



## A model inter-comparison study focussing on episodes with elevated PM10 concentrations

#### R. Stern<sup>1</sup>

- P. Builtjes<sup>1,2</sup>, M. Schaap<sup>2</sup>, R. Timmermans<sup>2</sup>, R.Vautard<sup>3</sup>, A. Hodzic<sup>4</sup>, M. Memmesheimer<sup>5</sup>, H. Feldmann<sup>5</sup>, E. Renner<sup>6</sup>, R. Wolke<sup>6</sup>, A. Kerschbaumer<sup>1</sup>
- (1) Freie Universität Berlin, Institut für Meteorologie, Germany
- (2) TNO, Apeldoorn, The Netherlands
- (3) LSCE/IPSL Laboratoire CEA/CNRS/UVSQ, 91191 Gif sur Yvette Cedex, France
- (4) LMD/IPSL Ecole Polytechnique 91198 Palaiseau Cedex, France
- (5) Rheinisches Institut für Umweltforschung an der Universität zu Köln, Germany
- (6) Leibniz-Institut für Troposphärenforschung, Leipzig, Germany





## **Status PM10 Modelling**

# Most models underestimate observed PM10 concentration levels !!!

## Result of several <u>long-term</u> model performance studies in Europe: EURODELTA, CITYDELTA, REVIEW OF THE UNIFIED EMEP MODEL





## Do we know the reasons ??

## **Severals suspects:**

- Underestimation or missing of sources
- Uncertainties in the treatment in aerosol chemistry and microphysics
- Inaccurate meteorological predictions

It is very difficult to attribute the PM underestimation to a certain source of error, in particular with long-term studies !!





## **Objective of this study**

# Assessment of the ability of models to reproduce PM10 concentrations under highly-polluted conditions

## Why episodes?

- It is easier to examine processes
- European air quality problem: Violation of the PM10 shortterm limit value (daily mean PM10 concentration > 50 µg/m3).

# Models as tools for air quality planning should be able to predict the high PM concentrations !!





# Five 3-d chemical transport models of different complexity

- CHIMERE, France
- LOTOS-EUROS (LOng Term Ozone Simulation-EURopean Operational Smog) model, The Netherlands
- EURAD (European Air Pollution Dispersion model), Germany
- LM-MUSCAT (Multi-Scale Chemistry Aerosol Transport ), Germany
- RCG (REM-CALGRID), Germany

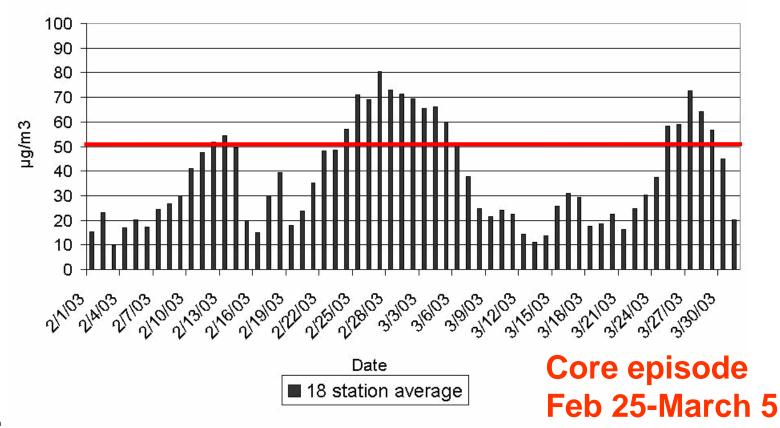




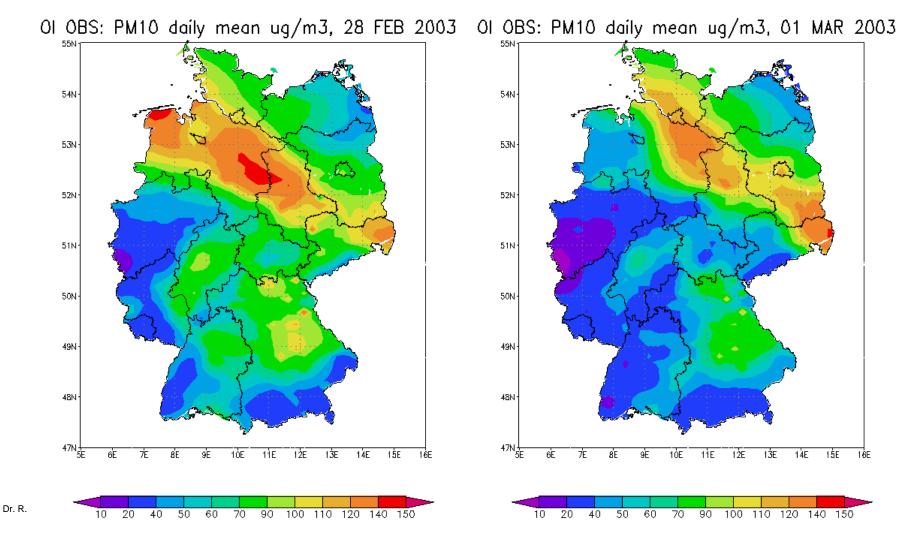
#### **Selected time period:**

#### January 15 thru April 5 2003 containing three distinct episodes NORTHERN GERMANY

Observed PM10 Daily Mean in Northern Germany Feb-March 2003



#### Troposphärische Umweltforschung Institut für Meteorologie Freie Universität Berlin Core episode: very pronounced PM10 concentration gradient across Germany over several days



