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Supplement of

The Chemical Mechanism Integrator *Cminor* v1.0: a stand-alone Fortran environment for the particle-based simulation of chemical multiphase mechanisms

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Contents

S1 Introduction

Dear user, we hope this documentation finds you well. This is the attempt of giving a useful manual to do simulations with Cminor. Handling this solver is believed by the authors to become (very) easy once it has been applied a couple of times. Setting up the first simulation might be overwhelming, considering the multiple files and their respective sections to set and choose. We recommend to use one of the given example simulation setups, then change the *.sys-file to a file containing the mechanism of your interest and change the rest accordingly. Before trying to describe all the technical necessities to conduct a simulation, let us give a summary of what we believe to be the most important aspects.

A simulation setup consists, beside the source code, of the following four files:

- *.sys-file,
- *.run-file,
- *.ini-file, and
- *.dat-file.

These files determine every aspect of the simulation of a chemical mechanism and each of those files can be changed without the need to recompile the entire model.

The *.sys-file contains the chemical reactions, each defined by four lines, where the last line is optional. A reaction is specified by a type (i.e., gaseous, aqueous, Henry, or dissociation) in the first line. This has several reasons, as aqueous might have to be considered multiple times for each droplet class, Henry and dissociation reactions are split up into a forward and backward reaction, etc. In the next line, the kinetics of the reaction are specified, i.e., which species react to which products. The third line sets the reaction constant, i.e., a speed of reaction, which might depend on several things (the reaction constant is constant with respect to the chemical species involved in the reaction, not generally constant). A large variety of reaction constants are implemented and the user can choose one of those freely. Alternatively, there is the possibility of using the SPECIAL reaction constant, where a specific reaction constant formula can be defined directly in the *.sys-file, on the cost of computation time. These three lines are mandatory to define a reaction. The last line is optional and called "FACTOR". This resembles a pseudo-reactant, which is essentially passive, i.e., abundant, and therefore does not change concentration. It is multiplied with the rate constant during the calculations.

In the *.run-file, simulation specifications are set. This means files, e.g. the *.sys-file to define the mechanism to simulate, integration time, meteorological parameters like liquid water content, all the numerical thresholds and tolerances and all output variables and settings.

The *.ini-file is, used to specify the initial values, i.e., initial concentrations of all species. If a species is not given, its value is set to zero. The *.ini-file includes emission and deposition rates, gaseous and aqueous initial values, aerosol composition and number, and the species to be tracked in the NetCDF files.

Last not least, the *.dat-file contains species-specific values like diffusion coefficients, molar masses, accommodation coefficients, and charge of aqueous species. These determine for example mass transfer coefficients, i.e. Henry reaction rates. Also, if RO2 factors are to be used, a list of the species to be considered to contribute has to be defined here.

In any case, the authors encourage every user to get in touch if problems are faced.

S2 Installation and testing

This section describes the installation process of Cminor. The code can be compiled on Linux and macOS machines.

1. Open a terminal and change to the Cminor home directory.

2. To clean the Cminor installation, delete the Cminor object files and the executable by typing "`make clean`".

3. Edit the M_DEF_[OS] file depending on the machine. Mac users have to specify the path to the NetCDF library folder and the path to the local include folder via
   `LocNCDF = /usr/local/lib`, and
   `LocINCL = /usr/local/include`.
   If you use a Linux machine, you just have to specify the `LocNCDF` path to the NetCDF library.

4. Make sure you have installed gfortran and mpif90. To test this, type "`which gfortran`" or "`which mpif90`".

5. Open the Makefile (the file is named Makefile) and specify the corresponding M_DEF_[OS] file (first line): "`IN1 = M_DEF_Mac`" or "`IN1 = M_DEF_Linux`".

6. Create the Cminor executable in either optimized (fast) or debugging (better error checking and tracebacks) with either one of the following commands:
   `make Cminor` (optimized version, -O3, see M_DEF_[OS]), or
   `make Cminor_dbg` (debugging version, -g -O0, see M_DEF_[OS]).

7. To run several examples type "`make test`" in the Cminor home directory. This will execute Cminor with six different mechanisms successively. The generated output is stored as NetCDF files in the NetCDF/ folder. To ensure your version of the code recreates the results of the simulations in the article, execute "`python3 PYTHONSCRIPTS/all_test_Cminor.py`", if Python is available. If the script finishes by printing "Everything went fine.", the test was successful. By executing "`python3 PYTHONSCRIPTS/plot_ncdf_overview.py`", if Python is available, the plots of Fig. 4 and Fig. 5 of the article are reproduced.

S3 Description of a chemical mechanism for atmospheric chemistry (*.sys-file)

S3.1 Head of the *.sys-file:

- All characters following a hash symbol (#) or the keyword "COMMENT" will be ignored.
- At the beginning of the document, a specification of the units for the initial data and rate constants, using the keywords UNIT GAS and UNIT AQUA, respectively, may occur. Default values are 0 for both phases. This is currently a dummy feature, as only the default values are available.

```
1: # ═══════════════════════════════════════════════════════════════
2: # ═══════════════════  CHEMISTRY  ═══════════════════════════════
3: # ═══════════  Output - Chemical Reaction Data  ═══════════════
4: # ═══════════════════════════════════════════════════════════════
5:
6: #  Created:              Wed June 14 12:00:00 2006 (AT)
7: #  Chemical Mechanism:  CAPRAM 3.0 (URBAN CASE)
8:
9: # ═══════════════════  Unit options  ═══════════════════════════
10:
11: UNIT    GAS     0   # Gas phase units  ( 0 = molec/cm3 )
12: UNIT    AQUA    0   # Aqueous phase units  ( 0 = mol/l)
```

S3.2 General directives for the reaction mechanism input structure

Syntax:

```
1: CLASS:  classname    #  Comment
2: Chemical equation
3: TypeName:  List of parameters
4: [FACTOR:  factor species ]  # optional
```

1. The input structure of one reaction consists of at least 3 lines.
2. The reaction description can begin anywhere on the line.
3. All characters following an hash symbol (#) or the keyword "COMMENT" will be ignored.
4. The length of an input line is limited to 400 characters.
5. Character "␣+␣" separates two reactants from each other. Note: Blank spaces are necessary!

6. Each reaction description must have "␣=␣" between the last reactant and the first product. All blank spaces, except the aforementioned and between reaction rate parameters, are ignored.

7. Each " *educts* = *products* " description must be contained on one line.

8. Blank lines in between the reaction structure will be ignored.

9. Line 3: The reaction rate parameters have to be separated from each other and from the reaction type description by at least one blank space; no blanks are allowed within the numbers themselves. The reaction rate parameter shall be taken from Sec. S4. The respective values have to be set either in exponential or floating point notation.

10. Colons are declared as separator for the parameter list.

11. SMILES notation for chemical species is allowed.

12. Line 4: FACTOR can be a special pseudo-species, see Sec. S3.5.

13. Higher order reactions can be declared as either: OH + OH = H2O2 or 2.0␣OH = H2O2

14. Blank spaces within species names are not allowed.

15. Examples: see Sec. S3.6

S3.3 Further syntax rules

Species names:

- the name has to start with one of these characters:

  "ABCDEFGHIJKLMNOPQRSTUVWXYZapsc[]()=+*"

- use arbitrary numbers, upper and lower case letters and SMILES notation conventions elsewhere, except special conventions as:

Special conventions:

aXXX	-	dissolved substance in aqueous phase (e.g., aO3)
XXXp	-	cation (e.g., H^+ = Hp , Fe^{3+} = Feppp)
XXXm	-	anion (e.g., OH^- = OHm)

Other conventions:

[species name] - passive species, is used for calculation of the rate constant, concentration will not be calculated dynamically (e.g., [O2])

(species name) - species acts like a dummy, concentration will not be calculated dynamically (e.g., (dummy))

$ - external species for microphysical properties (e.g., $N2, see Section S3.5)

S3.4 `CLASS` and `TYPE` dependencies

CLASS:	**Description**
`GAS`	gaseous reaction
`HENRY`	phase transfer reaction (equilibrium pseudo-reaction)
`DISS`	chemical dissociation (equilibrium reaction)
`AQUA`	aqueous reaction

The following table gives an overview about the possible combinations of reaction classes and types.

