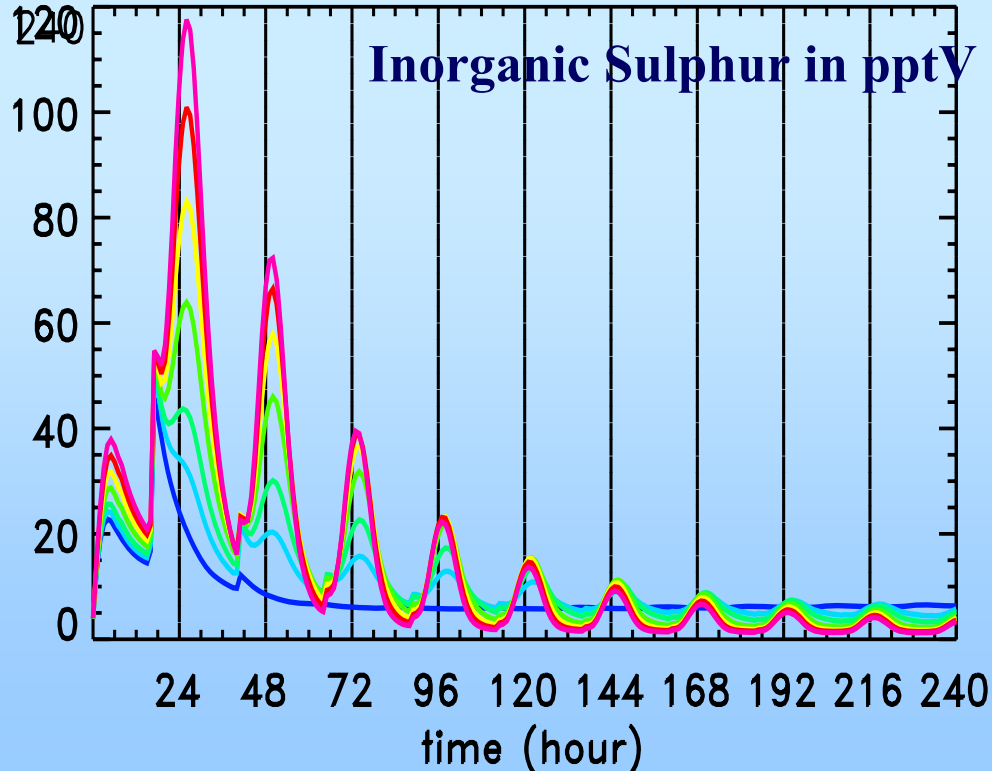
Emission of DMS in pptV/min: 0.00, 0.06, 0.12, 0.24, 0.36, 0.48, and 0.60

- **The RACM mechanism (Stochwell et al., JGR, 1997, 102, 25847) is coupled to ELCID mechanism in order to simulate the non-sulphur chemistry properly.**

- **The chemical composition of typically MBL aerosols was used to set up the scenarios.**

