Cloud-J v7.3e user guide for the EMEP model

Willem van Caspel

Last updated December 2023

Contents

1 Introduction

Cloud-J v7.3e calculates photolysis rates (J-values) at model run-time, taking into account the instantaneous modeled concentrations of O_3 and radiatively active aerosol (dust, sea salt, biomass burning). The implementation and evaluation of Cloud-J in the EMEP model is described in detail in van Caspel *et al.* (2023) (`https://doi.org/10.5194/gmd-16-7433-2023`). In the EMEP model, `CloudJ_mod.f90` provides the interface between the model and the Cloud-J modules contained in `ModsCloudJ_mod.f90`.

At its core, Cloud-J solves the radiative transfer equations each time that the code is called, evaluating the equations at eight Gaussian quadrature points. The solution is propagated to the default 18 Cloud-J wavelength bins. To turn the radiative transfer solution into photolysis rates, the Cloud-J module `ModsCloudJ_mod.f90` reads in cross-section data from the `FJX_spec.dat` input file. The cross-section data in this file represents the product of the quantum-yield and cross-section data. To construct the data for the 18 wavelength bins, high-resolution (1 nm) quantum yield and cross-section data are averaged across the solar irradiance spectrum using the Cloud-J supplied pratmo-code. The usage of the pratmo-code is described below, so that in the future it will hopefully be straightforward to add new photolysis reactions to the EMEP model. The relevant code and Python scripts are available from `https://github.com/metno/emep-mscw/tree/ecosx/box/CloudJ_doc` (for internal MET users only) and from the GenChem repository `https://github.com/metno/genchem/tree/master/box/input/cloudj_v7.3e/CloudJ_doc`, both of which also contain the standalone Cloud-J code. A good starting point for 1 nm resolution quantum-yield and cross-section data is provided by the Master Chemical Mechanism (MCM) webpage `https://mcm.york.ac.uk/MCM/rates/photolysis`.

2 Creating cross-section input data

The cross-section data read in by the Cloud-J code represents the molecular cross-section data multiplied with the quantum yield of the particular reaction. The product of these two parameters thereby yields the number of successful photolysis reactions, given some actinic flux density.

3 Assigning Cloud-J rates to EMEP reactions

In the Cloud-J module, the short and long-name of each of the photolysis reactions present in the `FJX_spec.dat` file are stored in the `TITLEJL(X_)` variable. Here `X_` represents the maximum number of cross-section data sets to be read in from the input file, set to 201 by default. Each cross-section dataset in `FJX_spec.dat` begins with a 6-character short-name, e.g. for `O3(1D)`, as

```
O3(1D)|
258a 4.897E-01 5.074E-01 5.270E-01 5.650E-01 6.556E-01 7.358E-01 O3(1D)
258b 7.768E-01 8.168E-01 9.000E-01 9.000E-01 8.985E-01 9.000E-01 O3(1D)
258c 8.951E-01 5.024E-01 1.438E-01 8.392E-02 0.000E+00 0.000E+00 O3(1D)
```

The first number on each of the rows ending with a, b, and c represents the temperature or pressure (when the first of the three rows begins with the letter 'p') at which the cross-sections have been evaluated in the pratmo-code. Some reactions have cross-section data available for a number of temperature and pressure ranges, which Cloud-J interpolates linearly between based on the atmospheric conditions in the EMEP model at the time of the photolysis code call. Note that the pressure-dependent rates are calculated using the standard atmospheric lapse-rate, thereby representing an interpolation in both temperature and pressure.

