[1]Steinbuch Centre for Computing (SCC)
[2]Institute of Meteorology and Climate Research (IMK)

Impact of acetone on ozone in the upper troposphere: Simulations with ICON-ART

M. Weimer[1,2], R. Ruhnke[2], J. Schröter[2], J. Eckstein[2], K. Deetz[2], B. Vogel[2], O. Kirner[1], P. Braesicke[2]

Introduction

Acetone is a precursor of OH and $\mathrm{HO_2}$ ($= \mathrm{HO_x}$), especially in the dry upper troposphere and lowermost stratosphere (UTLS). $\mathrm{HO_x}$ in turn is responsible for ozone depletion in this region. Therefore, acetone influences ozone depletion and has climatic impact in the UTLS region [1].

Here, we show results of the ICOsahedral Non-hydrostatic (ICON) modelling framework [2] and its online coupled extension for Aerosols and Reactive Trace gases (ART) [3] which are currently under development. We have included modules for handling emissions with ICON-ART and a new simplified OH chemistry. With this OH chemistry, the tracers' lifetime depends on space and time and the tracers' distribution then get more accurate than with a globally constant lifetime.

To investigate the impact of these new features on UTLS acetone, we compare our results with airborne measurements of the ongoing CARIBIC project [1]. We calculate annual cycles of UTLS acetone interpolated to the CARIBIC flight paths. In addition, we demonstrate the differences between offline and online biogenic emissions for acetone and demonstrate the impact of the simplified OH chemistry on the acetone lifetime.

Configuration of simulations

- two simulations with constant lifetime (acetone: 20 days) and two with OH chemistry, each with offline and online emissions
- resolution: R2B04 (~160 km)
- vertical: 90 model layers
- initialised with ERA-Interim on 01 January 2004, then free-running
- covering the CARIBIC period (2005 – 2015)
- output on 1°x1° lat-lon grid

Emission scenarios:

Offline

- monthly emission mass fluxes read from external files
- biogenic acetone emissions: MEGAN-MACC [5]

Online (biogenic)

- MEGAN2.1 [6] integrated in ICON-ART
- emission mass flux dependent on state of the atmosphere (T, SZA)

Acetone depletion via simplified OH chemistry + photolysis

OH concentration:

$$[\mathrm{OH}] = \frac{2\,[\mathrm{O(^1D)}]\,k_{\mathrm{H_2O}}\,[\mathrm{H_2O}]}{k_{\mathrm{CH_4}}\,[\mathrm{CH_4}] + (k_{\mathrm{CO,1}} + k_{\mathrm{CO,2}})\,[\mathrm{CO}]}$$

- OH concentration: steady-state of production due to react. of $\mathrm{O(^1D)}$ with $\mathrm{H_2O}$ and depletion with $\mathrm{CH_4}$ and CO

$$[\mathrm{O(^1D)}] = \frac{J_{\mathrm{O_3}}[\mathrm{O_3}]}{k_{\mathrm{O_2}}\,[\mathrm{O_2}] + k_{\mathrm{N_2}}\,[\mathrm{N_2}] + k_{\mathrm{H_2O}}\,[\mathrm{H_2O}]}$$

- $\mathrm{O(^1D)}$: steady-state of production due to photolysis of $\mathrm{O_3}$ and depletion due to quenching ($\mathrm{N_2}$ and $\mathrm{O_2}$), and react. with $\mathrm{H_2O}$

Reaction of acetone with OH:

$$\mathrm{CH_3C(O)CH_3 + OH \longrightarrow Products} \quad (k_{\mathrm{acetone}})$$

Photolysis of acetone:

$$\mathrm{CH_3C(O)CH_3 + h\nu \longrightarrow CH_3CO + CH_3} \quad (J_{\mathrm{acetone,1}})$$
$$\mathrm{CH_3C(O)CH_3 + h\nu \longrightarrow 2CH_3 + CO} \quad (J_{\mathrm{acetone,2}})$$

- calculation with online photolysis module in ICON-ART, based on Cloud-J [4]

Loss rate of acetone:

$$L_{\mathrm{acetone}} = k_{\mathrm{acetone}}\,[\mathrm{OH}] + J_{\mathrm{acetone,1}} + J_{\mathrm{acetone,2}}$$

Results

Figure 1: Profiles of the acetone lifetime

- pressure higher than 800 hPa: acetone lifetime dominated by OH chemistry
- pressure between 800 and 50 hPa: transition zone
- pressure lower than 50 hPa: acetone lifetime dominated by photolysis

→ mean tropospheric lifetime: 33 days (similar to 35 days by [7])

Figure 2: Offline and online emissions as time series

- offline: diurnal cycle is neglected (linearly interpolated between months)
- online: sensitive to calculation of LAI:
  LAI of ICON (red): emission twice as high as offline emissions
  LAI parametrised according to [8]: factor two lower than offline emissions

Figure 3: Acetone annual cycle of CARIBIC and ICON-ART

- CARIBIC: maximum acetone volume mixing ratio (VMR) during summer
  maximum acetone VMR of about 1100 pptv
- offline emissions with constant lifetime (20 days): underestimated by factor of 2.5 w.r.t. CARIBIC measurements
- offline emissions with OH chemistry: higher lifetime than 20 days → increase in acetone VMR
- online emissions with constant lifetime: higher emissions → reduced differences to CARIBIC measurements
- online emissions with OH chemistry: overestimation of the acetone VMR in the upper troposphere