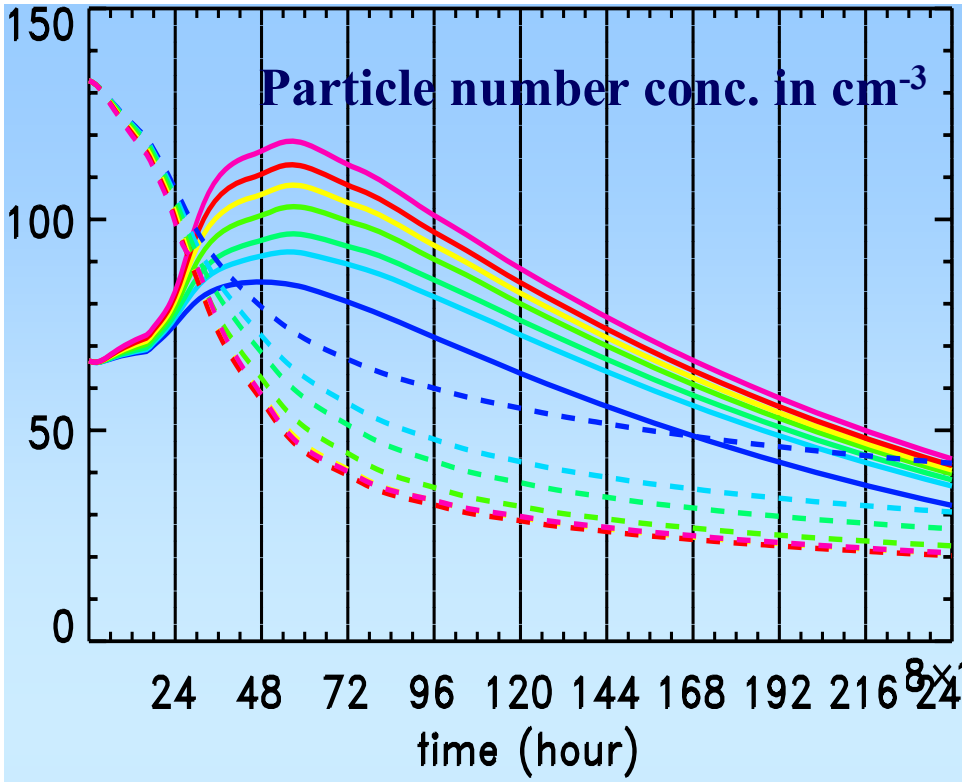


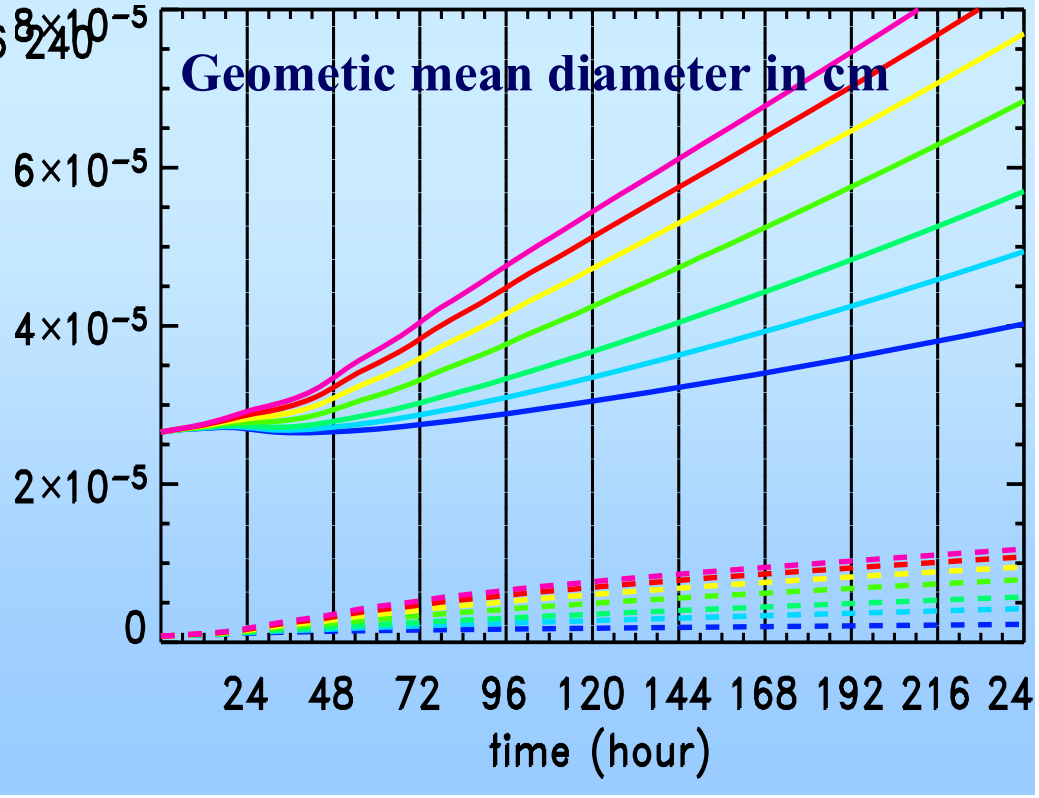
Emissions of DMS is varied from 0.0 (blue) to 0.6 (purple) pptV/min