In the original Cloud-J code, when Cloud-J is configured to run using a reduced number of 12 wavelength bins for tropospheric chemistry (i.e., the EMEP model), stratospheric photolysis reactions are not calculated in order to save CPU time. In this configuration, cross-sections whose first of the three rows begins with the letter 'x' are considered as being relevant to the stratosphere, and are omitted. However, in the EMEP Cloud-M implementation, stratospheric reactions have simply been removed from the Cloud-J input files, as the model is only ever used for tropospheric chemistry. Instead, the new 'e' flag is introduced, to distinguish between photolysis rates that are used in the EmChem family of chemistry schemes and those who aren't (which could be any other chemical mechanism). If for example a CRI-based chemistry scheme is used, all cross-sections and photolysis rates are used. When the 'p' flag is present, as it is for pressure-dependent rates, the reaction is included for all chemistry schemes, note that these reactions are quite rare and in most cases overlap with EmChem anyway. For Emchem19c, the verbose Cloud-J configuration (triggered by `USES%CLOUDJVERBOSE = T` in the EMEP config file) will for example output the following,

```
 >>> Photolysis calculations only for EmChem cross-sections/J-values.
 X-sects    1  O3      O2=O+O            e  3   218.0 258.0 298.0
 X-sects    2  O3(1D)  O3-total          e  3   218.0 258.0 298.0
 X-sects    3  H2COa   Qyld O3=O(1D)+O2  e  2   223.0 298.0
 X-sects    4  H2COb   NO=N+O            e  2   223.0 298.0
 X-sects    5  H2O2                      e  2   200.0 300.0
 X-sects    6  CH3OOH  H2CO=>H+HCO       e  1   298.0
 X-sects    7  NO2     H2CO=>H2+CO       e  2   200.0 300.0
 X-sects    8  NO3     H2O2=>OH+OH       e  2   190.0 298.0
```

```
X-sects    9  N2O5    CH3OOH=>CH3O+OH   e  2   233.0 300.0
X-sects   10  HNO2    NO2=>NO+O         e  1   300.0
X-sects   11  HNO3    NO3=NO2+O/NO+O2   e  2   200.0 300.0
X-sects   12  HNO4    N2O5=>NO2+NO3     e  1   300.0
X-sects   13  PAN     HONO=>OH+NO       e  2   250.0 298.0
X-sects   14  ActAld  HONO2=>OH+NO2     p  3   177.0 566.0 999.0
X-sects   15  MEKeto  HO2NO2=>HO2+NO2   p  2   177.0 999.0
X-sects   16  MGlyxl  ClNO3=Cl+NO3      p  3   177.0 566.0 999.0
X-sects   17  Glyxla  ClNO3=ClO+NO2     p  2   177.0 999.0
X-sects   18  Glyxlb                    p  2   177.0 999.0
X-sects   19  Glyxlc                    p  2   177.0 999.0
X-sects   20  Acet-a                    p  3   177.0 566.0 999.0
X-sects   21  BIACET  Cl2=>Cl+Cl        e  1   298.0
X-sects   22  GLYOX   HOCl=>OH+Cl       e  1   296.0
```

The above print-out shows the cross-sections for the photolysis rates that are included, along with their short-names and long-names, indicator ('e' for EmChem or 'p' for pressure-dependent), in addition to the number of cross-sections to interpolate between in temperature or pressure (2 or 3), and the temperature or pressure at which the cross-sections are calculated for.

The cross-section short-names are the same as those that appear in the `FJX_j2j.dat` file, which is used for the mapping of the cross-sections onto the J-values used for the chemistry-transport model (CTM). The latter is relevant to for example the photolysis of C_2H_5OOH, which uses the photolysis rate of CH_4OOH scaled by a factor of 0.5. The latter factor is included in `FJX_j2j.dat`. In the Cloud-J code, the `JLABEL(JVN_)` array holds the reactions (up to `JVN_` = 201 by default) available for use in the CTM, with the `JMAP(JVN_)` array containing the short-names of the cross-section data that they are mapped to. The first 10 characters of each entry in the `JLABEL(JVN_)` array contain the photolysed species for each reaction, e.g. $CH_3COC_2H_5$ for the reaction $CH_3COC_2H_5 \longrightarrow C_2H_5 + CH_3CO$, which is mapped onto the cross-section data with the name `MEKeto`. The number of photolysis reactions present in the `FJX_j2j.dat` input file is stored in the `NRATJ` variable, thus representing the full count of photolysis reactions that may be used by the CTM.

Once J-values have been calculated in the EMEP model, the photolysis rates corresponding to each of the reaction rates are output in the `VALJXX` array. Since some of the reaction rates are mapped onto other reactions, mapping the `VALJXX` output to the J-values is done as `VALJXX(:,JIND(J))`. Here the first ':' represents the altitude dimension, normally matching the vertical grid of the EMEP model, and `JIND(J)` contains the index in `TITLEJX(X_)` of each of the photolysis reactions `J` in `FJX_j2j.dat`. The scaling factors that map certain photolysis reactions onto others are contained in the `JFACTA(J)` array. Hence, once the index `J_emep` in the `FJX_j2j.dat` input file corresponding to a photolysis reaction in the EMEP model is found, the corresponding J-value can be retrieved from the Cloud-J output as `VALJXX(:,JIND(J_emep))*JFACTA(J_emep)`. Here the `JFACTA(J_emep)` array holds the mapping values from the `FJX_j2j.dat` input file, being for example 0.5 for the photolysis of $CH_3COC_2H_5$ described above.

