Influence of the choice of gas-phase mechanism on predictions of key gaseous pollutants during the AQMEII phase-2 intercomparison

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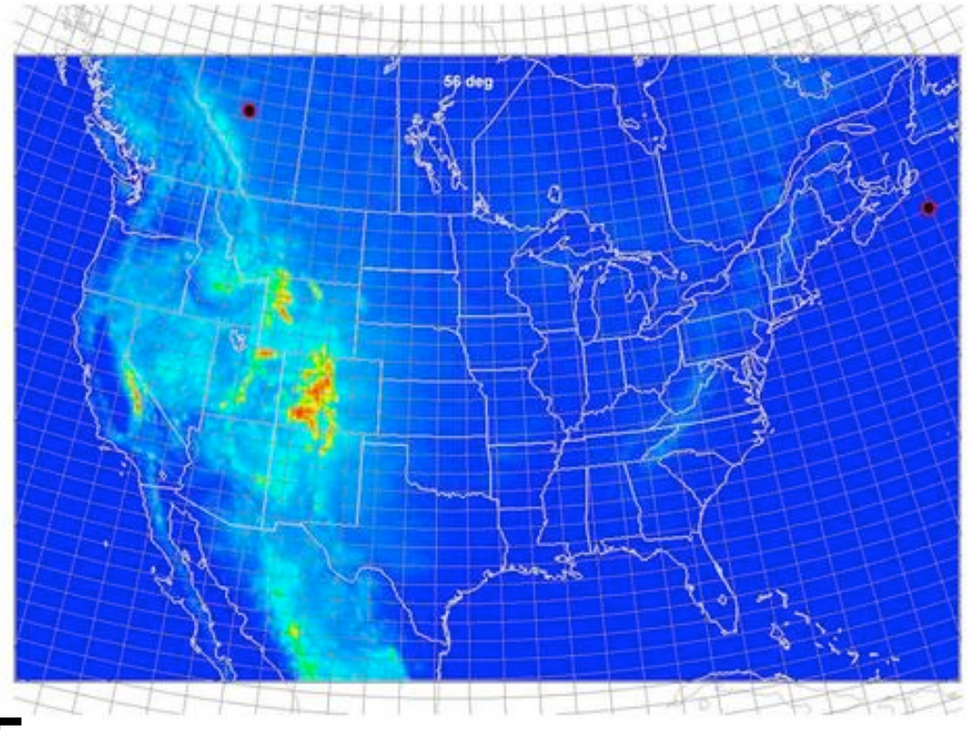
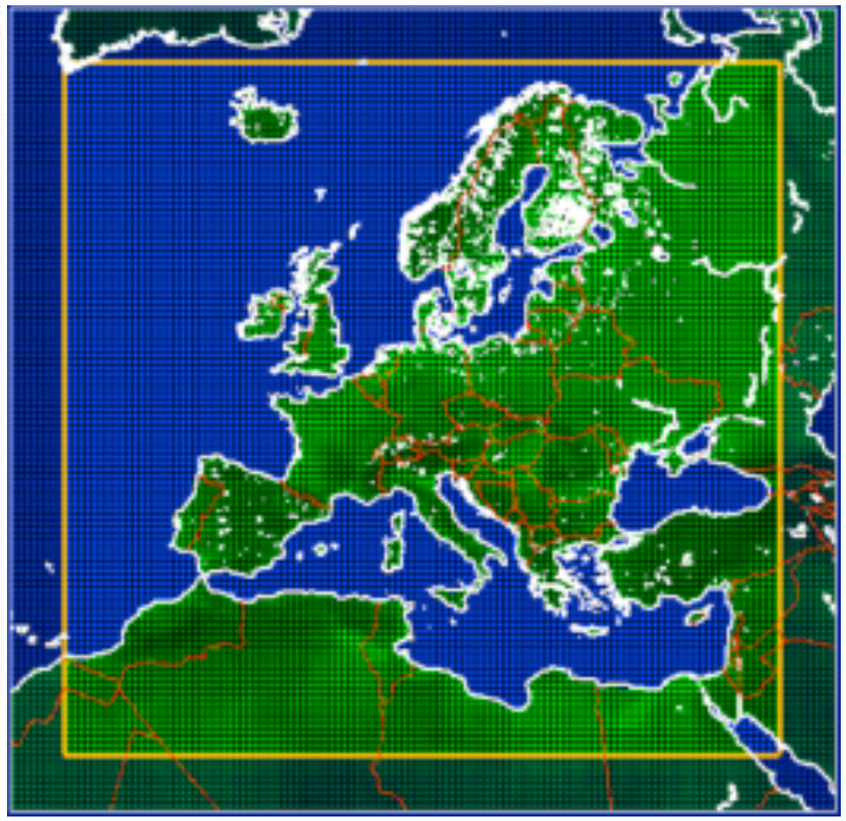
submitted to Atmospheric Environment