Emis. of SO_2 = 0.014 pptV/min for all the simulations

Gross and Baklanov, IJEP, 2004, 22, 51

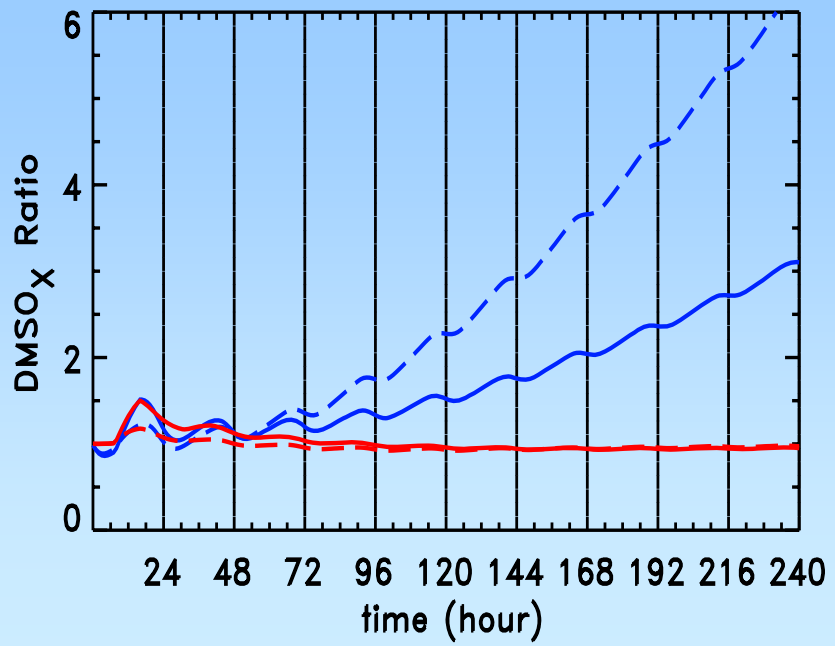


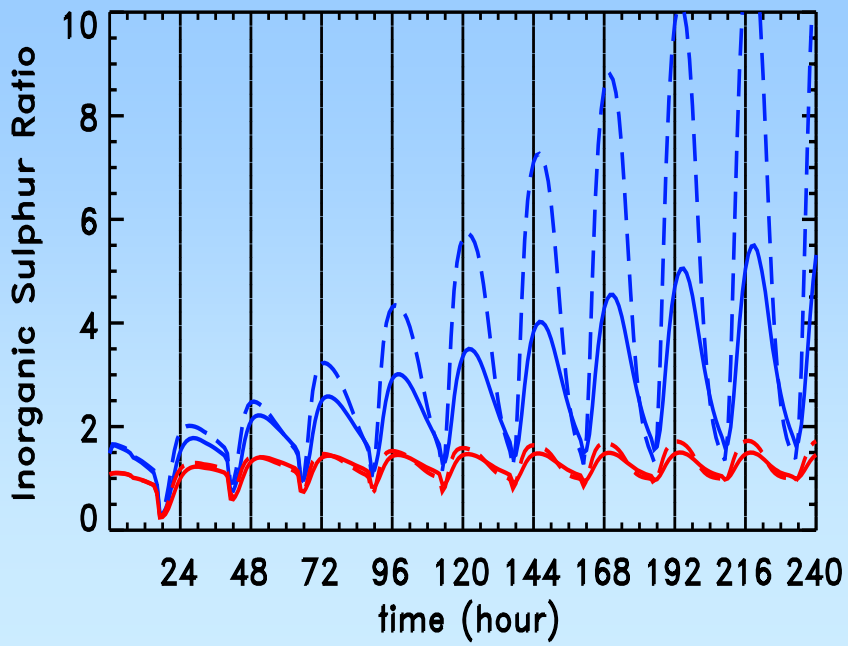


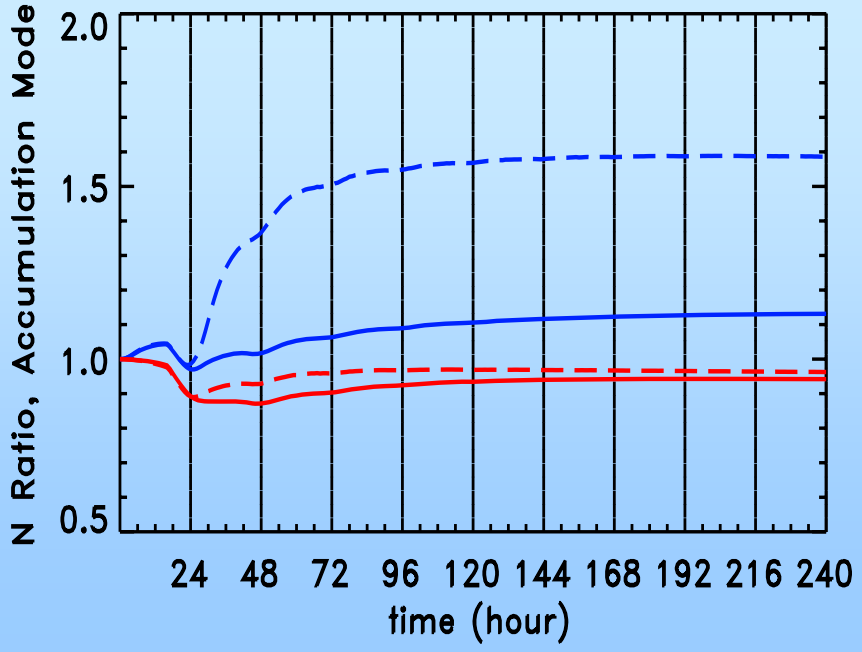
Solid line: accumulation mode