CLASS:	GAS	HENRY	DISS	AQUA
PHOTABC:	X			X
PHOTAB:	X			X
PHOTMCM:	X			X
PHOTO:	X			X
PHOTO2:	X			X
PHOTO3:	X			X
CONST:	X	X		X
TEMP:	X	X		X
TEMP1:	X	X		X
TEMP2:	X	X		X
TEMP3:	X	X		X
TEMP4:	X	X		X
S4H2O:	X			
T1H2O:	X			
TROE:	X			
TROEF:	X			
TROEQ:	X			
TROEQF:	X			
TROEXP:	X			
TROEMCM:	X			
SPEC1:	X			
SPEC2:	X			
SPEC3:	X			
SPEC4:	X			
SPEC1MCM:	X			
SPEC2MCM:	X			
SPEC3MCM:	X			
SPEC4MCM:	X			
SPEC5MCM:	X			
SPEC6MCM:	X			
SPEC7MCM:	X			
SPEC8MCM:	X			
SPEC9MCM:	X			
HOM1:	X	X		X
ASPEC1:				X
ASPEC2:				X
ASPEC3:				X
DTEMP:			X	
DTEMP2:			X	
DTEMP3:			X	
DTEMP4:			X	
DTEMP5:			X	
DCONST:			X	

Table S1. Overview of possible combinations for reaction types.

S3.5 Modification of a rate constant by FACTOR

CLASS: GAS

HO = HO2

TEMP1: K0: 5.5E − 12 E/R: 2000.0

FACTOR: $H2

The reaction rate constant k^* (here of type TEMP1) is modified by factor

$k^* \cdot [H_2]$.

In principle, all rate constants can be modified. Available factors are

$H2, $O2N2, $M, $O2, $N2, $H2O, $O2O2, $aH2O, $RO2, and $RO2aq.

The concentration of air molecules $[M]$ is calculated according to the pressure and temperature specified in the *.run-file using the ideal gas law. Default values for temperature and pressure are 280 K and 850 hPa in the atmospheric case, respectively. In the same manner, the concentration of water vapor is calculated by the specified relative humidity, with default value 0.6. If the adiabatic parcel option is activated, this value changes according to the current mixing ratio. The concentrations of N_2, O_2, and H_2 are then calculated according to the fractions 78.08%, 20.95% and 0.000055% of the dry air molecules, respectively. The factor $O2N2 refers to the product of the concentrations of O_2 and N_2, $O2O2 is the square of the O_2 concentration, [aH2O] is determined by the current LWC and [RO2] and [RO2aq] are the sum of all RO2 and RO2aq species' current concentrations. The names of the species to be considered RO2 and RO2aq need to be listed in the *.dat-file, see Sec. 7.

S3.6 Examples

```
# ================ MCM3.1 + CAPRAM ================
# = Please copy the data into your sys-file for =
# ================ chemical input. ================
#
#  ====================  Unit options  ====================

UNIT GAS     0   #     Gas phase units      (0 = molec/cm3 )
UNIT AQUA    0   #     Aqueous phase units (0 = mol/l)

#  ====================  Gas Phase  ====================

CLASS: GAS
HO = HO2
TEMP1:   K0:  5.5E-12      E/R:   2000.
FACTOR:   $H2

COMMENT chlorine chemistry -- propylene oxidation
CLASS: GAS  #G77
CL + CC=C = CC(O[O])CCL
TROE:    KO:  4.0E-28  N:   0.0   KINF:   2.8E-10  M:   0.00

#  ====================  Phase transfer  ====================

CLASS: HENRY
SO2 = aSO2
TEMP3:   K0:  1.23E0    B:   3.12E3

#  ====================  Aqueous phase  ====================

CLASS: AQUA      #Po00001
aCOO = aC[O] + aHO
PHOTMCM:     I:  8.625e-06  M:  1.043    N:  0.271
```

S4 Available reaction rate constants

Legend for the variables used in the following:

$\chi := \chi(t)$	zenith angle being time dependent
β	number of products - educts
γ	number of educts
$\varepsilon_{\mathrm{dust}}$	loss factor (where $0 \leq \varepsilon_{\mathrm{dust}} \leq 1$, usually: $\varepsilon_{\mathrm{dust}} = 1.0$)
T	current temperature
T_{ref}	reference temperature (298.15 K)
p	current pressure
p_0	standard atmospheric pressure (1013.25 hPa)
$[M]$	molecularity at current pressure and temperature (see Sec. 3.5)

Figure S1. Parameters for the calculation of reaction rates within atmospheric chemistry simulations.

S4.1 Creating custom-built rate constant functions

CLASS: *classname* # comment

chemical equation

`SPECIAL:` *formula* ; *number of variables*

- The formula has to be specified in one line and Fortran notation.
- The semicolon separates formula from the number of variables used in the formula.
- If the formula is supposed to be temperature-dependent, use "TEMP" where needed.
- Beside temperature, the formula may depend on educts and products of the reaction.
- The following mathematical operators and functions may be used:

  $+$, $-$, $*$, $/$, ^ or **, abs, exp, log10, log, sqrt, sin, cos, tan, asin, acos, atan, sinh, cosh, tanh.

- Note: the SPECIAL rate constant provides flexibility for newly developed rate constant types, but is less efficient to evaluate.

```
1: CLASS: GAS
2: RHO + RHOV + RHOL          =       RHO
3: SPECIAL: 2.2E-13*exp(620./TEMP)+1.9E-33*RHOV*abs(sqrt(RHO*RHO+RHOV*RHOV)); 3
```

S4.2 Gaseous and aqueous phase (CLASS: GAS or AQUA)

TYP: PHOTABC photolysis

$$\chi_z = C \cdot \chi$$
$$\chi_y = B \cdot \left(1 - \frac{1}{\cos(\chi_z)}\right)$$
$$\chi_x = \begin{cases} \exp(\chi_y), & \chi_y > -30,\ \chi_z < \pi/2 \\ 9.357 \cdot 10^{-14}, & \text{otherwise} \end{cases} \quad (1)$$
$$k = \begin{cases} \varepsilon_{\text{dust}} \cdot A \cdot \chi_x, & \chi < \pi/2 \\ 0, & \text{otherwise} \end{cases}$$

$A = j[\text{s}^{-1}], \quad B = b[-], \quad C = c[-]$

Parameter: A B C

Ref: Röth (1992)

TYP: PHOTAB photolysis

$$k = \begin{cases} \varepsilon_{\text{dust}} \cdot A \cdot \exp\left(-\frac{B}{\cos(\chi)}\right), & \chi < \pi/2 \\ 0, & \text{otherwise} \end{cases} \quad (2)$$

$A = a[\text{s}^{-1}], \quad B = b[-]$

Parameter: A B

TYP: PHOTMCM photolysis as used in MCM (M, N dimensionless parameter to describe shape of curve)

$$\chi_z = \exp\left(-\frac{N}{\cos(\chi)}\right)$$
$$\chi_y = \cos(\chi)^M$$
$$k = \begin{cases} \varepsilon_{\text{dust}} \cdot I \cdot \chi_y \cdot \chi_z, & \chi < \pi/2 \\ 0, & \text{otherwise} \end{cases} \quad (3)$$

$I = i[\text{s}^{-1}], \quad M = m[-], \quad N = n[-]$

Parameter: I M N

`TYP: PHOTO` KPP photolytic reaction

$$k = A \cdot \texttt{SUN}$$
$$A = a[\mathrm{s}^{-1}] \tag{4}$$

Parameter: A

Note: `SUN` is calculated according to the `Update_SUN()` procedure in KPP (github: KPP/util/UpdateSun.f).

`TYP: PHOTO2` KPP photolytic reaction

$$k = A \cdot \texttt{SUN}^2$$
$$A = a[\mathrm{s}^{-1}] \tag{5}$$

Parameter: A

Note: `SUN` is calculated according to the `Update_SUN()` procedure in KPP (github: KPP/util/UpdateSun.f).

`TYP: PHOTO3` KPP photolytic reaction

$$k = A \cdot \texttt{SUN}^3$$
$$A = a[\mathrm{s}^{-1}] \tag{6}$$

Parameter: A

Note: `SUN` is calculated according to the `Update_SUN()` procedure in KPP (github: KPP/util/UpdateSun.f).

