A note on the use of the two-stream delta-scaling approximation for calculating atmospheric photolysis rate coefficients

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Abstract. The delta-scaling approximation is widely used in radiative transfer models to improve the accuracy of calculated irradiances under strongly forward scattering conditions. At completion of the scaled radiative transfer calculations, the inverse transformation (or descaling) is applied to partition the total downward irradiance, which is assumed invariant with respect to the scaling/descaling transformation, into its diffuse and direct components. However, this invariance does not apply to the total downward actinic flux, and ambiguities arise when the actinic flux components are calculated from either scaled or descaled components of the irradiance. The problem is particularly severe in two-stream models because of the lack of angular resolution. We use a realistic example (the calculation of the photodissociation rate coefficient, or J value, for the photolysis reaction \( \text{NO}_2 \rightarrow \text{NO} + \text{O}^\left(3\text{P}\right) \)) to show in particular that, for high Sun and low surface albedo, the use of descaled J values leads to large overestimation of rather constant or modest increases of J values below a thin cloud or aerosol layer. Even larger overestimations of J values are found near the top of a thick cloud layer at high Sun when descaling is used. Use of scaled values appears preferable under most practical conditions, although they, too, suffer from inaccuracies generally associated with two-stream methods.

Introduction

Numerous approximate techniques have been developed to calculate radiation fields in vertically inhomogeneous, multiple scattering atmospheres. Among these are the computationally efficient and reasonably accurate two-stream methods [Paltridge and Platt, 1976; Meador and Weaver, 1980; Toon et al., 1989] which have found wide use in atmospheric modeling to calculate solar energy deposition rates, photolysis rates, and infrared cooling rates.

Models of atmospheric chemistry must include an accurate description of photodissociation processes driven by sunlight. This is done by calculating the photodissociation rate coefficient (J value) of each photoactive molecule:

\[
J = \int \lambda F(\lambda, \tau)\sigma(\lambda)\Phi(\lambda) d\lambda
\]

where \( \lambda \) is wavelength; \( \sigma(\lambda) \) is the absorption cross section of the molecule; \( \Phi(\lambda) \) is the photodissociation quantum yield; and \( F(\lambda, \tau) \) is the spectral actinic flux.

Actinic flux \( F(\lambda, \tau) \) is related but not equal to irradiance [Madronich, 1987]. The actinic flux concerns the probability of an encounter between a photon and a molecule, while the irradiance describes the flow of radiant energy through the atmosphere. Mathematically actinic flux and irradiance can be calculated, respectively, by integrating the radiances or intensity \( I(\tau, \theta, \phi) \) over all angles as follows:

\[
F(\tau) = \int_\theta \int_\phi I(\tau, \theta, \phi) \sin \theta d\theta d\phi
\]

\[
E(\tau) = \int_\theta \int_\phi I(\tau, \theta, \phi) \sin \theta \cos \theta d\theta d\phi
\]

where by simplicity we have suppressed the variable \( \lambda \) in the expressions of actinic flux, irradiance, and radiances.

Numerical difficulties arise when radiative transfer calculations encounter strongly forward-peaked scattering caused by, for example, atmospheric aerosol particles and cloud droplets, as accurate expansion of the phase function may require several hundred terms. To circumvent this difficulty, the so-called scaling transformations, or \( \delta \)-scaling, have been introduced [Shettle and Weinman, 1970; Joseph et al., 1976; Wiscombe, 1977]. It is assumed that the forward scattering peak can be represented by a Dirac \( \delta \)-function, while the remainder
of the phase function is expanded in Legendre polynomials as usual, i.e.,
\[ \tilde{p}(\cos \Theta) = 2f(1-\cos \Theta) + (1-f) \sum_{l=0}^{2N-1} (2l+1)\chi_l P_l(\cos \Theta), \]  
(4)
where \( \tilde{p}(\cos \Theta) \) is the phase function after scaling; \( \Theta \) is the scattering angle, which is a function of \( \theta \) and \( \phi \); \( P_l \) is the \( l \)th Legendre polynomial; \( \chi_l \) is the \( l \)th expansion coefficient; and \( 0 \leq f < 1 \) is the scaling factor which represents the strength of the forward scattering peak. We note that the above phase function is correctly nor-
malized to unity regardless of the value assigned to \( f \).
If \( f = 0 \), we retain the usual Legendre expansion. We usually determine \( f \) by setting \( f = \chi_{2N} \), but this is obviously not a unique choice.