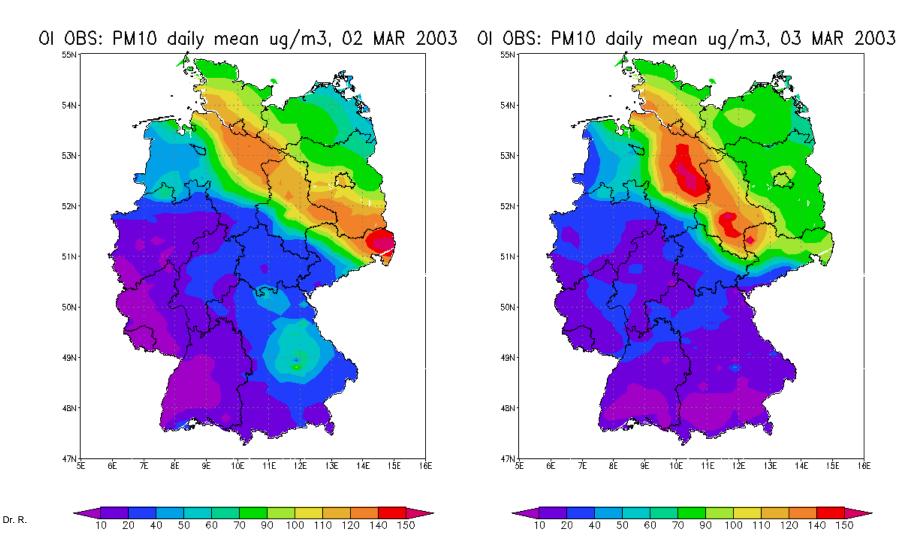


Troposphärische Umweltforschung

Institut für Meteorologie Freie Universität Berlin



# Observed daily mean PM up to 200 µg/m3 at rural background stations

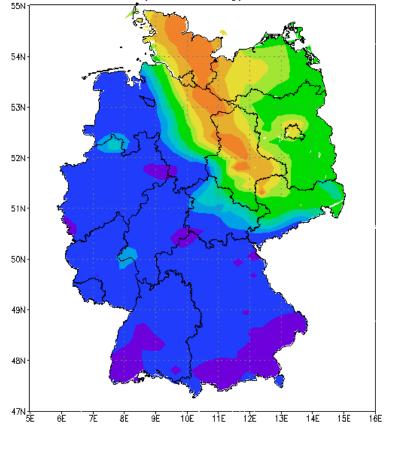




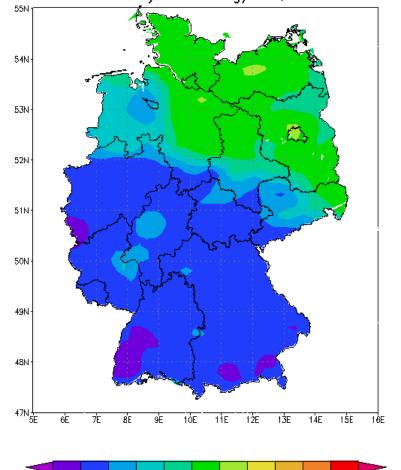


# High PM10 field is moved to the Northeast by a frontal system

OI OBS: PM10 daily mean ug/m3, 04 MAR 2003 OI OBS: PM10 daily mean ug/m3, 05 MAR 2003



100 110 120



90 100

110 120

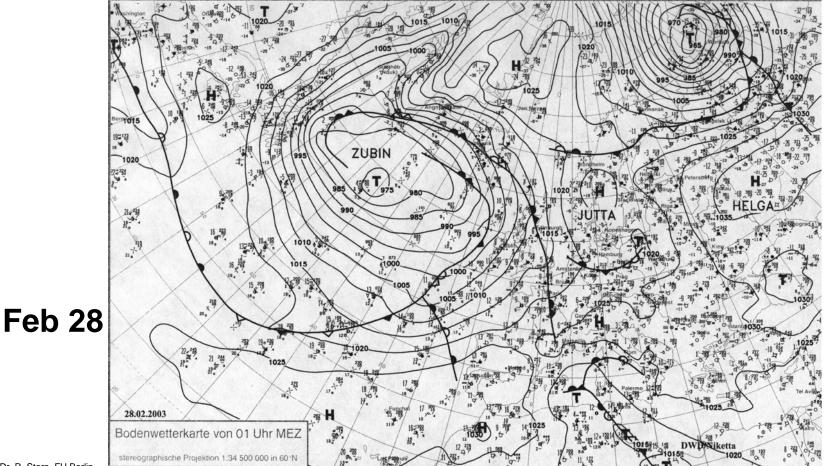
140 150

Dr. R.





# All episodes are connected with high pressure systems, stable conditions, low wind speeds





## **Model configurations**



#### All models used the same emissions data base including time factors

and height distribution and were applied on the regional scale

#### covering the central parts of Europe

- Horizontal resolutions between 25 and 35 km
- Vertical layers
  - EURAD: 15 below 2 km,
- LM-MUSCAT: 17 below 4 km
- CHIMERE: 8 up to 500 mb;
- RCG: 5 up to 3 km,

- LOTOS-EUROS: 4 up to 3 km.
- Different meteorological drivers
  - prognostic NWP model: MM5 (EURAD, CHIMERE), LM (LM-MUSCAT)
  - diagnostic interpolation scheme of observations coupled with a PBL model:

RCG, LOTOS-EUROS



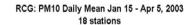


## Evaluation against observations at rura stations in the belt of high PM concentrations in Northern Germany

18 stations PM10, 4 stations PM2.5

4 stations: SO4, total nitrate (HNO3+NO3) and total ammonia (NH3+NH4)