NCAR

AQMEII phase 2 model intercomparison





WRF-chem (10x)
WRF-CMAQ (3x)
GEM-MACH
COSMO-MUSCAT
COSMO-ART
NMMB/BSC-CTM
ENVIRO-HIRLAM
MetUM/UKCA
Silam
RACMO2-LOTOS-EUROS
MEMO/MARS-aero
bolChem

period
full year 2010, optionally 2006

domain
EU / NA (or both)

emissions
TNO-MACC (updated)
EPA-NEI 2010 (2008 + updates)

http://4.bp.blogspot.com/_VX6ehGADPpA/Sh-IYFJegAI/AAAAAAAABio/6xwzAgFiw3A/s400/GoldenGunDuel.jpg

mechanism shootout

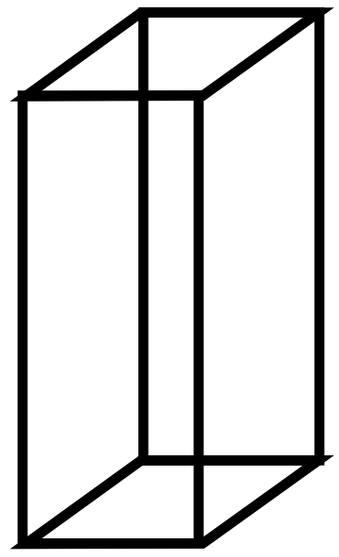
RADM2 (Stockwell et al., 1990)
RADMKA (Vogel et al., 2009)
RACM (Stockwell et al., 1997)
CBMZ (Zaveri and Peters, 1999)
CB05-TUCL (Yarwood et al., 2005 + updates)
CB05-Clx (Yarwood et al., 2005 + updates)
MOZART-4 (Emmons et al., 2010 + updates)

"2 1/2 decades of atmospheric chemistry"

mechanisms

KPP (Sander and Sandu, 2006)
using Rosenbrock solver

the “box”



the "box"

mechanisms

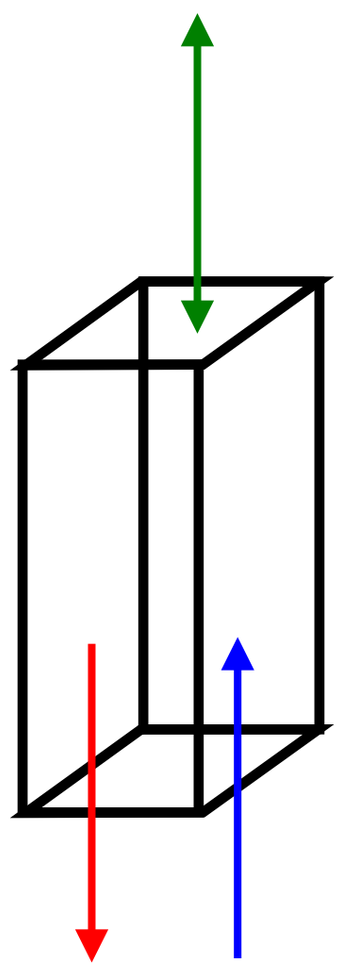
KPP (Sander and Sandu, 2006)
using Rosenbrock solver

photolysis rates

TUV
(Madronich and Flocke, 1997)
idealized diurnal cycle
(40°N, clear sky)

initial conditions

30 ppbv O_3, 120 ppbv CO,
1-10 ppbv NO_x, 1 ppbv SO_2



entrainment

mix against initial conditions
(same for all models, VOCs=0)

emissions

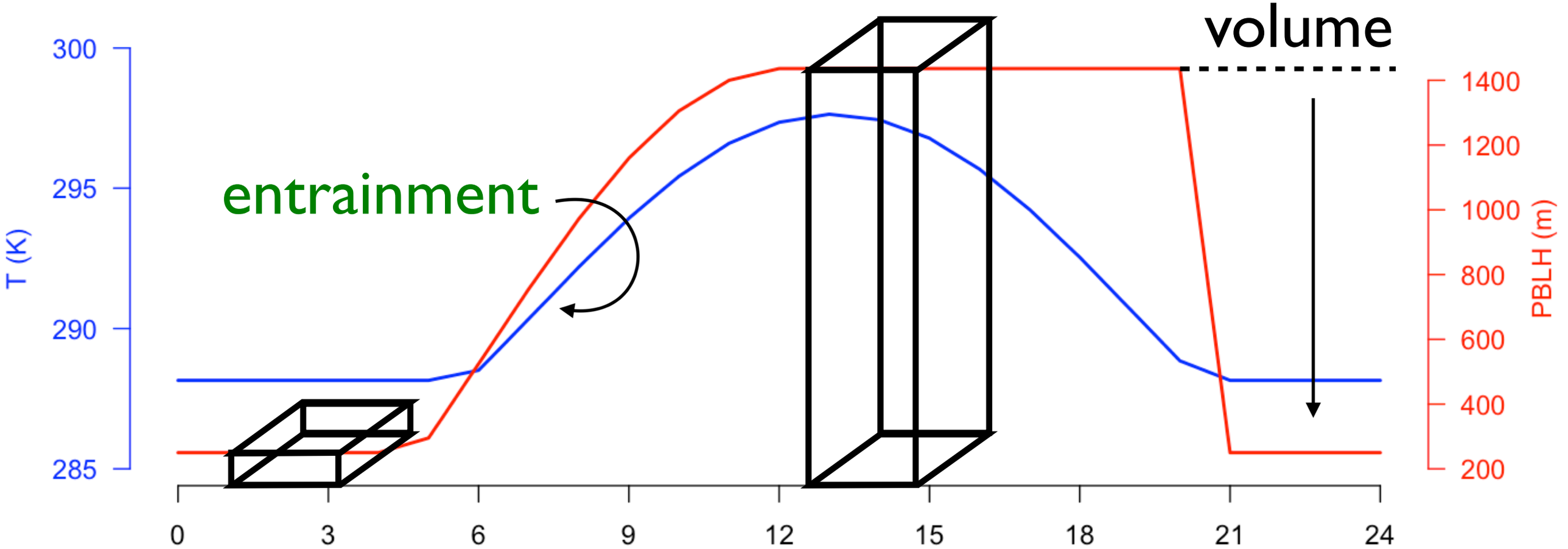
inorganics (NO_x, CO, SO_2, NH_3)
and biogenics (MEGAN)
from common base model