→ *Acetone annual cycle is well represented in the model, but questions remain about the lifetime of acetone in the atmosphere and about the magnitude of the acetone emissions*

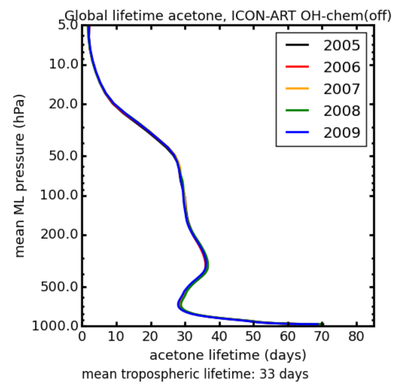

Figure 1: Profiles of annual mass-weighted mean acetone lifetime

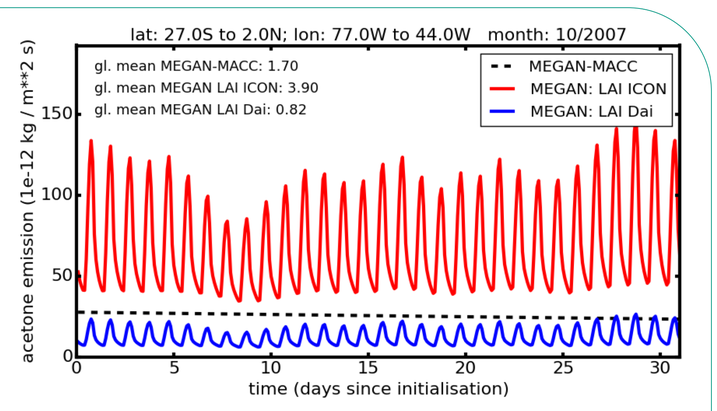

Figure 2: Time series of acetone emission mass flux density of offline (MEGAN-MACC, black dashed) and online biogenic emissions due to different calc. of leaf area index (LAI)

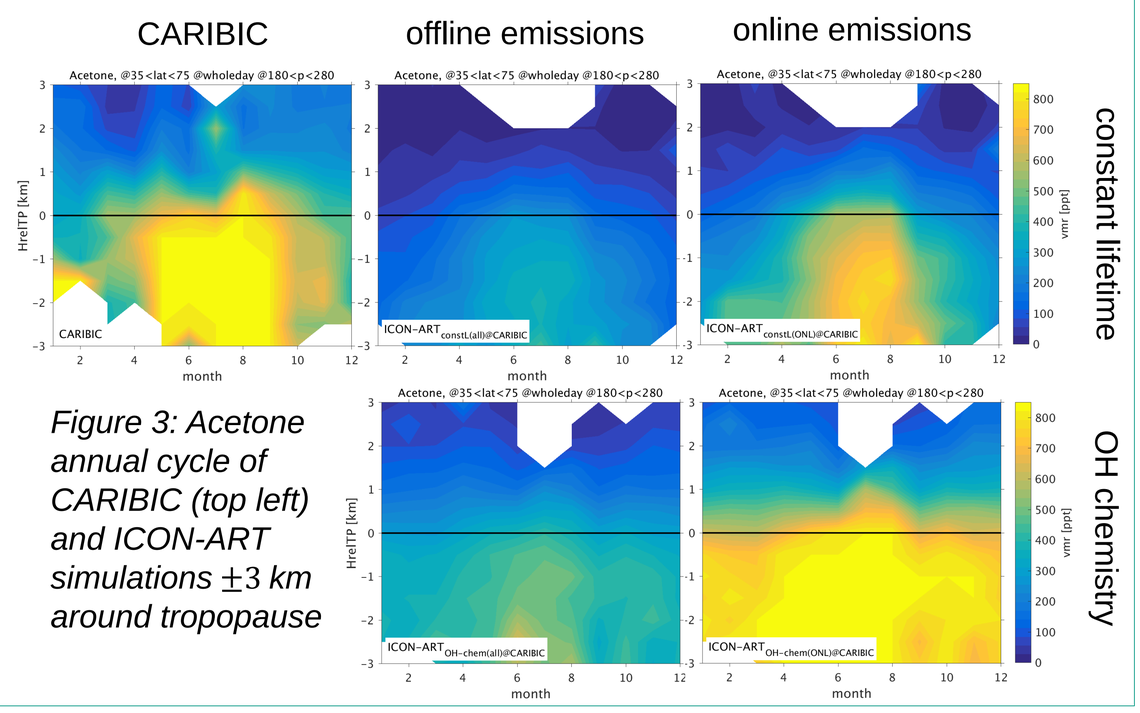

Figure 3: Acetone annual cycle of CARIBIC (top left) and ICON-ART simulations ±3 km around tropopause

References

[1] Neumaier et al. (2014), Geophys. Res. Lett., 41 (9), 3289–3297
[2] Zängl et al. (2015), Q.J.R. Meteorol. Soc., 141, 563–579
[3] Rieger et al. (2015), Geosci. Model Dev., 8, 1659-1676
[4] Prather (2015), Geosci. Model Dev., 8 (8), 2587–2595
[5] Sindelarova et al. (2014), Atmos. Chem. Phys., 14 (17), 9317–9341
[6] Guenther et al. (2012), Geosci. Model Dev., 5 (6), 1471–1492
[7] Arnold et al. (2005), J. Geophys. Res.: Atmosphere, 110, D22305
[8] Dai et al. (2004), J. Climate, 17, 2281–2299