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## Answer to comment of referee #1

### 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. When we talk about the "concentration" we mean the "number concentration" (in number of molecules per volume unit), just to clarify the used expressions. In addition, we have changed the variable name of the number of model layers of the emissions from  $k_{\text{emi}}$  to  $n_{\text{lev,emi}}$  and we refer to this number several times in our responses.

## 1 General comments

**Emissions of gas phase tracers, in particular surface emissions, can be brought into the model by at least three methods: (i) as a flux condition at the surface to vertical diffusion. In that case, a net flux on the surface is calculated by adding the dry deposition flux. The vertical diffusion distributes the tracer in the boundary layer. (ii) as a source term in the chemical kinetic equations in some appropriate model layers. (iii) as a tendency in some appropriate model layers near the surface. The authors chose method (iii) but should discuss the other methods also which are all associated with a certain operator splitting.**

Since we aim to follow the process splitting strategy of ICON (Rieger et al., 2015) we decided not to include emissions according to method (ii). Method (i) can only be used for one of the physics packages of ICON: Either the physics package for numerical weather prediction (NWP) or for climate projections (ECHAM physics). In case of method (iii) the algorithm for including the emissions follows the process splitting strategy as well as it is compatible with both the NWP and the ECHAM physics package.

Nevertheless, we performed a sensitivity test by including the emission mass fluxes in the NWP turbulence scheme. If the same variables are used as input, methods (i) and (iii) differ below 0.1 % and therefore are equivalent if the emissions are included into the lowest model layer, see Figure 1 herein.

We added the following sentence in Section 3: "Because of our aim to follow the process splitting concept of ICON (Rieger et al., 2015) and in order to be compatible with ICON for both numerical weather prediction and climate projections the emission mass flux densities are converted to volume mixing ratio and added to the tracer volume mixing ratios."

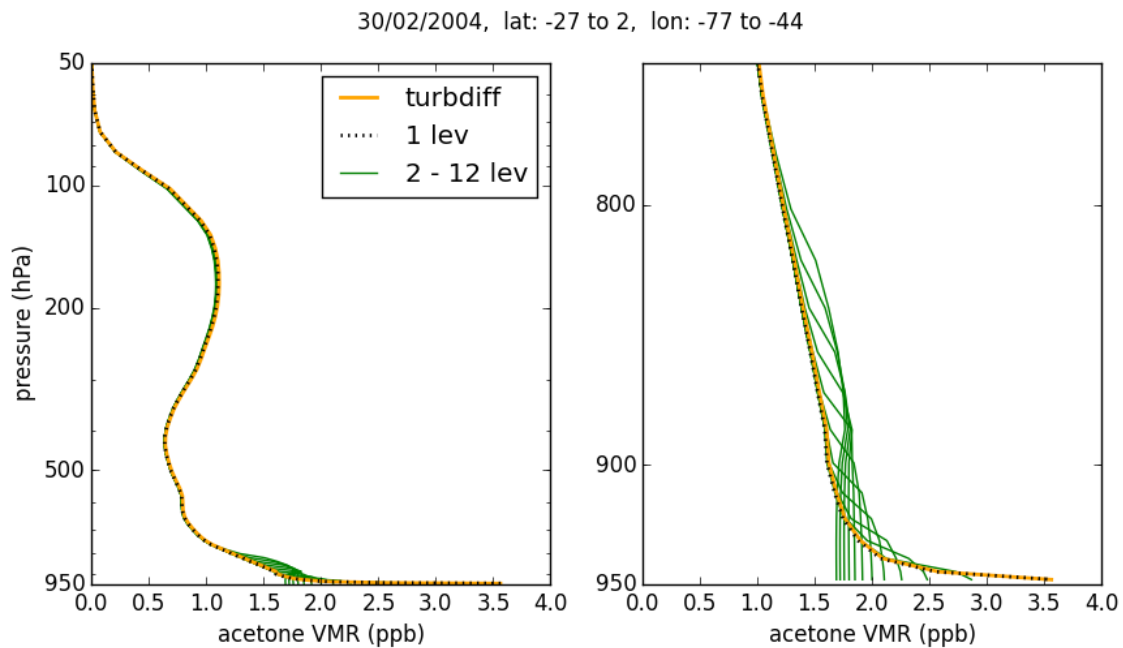


Figure 1: Spatially averaged profiles of the acetone VMR for different methods: adding emission fluxes to vertical turbulent diffusion (orange), method described in paper for  $n_{lev,emi} = 1$  (emission height of 20 m, black dotted) and 2 to 12 (emission height of 65 m to  $\sim 1500$  m above ground, green thin lines).