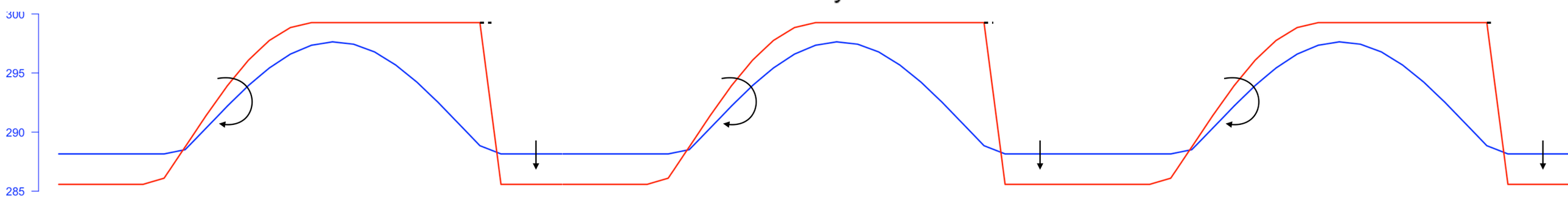
anthro. VOCs from
wrfchemi files of each group

deposition

simple first order loss
(inorganics only)

the "box": an idealized planetary boundary layer





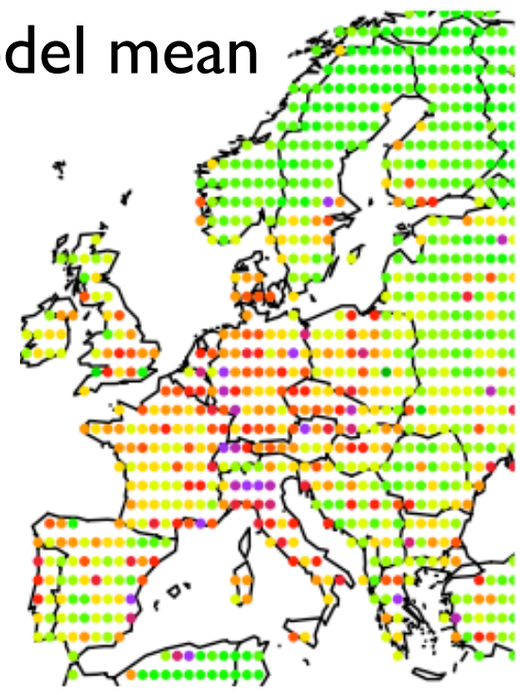
3 day long simulations

multi-model mean

O_3

- 50
- 40
- 30
- 20
- 10

ppbv



