Answer to comment of referee #2

A new module for trace gas emissions in ICON-ART 2.0: A sensitivity study focusing on acetone emissions and concentrations

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Dear referee,

Thank you for your review of the paper. In the following, you can find our answers to your comments which are in red.

1 General comments

As the title says, the emission module is claimed as the new topic, but readers cannot agree with this. Both the offline and online emission modules employ commonly-used techniques and are nothing new.

We have adapted the title as follows: "An emissions module for ICON-ART 2.0: Implementation and simulations of acetone"

Furthermore, descriptions of the off-line emission module are too technical and not suitable in the main text. I recommend to move most of the descriptions in Section 3.1 to a supplementary document as a sort of manual. Only descriptions of emission inventories used and Fig. 5 may be left in the main text.

As described on the website of GMD, our goal is reproducibility: "[...] ideally, the description should be sufficiently detailed to in principle allow for the re-implementation of the model by others, so all technical details which could substantially affect the numerical output should be described"
In addition, Referee #1 requested an even more detailed description of the module. That is why we think that this section is appropriate for the main text.

Comparing only with IAGOS-CARIBIC is not sufficient and more evaluation analyses are required. The evaluation only with the UT/LS data might be misleading, if the model vertical transport, which is often very uncertain, is wrongly simulated. Surface station data may be available and they should be compared with the simulated values in addition.

We have compared the OH-chem simulations with the surface observations of Hu et al. (2013) and included it in the paper, now Sect. 7.1, and discussed the results.

Furthermore, I cannot understand why the authors limited the IAGOS-CARIBIC data to the mid-latitude UT/LS region. I think tropical data and vertical profiles (if available) are also useful to evaluate the overall performance of the model.

We, of course, agree with this comment in principle. However, there are several issues: Firstly the PTRMS needs some time to stabilise, i.e. the first hour of the measurements after take-off generally is not a reliable measurement. Furthermore the PTRMS is switched off at $\sim 700\,\mathrm{hPa}$ to prevent damage of the turbo molecular pumps during landing.

As a second point, our aim was to create a climatology with a methodology similar to that shown in Jöckel et al. (2016).

Additionally, for a meaningful climatology we need sufficient number of measurement points. As the CARIBIC container is always mounted in Germany (Frankfurt or Munich), then flying to an intercontinental airport and coming back to Germany again, the data coverage over the mid-latitudes is much higher than over the tropics.

Furthermore, one more result with MEGAN-Online LAIsun, which is newly introduced in this study, is needed to be shown in the sensitivity test

We have included this test and have discussed the results in Sect. 7.3:

"As could be expected from Fig. 7, the annual cycles of acetone of constL(megan-onl,LAIsun) and OH-chem(megan-onl,LAIsun) are nearly identical with the respective offline emissions simulation except for slightly higher values in case of the LAIsun simulations. Thus, by parametrising the LAI according to Dai et al. (2004) the online biogenic emissions in ICON-ART are in good agreement with the offline data set MEGAN-MACC."

2 Minor comments

Title: As stated above, "a new module for trace gas emissions" seems inappropriate.

We have adapted the title (see above).

P.1, L.12: Insert a space between "dominated" and "concentrations"

We have changed this.

Introduction: What is the benefit of using ICON for atmospheric chemistry studies? Please discuss about that. Also, other previous studies in which similar icosahedral models (other than ICON) are used for atmospheric chemistry should be cited, for example, Suzuki et al. (2008), Elbern et al. (2010), Niwa et al. (2011), Goto et al. (2015)

We have increased the introductory part with respect to this and included the sentence:
"Recent work also includes the development of chemistry-climate models on icosahedral grids (Suzuki et al., 2008; Elbern et al., 2010; Niwa et al., 2011; Goto et al., 2015; Rieger et al., 2015)."

P.2, L.28: "to to" => "to"

We have changed this.

P.4, L.9: What is the overbar of rho?

It means that the air density is Reynolds-averaged (see Rieger et al., 2015).
We have included it in the paper.

P.8, L.14: I cannot understand the summation in Eq. (2).