**What concerns me most is the fact that the criteria are unclear according to which the number of lowermost layers are chosen into which the emissions are brought as a tendency.**

We have added a paragraph in Section 3.1.3 including Figure 1 in this answer. In this new paragraph, we describe that we select  $n_{lev,emi} = 1$  because the shown profiles are nearly identical above the height of around 750 hPa. Since our aim in the paper is the simulation of acetone in the UTLS region our results should be robust against other choices of  $n_{lev,emi}$ .

Our change in the manuscript:

"To investigate the differences in changes of  $n_{lev,emi}$  we perform sensitivity simulations of acetone by varying  $n_{lev,emi}$  between 1 and 12. These simulations are based on constL(megan-offl), see Sect. 6. In Figure 5, profiles of the acetone VMR are shown for the different choices of  $n_{lev,emi}$ .

In the case of  $n_{lev,emi} = 1$ , no emissions are included in the layers above. For larger values of  $n_{lev,emi}$  the VMR in the lowermost model layer decreases subsequently since the emissions are distributed into a larger column.

Above the specified emission height, all profiles converge each other and above around 750 hPa the influence of varying  $n_{lev,emi}$  is negligible. Because of our aim to simulate acetone in the UTLS region, the choice of  $n_{lev,emi}$  should make no difference. That is why we simply select  $n_{lev,emi} = 1$  for all used offline emissions."

**The authors should discuss this choice, prepare an appropriate sensitivity study varying the number of levels and perform a simulation with emissions given as a flux condition to the vertical diffusion equation.**

We have done that (see above and Figure 1 herein).

**In this latter simulation, the resulting tendencies in the lowermost model layers should be compared with the number of model layers used in method (iii). It would be particularly interesting to see the seasonal variation.**

As described above, we could show that both methods are equivalent. To avoid confusion, we focus on the method that has been chosen as the default implementation for ICON-ART.

**Discuss other chemistry general circulation models and what methods they use**

We have increased the introduction section of the paper with respect to this:

"Different approaches to include emissions in atmospheric modelling have been developed in the past and are used in current chemistry climate models: In the limited area chemistry model WRF-chem (Grell et al., 2005) emissions are treated as production terms in the chemical equations (McKeen et al., 1991). Emissions can be prescribed as a flux condition in the vertical diffusion, as e.g. in the Community Atmosphere Model (Lamarque et al., 2012; Neale et al., 2013) which is part of the Community Climate System Model (CCSM, Gent et al., 2011). This method is also used for emissions in the planetary boundary layer in the GEOS-Chem model (Bey et al., 2001) including the HEMCO module (Keller et al., 2014). Emissions in higher altitudes are brought into GEOS-Chem as a tendency in the respective height of the emissions (C. A. Keller, pers. comm., 2017). The MESSy interface (Jöckel et al., 2005) incorporated e.g. in the EMAC model (Jöckel et al., 2006) gives the possibility to choose the used method for including emissions into the model: Either emissions are prescribed as flux condition as described above or the increase of the tracer mixing ratio is calculated and added to the tracer (Kerkweg et al., 2006). The latter method is also used in the coupled limited area model COSMO-ART (COSMO: CONSORTIUM FOR SMALL-SCALE MODELLING, ART: AEROSOLS AND REACTIVE TRACE GASES, Vogel et al., 2009)."

## **2 Specific comments**

**p.2, l.26: The ICON model is not really "in development" anymore since the NWP physics is used for operational weather forecast.**

We have rephrased this sentence:

"ICON is a non-hydrostatic atmospheric model developed with the aim of providing a global model for both weather and climate (Wan et al., 2013; Zängl et al., 2015). Since January 2016, it is operationally used for global numerical weather prediction at German Weather Service (DWD). In July 2016, ICON also replaced the limited area model COSMO-EU (Baldauf et al., 2011) by a nested area over Europe."

