Outline

- Motivation, Challenge & Approach
- Model description
- Model validation
- Case study
- Conclusions
Motivation, Challenge & Approach

“Detection of long-term man-made trends in the concentration of greenhouse gases and aerosols related to climate change above natural variability and the corollary impacts of climate change on atmospheric composition.”

[GAW Strategic Plan 2008-2015]

Trends masked by inter-annual variability
- Understanding of inter-annual variability will help to obtain more robust and earlier trend estimates from observations

Lagrangian transport model with limited chemistry
- Focus on CO and CH$_4$
  - Largely controlling oxidizing capacity of the troposphere
  - About 1/3 of atmospheric CO produced from CH$_4$ oxidation
- Why Lagrangian?
  - transport times, age spectra
  - no numerical diffusion
  - computationally cheap
Based on FLEXPART V8.1 (Stohl, 2005, ACP)
- Global domain filled with 3 mio particles, carrying 7 CO, 7 CH$_4$ species, 1 inert air mass tracer
- Driven by 1° x 1° ECMWF analysis
- Initialised by NOAA MBL obs.; 3 year spin-up
Emission *uptake* by particles in ABL (7 source regions)

Clocks, counting the time since a particle left the ABL of a source region (lower limit of pollutant age)

CO and CH$_4$ degradation by OH (HTAP climatology for 2001)

Inter-particle mixing for particles in the same ABL box
Emissions

Gridded emission: 1° x 1°; distributed over atmospheric boundary layer
No trend, only variability in biomass burning

Anthropogenic: EDGAR 3.2, const.
Ox. Anth. NMVOC: EDGAR 3.2, const
Ox. Bio. NMVOC: EDGAR 3.2, monthly

Biomass burning: GFED 2.1, 8-daily
Ox. Biomass burning NMVOC: GFED 2.1

Gridded emission: 1° x 1°; distributed over atmospheric boundary layer
No trend, only variability in biomass burning

Anthropogenic: EDGAR 3.2, const.
Wild animals: (Houweling et al. 1999), const.
Termites: (Sanderson et al. 1996), const.
Ocean: (Houweling et al. 1999), const.
Rice: (Matthews et al. 1991), monthly
Wetlands: (Bergamaschi et al. 2007), monthly
Soil abs.: (Ridgewell et al. 1999) monthly

Biomass burning: GFED 2.1, 8-daily
Model Validation

- For which temporal resolution is particle statistics sufficient?
  - Original aim: monthly aggregates
  - Analysed: daily aggregates at receptor sites

- Ground based observations: GAW WDCGG

- Period: 2001-2006

- Compared time-series
  - CO
    - # Flask: 56
    - # Cont.: 15
  - CH4
    - # Flask: 56
    - # Cont.: 19
Model Validation

CO (Jungfraujoch, CH)

Daily simulations

Model Validation

CO (Jungfraujoch, CH)

PDF of obs - sim

In: 67 (%)
Model Validation
Taylor Diagrams: CO

Continuous

Flask

R: 0.6 – 0.9
Underestimation of variability

R: 0.6 – 0.95
Underestimation of variability
Model Validation

CO Bias

Negative bias in Northern hemisphere
More pronounced close to emissions

>> Emissions missing?!
Model Validation
CH$_4$ (Jungfraujoch, CH)

Global mean bias corrected: 35 ppb

Daily values

PDF of obs - sim

In: 74 (%)
Model Validation
Taylor plots: Daily CH$_4$

Continuous

Flask

R: 0.4 – 0.9

R: 0.6 – 0.9
Model Validation

CH$_4$ Bias

No median bias after global correction

Strong variability of bias in NH

>> over-estimating emissions in NH?!
Case Study
CO Anomaly, Mace Head (IR)

September 2002

Mace Head (MHD), −9.899°E, 53.328°N

Sept ‘02

CO anomaly (ppbv)

Africa
Asia
S. America
N. America
Australia
Europe
Oceans

Average September

AS, 55 (41 %)
AF, 5 (4 %)
EU, 42 (31 %)
NA, 22 (17 %)
AU, 3 (3 %)
SA, 6 (4 %)

S. Henne, et al.: Inter-annual variability of CO and CH₄, ESRL GMD Annual Conference 2010
Case Study
CO Anomaly, Mace Head (IR)

- CO anomaly (ppbv)
- Sept '09
- September 2002
- Average September

Mace Head (MHD), −9.899°E, 53.328°N

- A1: 2 – 1 d inside ABL
- A2: 1 – 0 d inside ABL
- A4: 0 – 1 d outside ABL
- A5: 1 – 2 d outside ABL
- A6: 2 – 4 d outside ABL
- A7: 4 – 7 d outside ABL
- A8: 7 – 15 d outside ABL
- A9: 15 – 30 d outside ABL
- A10: 30 – 60 d outside ABL
- A11: >60 d outside ABL
Monthly CO Anomaly: 2002-07

Total CO

Asian CO

European CO

Anomaly by source

Emission anomaly
Monthly CO Anomaly: 2002-08

Anomaly by source

Asian CO

European CO

Total CO
Monthly CO Anomaly: 2002-09

Anomaly by source

Asian CO

Emission anomaly

European CO

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Monthly CO Anomaly: 2002-10

Anomaly by source

Asian CO

European CO

Total CO

Emission anomaly
Month CO Anomaly: 2002-11

Total CO

Asian CO

European CO

Emission anomaly

Emission anomaly (%)

CO anomaly (ppb)
Monthly CO Anomaly: 2002-12

Anomaly by source

Asian CO

European CO

Total CO

Emission anomaly

Emission anomaly(%)
### Case Study

**Transport Event: Zugspitze**

- CONTRACE field experiment
- Pollution lifted by WCB over US east coast
- Transport for about 7 days in free troposphere towards Europe
- Influence on CO and O₃ in Alps documented

(Huntrieser, et al., 2005, JGR)
Case Study
Transport Event: ZUG & JFJ

Horizontal distance ca. 250 km

>> fine-scale features of plume are preserved
Summary & Conclusions

- Global domain filling Lagrangian approach for simulation of CO, CH$_4$

- Very satisfactory results for simulated daily CO and CH$_4$ wrt surface observations
  - Negative CO bias (missing emissions?)
  - Positive CH$_4$ bias and trend (overestimated emissions?)

- Inter-annual variability well simulated
  - Allows interpretation of variability in terms of emission vs. transport anomalies

- Inter-continental transport events traceable
Simulated monthly mean surface concentrations

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Thank you for your attention!