We have corrected the equation and explained all the symbols (of course the numbers of the equations herein differ from that used in the paper):

"Generally, the VMR is defined as fraction of the number of moles of the tracer (in our case the number of moles of the emission Δn_i) and the number of moles of (moist) air n_{air}:

$$\Delta X_{\mathrm{emi},i} = \frac{\Delta n_i}{n_{\mathrm{air}}} \tag{1}$$

The moles of the emission are calculated as the emission mass flux density E_i multiplied by the advective model time step Δt and the base area A of the grid box and divided by the molar mass of the species M_i:

$$\Delta n_i = \frac{E_i \, A \, \Delta t}{M_i} \tag{2}$$

The emission flux can be included into one or more lowest model levels to be specified in the LaTeX table, see Fig. 3. In the following, we will refer to this number as $n_{\mathrm{lev,emi}}$. The total number of model layers is stated as n_{lev}. In ICON, the lowest model layer has the highest index so that the index of the lowest model layer is $l = n_{\mathrm{lev}}$. For calculating the number of moles of the air we sum up the moles of air of the lowest $n_{\mathrm{lev,emi}}$ model layers using the ideal gas law:

$$n_{\mathrm{air}} = \sum_{l=n_{\mathrm{lev}}-n_{\mathrm{lev,emi}}+1}^{n_{\mathrm{lev}}} n_{\mathrm{air},l} = \sum_{l=n_{\mathrm{lev}}-n_{\mathrm{lev,emi}}+1}^{n_{\mathrm{lev}}} \frac{p_l \, V_l}{R^* \, T_l} = \frac{A}{R^*} \sum_{l=n_{\mathrm{lev}}-n_{\mathrm{lev,emi}}+1}^{n_{\mathrm{lev}}} \frac{p_l \, h_l}{T_l} \tag{3}$$

Accordingly, p_l, T_l, h_l and R^* stand for pressure, temperature and geometric height of the grid box and the universal gas constant, respectively.
With Eqs. (2) and (3) the VMR tendency of the emission $\mathrm{d}X_{\mathrm{emi},i}/\mathrm{d}t$, which is added to the tracer, is calculated according to:"

$$\frac{\mathrm{d}X_{\mathrm{emi},i}}{\mathrm{d}t} \approx \frac{\Delta n_i}{n_{\mathrm{air}}\,\Delta t} = \frac{E_i\,R^*}{M_i} \cdot \left(\sum_{l=n_{\mathrm{lev}}-n_{\mathrm{lev,emi}}+1}^{n_{\mathrm{lev}}} \frac{p_l\,h_l}{T_l} \right)^{-1} \tag{4}$$

P.9, L.1-2: These sentences are not clear to me.

We have separated the calculation of the number of moles of the emission from Eq. (2) and reformulated these sentences (see above). As can be seen, the number of moles Δn_i is independent of the emission height $n_{\mathrm{lev,emi}}$. We have reformulated the sentence:
"This method conserves mass of the emission since the calculated moles of the emission Δn_i are independent of the choice of $n_{\mathrm{lev,emi}}$ and therefore do not change if $n_{\mathrm{lev,emi}}$ is increased."

P.9, L.9: The biomass burning emission seems duplicated. The MACCity inventory includes biomass burning, while another explicit biomass burning data of GFED is also added.

We actually only use the anthropogenic dataset and have removed the "and biomass burning" in the paper.

P.11, L.7: "leaf area index" => "leaf area index (LAI)" P.11, L.8: Delete "(LAI)" P.11, L.11: "leaf area index" => "LAI"

We have changed that.

P.13, L.5: Why is the online emission so much higher than the offline one, although they are made by the same MEGAN?

The advantage of using online emissions lies in the much higher temporal resolution of the input parameters, in case of MEGAN especially the temperature. Thus, emissions are calculated every model time step in contrast to the offline emissions which usually have a monthly temporal resolution. Therefore, it is clear that differences in the emission output occur.
In addition, our configuration is different from that used by Sindelarova et al. (2014) as described in Sect. 3.1.4 and 3.2. The input parameters and metadata come from another model and we adapted the MEGAN model which is described in Sect. 3.2. Hence, although MEGAN in ICON-ART and in Sindelarova et al. (2014) are based on the same source code of Guenther et al. (2012), its implementation is model-specific.
We have included a new figure where the sensitivity of the MEGAN-Online emissions on LAI is demonstrated and discussed (in Section 3.2 in the paper):

"In order to investigate the influence of the parametrisation of LAI by Eq. (7) we show in Fig. 8 the distributions of LAI and LAI_{sun}, together with its influence on the acetone emission. As expected, large values in LAI (top panel) occur over the Amazon region in South America as well as in Central Africa where also the acetone VMR in Fig. 6 maximises. In addition, the forest areas in the east of Canada, northern Europe and Siberia show large values of the LAI. In these regions, the LAI is in the order of 3 to 6 $\mathrm{m}^2\,\mathrm{m}^{-2}$.