**p.3, Tab.1: The "official ICON grid number": Give a citation here.**

We have cited the web site where the global grids currently can be downloaded from: [http://icon-downloads.zmaw.de/dwd\\_grids.xml](http://icon-downloads.zmaw.de/dwd_grids.xml)

On this web site, the grids are called "official" grids.

**p.3, l.1: It should be clarified that Leuenberger et al. generalized the SLEVE coordinates**

We have done that.

**p.3, l.5 ff: The usual definition of volume mixing ratios is moles tracer per moles dry air. In ICON, tracers are defined per moist air for the horizontal transport. How do you treat these different definitions? How did you check mass conservation?**

We have reformulated Eqs. (2) and (3) so that it should be clear that the moles of air are calculated by the ideal gas law using pressure and temperature values of ICON. As these equations are then independent of the explicit molar mass of the air, the moles of the air are that of moist air.

With respect to the second question: The mass conservation of the tracers in ICON was discussed by Zängl et al. (2015). Since ICON-ART uses the ICON tracer structure to calculate the tracers this is also valid for ICON-ART.

The conversion to VMR in Sect. 3.1.3 is now formulated in the paper as follows (of course with other equation numbers):

"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  $\Delta n_i$ ) and the number of moles of (moist) air  $n_{\text{air}}$ :

$$\Delta X_{\text{emi},i} = \frac{\Delta n_i}{n_{\text{air}}} \quad (1)$$

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

$$\Delta n_i = \frac{E_i A \Delta t}{M_i} \quad (2)$$

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  $n_{\text{lev,emi}}$ . The total number of model layers is stated as  $n_{\text{lev}}$ . In ICON, the lowest model layer has the highest index so that the index of the lowest model layer is  $l = n_{\text{lev}}$ . For calculating the number of moles of the air we sum up the moles of air of the lowest  $n_{\text{lev,emi}}$  model layers using the ideal gas law:

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

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  $dX_{\text{emi},i}/dt$ , which is added to the tracer, is calculated according to:"

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

**p.5, Fig.2: "reset simulation year from boundary year back to current year": It's not so clear what you mean here. You would like to say that the last year for which emissions are given is repeated for all years later than this year. By the way, this may even be error prone repeating the last year without warning.**

We have adapted the figure accordingly. In the ICON output a message is given for each file that is read so that the user is able to trace which files are used during runtime.

**p.6, l.10: Emissions are interpolated by a nearest neighbour interpolation. A flux conserving interpolation would sound more natural since you like to have the same amount of tracer going into the atmosphere irrespective of the used horizontal model resolution. Discuss this.**

We have investigated the total mass fluxes of the emissions for different resolutions. The global mass fluxes differ below 1 %.

We have included the following sentence in the manuscript: "This method also conserves the total emission fluxes reasonably with a maximum deviation of 1 % in case of R2B04 and a less deviation for the other resolutions of Table 1 (not shown)."

**p.6, l.14: "for each time step" may sound like model time step here, but you mean times for which emissions are provided. Please, rephrase.**

We have done that: "Therefore the emission data have to be split into separate files according to their validity time."

**p.7, Fig. 3: Lines are cut and not completely displayed.**

That it is why it is called "extract" in the figure description. In addition, the important part, i.e. the tracer emission metadata, is shown completely.

**p.7, l.5: Fig. 3 shows LATEX not TEX code.**

We have replaced TeX by LaTeX.

**p.7, l.6: "the number of emission": is it emission sectors, emission types? Be more specific.**

We wanted to say "the number of emission types". We have included this word in the paper.

**p.7, l.7: "...account only the number..." an "if" is missing. "...is then used globally": Explain better that "the number" is then a globally applied emission mass flux.**

We have rephrased this sentence: "The standard value is taken into account only if the number of emission types is zero. Then it is used as the globally applied emission mass flux density. Otherwise [...]"

**p.8, l.2: "emission date" may be misinterpreted as the date when emissions are applied (actually, they are applied in every time step), but "emission date" refers to the date for which emission fluxes are provided. Please, rephrase.**

We have rephrased this sentence: "The first task of the module during runtime is to find the two dates closest to the simulation time where emission are available in the dataset."

