**Cloud-J write-up, Nov 1, 2023**

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***Variable List.***

w = wavelength (nm or µm)

T = temperature (K)

p = pressure (hPa)

z = altitude (cm)

M = air density (molecules cm-3)

X(w,T) = cross section (cm2 per molecule), the effective cross sectional area for absorbing a photon

q(w,T,p) = quantum yield, the fraction (0 to 1) of absorption events that lead to specific dissociation products

S(w) = mean intensity (photons cm-2 s-1 nm-1), which is the integral over 4π steradians (all directions) of the specific intensity (photons ster-1 cm-2 s-1 nm-1) coming from all directions. S is calculated with a somewhat costly radiative transfer (RT) computation that uses atmospheric distribution of scattering and absorbing gases, aerosols, and clouds plus the incoming solar radiation and the surface reflectance (albedo).

F(w) = top-of-atmosphere solar flux (photons cm-2 s-1 nm-1 on a flat surface normal to the solar beam), normalized to a sun-earth distance of 1 A.U. Seasonality or longer term variations in sun-earth distance is corrected in the J-value code. Changes in F(w) due to solar activity over the solar cycle are irregular and can be imposed externally to the J-value code.

OD(w,z) = optical depth (dimensionless) from top-of-atmosphere solar flux to atmospheric cell (altitude z) where J is wanted. OD is a path integral along the solar beam to the cell and will include the major absorbing and scattering (i.e., extincting) gases, their column densities, and their cross sections (X) including T dependence. Thus, OD(w,z) = ∑ext gases ODgas (Xext(z,T),w,z) over the major extincting gases. For the Earth’s atmosphere and focusing on tropospheric and stratospheric photochemistry, the major extinction is caused by O3 and O2. Additional extinction is provided by Rayleigh scattering from O2 and N2 plus some ultraviolet absorption by H2O. The absorption of specific NO lines below 200 nm by thermospheric NO decreases JNO and this is included parametrically with a correction to XNO. Absorption by NO2 in very highly polluted regions can slightly increase the local heating rate, but has little impact on J’s, and is not included in Cloud-J.

wi = mean wavelength of the ith micro-bin (nm). This is the finest resolution used to integrate over solar flux and cross sections. It is based on the high-resolution solar flux reference spectrum.

Fi = wavelength-integrated solar flux in the ith micro-bin (photons cm-2 s-1). Note that Fi is integrated over the width of the bin wi and hence the per nm in the continuous F(w) is gone.

Xi & qi = cross sections (cm2) and quantum yields (dimensionless) for an absorbing gas or J-value at the micro-bin wi. Usually this value is sampled at wi, rather than averaged over the micro-bin.

W\* = W(I) = a macro wavelength bin consisting of a set of wi micro-bins selected to have similar atmospheric opacities. Macro-bins are identified by an index I, and also possibly by a mean wavelength (nm), although the usefulness of a mean wavelength is marginal when a macro-bin comprises a discontinuous sequence of micro-bins. Macro-bins are the only wavelength bins used in Cloud-J since the micro-bins have already been averaged over to calculate the standard Cloud-J tables for FI, XI, qI.

FI = integrated solar flux in the Ith macro-bin.

XI = solar-flux weighted mean of Xi over the micro-bin wavelengths located in the Ith macro-bin.

qI = solar-flux & X weighted mean of qi over the micro-bin wavelengths located in the Ith macro-bin.

***Calculation of J-values.*** The quantity we want is the photodissociation (a.k.a. photolysis) rate denoted JAB (s-1), or sometimes JAB\_A+B to avoid ambiguity regarding the products.

AB + *hv*  → A + B (1)

This calculation requires integrating over all wavelengths with non-zero X and q.

J(T,p) = ∫ S(w) X(w,t) q(w,T,p) dw (2)

If we neglect scattering, we can simplify the RT calculation by replacing S(w) with the exponential decay of the solar beam F(w) e-OD(w).

J(T,p) = ∫ F(w) exp[–OD(w)] X(w,t) q(w,T,p) dw (3)

Suppose that there are a set of wavelengths W\* that have the same optical depth OD\*, then for the contribution to J from that set W\* we can move the constant exp[-OD(W\*)] outside of the integral

J\*(T,p) = exp[–OD\*] ∫W\* F(w) X(w,t) q(w,T,p) dw (4)

For the set of wavelengths w **∈** W\*, we can pre-calculate total flux G\* in the set of w\* (photons cm-2 s-1) as well as X\* (cm2) and q\* (dimensionless).