`TYP: CONST` simple constant

$$k = A$$
$$A = a[\left(\mathrm{cm}^{-3}\right)^{\beta-1} s^{-1}], \tag{7}$$

Parameter: A

`TYP: TEMP` temperature dependent reaction (Arrhenius expression)

$$k = A \cdot T^N \cdot \exp\left(-\tfrac{B}{T}\right)$$
$$A = a[\left(\mathrm{cm}^{-3}\right)^{\beta-1} s^{-1}], \qquad N = n\ [-] \qquad B = E/R = -\triangle H/R\ [K] \tag{8}$$

Parameter: A N E/R

TYP: TEMP1 temperature dependent reaction (Arrhenius expression)

$$k = A \cdot \exp\left(-\frac{B}{T}\right)$$
$$A = a[\left(\mathrm{cm}^{-3}\right)^{\beta-1} s^{-1}], \qquad B = E/R = -\triangle H/R\ [K] \tag{9}$$

Parameter: A E/R

TYP: TEMP2 temperature dependent reaction (Arrhenius expression)

$$k = K_0 \cdot T^2 \cdot \exp\left(-\frac{E/R}{T}\right)$$
$$K_0 = a[\left(\mathrm{cm}^{-3}\right)^{\beta-1} s^{-1}], \qquad E/R = -\triangle H/R\ [K] \tag{10}$$

Parameter: K_0 E/R

TYP: TEMP3 temperature dependent reaction Arrhenius expression in the format

$$k = A \cdot \exp\left[B\left(\frac{1}{T} - \frac{1}{T_{\mathrm{ref}}}\right)\right]$$
$$A = a[\left(\mathrm{cm}^{-3}\right)^{\beta-1} s^{-1}], \qquad B = E/R = -\triangle H/R\ [K] \tag{11}$$

Parameter: A B

TYP: TEMP4 temperature dependent reaction (Arrhenius expression)

$$k = A \cdot T \cdot \exp\left(-\frac{B}{T}\right)$$
$$A = a[\left(\mathrm{cm}^{-3}\right)^{\beta-1} s^{-1}], \qquad B = E/R = -\triangle H/R\ [K] \tag{12}$$

Parameter: A B

TYP: S4H2O (cp. the special reactions in RADM/ RACM by Stockwell et al. (1997))

$$k = C_1 \cdot \exp\left(\frac{C_2}{T}\right) + C_3 \cdot [M] \cdot \exp\left(\frac{C_4}{T}\right) \tag{13}$$

Parameter: A C_1 C_2 C_3 C_4

TYP: T1H2O

$$k = A \cdot [H_2O] \cdot \exp\left(-\frac{B}{T}\right)$$
$$A = a[\left(\mathrm{cm}^{-3}\right)^{\beta-1} s^{-1}], \qquad B = -\triangle H/R\ [K] \tag{14}$$

Parameter: A B

Note: $[H_2O]$ is prescribed or taken from meteorology (see Sec. 3.5).

S4.3 Only gas phase (CLASS: GAS)

TYP: TROE temperature and pressure dependent reaction (Troe)

$$k_1 = [M] \cdot K_0 \cdot \left(\tfrac{T}{300}\right)^{-N}$$
$$k_2 = k_\infty \cdot \left(\tfrac{T}{300}\right)^{-M}$$
$$k = \frac{k_1}{(1+k_1/k_2)} \cdot 0.6^{\left\{1+[\log_{10}(k_1/k_2)]^2\right\}^{-1}} \quad (15)$$

Parameter: K_0 N k_∞ M

TYP: TROEF temperature and pressure dependent reaction (Troe)

$$k_1 = [M] \cdot K_0 \cdot \left(\tfrac{T}{300}\right)^{-N}$$
$$k_2 = k_\infty \cdot \left(\tfrac{T}{300}\right)^{-M}$$
$$k = \frac{k_1}{(1+k_1/k_2)} \cdot F^{\left\{1+[\log_{10}(k_1/k_2)]^2\right\}^{-1}} \quad (16)$$

Parameter: K_0 N k_∞ M F

TYP: TROEQ Troe - equilibrium

$$k_1 = [M] \cdot K_0 \cdot \left(\tfrac{T}{300}\right)^{-N}$$
$$k_2 = k_\infty \cdot \left(\tfrac{T}{300}\right)^{-M}$$
$$k_3 = \frac{k_1}{(1+k_1/k_2)} \cdot 0.6^{\left\{1+[\log_{10}(k_1/k_2)]^2\right\}^{-1}} \quad (17)$$
$$k = \frac{k_3}{K_1 \cdot \exp(B/T)}$$

Parameter: K_0 N k_∞ M K_1 B

TYP: TROEQF Troe - equilibrium

$$k_1 = [M] \cdot K_0 \cdot \left(\tfrac{T}{300}\right)^{-N}$$
$$k_2 = k_\infty \cdot \left(\tfrac{T}{300}\right)^{-M}$$
$$k_3 = \frac{k_1}{(1+k_1/k_2)} \cdot F^{\left\{1+[\log_{10}(k_1/k_2)]^2\right\}^{-1}} \quad (18)$$
$$k = \frac{k_3}{K_1 \cdot \exp(B/T)}$$

Parameter: K_0 N k_∞ M 1 B F

`TYP: TROEXP` temperature and pressure dependent reaction (Troe)

$$
\begin{aligned}
k_1 &= [M] \cdot K_0 \cdot \exp\left(-\tfrac{B_1}{T}\right) \\
k_2 &= k_\infty \cdot \exp\left(-\tfrac{B_2}{T}\right) \\
k &= \frac{k_1}{(1+k_1/k_2)} \cdot F^{\left\{1+[\log_{10}(k_1/k_2)]^2\right\}^{-1}}
\end{aligned}
\tag{19}
$$

Parameter: K_0 B_1 k_∞ B_2 F

`TYP: TROEMCM` MCM version of temperature and pressure dependent reaction (Troe)

$$
\begin{aligned}
k_1 &= [M] \cdot K_1 \cdot \left(\tfrac{T}{298}\right)^{K_2} \exp\left(\tfrac{K_3}{T}\right) \\
k_2 &= K_4 \cdot \left(\tfrac{T}{298}\right)^{K_5} \exp\left(\tfrac{K_6}{T}\right) \\
F_c &= K_7 \cdot \exp\left(\tfrac{K_8}{T}\right) + K_9 \cdot \exp\left(\tfrac{T}{K_{10}}\right) \\
F_t &= \log_{10}(k_1/k_2) \,/\, [0.75 - 1.27 \cdot \log_{10}(F_c)] \\
k &= \frac{k_1}{(1+k_1/k_2)} \cdot F_c^{1/\left(1+F_t^2\right)}
\end{aligned}
\tag{20}
$$

Parameter: K_1 K_2 K_3 K_4 K_5 K_6 K_7 K_8 K_9 K_{10}

`TYP: SPEC1` (cp. the special reactions in RADM/ RACM by Stockwell et al. (1997))

$$
k = C_1 \cdot (1 + [M] \cdot C_2) \tag{21}
$$

Parameter: C_1 C_2

`TYP: SPEC2` (cp. the special reactions in RADM/ RACM by Stockwell et al. (1997))

$$
k = [M] \cdot C_1 \cdot \left(\tfrac{T}{300}\right)^{C_2} \tag{22}
$$

Parameter: C_1 C_2

`TYP: SPEC3` (cp. the special reactions in RADM/ RACM by Stockwell et al. (1997))

$$
\begin{aligned}
k_1 &= K_1 \cdot \exp\left(\tfrac{K_2}{T}\right) \\
k_2 &= K_3 \cdot \exp\left(\tfrac{K_4}{T}\right) \\
k_3 &= [M] \cdot K_5 \cdot \exp\left(\tfrac{K_6}{T}\right) \\
k &= k_1 + \frac{k_3}{1+k_3/k_2}
\end{aligned}
\tag{23}
$$

Parameter: K_1 K_2 K_3 K_4 K_5 K_6

TYP: SPEC4

$$k = C_1 \cdot \exp\left(\frac{C_2}{T}\right) + [M] \cdot C_3 \cdot \exp\left(\frac{C_4}{T}\right) \quad (24)$$

Parameter: C_1 C_2 C_3 C_4

TYP: SPEC1MCM (modified SPEC1 for MCM)

$$k = K_1 \cdot \left(1 + \frac{[M]K_2}{K_3 \cdot 300/T}\right) \quad (25)$$

Parameter: K_1 K_2 K_3

TYP: SPEC2MCM (modified SPEC2 for MCM)

$$k = K_1 \cdot \left(\frac{T}{300}\right)^{K_2} \cdot \exp\left(\frac{K_3}{T}\right) \quad (26)$$

Parameter: K_1 K_2 K_3

TYP: SPEC3MCM (modified SPEC3 for MCM)

$$k = K_1 \cdot \left(1 + \frac{[M]}{K_2}\right) \quad (27)$$

Parameter: K_1 K_2

TYP: SPEC4MCM (modified SPEC4 for MCM)

$$k = K_1 \cdot \left(1 + K_2 \cdot \exp\left(\frac{K_3}{T}\right) \cdot [H_2O]\right) \cdot \exp\left(\frac{K_4}{T}\right) \quad (28)$$

Parameter: K_1 K_2 K_3 K_4

Note: $[H_2O]$ is prescribed or taken from meteorology (see Sec. 3.5).