mechanism biases in O_3

against the multi-model mean

mechanism biases in O_3

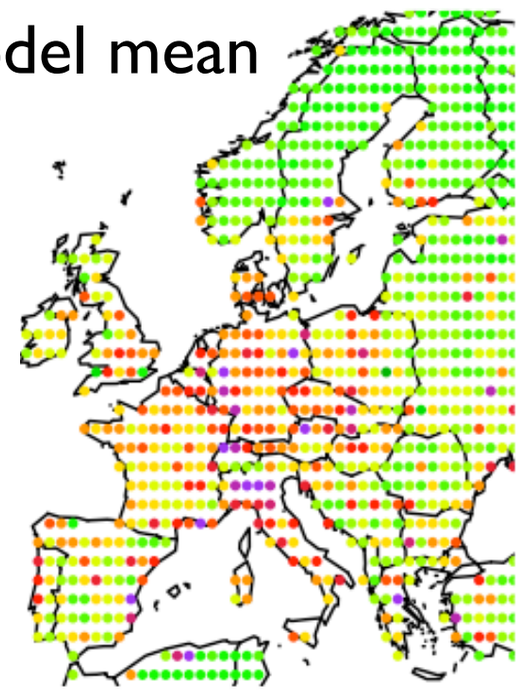
against the multi-model mean

multi-model mean

O_3

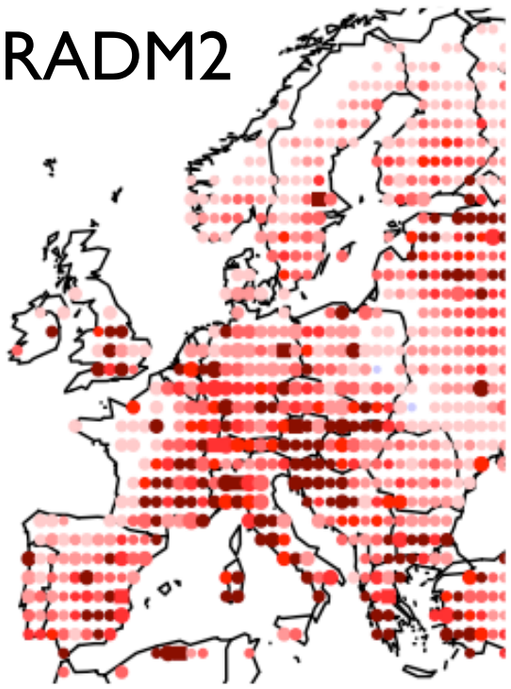
- 50
- 40
- 30
- 20
- 10

ppbv

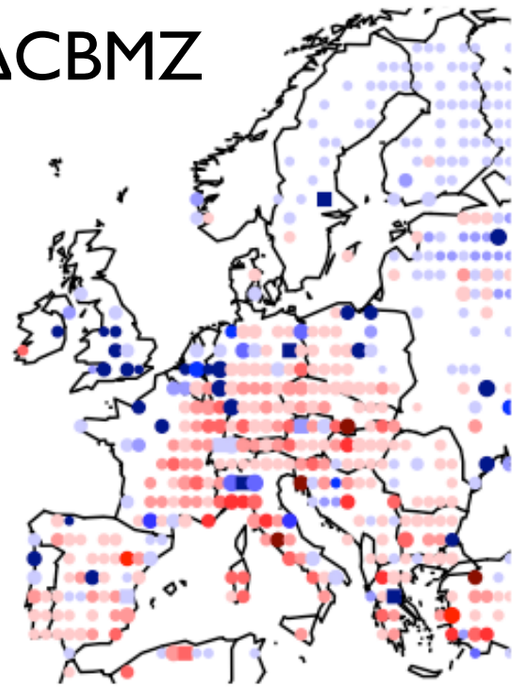


-1.8 0 1.8 ppbv

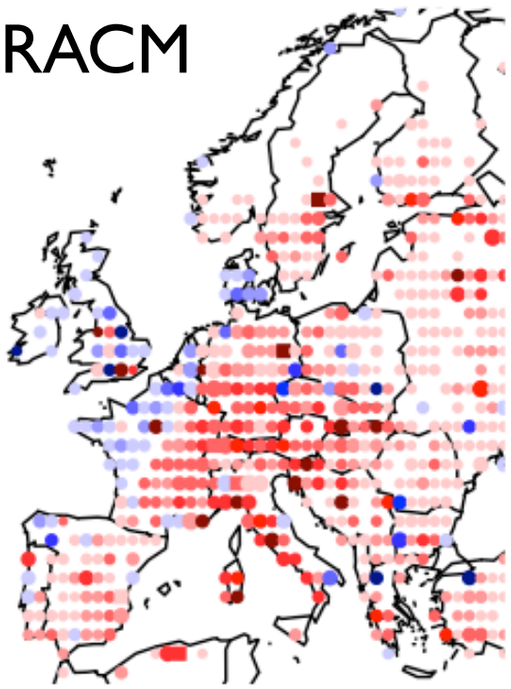