1 research site (Melpitz): SO4, NO3, NH4, EC, OM, NH3 In addition: NO2, SO2 at the 5 stations with PM components Focus on the 5 stations with PM composition data

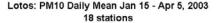


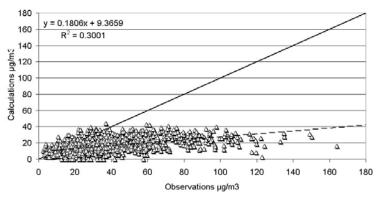
;m/gu

ulations

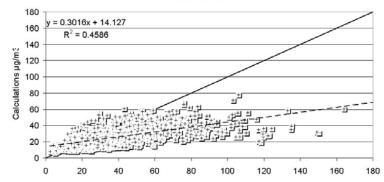
ole:

## Scatter of daily mean PM10 18 stations

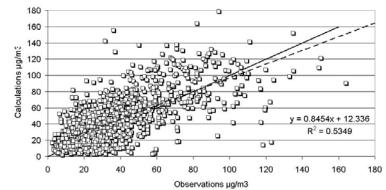




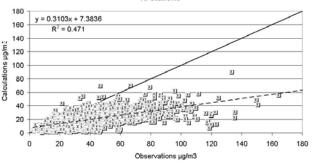
LM-MUSCAT: PM10 Daily Mean Jan 15 - Apr 5, 2003 18 stations



EURAD: PM10 Daily Mean Jan 15 - Apr 5, 2003 18 stations



Chimere: PM10 Daily Mean Jan 15 - Apr 5, 2003 18 stations

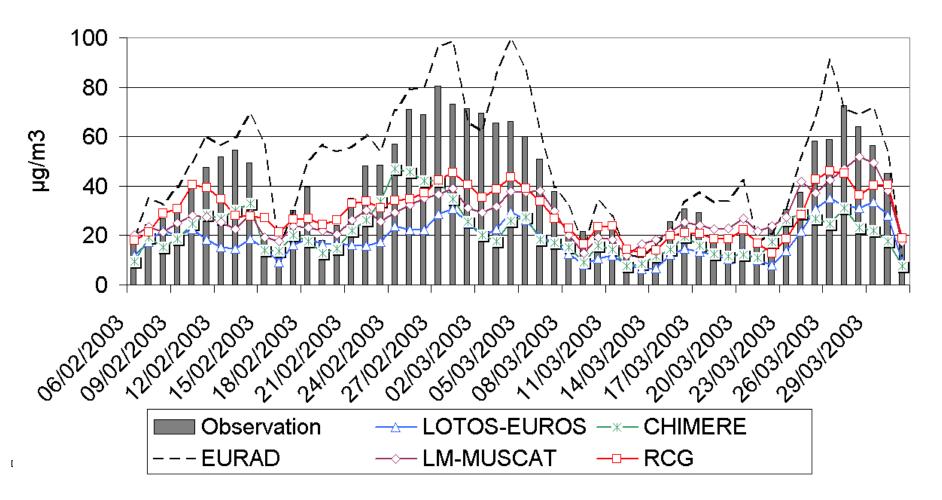


4 models (RCG, LOTOS, CHIMERE, LM-MUSCAT) clearly underestimate observed

#### TrUmF Troposphärische Umweltforschung All models recognize the Institut für Meteorologie Freie Universität Berlin episodes, but do not get the peak values (only EURAD)



PM10







# Analysis of precursor and primary PM concentrations

SO2: tendency to overestimate, in particular the low values (EURAD

more pronounced)

NO2: tendency to underestimate observed peak concentrations (EURAD partly strong overestimation)

NH3 : RCG, CHIMERE, LOTOS right order of magnitude, LM-MUSCAT and in particular EURAD overestimate (only 1 station)

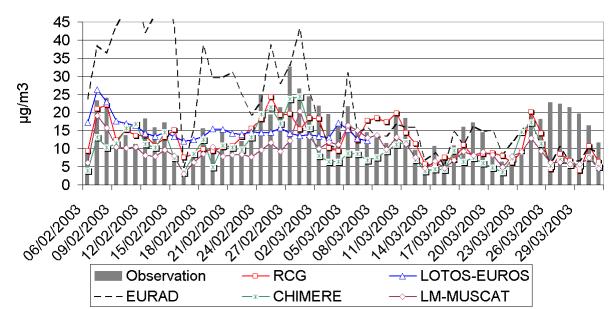
Dr. R. Stern, FU Be EC: All models strongly underestimate (only 1 station)

## Example: MELPITZ, but the picture is similiar at other stations

SO2

30 1 1 1 1 25 11 20 hg/m3 П  $\times$ 15 10 5 0 1210212003 1510212003 1810212003 2410212003 0210312003 08/03/2003 1410312003 2010312003 2610312003 06/02/2003 0310212003 2110212003 2710212003 0510312003 1103/2003 1710312003 2310312003 2910312003