TYP: SPEC5MCM

$$\begin{aligned} k_1 &= [M] \cdot K_1 \cdot 0.21 \cdot \exp\left(\frac{K_2}{T}\right) \\ k_2 &= [M] \cdot K_3 \cdot 0.21 \cdot \exp\left(\frac{K_4}{T}\right) \\ k &= k_1 \cdot (1 - k_2) \end{aligned} \quad (29)$$

Parameter: K_1 K_2 K_3 K_4

TYP: SPEC6MCM

$$\begin{aligned} k_1 &= K_1 \cdot \exp\left(\frac{K_2}{T}\right) \\ k_2 &= K_3 \cdot \exp\left(\frac{K_4}{T}\right) \\ k &= k_1 \cdot (1 - k_2) \end{aligned} \quad (30)$$

Parameter: K_1 K_2 K_3 K_4

TYP: SPEC7MCM

$$k_1 = K_1 \cdot \exp\left(\frac{K_2}{T}\right)$$
$$k_2 = K_3 \cdot \exp\left(\frac{K_4}{T}\right) \tag{31}$$
$$k = k_1 \cdot \left(K_5 - \frac{K_6}{1+k_2}\right)$$

Parameter: K_1 K_2 K_3 K_4 K_5 K_6

TYP: SPEC8MCM

$$k_1 = [M] \cdot K_1 \cdot 0.21 \cdot \exp\left(\frac{K_2}{T}\right)$$
$$k_2 = [M] \cdot K_3 \cdot 0.21 \cdot \exp\left(\frac{K_4}{T}\right) \tag{32}$$
$$k = \frac{k_1}{(1+k_2) \cdot T}$$

Parameter: K_1 K_2 K_3 K_4

TYP: SPEC9MCM

$$k_1 = K_1 \cdot [M] \cdot 0.21 \cdot \exp\left(\frac{K_2}{T}\right) / \left[1 + K_3 \cdot [M] \cdot 0.21 \cdot \exp\left(\frac{K_4}{T}\right)\right]$$
$$k_2 = K_5 \cdot [M] \cdot 0.21 \cdot \exp\left(\frac{K_6}{T}\right) / \left\{\left[1 + K_7 \cdot [M] \cdot 0.21 \cdot \exp\left(\frac{K_8}{T}\right)\right] \cdot T\right\} \tag{33}$$
$$k_3 = K_9 \cdot \exp\left(\frac{K_{10}}{T}\right)$$
$$k = \frac{k_1 \cdot k_3}{k_2 + k_3}$$

Parameter: K_1 K_2 K_3 K_4 K_5 K_6 K_7 K_8 K_9 K_{10}

TYP: HOM1

$$k = K_1 \cdot \exp\left(\frac{K_2}{T}\right) \cdot \exp\left(\frac{K_3}{T^3}\right) \tag{34}$$

Parameter: K_1 K_2 K_3

S4.4 Only aqua phase (CLASS: AQUA)

TYP: ASPEC1 H^+ times Arrhenius expression in the format

$$k = [H^+] \cdot A \cdot \exp\left[B \cdot \left(\frac{1}{T} - \frac{1}{T_{\text{ref}}}\right)\right] \cdot \left(1 + 13 \cdot [H^+]\right)^{-1}$$
$$A = a[\left(\text{cm}^{-3}\right)^{\beta-1} s^{-1}], \qquad B = -\triangle H/R \ [K] \tag{35}$$

Parameter: A B

Note: $[H^+]$ is the current concentration

TYP: ASPEC2 $(H^+)^B$ times Arrhenius expression in the format

$$k = [H^+]^B \cdot A \cdot \exp\left[C \cdot \left(\frac{1}{T} - \frac{1}{T_{\text{ref}}}\right)\right]$$
$$A = a[\left(\text{cm}^{-3}\right)^{\beta-1} s^{-1}], \qquad B = b[-], \qquad C = -\triangle H/R \ [K] \tag{36}$$

Parameter: A B C

Note: $[H^+]$ is the current concentration

TYP: ASPEC3 Arrhenius expression in the format

$$k = A \cdot \exp\left[-B \cdot \log_{10}\left([H^+]\right)\right] \tag{37}$$

Parameter: A B

Note: $[H^+]$ is the current concenctration

S4.5 Phase transfer (CLASS: HENRY)

According to the resistance model by Schwartz (1986). In the *.sys-file, only the Henry's Law constant is read, all other values (mass accommodation, gas phase diffusion coefficients, and molar mass being necessary for the mass transfer coefficient are defined in the *.dat-file).

Parameter: $K_0 = K_H[\text{M atm}^{-1}]$ $E/R = -\triangle H_{\text{sol}}/R$ [K]

S4.6 Dissociation (CLASS: DISS)

`TYP: DTEMP` (K_e temperature-dependent, K_- = constant)

$$K_e = K_+/K_- = A \cdot \exp\left[B \cdot \left(\tfrac{1}{T} - \tfrac{1}{T_{\text{ref}}}\right)\right]$$
$$K_- = C \tag{38}$$

Parameter: A B C

`TYP: DTEMP2` (K_e temperature dependent, K_- = temperature dependent)

$$K_e = K_+/K_- = A \cdot \exp\left[B \cdot \left(\tfrac{1}{T} - \tfrac{1}{T_{\text{ref}}}\right)\right]$$
$$K_- = C \cdot \exp\left[D \cdot \left(\tfrac{1}{T} - \tfrac{1}{T_{\text{ref}}}\right)\right] \tag{39}$$

Parameter: A B C D

`TYP: DTEMP3` (K_e temperature-dependent, K_- = constant)

$$K_e = K_+/K_- = A \cdot \exp\left\{B \cdot (T_{\text{ref}}/T - 1) + C \cdot [1 - T_{\text{ref}}/T + \log_{10}(T_{\text{ref}}/T)]\right\}$$
$$K_- = D \tag{40}$$

Parameter: A B C D

`TYP: DTEMP4` (K_e temperature-dependent, K_- = constant)

$$K_e = K_+/K_- = A \cdot \exp\left\{B \cdot (T/T_{\text{ref}} - 1) + C \cdot [1 - T/T_{\text{ref}} + \log_{10}(T/T_{\text{ref}})]\right\}$$
$$K_- = 1.0 \tag{41}$$

Parameter: A B C

TYP: DTEMP5 (K_e temperature-dependent, K_- = constant)

$$K_e = K_+/K_- = A \cdot (T/T_{\text{ref}})^B \cdot \exp\left[C \cdot \left(\frac{1}{T} - \frac{1}{T_{\text{ref}}}\right)\right] \tag{42}$$

$K_- = 1.0$

Parameter: A B C

TYP: DCONST (K_e and K_- = constant)

$$K_e = A$$
$$K_- = B \tag{43}$$

Parameter: A B

S5 Description of a chemical mechanism for combustion chemistry (*.sys-file)

The syntax format for combustion mechanisms is the widely used ChemKin format and can be viewed in the ChemKin documentation (Kee et al., 1996, p.40-63). The information about thermodynamic properties are listed in Kee et al. (1996) and Kee et al. (1990).