ΔRADM2



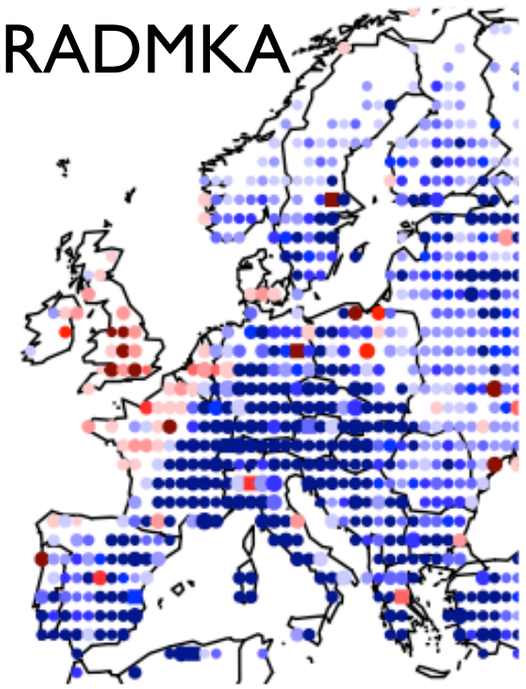
ΔCBMZ



ΔRACM



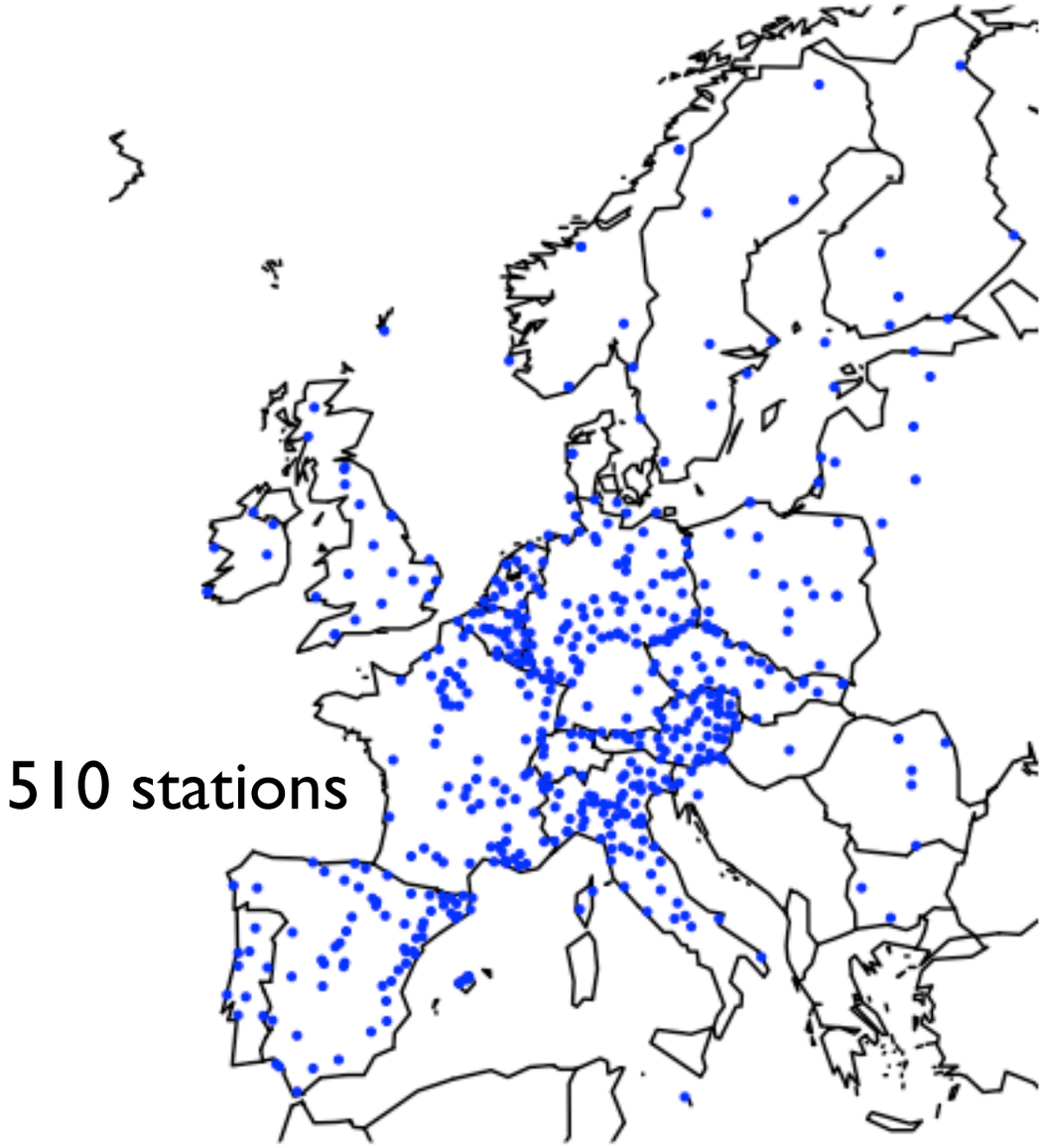
ΔRADMKA



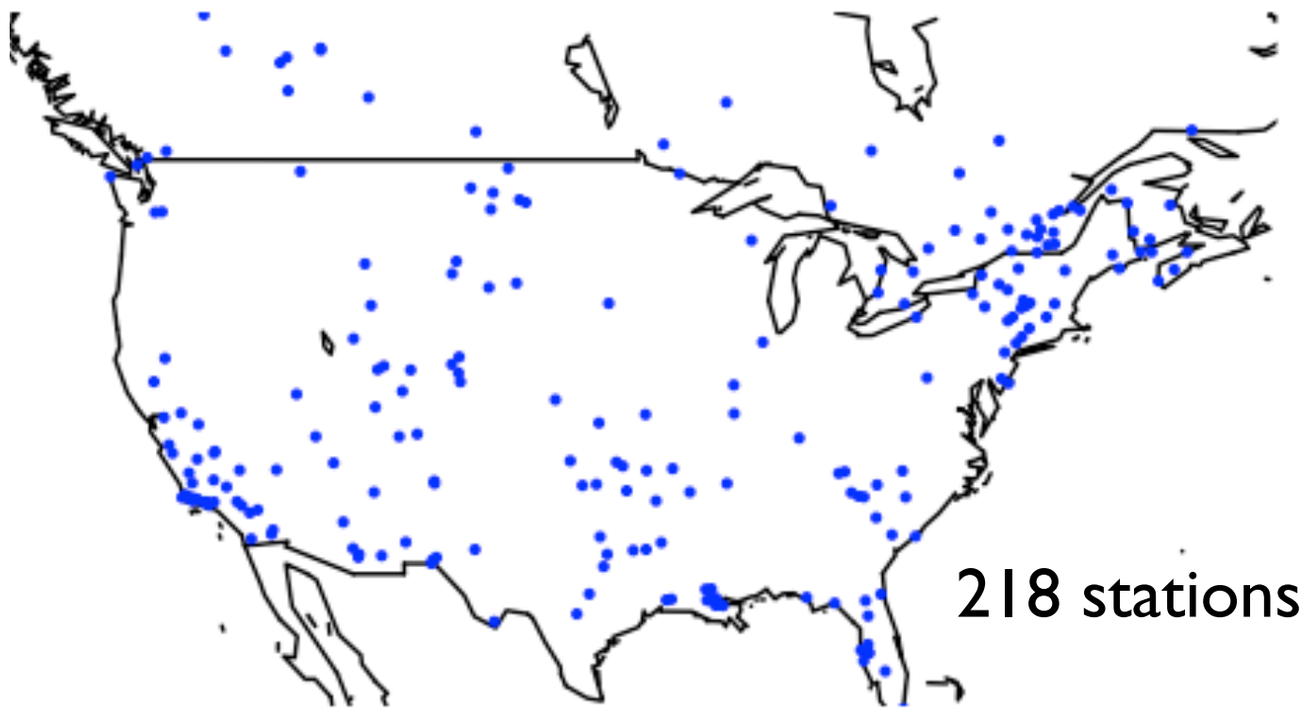
- mechanisms within 3.6 ppbv in avg. O_3
- different responses to clean, rural, or urban conditions
- high variability under strong biogenic VOC influence