Dashed line: nuclei mode

Gross and Baklanov, IJEP, 2004, 22, 51





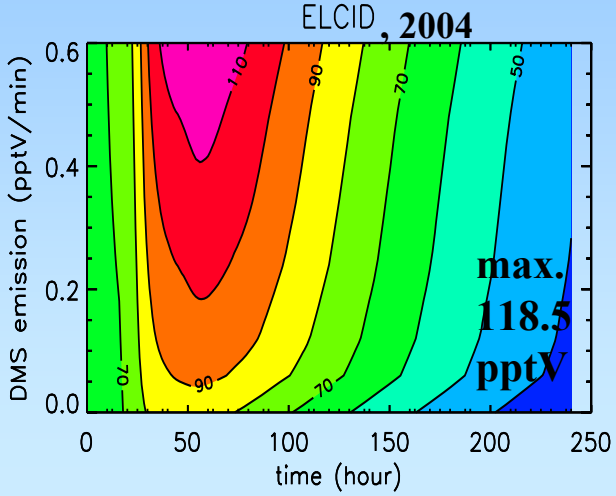


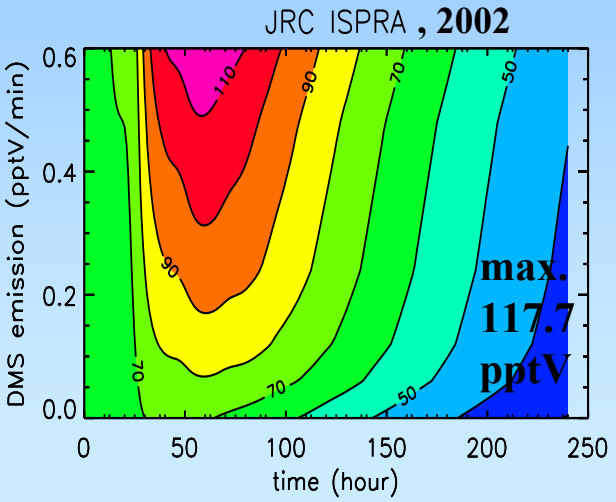
Dashed lines: DMS emis. = 0.36 pptV/min.
Solid lines: DMS emis. = 0.12 ppt./min.