**p.8, l.9: Just a remark: Is it necessary to have such a fixed time limit of 11 years? Volcanic emissions could be very irregular, farther away than 11-years and should not be interpolated?**

The module was created for the treatment of gas phase emissions from emission inventories. The used value of about 11 years is an arbitrarily chosen stop criterion that is far beyond the time resolution of commonly used inventories such as MACCity, MEGAN-MACC and GFED3. Volcanic emissions are treated in another way in ICON-ART (Rieger et al., 2015).

**p.8, l.13: "After interpolation the emission is converted to VMR ( $C_{emi,i}$ ): Rephrase "emission" to "emission flux"; furthermore, you mean a VMR tendency here, the symbol  $c$  is normally used for molarity, so  $dX_{emi,i}/dt$  would be the correct symbol. Devide eq. (2) by time.**

We have included "flux" and reformulated Eq. (2) with respect to this (see above).

**p.8, eq.(2): In mathematics, sums are not counting backwards. [...]**

We have changed that.

**p.9, l.1: Explain the choice of  $k_{emi}$  in more detail.**

We have included a paragraph discussing this choice (see above in our answers to the General Comments).

**p.9, l.5: Websites are not a good reference in general. If necessary, add in a footnote the date when you accessed the sites the last time. Similarly: p. 9, l. 15, p. 11, l. 25, p. 16 the footnote, p. 22, l. 3/5**

We have done that and where possible we have given another citation.

**p.11, l.17: Avoid to start a sentence with a mathematical symbol, in particular when it repeats the symbol of the last sentence**

We have adapted this sentence.

**p.11, l.28: "derived" instead of "defined".**

We have changed that.

**p.12, l.6: "sunlit leaves" instead of "sun leaves"**

We are aware that the notions "sun leaves" and "sunlit leaves" have different meanings: The term "sunlit leaves" (and "shaded leaves") is used for expressing that vegetation is either directly lit by sun or shaded by other vegetation, which is also used by Dai et al. (2004). The discrimination between "sun leaves" and "shade leaves" is a botanical discrimination.

We actually used the same terms as Dai et al. (2004, "sunlit leaves") and Guenther et al. (2012, "sun leaves") in the paper.

In the manuscript we now consistently follow the naming given by Dai et al. (2004) and rephrased the sentence:

"For standard conditions, we use the average Photosynthetic Photon Flux Density (PPFDS) of the values given by Guenther et al. (2012):  $PPFDS = 125 \mu\text{mol m}^{-2} \text{s}^{-1}$ ."

**p.13, fig.6: Explain better how you calculate the means. First, the word "global" is irritating, since you are calculating means over "S-America"? In fact, you calculated means over a rectangle in longitude and latitude with sea points contributing to the surface but not to the emissions.**

We have excluded the global means from this figure and included a table with the global mass fluxes (now Table 5 in the paper).

**p.15, l.7: "...due to Reactions (R5) and (R6)" you probably like to say that (R5) is the rate-determining step.**

Yes, we do. We have included this in the paper:

"Reaction 5 results in a cascade of fast reactions and finally in a production of CO and is the largest source for atmospheric CO (Jacob, 1999; Boucher et al., 2001; Seinfeld and Pandis, 2012,

pp. 46–47). Since Reaction 5 is the reaction with lowest reaction rate of this cascade the chemical production of CO can be estimated as follows:"

**p.17, l.20: ICON contains several time steps, which one do you mean?**

In the documentation of ICON, it is called the "advective" time step which is equivalent to the time step of the fast physics (see Rieger et al., 2015). We have adapted this in the paper where we previously referred to the "model time step".

**p.17, l.20: Output interval of 23 hours, meaning output at 0h, 23h (of next day), 22h (of next day), 21h (of next day), and so on?**

Yes, that is right. Please see next comment for an explanation.

**p.18, l.1: Temporal variability of OH at which time scale?**

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 adapted the 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."

**p.19, l.15/20/ Fig. 8: OH-chem(off): Better choose OH-chem(megan-off) or similar acronyms to make clear that it is not the OH-chemistry being switched off. Fig. 8: Show the free troposphere in an additional panel such that the interannual variability becomes visible.**

We have adapted the simulation names to e.g. constL(megan-offl) and OH-chem(megan-onl).

With respect to Figure 8: The interannual acetone lifetime in the free troposphere only differs by 1.7 days in the maximum. That is why we think that the current figure is appropriate for the paper and no additional figure is needed.