mechanism performance

at the location of measurements



510 stations



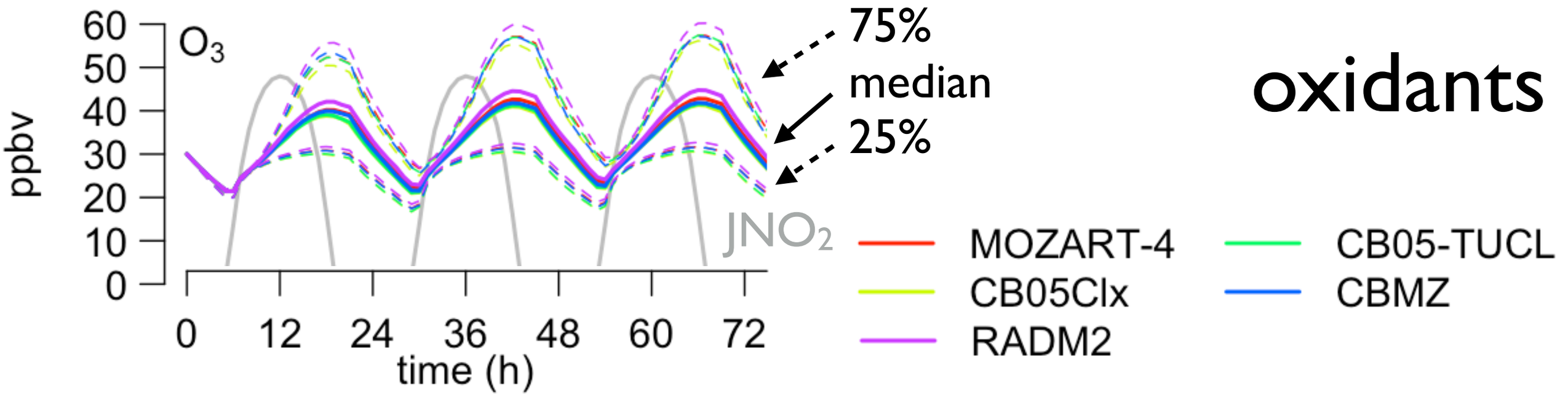
218 stations

location of surface measurements used in

Im et al., 2014: Evaluation of operational online-coupled regional air quality models over Europe and North America in the context of AQMEII phase 2. Part I: Ozone.