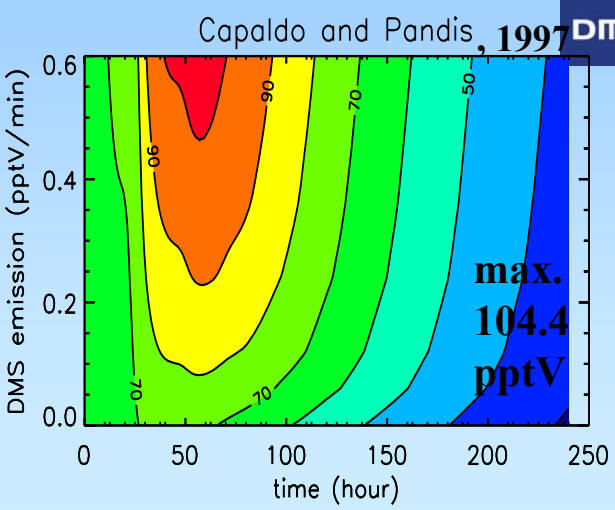
Blue lines: Russell et al. mch./ELCID mch.
Red lines: Saltelli and Hjort mch./ELCID mch.

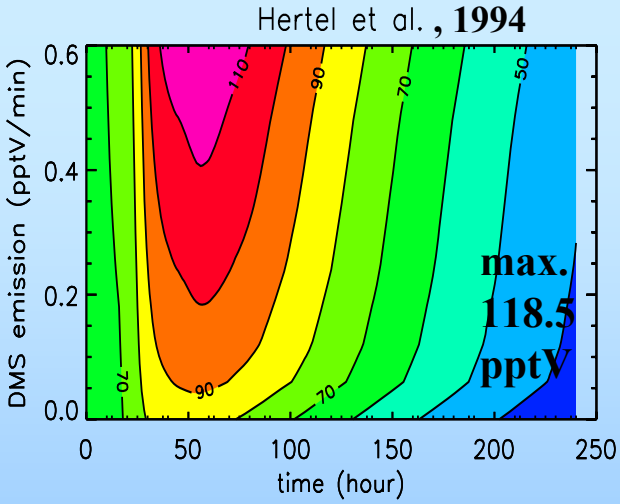
The results shown at the figures are identical with the main conclusions from the five studies are compared.

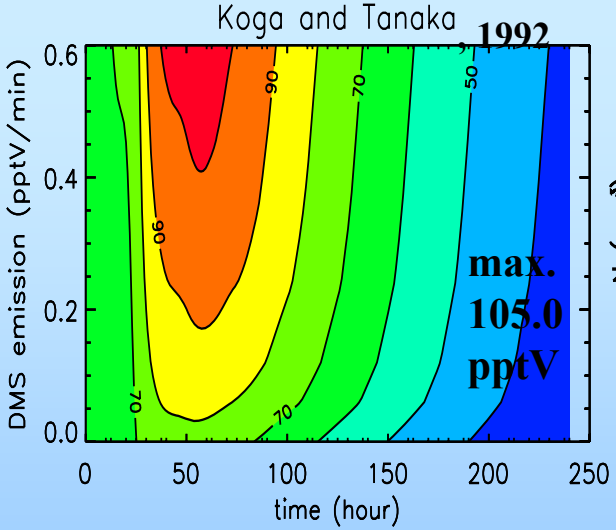
Particle number concentration (cm^{-3}), Accumulation mode