G\* = ∫w**∈**W\* F(w) dw (5)

X\*(T) = ∫W\* F(w) X(w,T) dw / ∫W\* F(w) dw (6)

q\*(T,p) = ∫W\* F(w) X(w,t) q(w,T,p) dw / ∫W\* F(w) X(w,T) dw (7)

Thus, we can simply calculate J\*(T,p) (sec-1) using the locally calculated OD\* which is a function of solar zenith angle (time of day) and overhead absorbers. In a scattering atmosphere OD\* will include also the effects of cloud, aerosol, and Rayleigh scattering. For each wavelength grouping W\*, we pre-calculate the values X\* (function of local temperature), q\* (function of both temperature and pressure or density), and G\* (function of sun-earth distance and solar activity). Because all the wavelengths included in W\* are assumed to have the same absorption and scattering throughout the atmosphere, we can do a single RT solution to get S\* and calculate J (equation 2). The J-value is then the sum over all the wavelength bins W\*.

J(T,p) = ΣW\* exp(-OD\*) G\* X\*(T) q\*(T,p) ) = ΣW\* S\* X\*(T) q\*(T,p) (8)

The critical design point in selecting the wavelengths to include in macro-bin W\* is “What counts as nearly the same opacity?” This decision is made based on a criteria for computational efficiency or relative error (i.e., compared to a reference calculation using finer macro-bins or the original source data in micro-bins).

***An Introduction to the Yale-Harvard-GISS-UCI photolysis codes.*** The calculation of photolysis rates requires a monochromatic **radiative transfer (RT) solver** that computes the mean specific intensity (S) throughout the atmosphere so that a **wavelength integrator** can sum the total J-values over the macro-wavelength bins. This paper focuses on the wavelength integrator and only a brief introduction to the RT solvers is given below.

The monochromatic solution of the **radiative transfer** equation in a scattering atmosphere via the Feautrier method (1964) started with Rayleigh scattering in planetary atmospheres (Prather, 1974). This symmetric scattering code was extended to asymmetric scattering and used by Cochran and Trafton (1978), also for planetary atmospheres. The extension to asymmetric scattering was essential for clouds and aerosols and is first documented in Jacob, Gottlieb, and Prather (1989) for calculating photochemistry in a polluted cloudy boundary layer. The first publicly released RT code using this method was documented in **Fast-J** (Wild et al., 2000), extended to the stratosphere **Fast-J2** (Bian and Prather, 2002), and then to fractional cloud fields with **Cloud-J** (Neu et al., 2007; Prather 2015) and to a spherical refracting atmosphere with **Solar-J** (Prather and Hsu, 2019; Hsu and Prather, 2021). The current public version of **Cloud-J** is posted at doi: 10.7280/D1Q398 (May 2023).

The J-value wavelength integrator began with a stratospheric chemistry model designated **Pratmo** (Logan et al., 1978; 1981; see applications in Prather and Remsberg, 1993; PhotoComp, 2010). The Pratmo photolysis code was the reference macro-bin code that Fast-J/J2 were built from. Pratmo has 77 sequentially and continuously ordered macro-bins from 177.5 nm to 850 nm. The first 15 bins roughly match the first 15 Schumann-Runge O2 bands, from SR(14,0) in 177.5-178.3 nm to SR(0,0) in 200.0-202.5 nm. The remaining 62 bins have varying widths from 1 nm near 300 nm where JO3\_O(1D)+O2 has a sharp cut-off in X and q to 100 nm in the visible region where the opacity, mainly from clouds and aerosols, is constant with wavelength. Each of 15 SR bands has very large, 1000:1 variations in XO2, with very narrow absorption lines (see Minschwaner et al., 1992), and thus each macro-bin is split into 3-to-6 mini-bins based on sorting the cross sections in each band (as originally done by Fang, et al., 1974). With the exception of NO photolysis in the δ(1,0) and δ(0,0) bands (see Minschwaner and Siskind, 1993), the cross sections of other species are assumed to be constant across these mini-bins. The mean solar intensity throughout the atmosphere has a separate RT calculation for each mini-bin; JO2 is evaluated for each mini-bin and summed; the mean solar intensity averaged over the macro-bin (SR band) is used with a single X value to calculate the other J’s. In all, there are 83 mini-bins plus the longer wavelength 62 macro-bins, and thus 145 RT calculations are needed in **Pratmo.** The Pratmo wavelength bins were selected at somewhat regular intervals with finer resolution when XO2 or XO3 varied rapidly with wavelength; there no attempt to optimize the number of bins based on opacity. Although included in some earlier versions, the Schumann-Runge continuum absorption of O2 (<175 nm) as well as Lyman-alpha (121.6 nm) are not included in the current Pratmo, and thus it cannot be used for photolysis rates above 70 km.