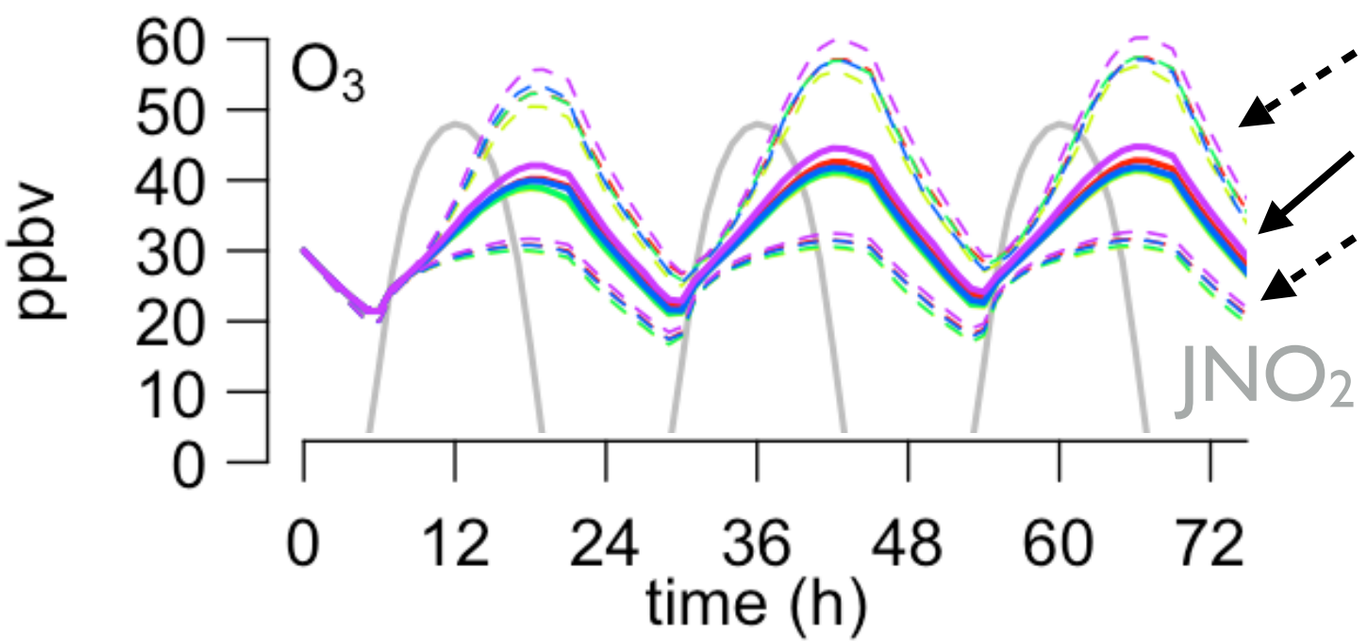
mechanism performance when compared against station network might be skewed due to station selection.

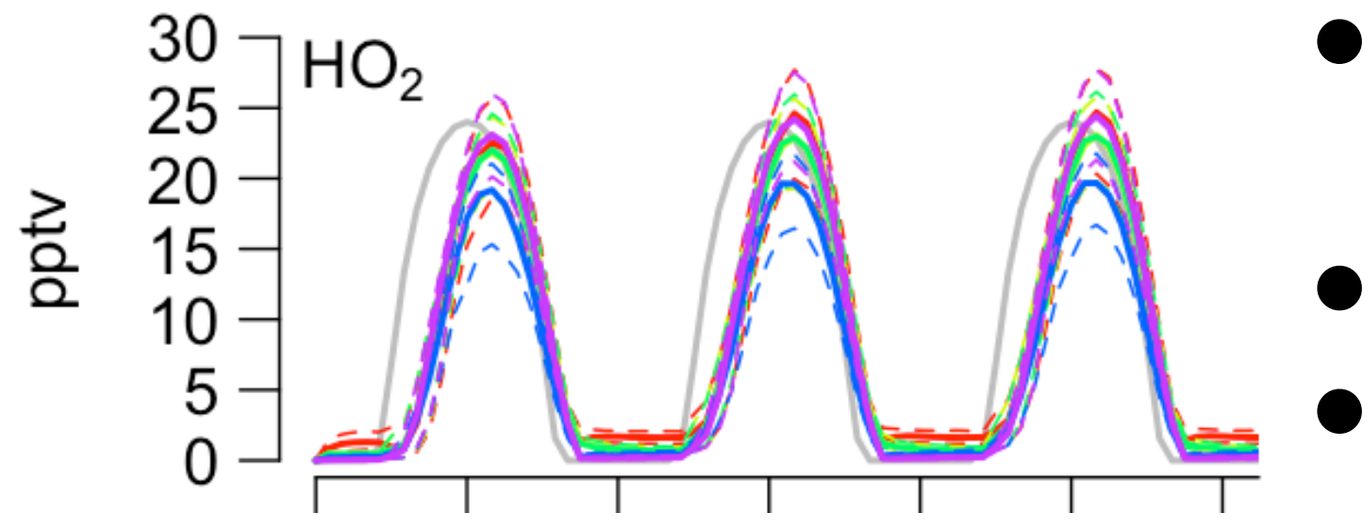
Run box model using emissions at these station locations.