**p.19, l.12 ff: Your reasons for the underestimation are pure hypotheses and more confusing than explaining your results. The reader is lost what refers to your simulations and what is speculation. Make it clear where your considerations are general and what concerns your situation.**

We have reconsidered our first argument because only the second and third ones of the listing are based on literature. Additionally, we have rephrased the remaining sentences:



"(1) We account for chemical production of acetone due to reaction of propane with OH but neglect the contribution of minor VOCs such as monoterpenes. The high impact of monoterpenes on acetone calculated by Khan et al. (2015) was recently challenged by Brewer et al. (2017). On the other hand, we neglect the weak uptake of acetone by the oceans and dry deposition which would decrease the acetone VMR slightly (Fischer et al., 2012; Khan et al., 2015). (2) [...]"

**p.20, fig.9: Remove unnecessary axis and titles of the plots. Create one color bar for all panels instead. Instead of a pressure range, give the geometric altitude of the tropopause.**

We have adapted the figure accordingly. As described in the text on p.19, 1.5-7 the pressure range given in the figures refers to the pressure range where measurement data is considered.

We have added this information to the figure description.

**p.20, 1.3ff: Describe your result first, then interpret/explain**

Due to a new sensitivity simulation, we have adapted the whole paragraph and included a description of the figure:

"The acetone VMR around the tropopause using MEGAN-Online *LAI* is shown in the rightmost column of Fig. 12."

**p.20, 1.13: Acetone life time of 1.5 years versus 28 days? This is worth an explanation.**

The main difference between these two values lies in their region and time scales they represent: The value of 28 days is a global annual mean value whereas the lifetime of 1.5 years stands for the northern winter mid-latitude average (35 to 75 °N).

In contrast to the global average, the low sun during (northern) winter decreases the photolysis rates related to OH and acetone in the mid-latitudes by one to two orders of magnitude. As the acetone loss rate in our simplified model is proportional to these photolysis rates, its lifetime increases accordingly.

We have included this sentence after p.20, 1.13: "This value is a mid-latitude (35 to 75° N) average for the months December to February in 2005 to 2015 in contrast to the global annual average mentioned above."

**p.21, 1.5 ff: The conclusions should contain information about the choice of  $k_{emi}$  and the sensitivity studies versus a flux condition in vertical diffusion.**

We have added a paragraph in the conclusions describing our choice of  $n_{lev,emi}$ :

"[...] the number of lowest model levels of the emission  $n_{lev,emi}$  has to be specified where we show a sensitivity test by varying this number. Differences only occur in the height of the emission itself.

Therefore, the tracer mixing ratio above the emission height  $n_{\text{lev,emi}}$  is independent of the choice of  $n_{\text{lev,emi}}$ . Since our focus in the results is the comparison to measurements in the upper troposphere and lowermost stratosphere (UTLS), we choose  $n_{\text{lev,emi}} = 1$ .

**p.22, l.1-5: Code availability: give a contact person only since there seems to be no icon license available at the site given (address not found!). The ART license is referring to a person anyhow.**

We have adapted the code availability paragraph by giving contact persons.

**Appendix A: "Concentration" should be either "molarity" or "number concentration" since concentration can be anything, mole fraction, volume mixing ratios, molality, molonity and the like. It's confusing to have a species index and a time step index on  $c$  in the same section. Put  $c_{i,t}(x)$  where  $x$  refers to the location,  $i$  to the species and  $t$  is time. Write the equations for an arbitrary integration step from  $t$  to  $t + \Delta t$ . From (A2) on you can, if you like, omit the index  $i$ . Mark the various solutions with superscripts <sup>(e)</sup> for explicit Euler and <sup>(pc)</sup> for the predictor-corrector method.**

We have clarified the expression concentration and adapted the indices of  $c$ : "We here refer to 'concentration' as an abbreviation of number concentration."

**p.23, l.9: Better: "If  $\Delta t$  becomes larger than...". What do you mean with "unstable"?**

We now give an example in the paper where we assume the chemical production  $P$  in Eq. (A6) to be zero, i.e.:  $P_* = P_n = 0$  and additionally  $\tau_* + \tau_n - \Delta t < 0$ . In this case, the concentration of the next time step  $c_{n+1}$  becomes negative which obviously shows that the numerical solution does not converge the physical solution of the differential equation.

**p.27, l.8: Give the page in Pandis/Seinfeld.**

We have added it for each citation in the main text.