The original **Fast-J** (Wilde et al., 2000) was designed for tropospheric photochemistry and hence focused on wavelengths from 291 to 850 nm. Because Fast-J was built for the more costly RT computation of multi-stream scattering in cloudy atmospheres, it was desirable to reduce the number of wavelength bins. Using Pratmo as the reference model, the 38 Pratmo macro-bins with wavelengths >291 nm were combined into 7 Fast-J macro-bins. All the Pratmo bins are mapped sequentially onto the Fast-J bins. The need to include stratospheric photolysis led to **Fast-J2** (Bian and Prather, 2002), and here the sorting of the remaining 24 macro-bins plus the 83 SR mini-bins results in non-sequential groupings. For example, from 233 to 291 nm, the only significant source of opacity is O3, and the cross section increases smoothly from about 1x10-18 cm2 at 215 nm to 11x10-18 cm2 at 255 nm and then falls back to 1x10-18 cm2 at 291 nm, see **Table 1**. Thus, the Fast-J2 selected 3 macro-bins that included a continuous wavelength section near the peak XO3 (233-276 nm) and then discontinuous regions on either side of the peak that had similar XO3. Similarly, the SR mini-bins are sorted into groups with similar XO2 and thus Fast-J2 macro-bin W(1) includes sections from SR(14,0) to SR(3,0) while W(5) includes only sections from SR(3,0) to SR(0,0) plus part of the O2 Hertzberg (202.5-206.5 nm). **Fast-J2** was designed and optimized to do J-values only for altitudes 0-64 km in the Earth’s atmosphere. Thus, because many of the SR mini-bins have large XO2 so that negligible solar flux reaches 64 km, Fast-J2 includes only 38 of the 83 mini-bins in its macro-bins.

**Fast-J2** originally calculated atmospheric scattering only in the Fast-J bins (>291 nm), and this required a parameterization for the small effects of Rayleigh scattering in the stratosphere (~6%). This code could not include the photochemical effects of enhanced stratospheric aerosols from either volcanoes or climate intervention. Thus, aerosol and cloud scattering were added for bins 1-11, and the name changed to **Fast-JX** or simply **Fast-J** using all 18 bins since version 5.3 (2005).

**Table 1.** Cloud-J wavelength bins W\* that combine non-neighboring wavelength regions with similar opacities (Bian & Prather, 2002).

|  |  |  |  |
| --- | --- | --- | --- |
| Cloud-J bin# | wavelength range | XO3(260 K) range^ | X\*O3(260 K) |
| 9 | 233.0 – 275.5 nm | 6.5–11.2–6.5 10-18 cm2 | 8.84 10-18 cm2 |
| 10 | 221.5 – 233.0 nm & 275.5 – 286.5 nm | 2.5–4.8 & 4.9–2.5 10-18 cm2 | 3.58 10-18 cm2 |
| 11 | 215.5 – 221.5 nm & 286.5 – 291.0 nm | 1.34–1.81 & 1.35–1.80 10-18 cm2 | 1.55 10-18 cm2 |
| ^ XO3 ranges do not match at the bin edges because they are derived from a reference photolysis code (pratmo) that was binned at a resolution of 2 to 6 nm over the range 200-327 nm. | | | |

The original Fast-J (Wild et al., 2000) had the ability to calculate multi-stream scattering in a single column plane-parallel cloud field. With the ability to map fractional cloud cover into a statistical mixture of single column atmospheres (Neu et al., 2007; Prather, 2015), **Cloud-J** version 7.3 was released in 2015. The Fast-J core was mostly unchanged, but it now averaged over a range of cloudy-clear atmospheres. Thus the tables of X’s and q’s did not change with the move to Cloud-J. Cloud-J made unique contributions to the problem of calculating J-values in a field of overlapping clouds: using different cloud-overlap algorithms, a low-cost method for explicitly calculating all possible atmospheres allowed for the clear definition of quadrature column atmospheres in the same way as micro-bins could be combined into macro-bins. Unfortunately, this increase the cost of averaged J-values by ~ 3x, but it significantly improved the accuracy.