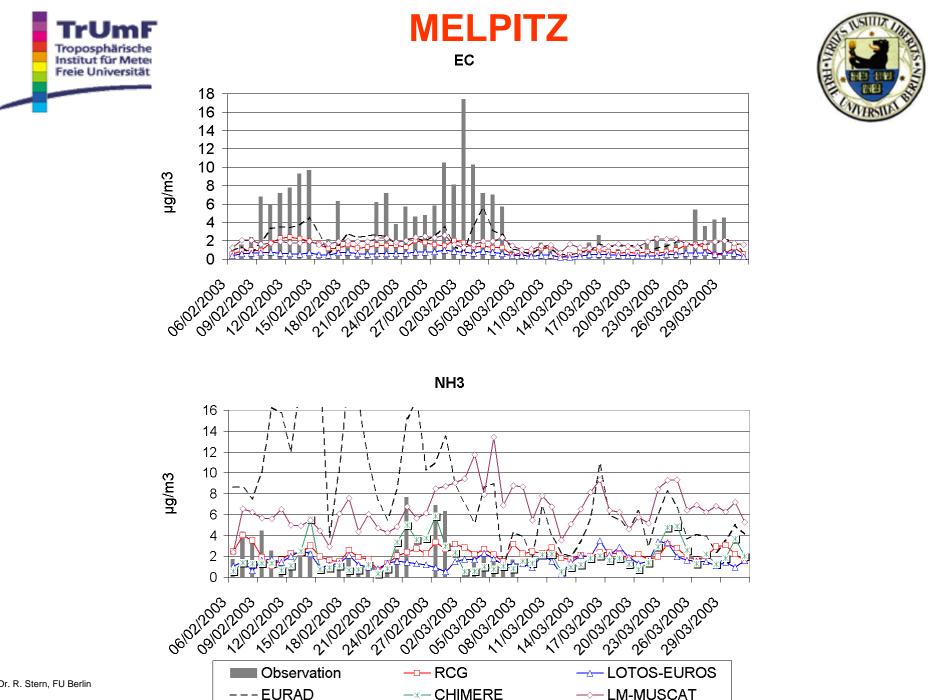
#### NO2





ſrUmF

Troposphärische L Institut für Meteor Freie Universität B







## Analysis of secondary PM components

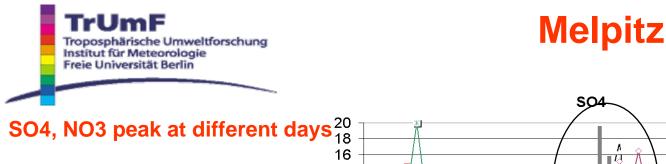
#### SO4: underestimation of the observed peaks, timing problems.

#### **LOTOS underestimates**

# NO3, NH4: underestimation of the observed peaks, timing problems. <u>EURAD strongly overestimates</u>

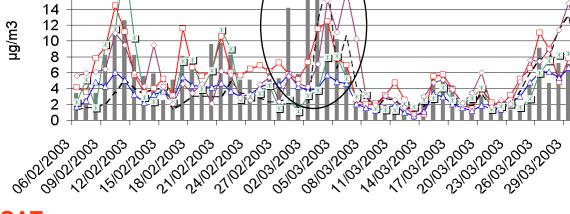
**OM : RCG underestimates, EURAD, LM-MUSCAT overestimate** 

(no data from CHIMERE, LOTOS, only 1 station)



Problems with the position of the front ?

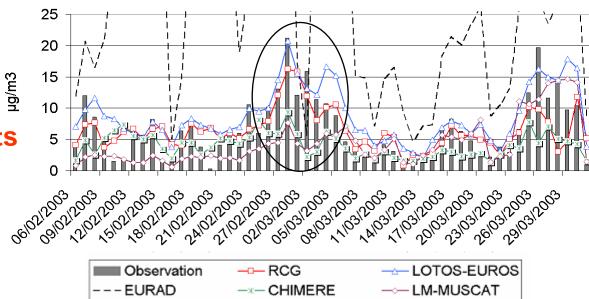
More pronounced for the prognostic drivers



EURAD, CHIMERE, LM-MUSCAT

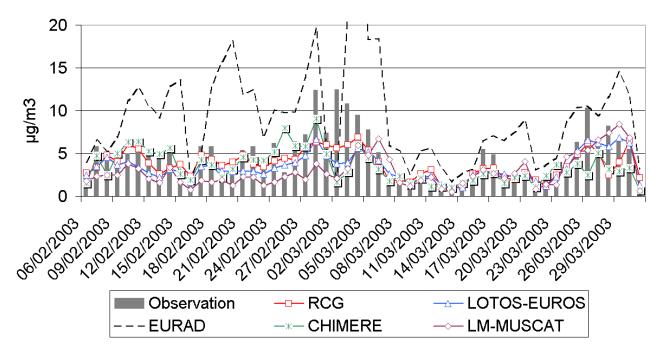
NO3

Quite a scatter







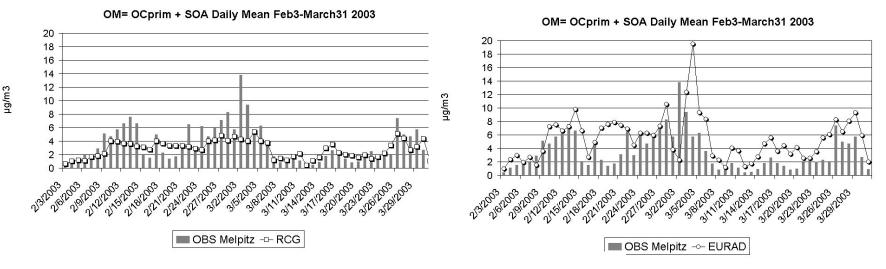


NH4

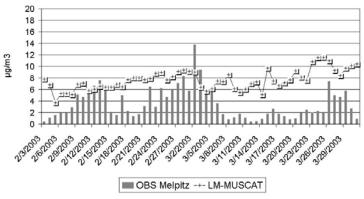








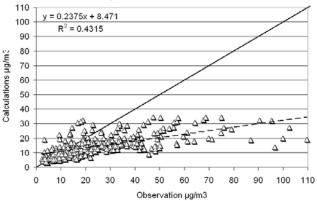




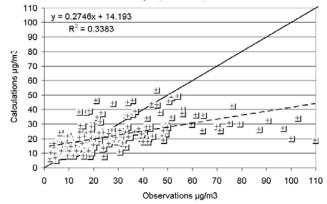
TrUmF Troposphärische Umweltforschung Institut für Meteorologie Freie Universität Berlin