### 3 technical corrections

We have corrected all the technical mistakes.

## References

Baldauf, M., Seifert, A., Förstner, J., Majewski, D., Raschendorfer, M., and Reinhardt, T.: Operational Convective-Scale Numerical Weather Prediction with the COSMO Model: Descrip-

- tion and Sensitivities, *Mon. Weather Rev.*, 139, 3887–3905, doi:10.1175/MWR-D-10-05013.1, 2011.
- Bey, I., Jacob, D. J., Yantosca, R. M., Logan, J. A., Field, B. D., Fiore, A. M., Li, Q., Liu, H. Y., Mickley, L. J., and Schultz, M. G.: Global modeling of tropospheric chemistry with assimilated meteorology: Model description and evaluation, *J. Geophys. Res.: Atmospheres*, 106, 23 073–23 095, doi:10.1029/2001JD000807, 2001.
- Boucher, O., Haigh, J., Hauglustaine, D., Haywood, J., Myhre, G., Nakajima, T., Shi, G., and Solomon, S., eds.: *Radiative Forcing of Climate Change. In: Climate Change 2001: The Scientific Basis. Contribution of Working Group I to the Third Assessment Report of the Intergovernmental Panel on Climate Change*, Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 2001.
- Brewer, J. F., Bishop, M., Kelp, M., Keller, C., Ravishankara, A. R., and Fischer, E. V.: A sensitivity analysis of key natural factors in the modeled global acetone budget, *J. Geophys. Res.: Atmospheres*, doi:10.1002/2016JD025935, 2016JD025935, 2017.
- Dai, Y., Dickinson, R., and Wang, Y.: A Two-Big-Leaf Model for Canopy Temperature, Photosynthesis, and Stomatal Conductance, *J. Clim.*, 17, 2281–2299, doi:10.1175/1520-0442(2004)017<2281:ATMFCT>2.0.CO;2, 2004.
- Fischer, E., Jacob, D. J., Millet, D., Yantosca, R. M., and Mao, J.: The role of the ocean in the global atmospheric budget of acetone, *Geophys. Res. Lett.*, 39, doi:10.1029/2011GL050086, L01807, 2012.
- Gent, P. R., Danabasoglu, G., Donner, L. J., Holland, M. M., Hunke, E. C., Jayne, S. R., Lawrence, D. M., Neale, R. B., Rasch, P. J., Vertenstein, M., Worley, P. H., Yang, Z.-L., and Zhang, M.: The Community Climate System Model Version 4, *J. Clim.*, 24, 4973–4991, doi:10.1175/2011JCLI4083.1, 2011.
- Grell, G. A., Peckham, S. E., Schmitz, R., McKeen, S. A., Frost, G., Skamarock, W. C., and Eder, B.: Fully coupled online chemistry within the WRF model, *Atmos. Environ.*, 39, 6957–6975, doi:10.1016/j.atmosenv.2005.04.027, 2005.
- Guenther, A., Jiang, X., Heald, C., Sakulyanontvittaya, T., Duhl, T., Emmons, L., and Wang, X.: The Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2.1): an extended and updated framework for modeling biogenic emissions, *Geosci. Model Dev.*, 5, 1471–1492, doi:10.5194/gmd-5-1471-2012, 2012.
- Jacob, D. J.: *Introduction to atmospheric chemistry*, Princeton Univ. Press, Princeton, NJ, 1st edn., 1999.
- Jöckel, P., Sander, R., Kerkweg, A., Tost, H., and Lelieveld, J.: Technical Note: The Modular Earth Submodel System (MESSy) - a new approach towards Earth System Modeling, *Atmos. Chem. Phys.*, 5, 433–444, doi:10.5194/acp-5-433-2005, 2005.

