Evaluation of Comprehensive Eulerian Chemistry and Transport Models Using Ozone Production Rates Determined by Direct Radical Measurements During BERLIOZ

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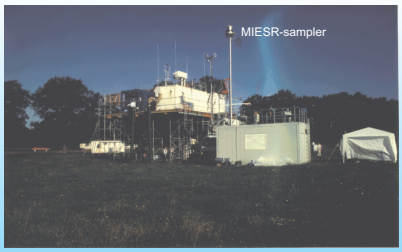
Motivation

To test the ability of 3D transport models to simulate the contribution of chemical and meteorological processes to the local O_3 budget correctly

Goal

To quantify the relative importance of chemistry and atmospheric transport for the local O_3 budget



Summary

☺ Simulated local concentrations are in rather good agreement with observations

☹ Simulated chemical O_3 production rates show significant differences of up to a factor of 4 compared to observations

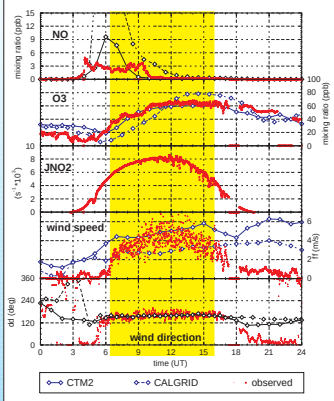
☹ Highest observed production rates for O_3 are <10 ppb/h [8]

😐 Transport compensates under- or overprediction of chemical production of O_3

➔ **correct O_3 forecasts in urban plumes may be fortuitous !**

Observed and simulated meteorology and concentration at Pabstthum

July 20th, 1998



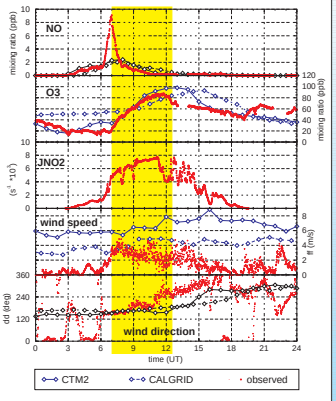
CTM2:

- wind speed too high at night, overestimated during daytime
- wind direction matches perfectly during MIESR observations
- O_3 overestimated before sunrise, little underestimation during MIESR observations
- Berlin plume at 6 UT

CALGRID:

- wind speed too high at night, underestimated during daytime
- wind direction matches perfectly during daytime
- O_3 overestimated before sunrise, maxima too high
- Berlin plume at 6 UT

July 21st, 1998



CTM2:

- wind speed considerable overestimated
- wind direction matches during MIESR measurements
- NO in time like observed, but maximum too low
- O_3 reasonable, maximum too high and too late

CALGRID:

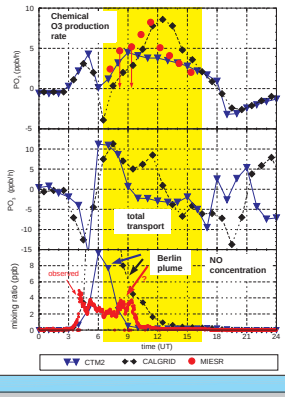
- wind speed overestimated
- wind direction matches during MIESR measurements
- NO in time like observed, but maximum too low
- O_3 maximum too high and too late, time development not catched

Synoptical situation

Complex vertical structure of PBL on both days - strong wind shear within the PBL - advection of different air masses at different heights

Observed and simulated O_3 production rates at Pabstthum

July 20th



MIESR measurements

$P(O_3) = k_1 \bullet [NO][HO_2] + \Sigma k_{2i} \bullet [NO][RO_2]_i$

$k_1 = 4.19 \bullet 10^{-12}$ $k_2 = 4.12 \bullet 10^{-12}$ (molecules m^{-3} s^{-1})

Models

Directly from the solution of the gas phase chemistry submodels

CTM2:

➔ underprediction of $P(O_3)$ during sunny hours

➔ stationary transport during MIESR measurements

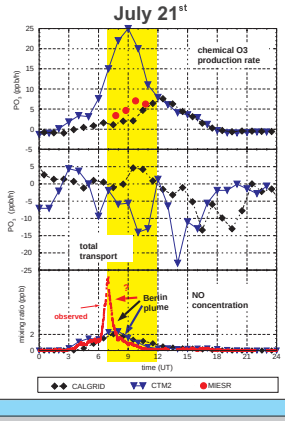
CALGRID:

➔ time development comparable to observations

➔ time shift of $P(O_3)$ maximum about 1.5 hours

➔ increased O_3 advection until noon

July 21st



CTM2:

➔ $P(O_3)$ a factor of 4 higher then observed

➔ strong local loss O_3 due to transport

CALGRID:

➔ slight underprediction of $P(O_3)$

➔ time shift of $P(O_3)$ maximum about 1.5 hours

➔ little O_3 transport

Model design

	CTM2	CALGRID
Driving meteorology	MM5 [1] 48 hour forecast	Europamodell [2] 36 hour forecast
Grid size	2 km	2 km
No. of vertical levels (within PBL, lowest)	23 (13, 30 m)	14 (12, 20 m)
Chemical model	RADM2 [3]	SAPRAC90 [4]
Emission data base	TFS-BERLIOZ 1998 [5]	1994 Umweltbundesamt [6]

Instrumentation

- NO, NO_2, NO_y, H_2O, O_3, CO, PAN, CH_4, met. Parameters, $J(NO_2)$
- VOC (C_2-C_{10})
- **HO_2, RO_2, NO_2, NO (det. lim. 1 ppt) with MIESR (Matrix-Isolation and Electron-Spin-Resonance, [7])**
- All samples at 11m above ground

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[2] Operational forecast model, German Weather Service

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[4] Carter, W., *A detailed mechanism for the gas-phase atmospheric reactions of organic compounds*, Atm. Env. 24A, 1990

[5] Friedrich, Universität Stuttgart, IER, priv. communication, 1999

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