In Cminor's ChemKin syntax, reversible reactions must be indicated by "<=>" between reactants and products, not by "=". Currently, Cminor is capable of simulating normal Arrhenius reactions (no auxiliary information), Arrhenius reactions with explicitly given reverse reaction coefficients (indicated by "REV"), third body collision reactants (only "+M", no specific species), and enhanced third body efficiencies and pressure-dependent reactions in Lindemann's or Troe's kinetic law forms, all in the usual ChemKin syntax.

S6 Initial data input (*.ini-file)

The *.ini-file contains all initial data including emissions.

S6.1 Head of the *.ini-file:

- All characters following a hash symbol "#" or the keyword "COMMENT" will be ignored.
- At the beginning of the document, the user has to specify the units for the initial data using the keywords UNIT GAS and UNIT AQUA. The default value is 0 for both phases. This is currently a dummy feature, as only the default values are available.

```
1: # ═══════════════════════════════════════════════════════════════
2: # ═══════════════════  CHEMISTRY  ═══════════════════
3: # ═══════════  Output - Chemical Reaction Data  ═══════════
4: # ═══════════════════════════════════════════════════════════════
5:
6: #  Created:              Wed June 14 12:00:00 2006 (AT)
7: #  Chemical Mechanism:  CAPRAM 3.0 (URBAN CASE)
8:
9: # ═══════════════════  Unit options  ═══════════════════
10:
11: UNIT  GAS   0   # Gas phase units ( 0 = molec/cm3 )
12: UNIT  AQUA  0   # Aqueous phase units ( 0 = mol/l)
```

S6.2 Initial and emission values of gaseous species

1. After the header lines, the next line specifies the gaseous input: BEGIN_GAS. END_GAS specifies the end of the gaseous input.

2. Initial values will be set between the keywords: BEGIN_INITIAL and END_INITIAL.

3. – **Atmospheric mechanisms:** Currently, Cminor only accepts the unit [molec cm^{-3}] for gas phase initial values, and [mol l^{-1}] for aqueous phase initial values.

   – **Combustion mechanisms:** For combustion mechanisms, Cminor currently only accepts the standard unit [cal mol^{-1}].

4. The syntax rule is: [*name_of_the_species*] blank-space [*value_in_exponential_notation*]

5. Emission rates will be set between the keywords: BEGIN_EMISS and END_EMISS.

6. Deposition rate constants will be set between the keywords: BEGIN_DEPOS and END_DEPOS.

```
1: BEGIN_GAS
2:     BEGIN_INITIAL       # Initial Concentrations [molec/cm^3]
3:       NO2               1.150e+11
4:       O3                2.290e+12
5:       HNO3              2.550e+10
6:       H2O2              2.550e+10
7:       [N2]              1.960e+19
8:     END_INITIAL
```

```
 9:
10:     BEGIN_EMISS          #  Emissions [molec/cm^3/sec]
11:       CH2OHCH2OH      2.01e+5
12:       HCHO            2.58e+5
13:       NH3             3.03e+6
14:       NO              1.01e+7
15:       SO2             3.27e+6
16:     END_EMISS
17:
18:     BEGIN_DEPOS          #  Deposition Rates [1/sec]
19:       HCL             1.0e-5
20:       HNO3            2.0e-5
21:       N2O5            2.0e-5
22:       NH3             1.0e-5
23:       NO2             4.0e-6
24:       O3              4.0e-6
25:     END_DEPOS
26: END_GAS
```

S6.3 Initial values of aqueous species (atmospheric mechanisms only)

1. Initial values of aqueous species are calculated using the liquid water content at time t_0.

2. The input begins with BEGIN_AQUA and ends with END_AQUA.

3. The user has to specify the molar mass, charge, soluble index, and the mass fraction of the corresponding species of each mode of the aerosol size distribution. The soluble index is not used currently and therefore can be any number. The values have to be set between the keywords: BEGIN_AFRAC+[*number_of_mode*] and END_AFRAC+[*number_of_mode*], demonstrated in the example below.

4. The declaration of the dry particle size distribution can be set between the keywords: BEGIN_SPEK and END_SPEK. It is assumed to be log-normal, specified by giving the mean radius, the total number, the density of aerosol, and geometric standard deviation. Please note the specific assumed probability density function given below. For a mono-disperse distribution, choose nDropletClasses=1, see Sec. S9. Each line in the SPEK section defines one mode, corresponding to one AFRAC section. If two AFRAC sections are equal, i.e., describe equal aerosols, a multi-modal distribution will be created, also explained in more detail below.

5. The first line after BEGIN_SPEK defines the number of modes to use. This is restrictive, i.e., this number is the number of AFRAC sections that are read, even if more AFRAC sections are present in the file. The

following lines contain the dry radius of a particle in [m], the number of particles and the densitiy in [g/m3] of the $i-$th AFRAC section. The numbers are separated by blankspace.

```
BEGIN_AQUA
    BEGIN_AFRAC1
        #-------------------------------------------------------------------------------
        #   Name        Mass Fraction
        #               [g/g]
        #-------------------------------------------------------------------------------
            NH4p        0.1567332754127
            HSO4m       0.8432667245873
    END_AFRAC1

    BEGIN_AFRAC2
            NH4p        0.1567332754127
            HSO4m       0.8432667245873
    END_AFRAC2

    BEGIN_AFRAC3
            NAp         0.3933747412
            CLm         0.6066252588
    END_AFRAC3

    BEGIN_SPEK
        3           # nModes  ...  number of input modes
        #-------------------------------------------------------------------------------
        #   Radius [m]      Number [#/m3]      Density [g/m3]      geometric
        #                                                          standard deviation
        #-------------------------------------------------------------------------------
            0.0039E-6       133.0E6            1800.0E3            1.929
            0.133E-6        66.6E6             1800.0E3            1.2337
            0.29E-6         3.06E6             2160.0E3            1.486
    END_SPEK
END_AQUA
```

The calculation of the actual concentration c_i^S, i.e., the initial concentration of species S in droplet class i, in [molec/cm^3] is done assuming log-normally distributed aerosols. The following probability density $\phi(r)$ and resulting cumulative distribution function $\Phi(r)$ are used, with μ being the mean radius, σ the standard deviation, and n the

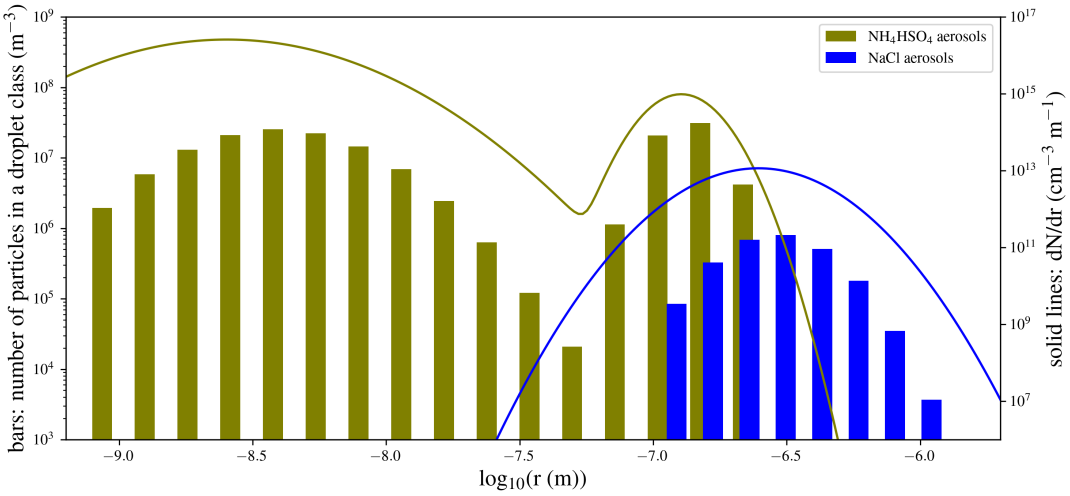

Figure S2. Generated droplet classes for the modes of the exemplary AQUA section above with nDropletClasses=24. Bars are centered around the radius of the aerosol class and the height is the number of particles in the respective class. Lines indicate the actual probability density function.

number of total aerosols, all of which are given in the SPEK section:

$$\phi(r) = \frac{n}{r \log\sigma \sqrt{2\pi}} \exp\left[-\frac{(\log r - \log\mu)^2}{2\log\sigma^2}\right],$$

$$\Phi(r) = \frac{1}{2}\left[1 + \mathrm{erf}\left(\frac{\log r - \log\mu}{\sqrt{2}\log\sigma}\right)\right].$$

The aerosol mass is determined by discretizing the aerosol distribution using logarithmically equidistant bins, one for each droplet class. To constrain the aerosol masses to a reasonable range, the aerosol distribution of each mode is bounded by determining the 0.5%-radius-quantile and the 99.5%-mass-quantile. If all modes contain the same aerosol type, this procedure is applied for the sum of these modes, using the respective smallest and largest quantiles of the individual modes. If the modes differ in aerosol type, the procedure is applied to each mode individually. The number of droplet classes are distributed equally among the modes. The remaining number of MOD(nDropletClasses, nModes) classes are assigned to the aerosol types one after another.