- Jöckel, P., Tost, H., Pozzer, A., Brühl, C., Buchholz, J., Ganzeveld, L., Hoor, P., Kerkweg, A., Lawrence, M.-G., Sander, R., Steil, B., Stiller, G., Tanarhte, M., Taraborrelli, D., van Aardenne, J., and Lelieveld, J.: The atmospheric chemistry general circulation model ECHAM5/MESSy1: consistent simulation of ozone from the surface to the mesosphere, *Atmos. Chem. Phys.*, 6, 5067–5104, doi:10.5194/acp-6-5067-2006, 2006.
- Keller, C. A., Long, M. S., Yantosca, R. M., Da Silva, A. M., Pawson, S., and Jacob, D. J.: HEMCO v1.0: a versatile, ESMF-compliant component for calculating emissions in atmospheric models, *Geosci. Model Dev.*, 7, 1409–1417, doi:10.5194/gmd-7-1409-2014, 2014.
- Kerkweg, A., Sander, R., Tost, H., and Jöckel, P.: Technical note: Implementation of prescribed (OFFLEM), calculated (ONLEM), and pseudo-emissions (TNUDGE) of chemical species in the Modular Earth Submodel System (MESSy), *Atmos. Chem. Phys.*, 6, 3603–3609, doi:10.5194/acp-6-3603-2006, 2006.
- Khan, M., Cooke, M., Utembe, S., Archibald, A., Maxwell, P., Morris, W., Xiao, P., Derwent, R., Jenkin, M., Percival, C., Walsh, R., Young, T., Simmonds, P., Nickless, G., O’Doherty, S., and Shallcross, D.: A study of global atmospheric budget and distribution of acetone using global atmospheric model STOCHEM-CRI, *Atmos. Environ.*, 112, 269 – 277, doi:10.1016/j.atmosenv.2015.04.056, 2015.
- Lamarque, J.-F., Emmons, L. K., Hess, P. G., Kinnison, D. E., Tilmes, S., Vitt, F., Heald, C. L., Holland, E. A., Lauritzen, P. H., Neu, J., Orlando, J. J., Rasch, P. J., and Tyndall, G. K.: CAM-chem: description and evaluation of interactive atmospheric chemistry in the Community Earth System Model, *Geosci. Model Dev.*, 5, 369–411, doi:10.5194/gmd-5-369-2012, 2012.
- McKeen, S. A., Hsie, E.-Y., Trainer, M., Tallamraju, R., and Liu, S. C.: A regional model study of the ozone budget in the eastern United States, *J. Geophys. Res.: Atmospheres*, 96, 10 809–10 845, doi:10.1029/91JD00052, 1991.
- Neale, R. B., Richter, J., Park, S., Lauritzen, P. H., Vavrus, S. J., Rasch, P. J., and Zhang, M.: The Mean Climate of the Community Atmosphere Model (CAM4) in Forced SST and Fully Coupled Experiments, *J. Clim.*, 26, 5150–5168, doi:10.1175/JCLI-D-12-00236.1, 2013.
- Rieger, D., Bangert, M., Bischoff-Gauss, I., Förstner, J., Lundgren, K., Reinert, D., Schröter, J., Vogel, H., Zängl, G., Ruhnke, R., and Vogel, B.: ICON-ART 1.0 - a new online-coupled model system from the global to regional scale, *Geosci. Model Dev.*, 8, 1659–1676, doi:10.5194/gmd-8-1659-2015, 2015.
- Seinfeld, J. H. and Pandis, S. N.: *Atmospheric Chemistry and Physics: From Air Pollution to Climate Change*, John Wiley & Sons, 2nd edn., 2012.
- Vogel, B., Vogel, H., Bäumer, D., Bangert, M., Lundgren, K., Rinke, R., and Stanelle, T.: The comprehensive model system COSMO-ART - Radiative impact of aerosol on the state of the atmosphere on the regional scale, *Atmos. Chem. Phys.*, 9, 8661–8680, doi:10.5194/acp-9-8661-2009, 2009.

- 
- Wan, H., Giorgetta, M. A., Zängl, G., Restelli, M., Majewski, D., Bonaventura, L., Fröhlich, K., Reinert, D., Rípodas, P., Kornbluh, L., and Förstner, J.: The ICON-1.2 hydrostatic atmospheric dynamical core on triangular grids - Part 1: Formulation and performance of the baseline version, *Geosci. Model Dev.*, 6, 735–763, doi:10.5194/gmd-6-735-2013, 2013.
- Zängl, G., Reinert, D., Rípodas, P., and Baldauf, M.: The ICON (ICOsahedral Non-hydrostatic) modelling framework of DWD and MPI-M: Description of the non-hydrostatic dynamical core, *Quart. J. Roy. Meteor. Soc.*, 141, 563–579, doi:10.1002/qj.2378, 2015.