**Solar-J** (Hsu et al., 2017; Prather & Hsu, 2019; Hsu & Prather 2021) was the next major development of the radiative transfer models at UCI. Solar-J is basically Cloud-J extended into the solar infrared wavelengths with additional RT diagnostics on fluxes and flux divergence (a.k.a. heating rates). The basic RT solution readily extends into the IR with the addition of cloud absorption in addition to scattering. The IR extension of Solar-J uses primarily the RRTMG-SW v4.0 code (Mlawer et al., 1997; Clough et al., 2005), but is also coded for some simpler versions from Grant & Grossman (1998) and Chou & Suarez (1999). Solar-J added improvements to Cloud-J in terms of spherical atmospheres, including now refraction and true geometric atmospheres (g is not constant with altitude). Merging with the solar heating codes led to shift the end of the visible photolysis bin edges from 412 to 485 nm and from 850 to 778 nm. This improved atmospheric heating rates in the UV-visible region with little change in J-values. All photolysis rates are still included in the new first 18 bins of Cloud-J and Solar-J version 7.6 and later.

The most recent addition to **Cloud-J** (version 8.0c, Prather, 2023) is the inclusion of UV absorption by gaseous H2O (Pei et al., 2019) made with a code change, and an alternative data tables with and without H2O cross sections. The impact of UV H2O absorption is to reduce JO3\_O(1D)+O2 and other UV-driven J’s by ~10% near the surface.

**Calculation of X’s and q’s** for Cloud-J uses a reference solar spectrum wi from 100 to 2100 nm at 0.05 nm resolution (40,000 points) with the solar irradiance (photons cm-2 s-1) given for each wi. The X’s and q’s for each photolysis rate are interpolated to the wi and summed to get the macro-bin values (equations 5-7). First, we calculate the Pratmo macro-bins, and then we calculate the Cloud-J macro-bins from the Pratmo bins. This double step provides us spectral data tables for the Pratmo since it is still in use. For each macro-bin W\* the sums look like

G\* = Σ w(i) **∈** W\* Fi where Fi = reference solar flux in the ith bin (wi ± 0.025 nm) (9)

X\*(T) = Σ w(i) **∈** W\* Fi X(wi,T) / G\* where X(wi, T) is interpolated to wi (10)

q\*(T,p) = Σ w(i) **∈** W\* Fi X(wi,T) q(wi,T,p) dw / X\*(T) where q(wi,T,p) is interpolated to wi (11)

For most species X and q are combined as that is the effective cross section for photodissociation.

qX\*(T,p) = Σ w(i) **∈** W\* Fi X(wi,T) q(wi,T,p) dw / G\* (12)

The code in the **Add\_Xs** directory provides the necessary reference files and examples of how to calculate a new cross section for a new species or for updated laboratory data. The basic operation is to map the reference micro-bins wi onto the selected macro-bins WI using equations 9-11 above. All the photolysis occurs in the first 18 macro-bins as described above. Macro-bins 19-27 in the IR are used only for heating rates, and some data are also generated to these bins. Thus for each species there is a set of tables with 18 numbers (some zero) with qX values provided for a set of temperature or a set of pressures.

Maintaining the two-step process in integrating q and X over wavelengths is cumbersome, but there is some logic in that the 76 primary UV+visible mini-bins in pratmo are all sequential in wavelength and each of these bins has an unambiguous mean wavelength and the q and X values can be plotted against the original data sources. The 11 UV bins in Fast-J are not sequential and are comprised of out-of-sequence UV pratmo bins. I once tried to skip the intermediate step, but the chance to make errors and complexity of code drove me back to the two-step process.