The number of particles in a specific class is determined by $\Phi(r_+) - \Phi(r_-)$, where r_+ and r_- are the lower and upper boundaries of the class in the density function. Each particle will be assumed to have the radius $\exp\left[\frac{1}{2}(\log r_- + \log r_+)\right]$. The example setup above (BEGIN_AQUA ... END_AQUA) with nDropletClasses=24 yields droplet classes as visualized in Fig. S2.

Finally, for a radius r_i of a specific droplet class i, the initial values, i.e., initial concentrations of a species S, are calculated out of the aerosol composition as

$$c_i^S = N_i \cdot F_S \cdot \frac{\frac{4}{3}\pi r^3 \cdot \rho}{m_{mol}} \cdot N_A.$$

Here, m_{mol} is the molar mass, $N_A = 6.02214 \cdot 10^{23}[\text{mol}^{-1}]$ is Avogadros-Constant, N_i the number of particles in droplet class i, F_S the mass fraction of species S in the aerosol, r_i the radius of the aerosol class and ρ the given density of the aerosol.

S6.4 Select diagnose species

Next to the initial data, a list of diagnose species has to be specified in the *.ini-file. Diagnose species denotes the species of which the concentrations are saved in the NetCDF file. The notation follows the same convention as above:

1. The input begins with BEGIN_DIAG and ends with END_DIAG.
2. The user has to specify one species per line.
3. The output routine generates a new NetCDF file for every run. Older files with the same file name will be overwritten. The user has to set the name of the NetCDF output file in the *.run-file, see Sec. S9.

Note, that *.ini-files containing two diagnose sections or repetitions of the same species in one diagnose section will lead to unwanted behaviour.

```
1: BEGIN_DIAG
2:          SO2
3:          SO4mm
4:          Hp
5:          OHm
6:          CC(C(=O)O)=C
7:          aCC(C(=O)O)=C
8: END_DIAG
```

S7 Additional information for atmospheric chemistry (*.dat-file)

The *.dat-file contains information necessary for the computation of the phase transfer.

Information about gas phase species:

1. The input begins with BEGIN_DATAGAS and ends with END_DATAGAS.
2. The user has to specify the mole mass m_{mol}, the mass accommodation coefficient α, and the specific gas phase diffusion coefficient D_g.

```
1: BEGIN_DATAGAS
2:          #-------------------------------------------------------
3:          #  Name          MolMass   Alpha          Dg
4:          #                [g/mol]
5:          #-------------------------------------------------------
6:          #
7:          SO2              64.0      0.11          0.0000128
8:          [O2]             32.0      0.1           0.0000112
9:          [N2]             28.0      0.0           0.0
10:         [H2O]            18.0      0.0           0.0
11: END_DATAGAS
```

Information about aqua phase species:

1. The input begins with BEGIN_DATAQUA and ends with END_DATAQUA.

2. The user has to specify the mole mass m_{mol} and charge.

```
1: BEGIN_DATAQUA
2:          #-------------------------------------------------------------------------
3:          # Name          MolMass      Charge
4:          #               [g/mol]
5:          #
6:          aSO2            64.00         0.00
7:          HSO3m           81.00        -1.00
8:          SO3mm           80.00        -2.00
9:          OHm             17.00        -1.00
10:         Hp              1.00          1.00
11:         [aH2O]          18.00         0.00
12: END_DATAQUA
```

Information about gaseous peroxy radical species:

1. The input begins with BEGIN_DATARO2 and ends with END_DATARO2.

2. The user has to specify the number of listed RO2 species and the names of the species that are to be considered RO2 line by line, see the following example.

3. For any reaction with FACTOR: $RO2 in the fourth line, the rate constant will always be multiplied with the sum of all current concentrations of the listed species.

```
BEGIN_DATARO2
            nRO2:  9
    BRC(BR)(BR)O[O]
    BRC(BR)O[O]
    BRCC(=O)O[O]
    BRCC(BR)O[O]
    BRCCO[O]
    BRCO[O]
    C(CC(C=O)C(C)(CO[N+](=O)[O-])O[O])O
    C(CC(C=O)[C](C)O[O])O
    C(CC(CC=O)C(C)(CO[N+](=O)[O-])O[O])=O
END_DATARO2
```

Information about aqueous peroxy radical species:

1. The input begins with BEGIN_DATARO2aq and ends with END_DATARO2aq.
2. The user has to specify the number of listed RO2aq species and the names of the species that are to be considered RO2aq line by line, see the following example.
3. For any reaction with FACTOR: $RO2aq in the fourth line, the rate constant will always be multiplied with the sum of all current concentrations of the listed species.

```
BEGIN_DATARO2aq
        nRO2aq:  8
   aBRCC(=O)O[O]
   aBRCC(O)(O)O[O]
   aC1C(=O)N(CO[O])C(=O)C1
   aC1C(=O)N(CO[O])CC1
   aC1C(=O)NC(O[O])C1
   aC1C(O[O])N(C)C(=O)C1
   aC=C(C(=O)O)C(=O)O[O]
   aC=C(C(=O)O)C(O)O[O]
END_DATARO2aq
```

S8 Additional information about the thermodynamic properties for combustion chemistry (*.dat-file)

The syntax rules for the thermodynamic data base using the ChemKin format is given in Kee et al. (1996, p.43-45).

S9 Specification of simulation parameters (*.run-file)

To specify the model run parameters, copy one of the example *.run-files and change the necessary information. The *.run-file is organised via Fortran namelists. All values have default values, so no value has to be listed and specified.

S9.1 Namelist SCENARIO

The first block contains some general information about the run.
Variables of Namelist SCENARIO are

- `Label` - (character(80), default: file name of the sys file) the name of the simulation run,
- `WaitBar` - (logical, default: `.TRUE.`) to plot a progress bar onto the screen if true,
- `combustion` - (logical, default: `.FALSE.`) to simulate a combustion mechanism using ChemKin syntax, and
- `Simulation` - (logical, default: `.TRUE.`) a trigger to do the numerical simulation of a mechanism.

Example:

```
1: &SCENARIO
2:          Label       = 'MCM32+CAPRAM40'
3:          WaitBar  = .T.
4:          Combustion   = .F.
5:          Simulation = .T.
6: /END
```

S9.2 Namelist FILES

The second block contains all the essential information about the paths of the mechanism, initial data, and other import. The file names do not have default values and the existence of the files is checked internally.
Variables of Namelist FILES are

- `SysFile` - (character(80)) to specify the path to the chemical mechanism,
- `DataFile` - (character(80)) to specify the path to
  - further information necessary for phase transfer calculations (troposphere), and
  - thermodynamic database for computing the polynomial fits to thermodynamic data (combustion),
- `MWFile` - (character(80)) to specify the molecular weights of species (combustion ONLY), and
- `InitFile` - (character(80)) to specify the initial values for species concentrations and emission values.

Example:

```
1: &FILES
2:          SysFile  = 'CHEM/MCM32_CAPRAM40_full.sys'
3:          DataFile = 'DAT/MCM32_CAPRAM40_full.dat'
4:          InitFile = 'INI/Urban2.ini'
5: /END
```

S9.3 Namelist TIMES

The third block contains the simulation time in seconds. `tBegin = 0.0d0` means that the simulation starts at midnight (crucial for mechanisms with photolytic reactions).
Variables of Namelist TIMES are

- `tBegin` - (real, default = 0.0) the point of time where the simulation starts, and
- `tEnd` - (real, default = 0.0) the point of time where the simulation ends.