For the used solar zenith angle of $10.3°$, the parametrisation according to Eq. (7) smoothes and reduces the LAI to values around $1\,\mathrm{m}^2\,\mathrm{m}^{-2}$ (Fig. 8B). Only for the less vegetated regions such as desserts (Sahara or Atacama), the distribution of LAI_{sun} shows nearly no response to the parametrisation of Dai et al. (2004).

In the MEGAN model the emission mass flux density is proportional to LAI (Guenther et al., 2012). That is why the resulting emissions in MEGAN-Online (Fig. 8C) depend linearly on the LAI for each shown plant type. The highest sensitivity on LAI can be seen for broadleaves in the tropics. Thus, the parametrisation of the LAI according to Dai et al. (2004) can lead to a reduction of the emission in the order of factor 2 to 3 in these regions.

To conclude, the correct treatment of LAI is crucial to get realistic results of the emissions in MEGAN. The parametrisation according to Dai et al. (2004) leads to emission flux densities in the same order of magnitude as in the offline data set MEGAN-MACC (see Fig. 7). Further investigation of this will be presented in Sect. 7."

P.14, L.4: What of Sander et al. (2011) is used?

We have adapted the sentence: "Cross sections and quantum yields are given in a tabulated form originating from Sander et al. (2011) and interpolated on given pressure and temperature values of Cloud-J."

Section 4.2: Is this reaction method for the stratosphere similar to those of other models?

The OH parametrisation as described in Section 4.1 is only valid for tropospheric conditions. In the paper, we are interested in UTLS acetone which is mainly driven by emissions at the surface. As shown in Fig. 7 (of the non-corrected manuscript) our definition of the UTLS region ranges high enough so that the stratospheric chemistry should not really disturb the simplified OH chemistry mechanism.

P.16, L11: "(IFS)" Please cite a paper and list it in Reference, not describing the URL in the footnote.

We have cited it.

P.16, L.20-P.17,L.1: "The air pressure corresponding ...in the CH4 VMR." This reason is not enough for the validity of using 1ppmv CH4 as the threshold.

We have rephrased the whole paragraph to clarify this:

"In Fig. 9, the zonal maximum of the air pressure where CH4 VMR decreases below $1\,\mathrm{ppmv}$ (blue dashed) is illustrated along with the zonal minimum of the WMO tropopause pressure (black solid). Additionally, the zonally averaged VMR of CH_4 at the tropopause is shown (red dotted) which ranges from 1.6 (Sounthern Hemisphere) to $1.68\,\mathrm{ppmv}$ (Northern Hemisphere). Due to its relatively long tropospheric lifetime, CH_4 is well-mixed in the troposphere and the CH_4 VMR does not decrease below $1\,\mathrm{ppmv}$. Above the tropopause, the CH_4 VMR decreases with height because of higher photolysis rates in the stratosphere.
As can be seen in Fig. 9, the lowest height where the CH_4 VMR decreases below $1\,\mathrm{ppmv}$ is clearly above the tropopause so that the OH mechanism is also applied in the lowermost stratosphere."

P.17, L.14: "110 to 261 and 373 to 528" Are they flight numbers? And where did the aircraft fly to? Please clarify.

Yes, they are the CARIBIC flight numbers. We have included a statistic of the destinations of the flights used for the climatologies. It can be found in Appendix B in the paper.

P.17, L.20-21: "All the simulations ...in the tracer concentrations" is not clear to me.

If the output interval is e.g. daily, we can only investigate OH concentrations at e.g. 00 UTC. However, the OH concentration strongly depends on the daily cycle and therefore also the compounds corresponding to the OH mechanism. That is why we chose an output interval less than daily.
We have rephrased this sentence: "All the simulations include an output interval of 23 hours. With this interval, we are able to see the impact of OH on acetone at different times of day without using too many resources."

Appendix A: Description of tau is needed somewhere.

We have added it.

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