The motivation for scaling is to modify a transfer equation with a strongly peaked phase function into a more tractable problem with a phase function that is much less anisotropic. In general, the more quadrature points (or streams) that are adopted in the radiation scheme, the more accurate is the calculated radiation field. Therefore special attention must be paid when lower stream approximations are used because of their lower angular resolution.

The \( \delta \)-scaling approximation is basically designed for accurately calculating irradiance incident on a certain surface. To complete \( \delta \)-scaling, the inverse transformation (or descaling) is usually applied, to repartition the direct and downwelling diffuse irradiance components. The total downward irradiance (direct + diffuse) is invariant before and after scaling, in accordance with energy conservation. The question is whether we need to do the same inverse transformation for the calculation of actinic flux.

We note that in contrast to the total downward irradiance, the total downward actinic flux is not conserved under the scaling or descaling transformations. The question then arises whether the actinic flux components (diffuse (upward + downward) and direct (downward), which are then summed to obtain the total actinic flux) should be computed from the scaled or descalled components of the irradiance. This is a particularly significant issue when using two-stream models, since these have very low angular resolution.

This paper examines the difference between scaled and descalled actinic flux numerically and physically, and compares the \( J \) values calculated with a two-stream delta-Eddington model [Joseph et al., 1976; Toon et al., 1989] with the results from a more sophisticated multiple-stream discrete ordinate method (DISORT) [Stamnes et al., 1988]. We cast our results directly in terms of the \( J \) value for the reaction
\[ NO_2 + h\nu \rightarrow NO + O(3P) \]  
(5)
which plays a central role in the chemistry of the atmosphere. Results for other atmospheric photoreactions were found to be qualitatively very similar.

The Delta-Scaling Approximation

The essential feature of scaling is to turn the original problem into one in which the scattering phase function appears considerably less anisotropic, the optical depth is reduced, and the single scattering albedo is artificially decreased [Joseph et al., 1976], i.e.,
\[ \tau = (1-\omega f)\tau \]
(6)
and
\[ \tilde{\omega} = \frac{(1-f)\omega}{1-\omega f} \]
(7)
where \( \tilde{\tau} \) and \( \tilde{\omega} \) represent the optical depth and single scattering albedo after scaling, and \( \tau \) and \( \omega \) are those before scaling. In the two-stream delta-Eddington approximation (for Heneyy-Greenstein phase function), we have
\[ f = g^2 \]
(8)
and
\[ \tilde{g} = \frac{g}{1+g} \]
(9)
where \( g \) is the asymmetry factor of the original problem and \( \tilde{g} \) is that after \( \delta \)-scaling.

From a physical point of view, the \( \delta \)-scaling approximation relies on the following premise: Those beams that are scattered through small angles contained within the forward peak are essentially not scattered at all. These beams are in fact added back to the original radiation field, resulting in a scaled optical depth smaller than the original optical depth. Therefore the directly transmitted solar irradiance is greater than in the unscaled (no \( \delta \)-scaling) problem. The scaled asymmetry factor is also less than the original value, since the angular distribution of those beams scattered outside the forward peak is less extreme in its angular dependence. Because of the phase function truncation (equation (4) for \( l > 2N-1 \)), the scaled direct irradiance actually contains some scattered beams of the radiation traveling in very nearly the same direction as the incident beam. Energy conservation is normally assumed, which requires that the total downward irradiance be the same whether scaling is used or not:

\[ E_d(\tilde{\tau}) + \mu_0 F^s e^{-\tilde{\tau}/\mu_0} = E_d(\tau) + \mu_0 F^s e^{-\tau/\mu_0}, \]
(10)
where the subscript \( d \) denotes the diffuse irradiance and superscript – the downward direction; \( \mu_0 \) is the cosine of the solar zenith angle, and \( F^s \) is the extraterrestrial solar flux incident at the top of the atmosphere. Once the scaled diffuse irradiance is calculated, we can always “recover” (descall) the unscaled downward diffuse irradiance by solving for \( E_d^-(\tau) \):
\[ E_d^-(\tau) = E_d^-(\tilde{\tau}) + \mu_0 F^s (e^{-\tilde{\tau}/\mu_0} - e^{-\tau/\mu_0}), \]
(11)
where all the quantities on the right are known. No such correction for the upward irradiance is necessary.
For two-stream approximations, the actinic flux is computed from the irradiance components. For the downward diffuse actinic flux,

\[ F_{d}^- (\tau) = \frac{1}{\tilde{\mu}} F_{d}^- (\tau); \]  