## PM2.5 daily mean

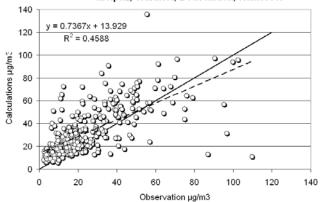
Lotos: PM2.5 Daily Mean Jan 15 - Apr 5, 2003 Melpitz, Waldhof, Deuselbach, Hannover



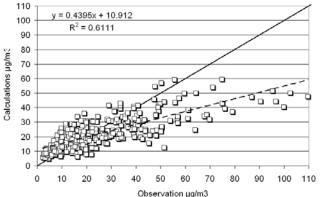
LM-MUSCAT: PM2.5 Daily Mean Jan 15 - Apr 5, 2003 Melpitz, Waldhof, Deuselbach

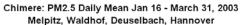


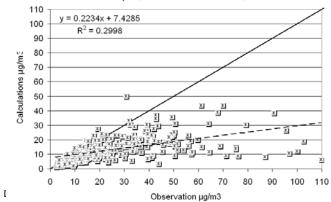
EURAD: PM2.5 Daily Mean Jan 15 - Apr 5, 2003 Melpitz, Waldhof, Deuselbach, Hannover



RCG: PM2.5 Daily Mean Jan 15 - Apr 5, 2003 Melpitz, Waldhof, Deuselbach, Hannover



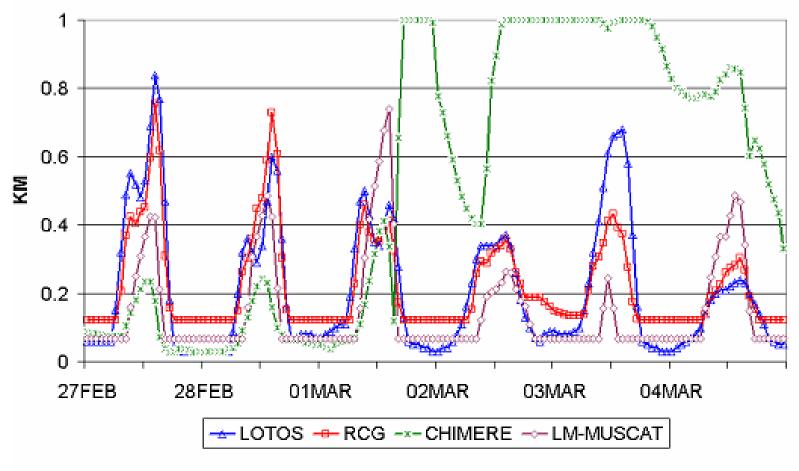






## Melpitz: Hourly Mixing heights core episode





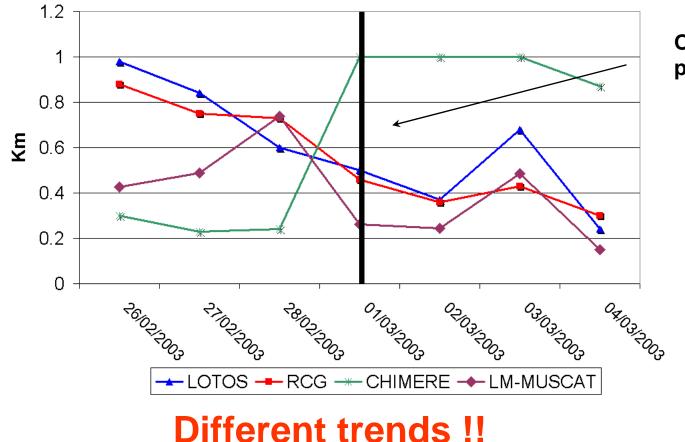
## Factor 2 to 5 difference !!



## Melpitz: Maximum daily mixing heights, core episode



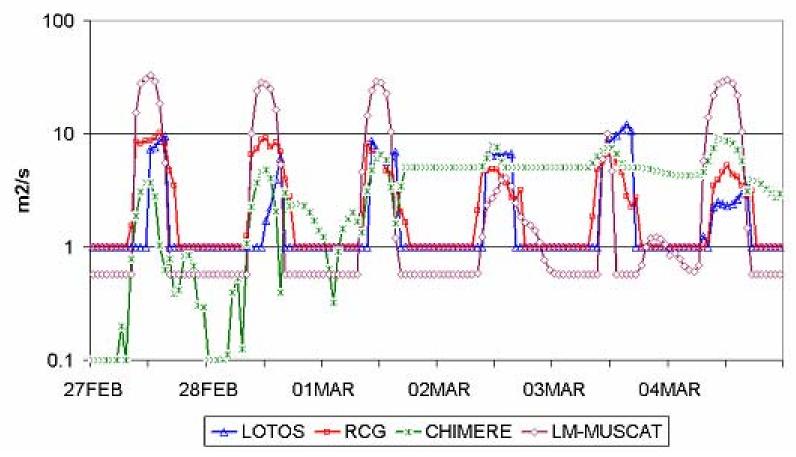
Maximum daily Mixing height



Observed PM peak

### Troposphärische Umweltforschung **Melpitz: Vertical exchange** coefficient Kz between layer 1 and layer 2: core episode





Factor 2 and more difference for meteorological key parameters !!