ppbv
60
50
40
30
20
10
0
O3
JNO2
75%
median
25%
0
12
24
36
48
60
72
time (h)
MOZART-4
CB05Clx
RADM2
CB05-TUCL
CBMZ

oxidants

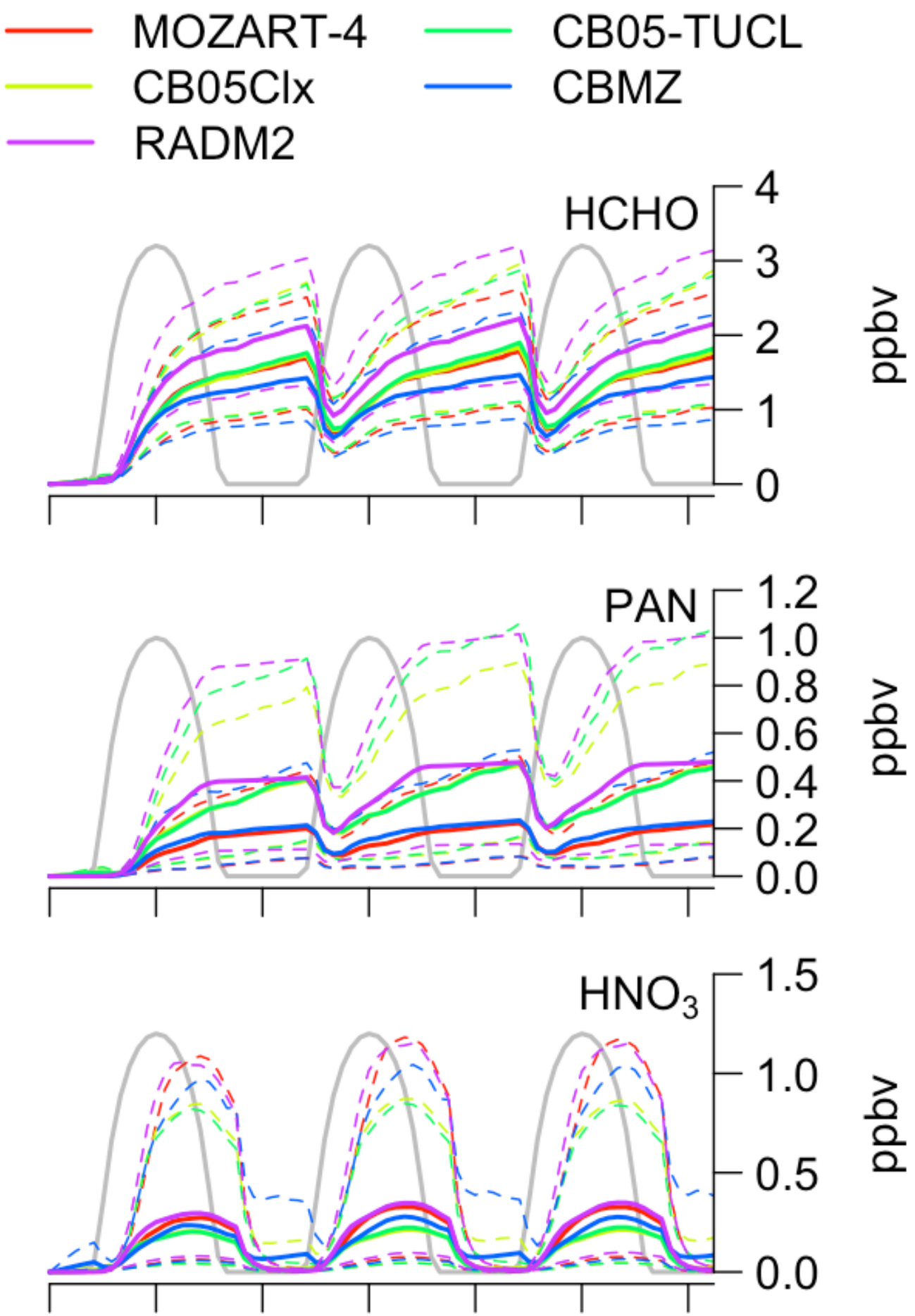
oxidants





- avg. O_3 within 5 ppbv, peak O_3 within 7-8 ppbv
- differences in HOx variab.
- nighttime chemistry (NO_3) needs more investigation

secondary products



VOC evaluation against satellite measurements?

"Remote" O_3 production?

Secondary inorganic aerosols (NH_3NO_3)?

Conclusions

- mechanisms agree on average O_3 within 3-5 ppbv, differences in peak O_3 7-8 ppbv
- good agreement in avg. HO_x, large differences in variability
- startling differences in key nighttime species as well as major secondary products

Implications

- mechanism form O_3 for different reasons, will hence react differently to changes in emissions (or climate)
- choice of gas-phase mechanism is a considerable source of uncertainty for other observables (oxidations, secondary products, particulate matter)