**Temperature and pressure dependence of photolysis**. Under higher temperatures, there is more internal energy (rotational and vibrational) available to aid photolysis and the cross sections can shift with wavelength. Cloud-J reads in 1 to 3 tables with monotonically increasing T values and interpolates between them. In some cases, the spectral data are only for one T that is used for all temperatures; in other cases we have a wealth of T data and can select up to 3 T values for the qX tables. Because O2 and O3 are so important in terms of the extinction of sunlight as well as their photolytic products, we use 3 tables with a wide range T’s reflecting atmospheric temperatures from 0 to 64 km altitude. Because both of the pathways for O3 photolysis (O(1D) and O(3P)) are important, we calculate qO3\_O(1D)+O2 separately in the tables and then interpolate and apply to the total JO3 in Cloud-J. It is important to select a broad range of T’s (IF there is lab data available) because Cloud-J only interpolates between the qX tables and does not extrapolate beyond the T’s. For many organic species, qX depends on pressure and temperature, under high pressures collisions can remove some of the energy and prevent photolysis even though a photon is absorbed. Cloud-J made a conscious decision to avoid two-dimensional qX tables. Thus for organics with a large pressure dependence on q (e.g., acetone, glyoxyl, methylvinyl ketone), we have provided 3 standard pressures (177, 566, 999 hPa) for the qX tables where the T values for each p are from a standard atmosphere lapse rate. This approach focuses on the troposphere where these organics are important; the stratospheric J-values will use qX values from the 177 hPa table.

The Cloud-J calculation adapts to the spectral data tables and other input flags: the number of qX tables for each J-value can vary from 1 to 3; the choice between T and p interpolation is triggered by a flag in the table; for troposphere only chemistry, the stratospheric species can be dropped from the calculation; and also for troposphere only, the number of macro-bins can be reduced from 18 to 8.

**Primary Data Sources.** For the most part, the cross sections and quantum yields are taken from critical reviews of photochemical data such as the expert committees at JPL (Burkholder et al., 2020) and IUPAC (Atkinson, 2004; 2006; IUPAC, 2023). They can also be taken from individual published papers when when new or alternative data is available. The values of X(w,T) and q(w,T,p) are based primarily on laboratory experiments across a range of wavelengths (w **∈** [w1:wN]) at one or two temperatures (T **∈** [T1, T2]) and pressures (p **∈** [p1, p2]). For each photolysis rate, the format of the recommended data tends to be unique, taken directly from individual laboratory studies. In some cases, only one temperature is reported; in others, cross sections for two or three temperatures are given. Usually a value is provided for near room temperature (273, 298 or 300 K) and some colder temperature. The wavelength values can be finely resolved points (<1 nm), coarsely resolved points (~10 nm), or an average value over a sequence of connected intervals. In some cases a polynomial, or rational polynomial fit to the cross section as a function of w, T, and p is given for the long-wavelength tail.

**Scientific decisions in calculating the qX tables**. The conversion of primary data tables to Cloud-J tables of X, q, and qX requires some scientific choices because the primary data is often incomplete or inconsistent. For example, from IUPAC (2023, Data Sheet PNOx4) the NO2 data includes cross sections reported at 5 nm intervals from 205 nm to 495 nm and for temperatures of 298 K and 220 K. The quantum yield for NO2 + *hv* → NO + O(3P) is measured at high wavelength resolution and reported as a constant (1.00) from 300-398 nm and then tabulated at 1 nm intervals from 399 to 415 nm but for temperatures of 298 K and 248 K. In the case with q < 1, the absorption results in fluorescence but no dissociation. JNO2\_NO+O is a very important rate throughout the troposphere and into the stratosphere, thus we need to calculate it for at least the range 200-300 K. First, do we assume that X(200) ~ X(220) or do we use the ratio X(220)/X(298) to extrapolate to X(200)? Second, we have q(298) which means we can readily calculate qX at 298 K; but with q(248), can we extrapolate to get q(220)? Now we have the wavelength issue. What do we use for X at w<205 and w>495 nm? Fortunately, JNO2 is driven by wavelengths 300-400 nm and so we can zero out qX outside of the 205-495 nm range. It is important to add a zero X value at the long-wavelength tail of published tables because otherwise a small value of X extended into longer wavelengths could be a large mistake.

Overall, one must decide the altitude range at which this J value is important for chemistry and then design the qX tables to meet this need. For example, photolysis of VOCs is complex because they have q values that decrease with increasing p. Cloud-J photolysis sets up 3 wavelength tables of qX to cover altitudes from the surface to tropopause. Throughout the stratosphere the J values just use the tropopause tables (p = 177 hPa). This works out well because VOCs are primarily in the troposphere, and disappear rapidly in the stratosphere with little consequence. Thus using qX evaluated at 177 hPa is a safe solution for stratospheric chemistry of VOCs. One could always extend Cloud-J with 2D interpolation in tables, but that would become cumbersome and costly to maintain. It would also be more difficult to maintain and test.

**Cloud-J references (reverse chronological) plus some cross –section sources**

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