Institut für Meteorologie Freie Universität Berlin





## > All models recognize the embedded episodes but 4 out of 5 model underestimate the PM peak concentrations

Large differences between models in the aerosol precursor concentrations and

primary PM components point to different transport/mixing characteristics (PBL key

parameter, vertical exchange mechanism, grid layout)

t für Meteorologie

Freie Universität Berlin

 $\geq$ Different SIA formation might be more related to differences in the input to the

aerosol modules (concentrations, met. variables) than to different module formulations

(EURAD: NO3, NH4 overestimation due to NH3 overestimation or module problem or both?)

#### EURAD has the PM peaks right but for the wrong reason (NO3, NH4 overestimation) Dr. R. Stern, FU Berlin





## Analysis of <u>observed</u> PM10 composition at Melpitz

- During episodes there is an increase of primary <u>and</u> secondary pollutants
- > SIA (fine mode) contribution to total PM10 decreases with

increasing PM10 concentrations

(days with PM10 < 50 µg/m3 55%, > 50 µg/m3 46%)

>EC, OM, unacc. mass contributions to total PM10 increase with increasing PM10

## Dr. R. Stern, FU Berlin Episodes seem not to be LRT dominated

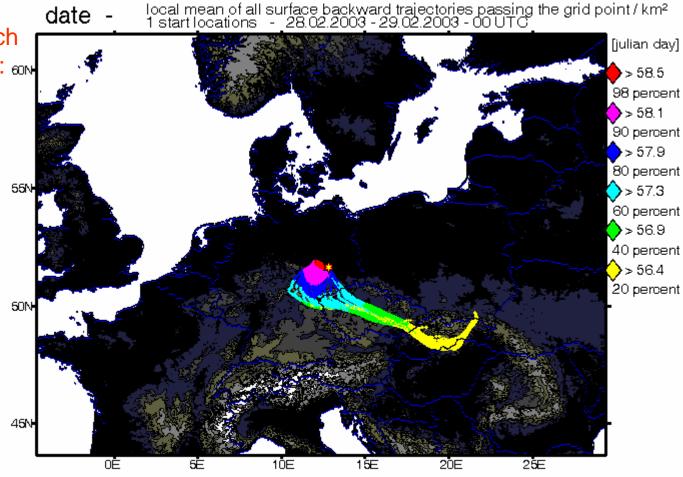
TrUmF Troposphärische Umweltforschung Institut für Meteorologie Freie Universität Berlin

# Backward trajectory bundle at Melpitz in the FS Saxonia



Trajectory started each 10 min over 72 hours: and

All trajectories stay about 2 days over Saxonia indicating stagnant conditions







# Analysis of PM10 peak underestimation of the models

Increasing underestimation of primary and secondary

pollutants with increasing PM10 concentrations

>Underestimation of EC, unaccounted mass is considerably larger than the underestimation of SIA at days with observed PM10 > 50µg/m3

Have we problems to simulate stagnant weather conditions ? Parameterization of the stable boundary layer is still poor !!!