Example:

```
1: &TIMES
2:          tBegin = 0.0d0
3:          tEnd   = 172800.0d0
4: /END
```

S9.4 Namelist METEO

The fourth part of the .run file consists of meteorological information.
Variables of Namelist METEO are

- `pHSet` - (logical, default = `.TRUE.`) if true, the H^+ concentration at the beginning is calculated in equilibrium with the specified initial values; if false, the pH is set to 7 at the beginning,
- `LWCLevelmin` - (real, default = $2.0e-8$) in [l m^{-3}] to specify the minimal liquid water content in the simulation (only relevant if the adiabatic parcel option is off),
- `LWCLevelmax` - (real, default = $3.0e-4$) in [l m^{-3}] to specify the maximal liquid water content in the simulation (if `LWCLevelmax` < `LWCLevelmin` → cloud and non-cloud phases are swapped, see pseudo LWC function) (only relevant if the adiabatic parcel option is off),
- `dust` - (real, $\in [0,1]$, default = 1.0) a parameter for reducing the intensity of solar radiation, which is usually 1 (it acts as a factor multiplied onto PHOTAB, PHOTABC and PHOTMCM rate constants),

- `idate` - (integer, default = 011027) format YYMMDD to specify the simulation date (used for solar radiation calculation),
- `rlat` - (real, default = 50.65) the latitude (used for solar radiation calculation),
- `rlon` - (real, default = 10.77) the longitude (used for solar radiation calculation),
- `Temperature0` - (real, default = 280) the initial temperature in [K],
- `Pressure0` - (real, default = 85000) the initial pressure of the system in [Pa],
- `activation_radius` - (real, default = $1.0e-12$) a minimal dry radius under which particles are not activated and aqueous reactions are suppressed (only considered if `adiabatic_parcel` is false),
- `nDropletClasses` - (integer, default = 1) the number of droplet classes to create, see Subsec. S6.3, if no aqueous species are present this does not affect the simulation,
- `adiabatic_parcel` - (logical, default = `.FALSE.`) the switch to activate additional solving of adiabatic parcel equations and droplet condensation equation,
- `RH0` - (real, default = 0.6) the relative humidity at beginning of simulation,
- `updraft_velocity` - (real, default = 1.0) the constant upward velocity of the parcel in [m s^{-1}].
- `alpha_H2O` - (real, default = 0.0415) accommodation coefficient of water, and
- `beta_H2O` - (real, default = 1.0) relaxation radius coefficient of water

Example:

```
&METEO
        pHSet          = .T.
        LWCLevelmax = 2.0d-4
        LWCLevelmin = 3.0d-8
        Dust           = 0.5d0
        idate          = 010621
        rlat           = 45.0d0
        rlon           = 0.0d0
        Temperature0= 280.0d0
        nDropletClasses = 5
/END
```

S9.5 Namelist NUMERICS

The fifth part contains information for the numerical solver.
Variables of Namelist NUMERICS are

- `RtolROW` - (real, default = $1.0e-5$) the relative tolerance for integration scheme,
- `AtolGas` - (real, default = $1.0e-7$) the absolute tolerance for all gaseous species,
- `AtolAqua` - (real, default = $1.0e-7$) the absolute tolerance for all aqueous species (ONLY tropospheric systems),
- `AtolTemp` - (real, default = $1.0e-7$) the absolute tolerance for temperature (ONLY combustion systems),
- `AtolWaterMass` - (real, default = $1.0e-15$) the absolute tolerance for q_l (if adiabatic parcel option is on),
- `Atolq` - (real, default = $1.0e-5$) the absolute tolerance for q_v (if the adiabatic parcel option is on),
- `Atolz` - (real, default = $1.0e-1$) the absolute tolerance for height of parcel (if the adiabatic parcel option is on),
- `AtolRho` - (real, default = $1.0e-3$) the absolute tolerance for density of parcel (if the adiabatic parcel option is on),
- `Error_Est` - (integer, default = 2) the choice of error estimator, 2 = euclidean like norm, else: maximum norm,
- `minStp` - (real, default = $1.0e-20$) the minimum step size in seconds for integration scheme,
- `maxStp` - (real, default = 250.0) the maximum step size in seconds for integration scheme (set to ≤ 0 to allow arbitrarily large time steps),
- `ODEsolver` - (character(80), default = `'Rodas3'`) the path to solver coefficients for Rosenbrock methods (stored in the METHODS/ folder and will be included at compile time), and
- `Ordering` - (logical, default = `.TRUE.`) if true, Markowitz ordering strategy to preserve sparsity is used; if false, no reordering is done during LU factorization.

Example:

```
1: &NUMERICS
2:          RtolROW   = 1.0d-3
3:          AtolGas   = 1.0d-7
4:          AtolAqua  = 1.0d-7
5:          Error_Est = 2
```

```
6:          minStp   = 1.0d-30
7:          maxStp   = 600.d0
8:          ODEsolver = 'METHODS/TSRosWRodas3.ros'
9: /END
```

S9.6 Namelist OUTPUT

The last part contains parameter for the output procedures.
Variables of Namelist OUTPUT are

- `NetCdfPrint` - (logical, default = `.TRUE.`) a trigger to write data to NetCDF file every s seconds; if `StpNetCDF` $< 0 \rightarrow$ write every time step,
- `NetCdfFile` - (character(80), default=`''`) the path to NetCDF output file, note that older files with same name will be overwritten, if no file name is given, no NetCDF output will be generated,
- `StpNetCDF` - (real, default = -1.0) writing data to NetCDF file every s seconds, if `StpNetCDF` $< 0 \rightarrow$ write every time step,
- `FluxDataPrint` - (logical, default=`.FALSE.`) trigger to write all fluxes at to a binary file (every `StpFlux` seconds) (only for analysis purposes),
- `StpFlux` - (real, default=-1.0) time step for writing reaction rates data to binary file, if `StpFlux` $< 0 \rightarrow$ write every time step,
- `ConcDataPrint` - (logical, default=`.FALSE.`) trigger to write all concentrations to a binary file (every `StpConc` seconds) (only for analysis purposes),
- `StpConc` - (real, default=-1.0) time step for writing (all) concentrations data to binary file, if `StpConc` $< 0 \rightarrow$ write every time step (only for analysis purposes),
- `MatrixPrint` - (logical, default=`.FALSE.`) to print all matrices for the simulation run to formatted file (for investigation of sparsity patterns, plotting can be done via `PYTHONSCRIPTS/plot_sparsity_pattern.py`), and
- `DropletClassPrint` - (logical, default=`.FALSE.`) to output some information about the droplet classes at initialization, like dry and wet radius.

Example:

```
1: &OUTPUT
2:          NetCdfFile   = 'NetCDF/MCM32+C40_full.nc'
3:          StpNetCDF    = 600.0d0
```

```
4:          StpFlux      = -1.0d0
5:          MatrixPrint  = .F.
6: /END
```