In the chemistry schemes of the EMEP model, photolysis rates are stored in the `rcphot` array, and applied to the chemistry using a set of photolysis rate indices. All photolysis rate indices are initialized as a negative numbers from EMEP version rv4.51 onward. When the Cloud-J code is initialized (upon the first call to the module), each of the photolysis indices present in the chemistry files is assigned a positive number corresponding to its index in the Cloud-J input files. The EMEP code then loops over all the photolysis indices to verify that each of the rates used in the chemistry scheme were indeed found. If one or more reaction rates are missing, the model stops. Note that this

fail-safe behavior is not present in GenChem, where both the Cloud-J and MCM-based photolysis systems can be used.

4 Defaults in the EMEP model

The default cloud-averaging scheme is the Briegleb scheme, which modifies the in-cloud optical depth by a factor of $f^{(\frac{3}{2})}$. This scheme is comparatively inexpensive computationally wise, and does not degrade surface air quality simulation results over more demanding cloud-averaging schemes. Nevertheless, a choice of seven schemes for averaging over broken over-lapping cloud fields is available, which can be configured using the `CLDFLAG` parameter set in `CloudJ_mod.f90`.

The cross-section data included in EMEP and boxChem is based on the standard data provided along with the Cloud-J v7.3e code up until reaction 73 in `FJX_j2j.dat`, or as described in van Caspel *et al.* (2023) for a select few reactions. The remaining molecular data are taken from the GEOS-Chem Cloud-J input data (see e.g. `http://wiki.seas.harvard.edu/geos-chem/index.php/FAST-JX_v7.0_photolysis_mechanism` and `https://github.com/geoschem/Cloud-J`). The latter concern almost exclusively VOC photolysis reactions, and are for example used in the EMEP implementation of the CRIv2R5 chemical mechanism. Alternatively, cross-section data from the UCI implementation of Cloud-J (or rather, its pre-decessor code Fast-J) is also available on the github page.

5 Quick guide to creating new J-values

The steps below can be followed using the codes and scripts available on Github `https://github.com/metno/emep-mscw/tree/ecosx/box/CloudJ_doc` (for internal MET users only) or GenChem `https://github.com/metno/genchem/tree/master/box/input/cloudj_v7.3e/CloudJ_doc`. The github page(s) contain example scripts for a range of reactions, with the below steps following that of the BIACET photolysis reaction.

- Create quantum-yield and cross-section input data on a 1 nm resolution and input these in the pratmo-code, to calculate the averages over the 18 Cloud-J wavelength bins. For example based on molecular data from literature, or from 1 nm resolution data based on the MCM website (`https://mcm.york.ac.uk/MCM/rates/photolysis`). Each pratmo-code script is unique to the newly added reaction, but can be based on a pre-existing template or reaction rate. For BIACET, the pratmo-code has the name `addX_BIACET.f`. In this file, the subroutine BIACET (lines 180-250) handles the reading in of cross-section and quantum yield data specific to this reaction. A 6-character short-name of the reaction should be specified (line 203). Compiling and running the file then generates the input data for the below Cloud-J input file.

- Add the 18-bin cross-section data to the bottom of the `FJX_spec.dat` file along with the user defined 6-character short-name. The output from the above script should be *exactly* copied here, as the Cloud-J Fortran code is sensitive to line length.

- Define a photolysis reaction in `FJX_j2j.dat` with the first 10 characters defining the photolysed species, while also referring to the 6-character name defined in `FJX_spec.dat` inside the `//` brackets at the end of each line.

- Define a photolysis rate index in `CloudJ_mod.f90` (lines 764 to 827) and assign it using the first 10-characters of the reaction name defined in `FJX_j2j.dat`. The name of the photolysis index should match the name as configured in GenChem.

If the newly created photolysis index exists in the chemical mechanism scheme used to run the EMEP model, it will now be used. If it isn't found, the model will crash.