## Troposphärische Umweltforschung Vertical temperature gradient at Feb 28, 12 UTC, 2003



5

4

З

2

1

Û

-1

-2

-3

 $^{-4}$ 

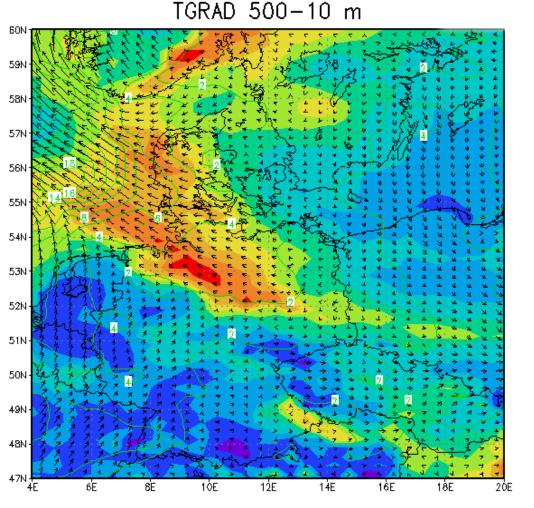
-5

Green, yellow, red: areas with stable conditions

**FrUmF** 

Freie Universität Berlin

**BASED ON OBSERVATIONS** 



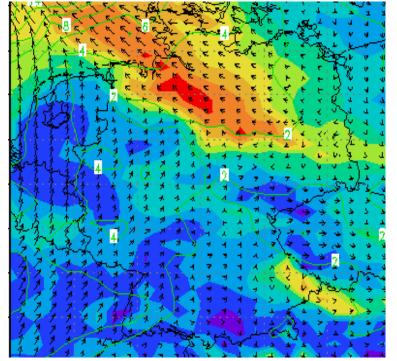
20



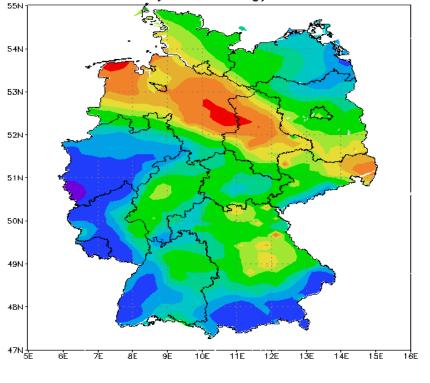


**Stability pattern at Feb 28** 

#### Observed PM10 concentration field at Feb 28, 2003



OI OBS: PM10 daily mean ug/m3, 28 FEB 2003



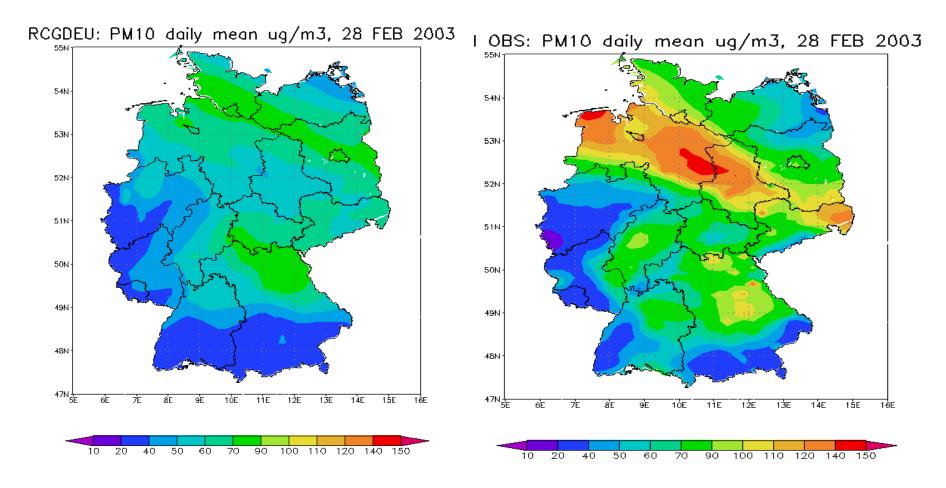
10 20 40 50 60 70 90 100 110 120 140 150





#### Observed PM10 concentration field Feb 28, 2003

Calculated PM10 concentration field at Feb 28, 2003 REM-CALGRID model









(European?) Models seem currently not to be able to simulate high PM10 concentrations (> 50 µg/m3), which are observed mostly in winter time





## Conclusion

# The underestimation cannot be attributed to one single source of error

Missing sources (unaccounted mass) or underestimation (probably EC

emissions)

>Uncertainties for key boundary layer parameters for inversion induced winter

episodes (underestimation of the strength of inversions, overestimation of mixing

heights, missing calms with false wind directions etc.)

The error source "meteorological processes" seems to have (at least) equal importance as the error sources "emissions" or "physical/chemical processes"



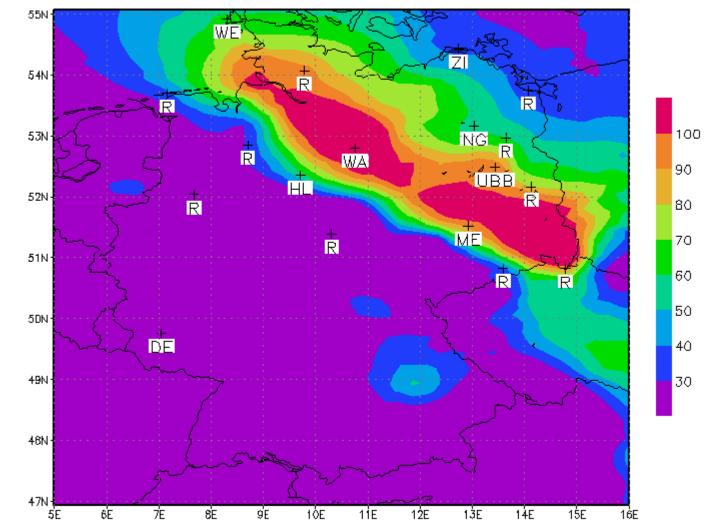


## **Additional Material**

TrUmF Troposphärische Umweltforschung Institut für Meteorologie Freie Universität Berlin



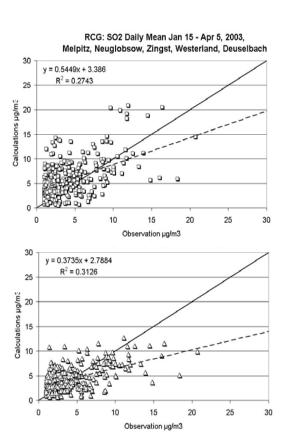
#### Observed PM10 concentration field at March 2, 2003 station labels indicate the location of the stations used for model evaluation



Dr. R. Stern, FU Berl

GrADS: COLA/IGES

**TrUmF** Troposphärische Umweltforschung Institut für Meteorologie Freie Universität Berlin



EURAD: SO2 Daily Mean Jan 15 - Apr 5, 2003, Melpitz, Neuglobsow, Zingst, Westerland, Deuselbach 0 Ó 80 0 00 08 y = 2.2334x + 5.6719 2000 0 R<sup>2</sup> = 0.2985 0 10 20 30 40 50 60 70 80 90 Observation µg/m3 Chimere: SO2 Daily Mean Jan 16 - March 31, 2003, Melpitz, Neuglobsow, Zingst, Westerland, Deuselbach y = 0.895x + 1.3451 R<sup>2</sup> = 0.4509 X