(12)

while the direct actinic flux is

\[ F_{dir}^- (\tau) = F^a e^{-\tau / \mu_0}, \]  

(13)

where \( \tilde{\mu} \) is an effective cosine-like parameter in the range \( 0 \leq \tilde{\mu} \leq 1 \). The angle \( \theta = \cos^{-1} \tilde{\mu} \) can be considered to be the mean inclination of the effective direction of diffuse radiation from the vertical. By using the same \( \tilde{\mu} \) before and after scaling, equation (12) can then be rewritten as

\[ F_{d}^- (\tau) = \tilde{F}_{d}^- (\tau) + \frac{\mu_0}{\tilde{\mu}} \left( e^{-\tau / \mu_0} - e^{-\tau / 2\mu_0} \right). \]  

(14)

As a result, the total downward actinic flux after scaling is

\[ F_{tot}^- (\tau) = \tilde{F}_{d}^- (\tau) + k^a e^{-\tau / \mu_0}. \]  

(15)

So the recovered or descaled total downward actinic flux is

\[ F_{tot}^- (\tau) = \tilde{F}_{d}^- (\tau) + F^a \left( \frac{\mu_0}{\tilde{\mu}} e^{-\tau / \mu_0} + (1 - \frac{\mu_0}{\tilde{\mu}}) e^{-\tau / 2\mu_0} \right). \]  

(16)

Thus the total downward actinic flux after scaling is different from the descaled total downward actinic flux unless \( \tilde{\mu} = \mu_0 \), despite downward irradiance conservation before and after \( \delta \)-scaling. For atmospheric chemistry, the total actinic flux

\[ F_{tot} (\tau) = F_{tot}^- (\tau) + F_{d}^+ (\tau), \]  

(17)

where superscript + denotes upward direction, is of greatest interest for calculating photolysis rate coefficients and is used in the following calculations.

Model Description

For reasons of practical interest to atmospheric chemistry, we show results for \( J \) values rather than the more abstract actinic flux. We have made calculations for different photolysis rates which have shown very similar results. Here we show only the calculations for

\[ NO_2 \rightarrow NO + O(3P) \quad J_{NO_2} \]  

(18)

at two different solar zenith angles and two different surface albedos. Both the two-stream and DISORT models include the effects of absorption by molecular oxygen and ozone, Rayleigh scattering by air molecules, and absorption and scattering by aerosol particles and water cloud droplets. The vertical profile of aerosol attenuation coefficients from Ellerman [1968] is used to represent typical continental tropospheric aerosols. A vertically homogeneous cloud layer is introduced between 3 and 5 km. An asymmetry factor of 0.87 is used for water clouds in the wavelength range (280-420 nm) [Hu and Stamnes, 1993] and a single scattering albedo of 0.9999 is adopted as a first approximation. The vertical profiles of pressure, temperature, and ozone are from the U.S. Standard Atmosphere [1976], and the total ozone column abundance is scaled to 300 Dobson units. Rayleigh, oxygen, and ozone cross sections are from World Meteorological Organization [1986], with the exception of ozone absorption in the Hartley region and the Huggins bands, where high-resolution temperature-dependent data are as given by Molina and Molina [1986]. The extraterrestrial solar flux incident at the top of the atmosphere is adopted from Van Hoven et al. [1988] for 120-350 nm and Neckel and Labs [1984] beyond 350 nm. The cross sections and quantum yield for \( NO_2 \) are from DeMore et al. [1994]. Spectral integration is carried out with 1-nm resolution over 280-420 nm. The effective cosine zenith angle \( \mu = 0.5 \) (as appropriate for the Eddington approximation [Meador and Weaver, 1980; Toon et al., 1989]) is used in the two-stream model to convert diffuse irradiances to diffuse actinic fluxes.

Results

The model DISORT has previously been shown to give good agreement with measurements [Stamnes et al., 1991; Shetter et al., 1992; Zeng et al., 1994]. The eight-stream DISORT is employed, which yields an accuracy in computed \( J \) values better than 0.5% relative to higher streams [Petrova and Volkovskikh, 1995]. Therefore the results from the eight-stream DISORT are considered as a benchmark for comparisons.