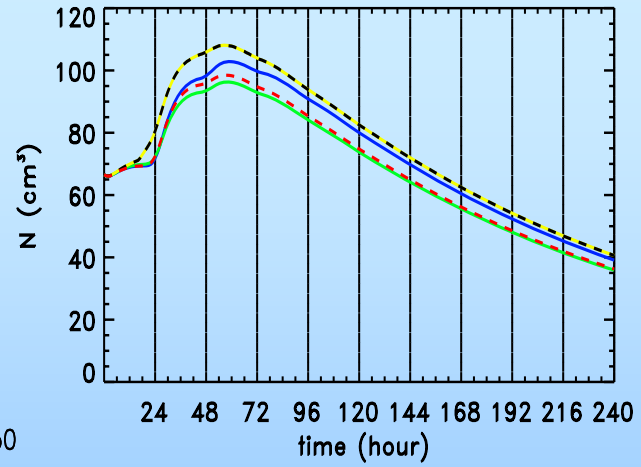












Contour levels from 10 to 120 cm^{-3}, increment interval 10 cm^{-3}

DMS emis. = 0.36 ppt/min: ELCID: —
JRC ISPRA: —, Cap&Pan: —
Hertel et al.: — , Kog&Tan: —

Summary, the simulations Showed

- DMS can roughly contribute from 13% to 27% (summer period) and 3% to 13% (winter period) of the formation of non sea salt aerosols.
- DMS can roughly contribute from 10% to 18% (summer period) and 1% to 10% of the total formation of aerosols.
- Too simplified DMS chemistry [DMS(g)+HO(g)->SO2(g)-> H2SO4(l)] create too many new accumulation mode particles.
- The DMS mechanism comparison showed that all five mechanism gave all most the same amount of aerosols. We assume that the small difference simulated by the five different mechanisms cannot be observed if the mechanisms are applied to 3-D modelling, i.e. we conclude that the five mechanisms are equally good for 3-D modelling.

However, all these DMS mechanisms are based on the same guessed rates and reactions, i.e. the same amount of uncertainty.

DMS chemistry, Resent Results

- A resent ab initio/DFT study (Gross et al., JPC A, 2004) shows:
  1. $DMSOH + O_2 \rightarrow DMSO + HO_2$ (the dominant channel)
  2. $DMSOH + O_2 \rightarrow DMS(OH)(OO)$ (occur, minor channel)
  3. $DMSOH + O_2 \rightarrow CH_3SOH + CH_3O_2$ (does not occur)

  However, in DMS mechanisms channels 1 and 2 are often considered to be equal important, and channel 3 is included.
- Simulations of DMS chamber experiments (which were performed at different temperatures and NO_X concentrations) indicate that we still not fully understand the chemistry of the additional DMS+HO channel. *Important chemical mechanisms are missing.* (Gross and Barnes, unpublished results).

Conclusion

- The DMS chemistry is still highly uncertain.
- Many parameters used to described the mass transport of $DMSO_X$, MSA, MSIA etc. to aerosols and its aerosol physics are still uncertain/unknown.

- No conclusion yet !

- It’s task for future work (maybe your?)!

Recommended literature:

- Baklanov, A., A. Mahura, R. Sokhi (eds.), 2008: Integrated systems of meso-meteorological and chemical transport models, Materials of the COST-728/NetFAM workshop, DMI, Copenhagen, 21-23 May 2007, 183 pp. Available from: http://www.cost728.org
- Korsholm U.S., A. Baklanov, A. Gross, A. Mahura, B.H. Sass, E. Kaas, 2008: Online coupled chemical weather forecasting based on HIRLAM – overview and prospective of Enviro-HIRLAM. *HIRLAM Newsletter*, 54: 1-17.
- Zhang, Y., 2008: Online-coupled meteorology and chemistry models: history, current status, and outlook. *Atmos. Chem. Phys.*, 8, 2895–2932
- Baklanov, A. and U. Korsholm: 2007: On-line integrated meteorological and chemical transport modelling: advantages and prospective. In: ITM 2007: 29th NATO/SPS International Technical Meeting on Air Pollution Modelling and its Application, 24 – 28.09.2007, University of Aveiro, Portugal, pp. 21-34.
- Gross, A., Baklanov A., 2004: Modelling the influence of dimethyl sulphide on the aerosol production in the marine boundary layer, *International Journal of Environment and Pollution*, 22(1/2): 51-71.