90

80

70 60

50

20

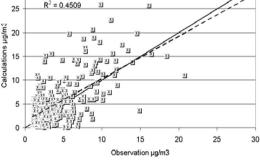
10 0

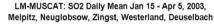
30

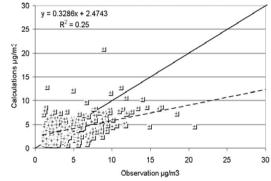
10 40

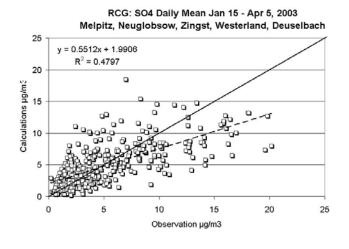
00 Calc



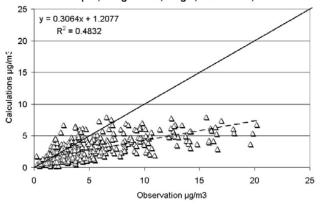




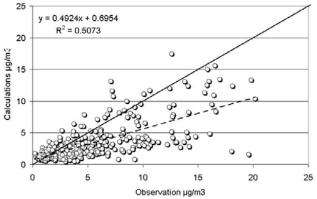




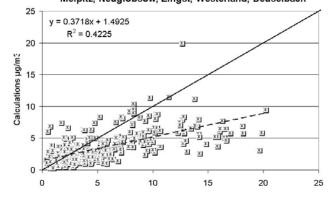
LOTOS-EUROS: SO4 Daily Mean Jan 15 - Apr 5, 2003 Melpitz, Neuglobsow, Zingst, Westerland, Deuselbach



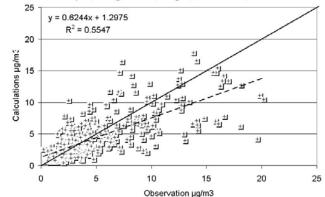
EURAD: SO4 Daily Mean Jan 15 - Apr 5, 2003 Melpitz, Neuglobsow, Zingst, Westerland, Deuselbach

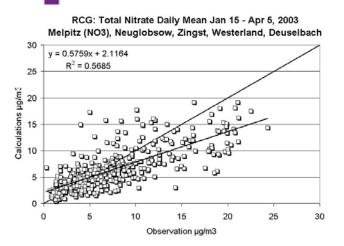


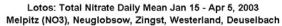
Chimere: SO4 Daily Mean Jan 16 - March 31, 2003 Melpitz, Neuglobsow, Zingst, Westerland, Deuselbach

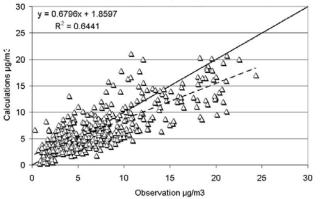


LM-MUSCAT: SO4 Daily Mean Jan 16 - March 31, 2003 Melpitz, Neuglobsow, Zingst, Westerland, Deuselbach

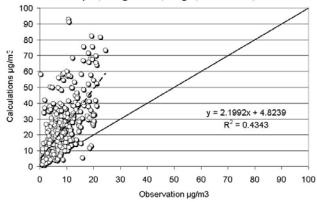




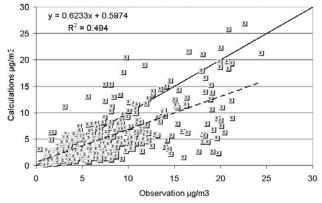




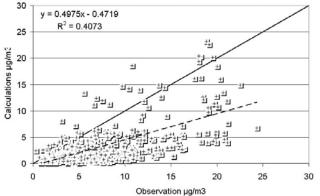
EURAD: Total Nitrate Daily Mean Jan 15 - Apr 5, 2003 Melpitz, Neuglobsow, Zingst, Westerland, Deuselbach

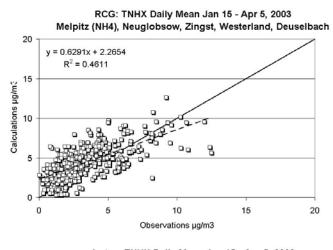


Chimere: Total Nitrate Daily Mean Jan 15 - Apr 5, 2003 Melpitz (NO3), Neuglobsow, Zingst, Westerland, Deuselbach



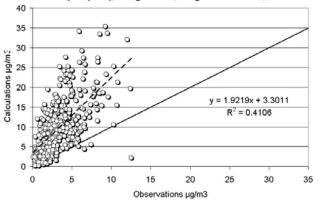
LM-MUSCAT: Total Nitrate Daily Mean Jan 15 - Apr 5, 2003 Melpitz (NO3), Neuglobsow, Zingst, Westerland, Deuselbach



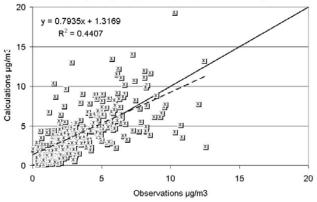


Lotos: TNHX Daily Mean Jan 15 - Apr 5, 2003 Melpitz (NH4), Neuglobsow, Zingst Westerland, Deuselbach 20 y = 0.5635x + 1.4237 R<sup>2</sup> = 0.5193 Calculations µg/m<sup>5</sup> A Δ 5  $\Delta$  $\Delta$ 0 0 10 5 15 20 Observations µg/m3

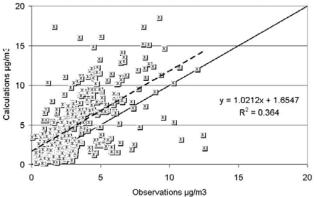
EURAD: TNHX Daily Mean Jan 15 - Apr 5, 2003 Melpitz (NH4), Neuglobsow, Zingst Westerland, Deuselbach



Chimere: TNHX Daily Mean Jan 15 - Apr 5, 2003 Melpitz (NH4), Neuglobsow, Zingst Westerland, Deuselbach



LM-MUSCAT: TNHX Daily Mean Jan 15 - Apr 5, 2003 Melpitz (NH4), Neuglobsow, Zingst Westerland, Deuselbach



ACT NIT