S10 Exemplary simulation setups for combustion systems and atmospheric (multi-phase) chemistry mechanisms

mechanism	Chapman	RACM+CAPRAMv2.4	MCMv3.2+CAPRAMv4.0α
class	pure gas phase	multiphase (gas, aqua)	multiphase (gas, aqua)
scenario	no emissions	urban	urban
droplets	no cloud	mono-disperse	mono-disperse
adiabatic parcel	off	off	off
T_0	280 [K]	280 [K]	280 [K]
p_0	1 [atm]	1 [atm]	1 [atm]
n_s	7	250	10,196
n_r	10	787	23,098
tolerances	$\text{tol}_r = 10^{-4}$ $\text{tol}_{a,\text{gas}} = 10^{-5}$	$\text{tol}_r = 10^{-4}$ $\text{tol}_{a,\text{gas}} = 10^{-5}$ $\text{tol}_{a,\text{aqua}} = 10^{-5}$	$\text{tol}_r = 10^{-4}$ $\text{tol}_{a,\text{gas}} = 10^{-5}$ $\text{tol}_{a,\text{aqua}} = 10^{-5}$
reference	Sandu and Sander (2006)	Stockwell et al. (1997)	Jenkin et al. (2012)

mechanism	ERC n-heptane	LLNL n-heptane	LLNL methyl-decanonate
class	gas phase combustion	gas phase combustion	gas phase combustion
environment	constant volume	constant volume	constant volume
T_0	750 [K]	750 [K]	750 [K]
p_0	2 [Bar]	2 [Bar]	2 [Bar]
n_s	29	160	2,878
n_r	104	1,540	16,831
tolerances	$\text{tol}_r = 10^{-4}$ $\text{tol}_{a,\text{gas}} = 10^{-15}$ $\text{tol}_{a,\text{temp}} = 10^{-1}$	$\text{tol}_r = 10^{-4}$ $\text{tol}_{a,\text{gas}} = 10^{-15}$ $\text{tol}_{a,\text{temp}} = 10^{-1}$	$\text{tol}_r = 10^{-4}$ $\text{tol}_{a,\text{gas}} = 10^{-15}$ $\text{tol}_{a,\text{temp}} = 10^{-1}$
initial mixture	$[C_7H_{16}] = 15.3846$ % $[O_2] = 17.7689$ % $[N_2] = 66.8465$ %	$[C_7H_{16}] = 15.3846$ % $[O_2] = 17.7689$ % $[N_2] = 66.8465$ %	$[C_{11}H_{22}O_2] = 8.43882$ % $[O_2] = 19.2275$ % $[N_2] = 72.3337$ %
reference	Patel et al. (2004)	Seiser et al. (2000)	Herbinet et al. (2008)

Table S2. Initial conditions and scenarios of six different chemical mechanisms.

S11 Table of variables and parameters with description, values, and references

Parameter	Description	Value	Unit
a	surface tension parameter in Kelvin term of Köhler curve	$2\sigma/(\rho_w R_v T)$	m
a_{ij}	Rosenbrock method parameter	see Sec. 4.1	-
b_i and b'_i	fixed time points to change piecewise linear LWC behaviour	-	hours
$\mathbf{c}$ (c_j)	vector of all concentrations (concentration of species j)	-	[species unit], e.g., molec cm^{-3} or mol m^{-3}
c_j^{emis}	sum of constant sources and sinks of species j	-	[species unit] s^{-1}
c_{pa}	heat capacity of air at constant pressure	1004.0	J kg^{-1} K^{-1}
$\bar{c}_v(\mathbf{c},T)$	mass-averaged specific heat	$\sum_{j=1}^{n_S}(c_j \partial U_j(T)/\partial T)$	J kg^{-1} K^{-1}
$\mathbf{C}_v$ $(C_{v,j})$	constant volume specific heat (of species j)	$\partial U_j(T)/\partial T$, $j = 1,\ldots,n_S$	J mol^{-1} K^{-1}
c_{va}	heat capacity of air at constant volume	717.0	J kg^{-1} K^{-1}
D_i	water mass uptake coefficient of droplet class i	$\frac{4\pi r_i^2}{F_k+F_d}$	kg s^{-1}
D_c	diagonal matrix of concentrations	diag($\mathbf{c}$)	[species unit]
D_r	diagonal matrix of rates	diag($\mathbf{r}$)	[species unit] s^{-1}
D_v	molecular diffusivity of water vapor in air	Hall and Pruppacher (1976, (13))	m^2 s^{-1}
d_{ij}	Rosenbrock method parameter	see Sec. 4.1	-
$e_s(T)$	saturation vapor pressure over water	Flatau et al. (1992, Tab. 4)	Pa
F_d	coefficient for diffusion of vapor during condensation	see Sec. 3.3	m s kg^{-1}
F_k	coefficient for latent heat release during condensation	see Sec. 3.3	m s kg^{-1}
g	gravitational constant	9.81	m s^{-2}
h	numerical step size	-	-
J	(approximate) Jacobian matrix of the system	variable, see text	-

K	Kelvin term of Köhler curve	$\exp(a/r)$	-
$\mathbf{K}$ ($\mathbf{K}_i$)	vector of rate constant derivatives divided by rate (or its entries)	$\partial k_i/\partial T \cdot k_i^{-1}$, $i = 1, \ldots, n_R$	K^{-1}
$k^{(i)}$	in Sec. 3: rate constant of reaction i	-	[species unit]$^{\mathrm{n_{educts}}-1}$ s^{-1}
	in Sec. 4: value of stage i solution of numerical procedure		units of prognostic variables
k_T	thermal conductivity of air	Yau and Rogers (1996, Tab. 7.1)	J m^{-1} s^{-1} K^{-1}
L_v	latent heat of condensation	$2.5104\mathrm{e}+6$	J kg^{-1}
LWC(t)	liquid water content	-	l m^{-3}
M_w	molar weight of water	0.018015	kg mol^{-1}
m_j	Rosenbrock method parameter	see Sec. 4.1	-
n_D	number of droplet classes, i.e. super droplets	-	-
n_i	number of (identical) droplets in droplet class i	-	kg^{-1}
n_R	number of reactions in a mechanism	-	-
n_S	number of species in a mechanism	-	-
n_s	moles of solute in a droplet	-	mol
n_w	moles of water in a droplet	-	mol
p	pressure	-	Pa
q_v	water vapor mixing ratio	-	kg kg^{-1}
q_l ($q_{l,i}$)	liquid water mixing ratio (of droplet class i)	-	kg kg^{-1}
$q_S(T)$	saturation mixing ratio	$\frac{R_a e_S(T)}{R_v(p - e_S(T))}$	kg kg^{-1}
R	Raoult term of Köhler curve	$n_w/(n_s + n_w)$	-
R_a	gas constant of dry air	287.1	J kg^{-1} K^{-1}
R_v	gas constant of water	461.5	J kg^{-1} K^{-1}
r (r_i)	in Sec. 3.3. & Sec. 4.8: radius of droplet (of droplet class i)	-	m
	elsewhere: reaction rate vector (reaction rate of reaction i)		[species unit] s^{-1}
r_α and r_β	relaxation radii	see Sec. 3.3	m

S	relative humidity	$q/q_S(T)$	-
S_{eq} ($S_{\text{eq},i}$)	equilibrium relative humidity over solution droplet (of droplet class i)	see Sec. 3.3	-
S_j	j-th species	e.g., O_3	-
T	temperature	-	K
T_{ref}	reference temperature	298.15	K
t_n	time point number n	-	s
$U(T)$ ($U_j(T)$)	polynomial fit of molar internal energy (of species j)	-	J mol^{-1}
$\mathbf{u}^{(i)}$	modified i-th stage vector of Rosenbrock method	see Sec. 4.1	[species unit]
$\tilde{u}^{(i)}$ ($\tilde{\mathbf{u}}^{(i)}$)	modified last entry of i-th stage vector of Rosenbrock method corresponding to other variables than species	see Sec. 4.4 and Sec. 4.8	see Sec. 4.4 and Sec. 4.8
$\mathbf{y}_n$	numerical solution of ODE system at time point number n	-	-
α_{H2O}	accommodation coefficient of water	0.0415	-
α_i	Rosenbrock method parameter	see Sec. 4.1	-
β_{H2O}	relaxation radius coefficient of water	1.0	-
γ	Rosenbrock method parameter	see Sec. 4.1	-
θ	possible dependencies of rate constants	-	-
ν^e (ν^e_{ij})	stoichiometric coefficient matrix of educts (coefficient of species j as an educt in reaction i)	-	-
ν^p (ν^p_{ij})	stoichiometric coefficient matrix of products (coefficient of species j as a product in a reaction i)	-	-
ν (ν_{ij})	matrix of differences of stoichiometric coefficients (difference of coefficients of species j in reaction i)	$\nu^p_{ij} - \nu^e_{ij}$	-
ρ	average reactor density (of combustion systems)	sum of mole concentrations times molar weights	kg m^{-3}
ρ_a	mass density of air	$\frac{p}{R_a T}$	kg m^{-3}
ρ_w	mass density of liquid water	1000.0	kg m^{-3}

σ	surface tension between liquid water and air	Vargaftik et al. (1983, Tab. 1)	J m^{-2}

Table S3: Description of variables used in the article.

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