We should note that there is a significant difference between the eight-stream DISORT and two-stream model even when the effects of aerosols and clouds are not included so that \( \delta \)-scaling is irrelevant. Therefore we do not consider the absolute differences in \( J \) values calculated from the two-stream model compared with the benchmark, but instead examine the changes in \( J \) values as clouds with various optical depths are introduced, for various solar zenith angles and surface albedos.

Figure 1 shows the relative changes in \( J \) at the ground surface with various cloud optical depths relative to cloud-free sky conditions at two different solar zenith angles (30° and 70°) and two different surface albedos (0.1 and 0.9). Dotted lines are calculated by the two-stream delta-Eddington approximation with scaled actinic fluxes, while the short-dashed lines show the results with actinic fluxes after descaling. These should be compared with results from the eight-stream DISORT method (solid lines). For comparison, we also show \( J \) values calculated from the two-stream model with no \( \delta \)-scaling at all (long-dashed lines), which unequivocally shows that large errors are introduced if no \( \delta \)-scaling is used. Next, we compare scaled and descaled \( J \) values from the two-stream model with the presumably more
**Figure 1.** Changes in $J$ values at the surface, for $NO_2 \rightarrow NO + O(^3P)$, relative to cloud-free conditions, for various cloud optical depths. Solid lines are $J$ values calculated from a discrete-ordinate-method radiative transfer model (benchmark), dotted lines from a two-stream model with scaled actinic fluxes, and short-dashed lines from the same model as "scaled" but with descaled actinic fluxes. The long-dashed lines are the $J$ values calculated from a two-stream model without any $\delta$-scaling at all (see text). SZA represents solar zenith angle, and albedo is the surface albedo. Note the changes of vertical scale between panels.

Accurate results of DISORT, $J$ decreases monotonically with cloud optical depth for low surface albedo (Figure 1a) and has a rather modest increase at high surface albedo (Figure 1b) when an optically thin layer of cloud develops at high Sun, but then decreases with increasing cloud optical depth. Descaled values overestimate $J$ by more than 12% when the cloud is optically thin at high Sun and low surface albedo. Scaled values also overestimate $J$ when the cloud becomes optically thick, but they are relatively closer to the benchmark when the cloud layer is relatively thin at high Sun. At low Sun, the difference between scaled and descaled $J$ values is very small. The major difference between them lies at high Sun when cloud layer is relatively thin (optical depth less than 7-8); the disagreement between the two-stream and DISORT becomes relative constant (about 8%) for larger cloud optical depths. Therefore, from a practical point of view, it appears advantageous to use the scaled actinic fluxes, although for larger optical depths these, too, become inaccurate.

We also calculated the vertical distribution of $J$ for a surface albedo of 0.1 with thin (optical depth of 1) and thick (optical depth of 10) clouds and different solar zenith angles (Figure 2). $J$ is overestimated by descaled values by as much as 18% near the top of a thick cloud layer at high Sun (Figure 2b), while scaled values are in reasonably good agreement (3% or better) with DISORT within and above the cloud layer. The scaled values are consistently more accurate than descaled values under the cloud layer at high Sun. Descaled values become better under the cloud layer when the solar zenith angle is larger than 60° (when $\mu_0 > \mu$), but the difference between scaled and descaled values is small (less than 4% below a thin cloud; Figure 2c). Under the cloud layer, the two-stream method overestimate $J$ by about 2-10% even if scaled actinic flux is used.

**Conclusions**

The $\delta$-scaling increases the direct component of actinic flux and decreases the diffuse component, while descaling converts some parts of the direct light back to diffuse radiation. At high Sun, under an optically thin layer of cloud, descaling makes the diffuse component artificially too large because $\mu_0 \gg \mu$, $\tau < \tau$, and both $\tau$ and $\tau$ are relatively small (see equation (14)). As clouds become optically thick, the solar radiation penetrating through the cloud layer is mostly diffuse, and
the difference between scaled and descaled actinic flux becomes smaller. Therefore when computational time is important and hence a two-stream method is preferred, although descaled irradiances are usually reported for lower stream radiative transfer models, our calculations with two-stream approximations demonstrate that the scaled actinic flux is more reasonable for computing photolysis rates.

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References


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